PHYSICAL REVIEW B VOLUME 30, NUMBER 11 1 DECEMBER 1984

Transport properties of uranium monochalcogenide and monopnictide single crystals

J. Schoenes, B. Frick, and O. Vogt

Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule Zürich, CH-8093 Zürich, Switzerland

(Received 14 May 1984)

The temperature dependence of the electrical resistivity and the Hall resistivity in magnetic fields up to 100 kOe is reported for US, USe, UTe, UP, UAs, and USb single crystals. The temperatures of the various first- and second-order magnetic phase transitions are derived as singularities in $\rho(T)$ and $d\rho(T)/dT$, respectively. The magnetic part of the resistivity of the uranium monochalcogenides (UX) is proportional to $T^2\exp(-\Delta/k_BT)$, as expected for electron-magnon scattering. The value of the spin-disorder resistivity of all UX compounds can be interpreted with the same exchange integral. In UTe and USb, a logarithmic decrease of $\rho(T)$, known as the Kondo anomaly, is observed for $T>T_{C,N}$. This behavior is discussed in the framework of existing theories and experiments on intermediate-valence and dense Kondo systems. Thus, the ferromagnetic ordering of UTe at 102 K is a new high T_C for a Kondo material.

INTRODUCTION

The uranium monochalcogenides and monopnictides have attracted, over the last two decades, increasing attention from both experimental and theoretical solid-state physicists. Uranium monochalcogenides (UX, $X={}_{16}$S, ${}_{34}$Se, ${}_{52}$Te) and uranium monopnictides (UZ, $Z={}_{7}$N, ${}_{15}$P, ${}_{33}$As, ${}_{51}$Sb, ${}_{83}$Bi) crystallize with the fcc NaCl-type structure. The UX compounds show ferromagnetic ordering, with T_C decreasing from 177 to 102 K with increasing anion atomic number. The monopnictides order antiferromagnetically with T_N increasing with increasing anion atomic number.[1] The dominant and most puzzling question is the degree of localization of the 5f electrons. Photoelectron spectroscopies, such as x-ray photoemission spectroscopy[2] (XPS) and ultraviolet photoemission spectroscopy[3–5] (UPS), using excitation energies above 40 eV, have been interpreted in terms of a $\approx$2-eV-wide f-d band at ϵ_F for the light UX and UZ compounds. For the heavier compounds, Reihl, Mårtensson, and Vogt,[5] using UPS, term the f state "quasilocalized" in USe, UTe, and USb, while Baer,[2,6] using XPS, reserves a localized character for the f electrons in UTe. Optical[7] and magneto-optical spectroscopies,[8] using energies in the (0.1–5)-eV range, show a rather continuous narrowing of the f state with no final-state splittings. However, the decrease of the magneto-optical response in UTe points to a substantial decrease of the spin-orbit splitting which may reflect a partial quenching of the orbital momentum due to strong f-d hybridization.

Inelastic neutron scattering,[9,10] using still lower excitation energies, finds branches of well-defined magnetic excitations in UTe and USb and a broad featureless frequency response in US and UAs. Specific-heat data below 10 K extrapolate to an electronic $\gamma(0)$ term which decreases nearly linearly as function of lattice constant from $\approx$23.3 mJ/K^{2}mol for US (Ref. 11) to 14.3 mJ/K^{2}mol for USe (Ref. 12) to 10.3 mJ/K^{2}mol for UTe (Ref. 12). In the pnictide series, $\gamma(0)$ peaks at UAs, but the values reported by different authors for USb vary from 0.2 (Ref. 12) to 9 mJ/K^{2}mol (Ref. 13). Thus, in addition to the inherent difficulty in relating high-energy techniques measuring excited states to low-energy techniques probing ground-state properties, there also exist significant differences between results of the same class and even the same type of experiments.

With the aim of shedding more light on the ground-state properties of the uranium chalcogenides and pnictides, we have performed electrical-resistivity measurements as a function of temperature (2–350 K) and magnetic field (0–100 kOe) for US, USe, UTe, UP, UAs, and USb single crystals. Except for US,[14,15] all prior measurements on some of these materials have been performed on powders or sintered polycrystalline samples.[16–21] We also report Hall-effect measurements as functions of temperature and field for some of the samples.

