

Hall effect in the heavy-fermion superconductor UPt_3

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The Hall effect of a UPt_3 single crystal has been determined for temperatures from 2–300 K. The total Hall signal is decomposed into the normal and anomalous parts. The anomalous part is very large and positive with a pronounced maximum near 30 K reaching $2.8 \times 10^{-3} \text{ cm}^3 (\text{A s})^{-1}$. The normal part is smaller and negative and corresponds in a one-band model to about one electron per formula unit. The change of sign of the temperature derivative of the anomalous Hall effect is related to the transition into the heavy-fermion state.

Recently, the Hall effect of heavy-fermion and intermediate-valence cerium and uranium systems has attracted considerable interest from both experimental and theoretical workers.^{1–6} Generally, for $T > 10\text{--}20$ K, a remarkably large and positive Hall coefficient is observed. Below this temperature the Hall coefficient often decreases and sometimes changes its sign. In a simple resonant-level Fermi-liquid model for the anomalous Hall constant of mixed-valence and Kondo-lattice systems Coleman, Anderson, and Ramakrishnan⁶ have shown that the sign of the anomalous Hall constant is determined by an interference term between the d and f scattering channels. For Ce^{3+} ions and temperatures exceeding the renormalized width of the resonance level, a positive anomalous Hall constant is predicted in agreement with the experimental data. At low temperatures coherence effects become important and a single-impurity-based description should no longer apply.

UPt_3 is one of the few heavy-fermion systems which become superconducting at low temperatures (0.5 K).^{7,8} Its resistivity in the normal state differs qualitatively from that of other heavy-fermion superconductors, like $CeCu_2Si_2$ (Refs. 1 and 9) and UBe_{13} (Ref. 10), by the absence of a maximum. Instead, the resistivity $\rho(T)$ decreases continuously with decreasing temperature with, however, a net acceleration below ≈ 30 K. The change of sign of $\delta\rho/\delta T$ is generally associated with the onset of coherent scattering and the transition into the heavy-fermion state. Hall-effect measurements have been performed for $CeCu_2Si_2$,^{1,4,5} UBe_{13} ,^{2,3} and $CeCu_6$.² For none of these has a decomposition into the normal and anomalous Hall effects been performed, and consequently, the total Hall effect has been interpreted on a qualitative basis. In this paper we present Hall data obtained on a UPt_3 single crystal which allow, for the first time, a decomposition into the normal and anomalous parts. While the anomalous part is positive for all temperatures with a pronounced maximum near 30 K, the normal Hall effect is found to be negative and to correspond in a one-band model to approximately one electron per formula unit.

UPt_3 crystallizes in the hexagonal Ni_3Sn structure with space group pb_3/mmc . The investigated sample had the dimensions $3 \times 0.25 \times 1.2 \text{ mm}^3$ in the directions of the a , b ,

and c axes, respectively. The resistivity and the Hall effect were measured with the ac van der Pauw method. For the Hall-effect measurement the magnetic field was applied along the b axis, the current flowed along the a axis, and the Hall voltage was determined in the c direction. Because the same spring-loaded tungsten electrodes were also used to measure the resistivity, we have obtained an average value between the resistivity in the a and the c direction. Magnetic fields up to 100 kOe could be produced with a split-coil superconducting magnet.

Figure 1 displays the result of the Hall-effect measurements obtained in an applied field of 40 kOe. The different symbols indicate various runs on the same sample. We observe a positive Hall effect as clearly controlled by a Hall measurement on a pure gold reference. The total Hall effect shows a maximum at 29 K and a steep decrease at low temperatures. For temperatures above ≈ 50 K the temperature dependence of the Hall effect resembles that of the susceptibility along the a axis as can be realized from the susceptibility data¹¹ which are also shown in Fig. 1. To separate the normal R_0 from the anomalous Hall-effect R_s , we make the ansatz¹²

$$\rho_H = R_0 B + 4\pi M R_s .$$

For $T \gg T_C, \Theta$ we take $B = H$, $M = \chi H$, and $\chi = C/(T - \Theta)$, giving

$$R_H = \frac{\rho_H}{H} = R_0 + 4\pi \frac{C}{T - \Theta} R_s .$$

Figure 2 shows a plot of $R_H(T - \Theta)$ vs $T - \Theta$, with $\Theta = -50$ K as derived from the $\chi^{-1}(T)$ curve for $T \leq 300$ K. We obtain a good fit for $50 \text{ K} \leq T \leq 300$ K with a negative slope, indicating a negative sign of R_0 . Thus the ordinary Hall effect is dominated by electron contributions. The fit value $R_0 = -4.0 \times 10^{-4} \text{ cm}^3 (\text{A s})^{-1}$ corresponds in a one-band model to 1.06 electrons per formula unit. This one conduction electron per UPt_3 is very suggestive for the existence, in the normal state, of a rather simply shaped conduction band crossing E_F . Of course, one might argue that this value is the accidental result of a combination of various electron and hole surfaces with different carrier velocities and relaxation times, but as we will show the ob-

