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Hall-effect and resistivity study of the heavy-fermion system URu_2Si_2

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Hall-effect measurements from 2–300 K and resistivity measurements up to 1200 K are presented for different orientations of URu_2Si_2 single crystals. For **H**||**c** a decomposition into an ordinary and an extraordinary Hall effect allows an estimate of the temperature dependence of the carrier concentration and provides evidence for the existence of a reconstruction of the Fermi surface at 17 K and the onset of coherence near 70 K. A third characteristic temperature, the single-impurity Kondo temperature $T_K \approx 370$ K, is derived from the resistivity data at high temperatures.

A characteristic feature of heavy-fermion systems, in contrast to dilute impurity systems, is the onset of coherence below a certain temperature T_0.[1] While heavy quasiparticles, as determined from the electronic part of the specific heat, are also found in some dilute compounds, the decrease of electron scattering below T_0, as is apparent in electrical transport properties, is unique to correlated, dense systems. Recently, the Hall effect of heavy fermions has attracted considerable interest as a result of its magnitude, which is 100 times that seen in normal metals, and its strong temperature dependence, which shows in a striking manner this onset of coherence. To date, Hall data have been reported for the superconducting heavy-fermion compounds, UPt_3,[2-4] UBe_{13},[5,6] $CeCu_2Si_2$,[7,8] and the antiferromagnetic heavy-fermion compound U_2Zn_{17}.[9] In all cases except Ref. 4 the Hall effect was found to be positive and to increase with decreasing temperature. In UPt_3, UBe_{13}, and U_2Zn_{17} a maximum develops below which the Hall effect decreases if the temperature is further decreased. After a separation of the total Hall effect into an ordinary and an extraordinary part, quantitative evidence was found in UPt_3 that this decrease of the Hall effect is much faster than the decrease of the susceptibility, and therefore has to be related to a decrease of the scattering due to coherence.[2]

URu_2Si_2 is the only heavy-fermion system in which it is claimed that both a magnetic and a superconducting transition, at $T_N = 17$ and $T_C = 0.8$ K, respectively, exist.[10,11] The origin of the 17-K anomaly, however, is debated, and nuclear-magnetic-resonance and specific-heat measurements have been interpreted in terms of formation of a charge- or spin-density wave opening a gap of 11 meV over a portion of the Fermi surface.[12,13] Inelastic neutron scattering results for $T > T_N$ show an inelastic excitation at 6 meV.[14] The susceptibility χ is highly anisotropic,[10] with a Curie-Weiss behavior for $T > 150$ K in the c direction and a temperature-independent and much smaller value for **H**||**a**. For **H**||**c**, χ_c peaks around 60 K and its temperature derivative peaks at 17 K. The resistivity of a polycrystalline sample shows a small peak at 17.2 K and a very broad maximum near 70 K.[12]

To shed more light on the phase transition at 17 K and to study the onset of coherence, we have performed the first Hall-effect measurements on URu_2Si_2. Because we used single crystals we were also able to perform the first investigation of the anisotropy of the Hall effect in a heavy-fermion system. Finally, to derive the parameters of the single-impurity Kondo behavior, the electrical resistivity was measured for temperatures up to 1200 K. The Hall resistivity was determined on three oriented single crystals using the ac van der Pauw method.[15] Four tungsten tips were pressed onto the polished surface close to the edges. Fields up to 100 kOe were produced with a split-coil superconducting magnet and the temperature was varied between 2 and 300 K. For the resistivity measurements above 300 K, pressure contacts were made to each of the four edges of a thin crystal plate cut perpendicular to the c axis.

Figure 1 displays the total Hall effect R_H for **H**||**c** and **H**||**a**. Also shown, for comparison, are the respective dc susceptibilities.[10] For $T > 20$ K we recognize clear similarities, namely, a temperature-independent behavior for **H**||**a**, and an increase of $R_{H,c}$ and χ_c with decreasing temperature down to ≈ 70 K for **H**||**c**. Below 20 K the behavior of R_H and χ differs markedly for both field directions. While the phase transition at 17 K is barely resolved in the susceptibility, it gives rise to a huge peak in the Hall effect for **H**||**c** as well as for **H**||**a**. The similarity of R_H and χ in the paramagnetic phase suggests the empirical ansatz:

$$R_H(T) = R_0 + 4\pi\chi(T)R_s \ , \tag{1}$$

where R_0 and R_s are temperature-independent ordinary and extraordinary Hall coefficients. A separation of R_0 and R_s requires that R_H and χ be temperature dependent. This is only the case for **H**||**c** and we have derived the two Hall coefficients for this case. In a first fit we have plotted $R_{H,c}$ versus the Curie-Weiss susceptibility per formula unit $\chi_{CW} = \mu_{eff}^2/[3k_B(T-\Theta)]$ for $T > 150$ K. The constants $\mu_{eff} = 3.51\mu_B$ and $\Theta = -65$ K are taken from the susceptibility fit of Palstra *et al.*[10] Alternatively we can plot $R_{H,c}(T-\Theta)$ vs $T-\Theta$ and we also obtain a straight