EXPERIMENTAL DETAILS

For the measurement of the electrical resistivity and the Hall effect, the four-point ac van der Pauw method has been used. The samples were [100]-oriented platelets with typical dimensions of $2\times2\times0.5$ mm^3 which were cleaved out of larger crystals and which were sometimes polished. Electrical contacts were made with tungsten tips pressed onto the crystals near the edges. Whenever possible, preference was given to a square arrangement of the contacts to minimize the zero-field signal in the Hall configuration. Magnetic fields up to 100 kOe were generated by a split-coil superconducting magnet, and the sample was rotated by 180° to allow the measurement in the reversed field direction. The data points were either collected by manually zeroing the ac-bridge output or by taking a computer-controlled measurement of the potential drop over the sample. The error on the absolute resistivity values in our geometry is estimated to be less than 10%, which is small compared to the variation from sample to sample due to the Schottky defects introduced by growing single crystals at temperatures exceeding 2000 °C.[22] The accuracy of the temperature measurement is 1 K over the entire temperature range.

RESISTIVITY OF URANIUM MONOCHALCOGENIDES

The temperature dependence of the resistivity ρ in zero field of US, USe, and UTe single crystals is displayed in Fig. 1. At first, the three curves may appear to be quite similar, with a strong increase of the resistivity with increasing temperature for $T < T_C$ and a much weaker temperature dependence above T_C. However, the quantitative analysis which we present reveals very substantial differences.

US. The residual resistivity ρ_0 for the five investigated US samples varied between 19 and 45 $\mu\Omega$ cm, with most samples having a value near 40 $\mu\Omega$ cm. Above 240 K, and up to the limit of our measurements at 350 K, the resistivity increases linearly with temperature. The slope gives $c_{\text{ph}} = 0.092$ $\mu\Omega$ cm/K. To extract the magnetic contribution to the resistivity we must subtract the phonon contribution also at lower temperatures. For this we have extrapolated the phonon contribution to zero temperature. In view of a Debye temperature in the range of 200 K, this procedure may appear questionable. However, resistivity measurements on nonmagnetic ThS powders[21] and ThSb single crystals[23] show a linear temperature dependence of ρ down to ≈ 20 K, indicating important low-energy phonon scattering. After subtraction of the residual resistivity and the electron-phonon—scattering part, the resistivity below ≈ 100 K can be fitted with a T^2 law, i.e., we have

$$\rho = c_m T^2 + c_{\text{ph}} T + \rho_0 \ . \tag{1}$$

The ordering temperature manifests itself by a maximum in $d\rho(T)/dT$ at (177±1) K, in perfect agreement with magnetization measurements.[24]

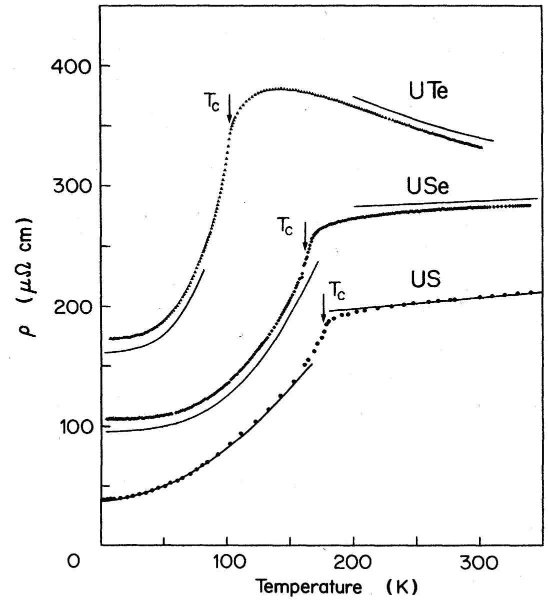

FIG. 1. Temperature dependence of the electrical resistivity ρ of three typical single crystals of US, USe, and UTe in zero field. The arrows indicate the ordering temperature as determined by the temperature of largest $d\rho(T)/dT$. The fits (solid lines) have been shifted by a constant $\Delta\rho$ for clarity.

USe. The residual resistivity of the USe samples was found to be a factor of about 2 larger than that of US, the variation between six investigated samples being of the same order (82—105 $\mu\Omega$ cm). As for US, the resistivity increases linearly with temperature above 240 K. However, the slope c_{ph} is smaller than in US (Table I). This is in contrast with the situation in the respective thorium chalcogenides,[21] where the material with the smaller Debye temperature has the larger c_{ph}, as expected. The anomaly for USe may be due to an additional resistivity term with $d\rho(T)/dT < 0$, as is unambiguously observed in UTe.