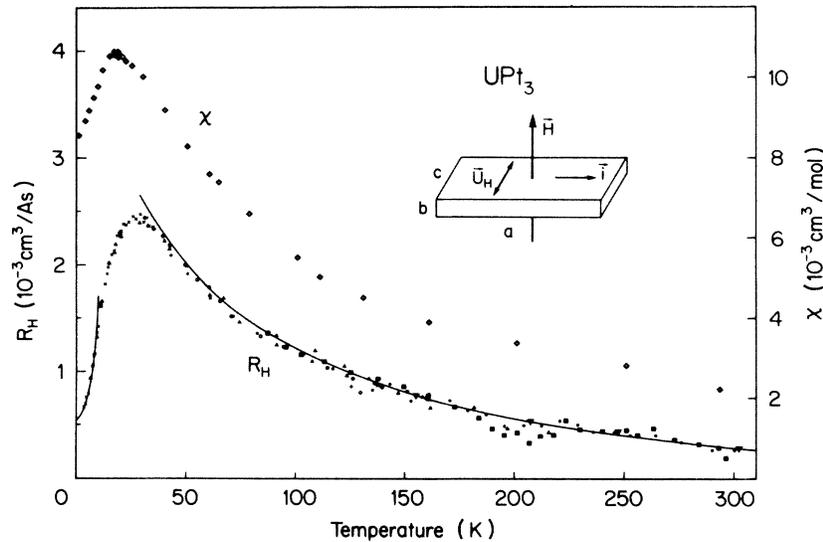


FIG. 1. Total Hall effect of UPt_3 as measured in a field of 40 kOe. The inset shows the arrangement of the electrodes. The full lines are fits (see text). Also shown for comparison are the susceptibility data in the basal plane after Ref. 11.

servation of one electron per formula unit is compatible with the results of a recent band-structure calculation and an estimate of the relaxation time. From optical data¹³ and also from a band-structure calculation¹⁴ we have evidence that the Pt d band is nearly completely filled and therefore we expect little, if at all, hole conduction from these carriers. Besides these 30 Pt $5d$ bands per unit cell located merely between -0.5 and -7 eV, the band-structure calculation of Oguchi and Freeman¹⁴ shows two Pt sp bands near -8 eV. Obviously, these two bands are shifted to low energies due to bonding and do not contribute to the density of states at the Fermi energy E_F . A third Pt sp band, however, which is centered around the K point of the Brillouin zone is within 1 eV of E_F and would cut E_F if we freeze the U f electrons. Each of these bands contains two electrons per unit cell, i.e., one per formula unit (f.u.), and we assign the experimentally determined one $e/f.u.$ to this third conduction band.

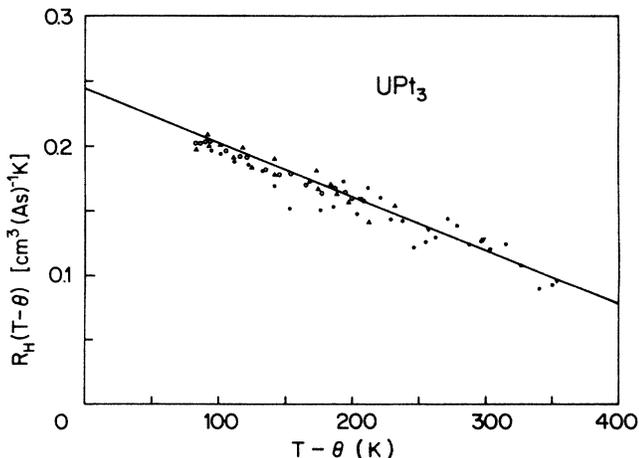


FIG. 2. The decomposition of the total Hall effect of UPt_3 into the normal and anomalous parts for $T > 50$ K.

An interesting question remains to be answered: Why is the Hall effect insensitive to details of the Fermi surface of UPt_3 in contrast to general belief and our knowledge of other materials?¹² To give an answer we compute the relaxation time,

$$\tau_{a,b} = \sigma_{a,b} m^* / ne^2,$$

where n is the free-carrier concentration and m^* is the effective mass. Setting $m^* = m$ and taking the room-temperature conductivity in the a,b plane of $3.75 \times 10^{15} \text{ s}^{-1}$, we obtain $\tau_{a,b} = 10^{-15} \text{ s}$. This is 20 times less than in Cu and corresponds to the remarkably large damping of $\hbar\gamma = 0.63 \text{ eV}$. Thus, the strong scattering of the “free carriers” in UPt_3 allows the Hall effect to measure the carrier concentration independently of any structure in the conduction band on the scale of a few hundreds of a meV.

As we recognize from the anomalous part of the Hall effect and also the susceptibility data, the $5f$ electrons appear at higher temperatures as localized scattering partners of the conduction electrons. The above counting procedure of Pt and U electrons together with the experimental result for the normal Hall effect then provides evidence that uranium is in a $5f^3$ state in this temperature range.