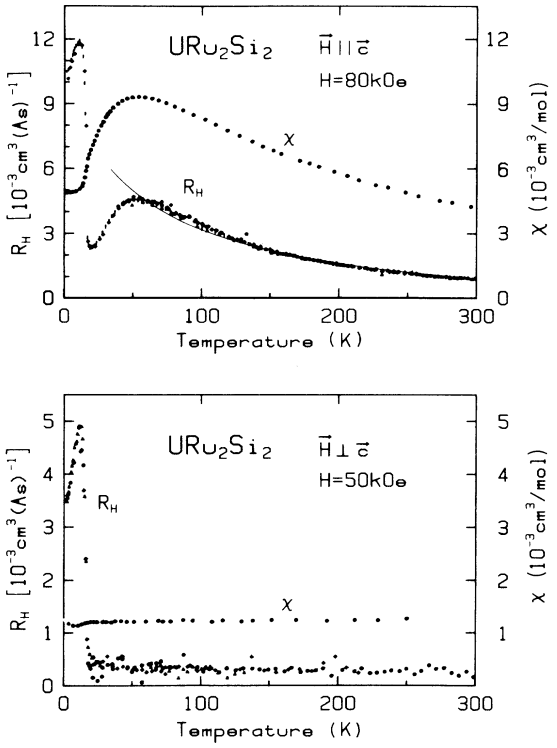

FIG. 1. Temperature dependence of the total Hall effect R_H and the susceptibility (after Ref. 10) of URu_2Si_2. The magnetic field is applied parallel to the c axis (top) and perpendicular to the c axis (bottom). The full line is a fit with Eq. (1).

line, if we limit again the data to temperatures exceeding 150 K. From these fits one derives $R_0 = -(0.97 \pm 0.1) \times 10^{-3}$ cm^3 (As)$^{-1}$ and $R_s = (1.72 \pm 0.08) \times 10^{-3}$ cm^3 (As)$^{-1}$. The full line in Fig. 1 shows the fit of R_H with these parameters. It is interesting to note that both R_0 and R_s have the same signs as in the heavy-fermion system UPt_3 and that they are roughly twice as large.[2] In a one-band model with a spherical Fermi surface the observed ordinary Hall coefficient corresponds to 0.5 electrons per formula unit. Contrary to UPt_3 where band-structure calculations were available and the one-electron-per-formula unit derived from the Hall data could be assigned to a Pt sp band, we cannot compare the data for URu_2Si_2 due to the lack of a band-structure calculation. However, band-structure calculations have been performed for the isomorphous compounds $CeCu_2Si_2$ and $LaCu_2Si_2$ (Ref. 16) and should also become available for URu_2Si_2. It would be very helpful for the understanding of the Fermi surface in the normal state to have such a calculation for the case of f electrons frozen into the core to mimic the case of localized f electrons in the paramagnetic phase.

The assumption of temperature-independent coefficients R_0 and R_s in Eq. (1) has proven useful in many magnetic rare-earth metals[17] and actinides[18] and particularly also in UPt_3 (Ref. 2). Some microscopic theories,[19,20] on the other hand, rather suggest an ansatz:

$$R_H(T) = R_0 + 4\pi\chi(T)\rho_{\text{magn}}(T)r_s \ . \qquad (2)$$

A fit of our data for URu_2Si_2 with Eq. (2) gives a poorer agreement over a smaller temperature interval than with Eq. (1).[21] Also R_0 would become three times smaller for a fit with Eq. (2) than for a fit with Eq. (1) giving, in combination with our resistivity data to be discussed later, an unlikely high Kondo temperature of 960 K. A remarkable feature for both fits is that the measured Hall effect exceeds below about 150 K the value that one extrapolates from the fit parameters at higher temperatures. To visualize this we plot in Fig. 2, $R_s^*(T) = [R_H(T) - R_0]/4\pi\chi(T)$. The relative increase of $R_s^*(T)$ between 150 and 50 K is considerably larger than the increase of the resistivity and this has to be attributed, at least partially, to a change of R_0. Thus we argue that the absolute value $|R_0|$ decreases smoothly from its constant negative value for $T > 150$ K to reach a constant value again near 70–80 K. If we assume that the deviation of $R_s^*(T)$ from a constant is entirely due to R_0, one derives as upper limit of the new conduction-electron concentration of 0.7 e/f.u. Below 20 K, R_0 varies a second time to give a large positive contribution to the shooting up of $R_H(T)$ between 20 and 10 K. Since the extraordinary Hall effect should go to zero for $T \to 0$ we can estimate that the ordinary Hall effect at low temperatures will be equal to the total Hall effect at the lowest temperature, i.e., $R_0(2-10\ \text{K}) \approx 10^{-3}$ cm^3(As)$^{-1}$. Using the simple one-band model this corresponds to 0.05 holes per formula unit.