Up to about 150 K the resistivity of USe fits the expression

$$\rho = c_m T^2 e^{-\Delta/k_B T} + c_{\text{ph}} T + \rho_0 \ . \tag{2}$$

The Curie temperature is deduced from the maximum in $d\rho(T)/dT$ to be (160±1) K (Fig. 2), which agrees with magnetic data.[1]

UTe. The residual resistivity of UTe is found to be ≈ 170 $\mu\Omega$ cm, which greatly exceeds the values observed for US and USe. Yet, most striking is the appearance of a maximum in the resistivity at 140 K. The decrease of the resistivity of UTe with increasing temperature from 205 K up to the high-temperature limit of our measurements can be fitted with a term

$$\rho_K \propto c \ln(k_B T / E_F) \ . \tag{3}$$

Below the ordering temperature, which we derive from the maximum in $d\rho(T)/dT$ to be 102 K (Fig. 2), $\rho(T)$ is extremely steep. If we try to fit the data with a power law, good agreement is achieved for $T < 90$ K with

$$\rho - \rho_0 \propto T^{3.0} \ . \tag{4}$$

Alternatively, a fit with Eq. (2) gives even a slightly better

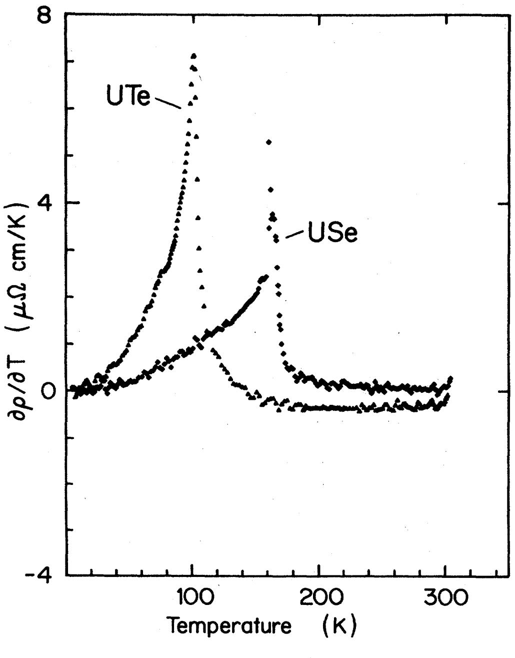

FIG. 2. Temperature derivative of the electrical resistivity of USe and UTe. The maxima give the ordering temperatures.

TABLE I. Results and fit parameters from our transport measurements. For the chalcogenides, ϵ_F is set and the exchange is computed. For the pnictides, the exchange values are set, and ϵ_F and the number of conduction electrons per formula unit are computed. The brackets for $c_{\rm ph}$ of UTe and USb indicate that these values are estimates from the low-temperature fits of the resistivity. (f.u. represents formula unit.)

	N	T_C, T_N	ρ_0	$c_{\rm ph}$	ρ_m	c_m	Δ/k_B	I, $S(S+1)$	I, $J(J+1)(g-1)^2$	I, $\frac{4J(J+1)}{(2J+1)^2}$	ϵ_F	z
	(10^{22} cm^{-3})	(K)	($\mu\Omega$ cm)	($\mu\Omega$ cm/K)	($\mu\Omega$ cm)	($\mu\Omega$ cm/K^2)	(K)	(eV)	(eV)	(eV)	(eV)	e/f.u.
US	2.42	177	35.8	0.092	142	0.0036	0	0.19	0.27	0.37	1	1
USe	2.11	160	105	0.044	163	0.010	135	0.19	0.27	0.37	1	1
UTe	1.71	102	172	(0.07)	$\simeq 200$	0.033	95	0.19	0.27	0.37	1	1
UP	2.29	119, 23	56		$\simeq 200$			0.19	0.27	0.37	0.75	0.65
UAs	2.08	123, 63	$\simeq 7$		$\simeq 270$	3.5 $\mu\Omega$ cm/K		0.19	0.27	0.37	0.61	0.48
USb	1.68	215	90	(0.14)	$\simeq 1040$	0.22	50	0.19	0.27	0.37	0.20	0.09
						5.1×10^{-5} $\mu\Omega$ cm/K^4	0					

agreement if Δ and $c_{\rm ph}$ are taken to be free parameters. If we set $\Delta = 165$ K to correspond to the spin-wave gap found at Γ in UTe,[10] the least-squares error is larger than for the fit using Eq. (4). Figure 1 shows the best fit using Eq. (2), and Table I gives the corresponding fit parameters.