The intersection of the straight line in Fig. 2 with the ordinate gives, for the extraordinary Hall effect $4\pi CR_s$, a value of $0.244 \text{ cm}^3 \text{ K (As)}^{-1}$. With an effective moment of $2.6\mu_B / (\text{U atom})$, we compute $R_s = 0.97 \text{ cm}^3 (\text{As})^{-1}$ which is ≈ 2500 times larger than R_0 and of opposite sign. This positive sign of R_s is in agreement with the theory of Coleman *et al.*⁶ for $T \gg \Delta^*$, where Δ^* is the renormalized width of the $5f$ resonance level. On the other hand, a positive sign of the anomalous Hall coefficient is common in uranium compounds as has been shown in ferromagnetic US and USe and in antiferromagnetic USb.¹⁵ The positive sign in these later compounds has been interpreted in terms of the negative spin polarization of the uranium-derived d conduction electrons which is also supported by the observation of a Kondo effect in UTe, USb, and some magnetically diluted systems.

While the Curie-Weiss ansatz for the temperature depen-

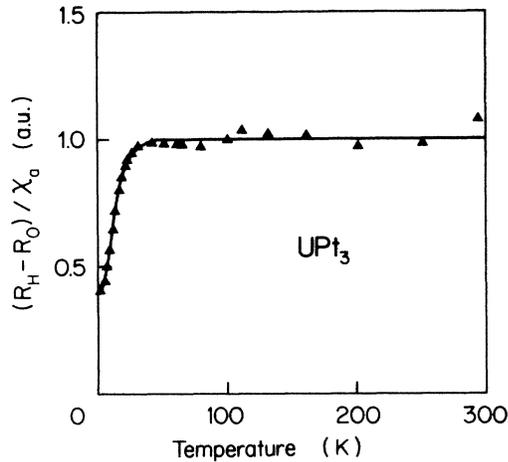


FIG. 3. The ratio of the anomalous Hall effect to the susceptibility in the basal plane of UPt_3 . (a.u. = arb. units.)

dence of R_x describes the experimental data very well above ≈ 50 K (full line in Fig. 1) we also recognize in Fig. 1 that the Hall effect peaks at a temperature 10 K higher than the susceptibility. If we assume that the normal Hall contribution also remains small below 50 K we may separate from the anomalous Hall effect the factor which is proportional to the susceptibility. Figure 3 shows the result of dividing the anomalous part of the Hall effect by the experimental $\chi_a(T)$ values.¹¹ As expected, we obtain a horizontal line where the Curie-Weiss ansatz applies, but below 40 K, R_x decreases noticeably. This enhanced decrease of the anomalous Hall effect, compared with the susceptibility, is viewed as a clear sign of the onset of coherence for the scattering of the conduction d electrons on the f moments.

Figure 4 displays a plot of the Hall data against T^2 . We obtain a good fit up to about 100 K^2 . It is remarkable that this temperature range is nearly one order of magnitude larger than that of the T^2 dependence for the resistivity. According to a theory of Volonshinskii, a quadratic temperature dependence of the anomalous Hall effect results for a mixed spin-orbit interaction if spin disorder is the scattering mechanism.¹⁶ However, the applicability of this model to the Fermi-liquid state is open and more theoretical efforts would be very welcome.

In summary, we have presented the first Hall data on a

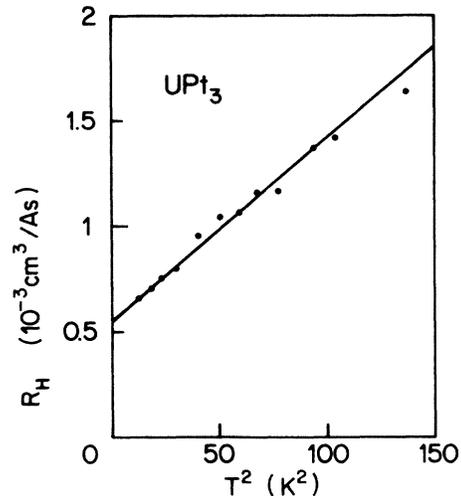


FIG. 4. Low-temperature fit of the total Hall effect of UPt_3 .

heavy-fermion system that could be unambiguously decomposed into the normal and anomalous parts. The latter contribution shows the onset of coherence much clearer than does the resistivity, suggesting that the resistivity is determined by various kinds of carriers which do not all participate in the formation of the coherent state. The separation of the two Hall contributions also allowed an estimate of the "free-carrier" concentration and an assignment of these electrons to a Pt-derived sp band crossing E_F . The size of the anomalous part can then be compared with data for normally behaving rare earths. We find in UPt_3 a value of $\approx 1 \text{ cm}^3 (\text{As})^{-1}$ compared to $-4 \times 10^{-3} \text{ cm}^3 (\text{As})^{-1}$ in terbium, or $4.4 \times 10^{-4} \text{ cm}^3 (\text{As})^{-1}$ in the basal plane of erbium.^{12,17} This huge signal in UPt_3 is, among others, a consequence of the large spin-orbit splitting in uranium which manifests itself in record magneto-optical effects in some ferromagnetic uranium compounds.¹⁸

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