The physical picture emerging from this data analysis is then the following. For $T > 150$ K, URu_2Si_2 is in the single-impurity Kondo regime with localized f electrons. Starting at 150 K and extending down to 70 or 80 K the f electrons delocalize due to a strong hybridization with conduction electrons. We note that this is also the temperature where χ deviates from the Curie-Weiss behavior.[10] The hybridization goes together with a move-up to near the Fermi energy of those f states with the correct symmetry and leads to an increase of the total conduction-electron concentration. Below 70 K the total and the extraordinary Hall effect decrease faster than the susceptibility indicating the onset of coherence which increases the

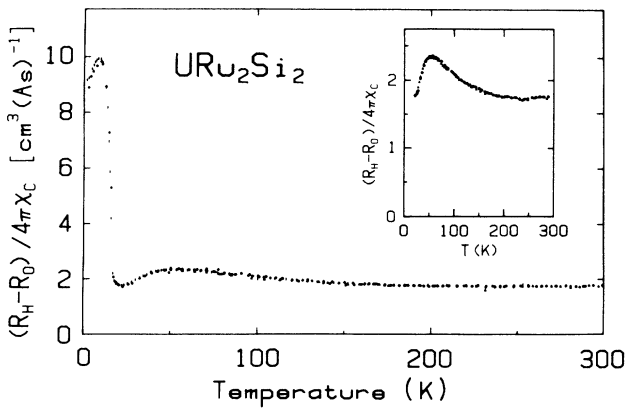

FIG. 2. The ratio of the extraordinary Hall effect (assuming a temperature-independent ordinary Hall effect) to the susceptibility for H parallel to the c axis. The inset shows an enlargement in the neighborhood of the coherence temperature.

mobility of the formed quasiparticles. This regime of increasing coherence is interrupted by the phase transition near 17 K where the Fermi surface is reconstructed. The superstructure is expected to reduce the Fermi surface by a partial gapping in agreement with the very low concentration of 0.05 holes that one may derive in the one-conduction-band picture. We note, however, that this latter value should not be taken too literally. In fact, various electron and hole pockets are expected to exist and the increase of the carrier mobility at low temperatures will make the Hall effect sensitive to details of the Fermi surface contrary to the situation at high temperatures. An estimate of the carrier damping using the expression $\hbar\gamma=\hbar ne^2/(\sigma m^*)$ with at 10 K, $n=6\times10^{20}$ cm^{-3}, $\sigma=3\times10^{16}$ s^{-1}, and $m^*=30$ m gives $\hbar\gamma\approx10^{-4}$ eV, compared to 0.15 eV that we calculate at 300 K with $n=6\times10^{21}$ cm^{-3}, $\sigma=3\times10^{15}$ s^{-1}, and $m^*=2$ m. Below 10 K the reconstruction of the Fermi surface is terminated and the decrease of the Hall effects reflects again the increase of coherence.

To obtain additional information on the single-impurity Kondo regime, we have performed resistivity measurements up to 1200 K. Figure 3 shows the results for the same crystal that has been used for the Hall measurements with H$\|$c, i.e., the resistivity is determined in the isotropic (ab) plane. The resistivity is dominated by a maximum near 70 K, marking the onset of coherence for lower temperatures. Above 250 K the resistivity can be fitted with the expression

$$\rho(T)=\rho_0+c_{ph}T+\rho_m(T)\ , \tag{3}$$

where ρ_0 is the residual resistivity amounting to 22 $\mu\Omega$ cm, and c_{ph} is the coefficient of the electron-phonon contribution equal to ≈0.09 $\mu\Omega$ cm/K. The latter value can be compared to $c_{ph}=0.118$ ($\pm2\%$) $\mu\Omega$ cm/K that we have determined in the reference material $ThRu_2Si_2$. $\rho_m(T)$ is the magnetic contribution obeying the Kondo law:

$$\rho_m(T)=b+c\ln T\ . \tag{4}$$

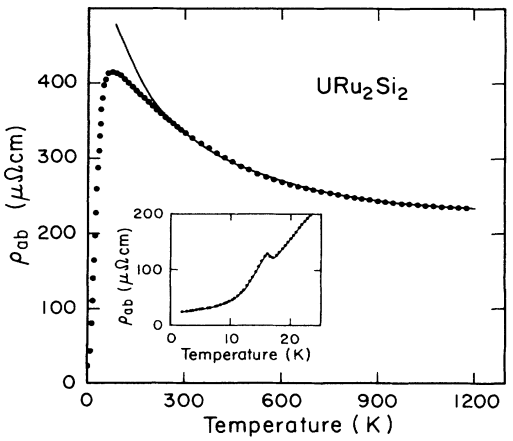

FIG. 3. Temperature dependence of the resistivity in the (ab) plane of URu_2Si_2. The full line is a fit with Eq. (3). The inset shows an enlargement near the transition at 17 K and the determination of ρ_0.

For a degenerate free-ion ground state with total momentum j the slope of the ρ_m vs $\ln T$ plot is[22]

$$c=2\pi m^* v n^2(E_F)J_{sf}^3 4j(j+1)/ze^2\hbar\ , \tag{5}$$

where v is the volume per formula unit, $n(E_F)$ is the density of states at E_F per spin, J_{sf} is the s-f or d-f exchange constant, and z is the conduction carrier concentration per formula unit. The single-impurity Kondo temperature T_K is given by

$$T_K=D\exp\{-[(2J+1)\,|J_{sf}|\,n(E_F)]^{-1}\}\ . \tag{6}$$

Here D is an effective bandwidth which can be related to T_K and the temperature $\tilde{T}$ for which $\rho_m(\tilde{T})=0$ by[22]

$$\tilde{T}T_K=0.77D^2/k_B^2\ . \tag{7}$$

With T in kelvin our fit of the data with Eq. (4) gives $b=1025$ $\mu\Omega$ cm and $c=130$ $\mu\Omega$ cm. Assuming for uranium in URu_2Si_2 a f^3 configuration with $j=\frac{9}{2}$ and an effective mass of the conduction electrons of $m^*=2$ m, we compute from Eq. (5) the product $n^2(E_F)J_{sf}^3/z$. With the relation for an ideal electron gas $n(E_F)=3z/4E_F$, and $z=0.5$ e/f.u., as obtained from the Hall data, we then compute $J_{sf}=-0.15$ eV. The combination of Eqs. (6) and (7) finally allows us to calculate $T_K\approx370$ K. This value is considerably larger than the temperature $T_0\approx70$ K below which coherence sets in, and thus clearly indicates that T_K and T_0 have to be distinguished in URu_2Si_2. We emphasize that the situation is different in $CeCu_2Si_2$, where one obtains from a similar fit to data of Ref. 23, $T_K=10$ K and $T_0=9$ K.

A puzzling question is how these two characteristic temperatures are related to each other. In a study of the Kondo lattice by the functional-integral method Lacroix has derived the following relation:[24]

$$T_0\approx D(T_K/D)^{3/2}\ , \tag{8}$$

giving with our fit parameters for T_K and D a coherence temperature $T_0=210$ K. Thus, the experimentally observed order, $T_0<T_K$ is correctly predicted, but the difference between T_0 and T_K is larger than one derives from Eq. (8). The same Kondo-lattice model also predicts a change of the f density of states. For $T>T_K$, i.e., in the single-impurity regime, the f states form a Kondo resonance with Lorentzian shape at E_F, while for $T<T_0$ a pseudogap is formed. We note, in fact, that our T_K scales well with $\gamma(0)=65$ mJ/mole K^2,[12] while a fit of the same Sommerfeld relation with T_0 requires an approximately five times smaller density of states. Very recently, Doniach[25] has taken into account anisotropy by treating the degenerate Kondo lattice with angle-dependent hybridization matrix elements. Below a critical value of the effective Schrieffer-Wolff coupling constant a spin-density-wave instability is obtained. From the strong anisotropy of χ, ρ, R_H, and the observed striking change of the Hall effect below 17 K, which we have related to a strong reconstruction of the Fermi surface, it appears that the 17-K transition should be interpreted in these terms rather than as transition into an itinerant type of antiferromagnetic order.

In conclusion, we have shown that although URu_2Si_2 is

only a moderately heavy-fermion compound, its transport properties show remarkable features. In particular, the rather large width of the Kondo resonance allows us to distinguish very clearly the single-impurity Kondo temperature and the coherence temperature. Together with the spin-density wave transition at 17 K and the superconducting transition near 1 K, URu_2Si_2 possesses four characteristic temperatures which shall challenge both theorists and experimentalist also in the future.

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