HALL RESISTIVITY OF URANIUM MONOCHALCOGENIDES

The Hall-resistivity measurements were extremely difficult. Upon cycling the field or the temperature, many samples broke and in some cases the electrical contacts became unstable. Thus we did not succeed in measuring a full temperature cycle in both field directions for any one of the five UTe single crystals. Furthermore, quantitative differences were observed for various samples of the same material, and we would like to restrict ourselves here to a semiquantitative discussion of the results. Figure 3 shows the total Hall resistivity ρ_H as obtained for a US single crystal as a function of temperature for fields of 10, 20, 40, and 60 kOe. The Hall resistivity is positive over the entire temperature range, with a maximum close to T_C. For $T \le T_C$, the Hall resistivity is dominated by the extraordinary Hall effect. The low-temperature values of ρ_H depend strongly on the residual resistivity of the very same sample, but for all five investigated samples, ρ_H (4.2 K) decreases for $H > 10$ kOe with increasing field. Assuming the validity of the empirical formula[25]

$$\rho_H = R_0 B + R_s M \, , \tag{5}$$

where R_0 and R_s are the ordinary and extraordinary Hall coefficients, respectively, and $\vec{B} = \vec{H} + 4\pi\vec{M}$, the negative R_0 indicates the dominance of electron conduction. The positive sign of the extraordinary Hall effect, on the other hand, is indicative of the negative spin polarization of the conduction electrons.

Qualitatively similar results are obtained for USe (Fig. 4). With the aim of also analyzing the paramagnetic tem-

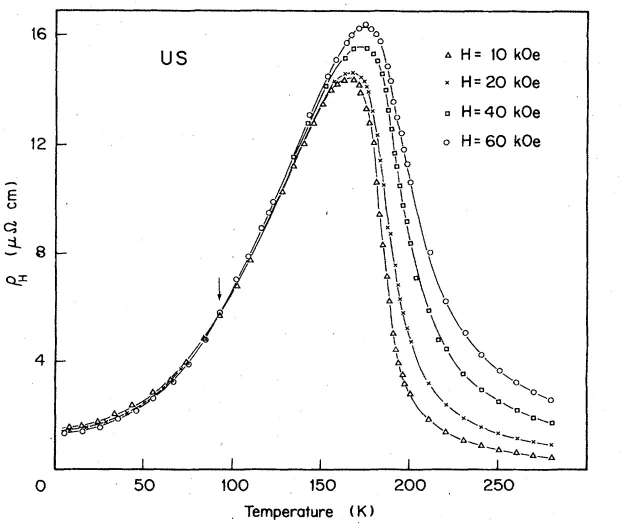

FIG. 3. Total Hall resistivity of a US single crystal as a function of temperature for fields ranging from 10 to 60 kOe.

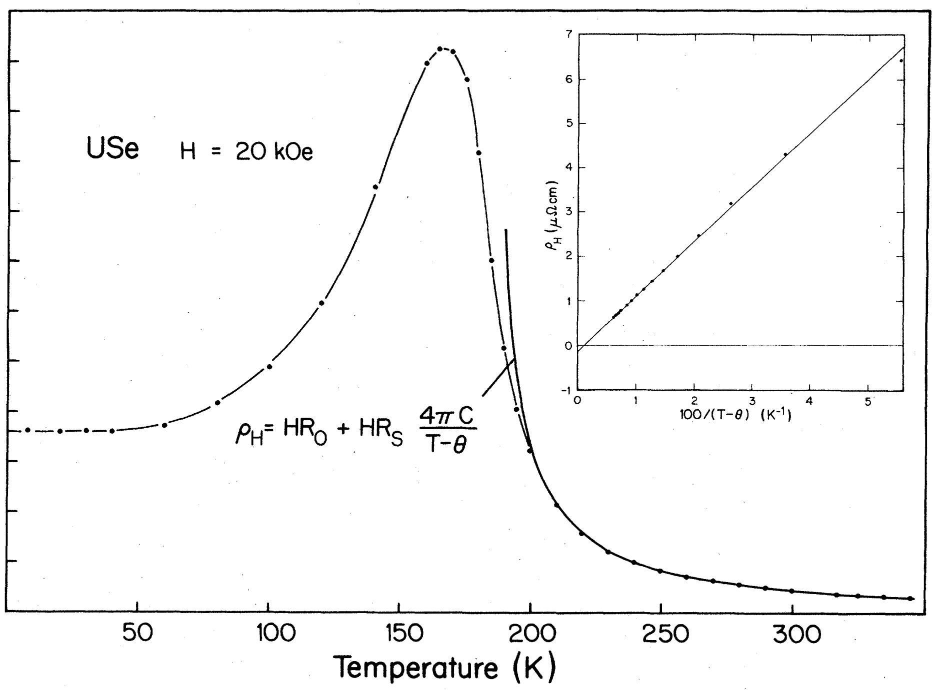

FIG. 4. Total Hall resistivity of a USe single crystal as a function of temperature, and a fit of the data in the paramagnetic temperature range. The inset shows the separation of the total Hall resistivity of USe in its ordinary and its extraordinary parts for $T > T_C$ using Eq. (6). The fit parameters are $\Theta = 182$ K, $HR_0 = -0.12$ $\mu\Omega$ cm, and $HR_s \times 4\pi C = 123$ $\mu\Omega$ cmK.

perature range, these measurements have been extended to the highest temperature which could be attained in our low-temperature cryostat.

For $T \gg T_C$, the magnetization is small compared to the induction, and we may replace, for the ordinary part of the Hall effect, B with H. If the magnetization obeys a Curie-Weiss law, the total Hall effect can be expressed as

$$\rho_H = R_0 H + R_s \frac{4\pi C}{T-\Theta} H , \qquad (6)$$

where Θ is the paramagnetic Curie temperature and C denotes the Curie constant. The inset of Fig. 4 shows a fit of the high-temperature Hall data for USe in which we have chosen Θ so as to give the best fit. The inset of Fig. 4 indicates that the ordinary Hall coefficient is also negative for $T \gg T_C$ although the total Hall effect is positive. This result is in contrast to the existing data in the literature for R_0, which were collected by Grunzweig-Genossar *et al.*[1] and which were all positive. On the other hand, the inset of Fig. 4 clearly shows that a derivation of an exact value for the ordinary Hall coefficient and, with it, possibly a carrier concentration, is imprecise and must await Hall measurements at much higher temperatures. As an indication of the order of magnitude, we mention that in a one-band model the fitted R_0 corresponds to 0.3 *e*/f.u. (where f.u. represents formula unit).

RESISTIVITY OF URANIUM MONOPNICTIDES AND MAGNETIC PHASE DIAGRAMS

The temperature dependence of the electric resistivity of UP and UAs single crystals is displayed in Figs. 5 and 6, respectively. For both compounds the curves are marked by two first-order magnetic phase transitions.[26] In UP the transition from paramagnetism to the single-$\vec{k}$ antiferromagnetic phase is derived to occur at $T_N = (119 \pm 1)$ K. The transition from single to double $\vec{k}$ manifests itself as a steep jump at (23 ± 1) K. Owing to the small temperature interval of the double-$\vec{k}$ phase, a fit

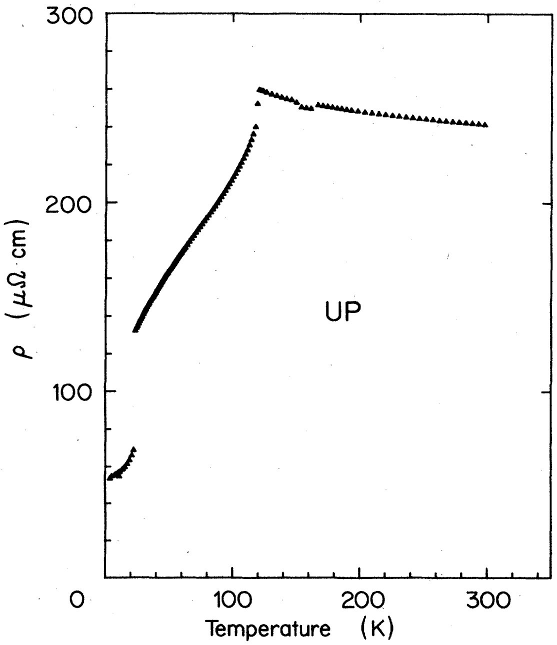

FIG. 5. Temperature dependence of the electrical resistivity of UP in zero field.

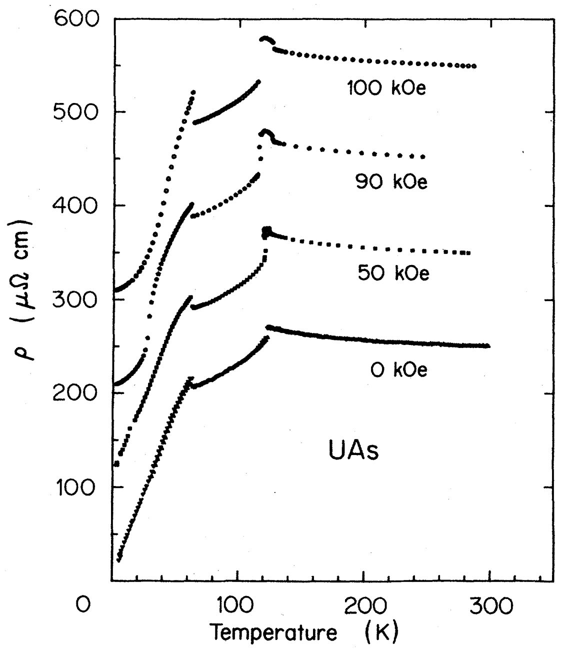

FIG. 6. Temperature dependence of the electrical resistivity of UAs for magnetic fields of 0, 50, 90, and 100 kOe along the [001] direction. Consecutive curves are shifted by 100 $\mu\Omega$ cm for clarity.

of the temperature dependence of ρ relies critically on the exact choice of the residual resistivity. However, the measurements clearly show that the increase of ρ with T is stronger than linear. The application of a magnetic field of 50 kOe depresses the resistivity in the two antiferromagnetic phases by $\approx$5–6 $\mu\Omega$ cm and has no sizeable effect in the paramagnetic temperature range. The zero-field-resistivity curve of UAs (Fig. 6) shows the paramagnetic–to–single-$\vec{k}$ antiferromagnetic phase transition at $T_N = (123 \pm 1)$ K in a sudden drop of the resistivity similar to that in UP. The transition to the double-$\vec{k}$ structure in UAs, however, is marked by a sudden increase in the resistivity at (64 ± 1) K leading to a maximum of the resistivity at (63 ± 1) K. This behavior is reminiscent of the heavy rare-earth metals Dy and Ho,[27] which are known to have spiral spin structures which introduce new boundaries in the Brillouin zone. As pointed out by Rossat-Mignod *et al.*,[26] the difference between the low-temperature phase transition in UP and UAs is that in the former material no change in the wave-vector value occurs, whereas in UAs it changes from 1 to $\frac{1}{2}$. Below 50 K the resistivity of UAs varies linearly with temperature, which again is similar to the behavior found in some rare-earth metals. This behavior has been explained in a theory of Elliott and Wedgwood[28] which takes into account the formation of new Brillouin-zone boundaries and a distortion of the Fermi surface due to the different periodicity of the spin structure and the lattice.

Above 30 kOe a single-$\vec{k}$ ferrimagnetic phase has been shown to occur between the paramagnetic and the single-$\vec{k}$ antiferromagnetic phases.[26] The reduced wave-vector value $k = \frac{2}{3}$ of this new phase again produces an enhancement of the resistivity which extends to an increasing temperature interval with increasing field (Fig. 6). Thus we deduce, for 100 kOe, a width of 12 K for this ferri-

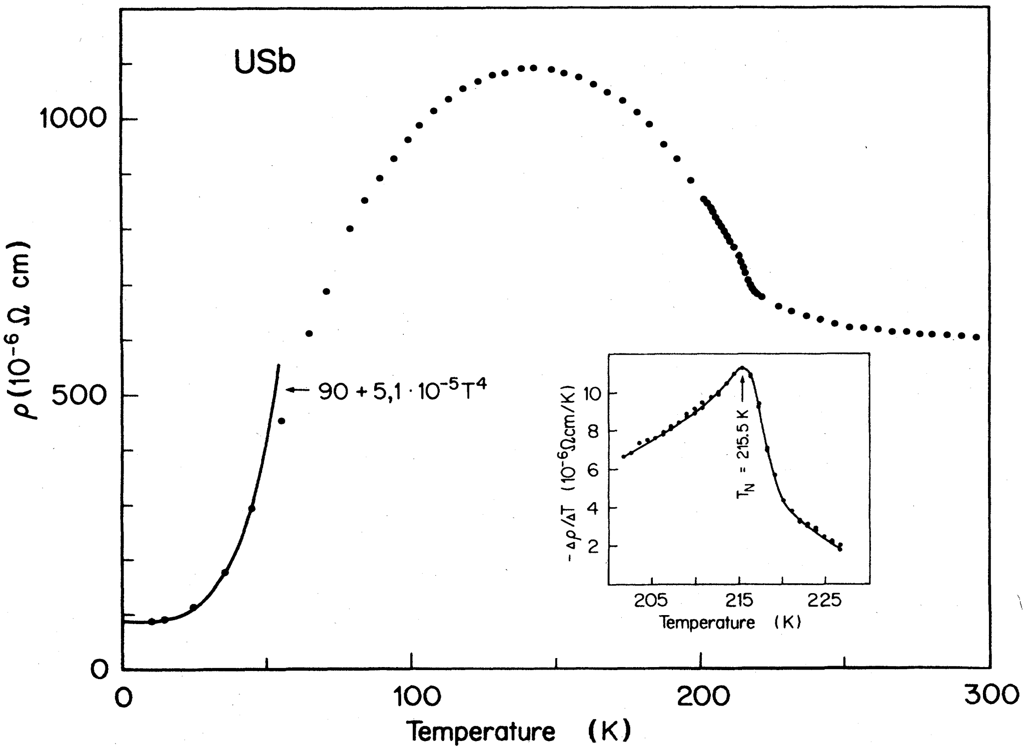

FIG. 7. Temperature dependence of the electrical resistivity of USb in zero field, and a T^4 power-law fit at low temperatures. Note that a $T^2 \exp(-\Delta/T)$ fit is also possible (see text). The inset shows the derivation of T_N from the resistivity data.

magnetic phase, in good agreement with magnetic and neutron data.[26]

The low-temperature phase transition in UAs is only altered in the vicinity of 90 kOe. Hence the resistivity for $T < 50$ K remains unchanged upon application of a field of 50 kOe. At 100 kOe the double-$\vec{k}$ antiferromagnetic phase is substituted for by a double-$\vec{k}$ ferrimagnetic phase with $k = 0.66$. This manifests itself both in a lower resistivity at low temperatures and in a larger peak immediately at the phase transition.

The temperature dependence of the resistivity of USb (Fig. 7) is markedly different from that of UP and UAs. It is dominated by a very large, broad maximum centered at 140 K. The value of more than 1000 $\mu\Omega$ cm in this maximum is typical for a semimetal. As shown in the inset of Fig. 7, the Néel temperature appears as a maximum of $-\Delta\rho/\Delta T$ at (215.5 ± 1) K. The low-temperature part of the resistivity follows a T^4 power law up to 45 K, as indicated by the fit in Fig. 7. However, as for UTe, a fit with Eq. (2) is also possible and a smaller least-squares error can be obtained due to the larger number of free parameters. Table I gives these parameters for both fits. The residual resistivity of the six investigated samples varied between 90 and 160 $\mu\Omega$ cm. The increase of the resistivity below T_N by more than 400 $\mu\Omega$ cm and the following decrease by about 1000 $\mu\Omega$ cm down to 4 K is quite unique and may have its origin in the semimetal-like Fermi surface, which is extremely sensitive to the existing spin order. Resistivity measurements in fields up to 100 kOe showed only very minor changes compared to the zero-field measurement, which is explained by the fact that the magnetization also does not change substantially.

HALL RESISTIVITY OF USb

The Hall-resistivity measurements (Fig. 8) show an interesting temperature dependence. With the temperature decreasing from 300 K, we observe first an increase of the positive Hall resistivity down to ≈ 210 K, and then the Hall resistivity decreases and becomes negative below ≈ 110 K. Because the Hall resistivity ρ_H was found to increase linearly with magnetic field over the entire available field range, a breakdown into an ordinary and an extraordinary part is not possible in the ordered phase. If we compare the results at lowest temperature for USb with that for $U_{0.9}Th_{0.1}Sb$,[23] in which the spins can be aligned with our fields, one finds $R_H(\mathrm{USb})/R_0(\mathrm{U_{0.9}Th_{0.1}Sb}) \approx 5$. In a one-band model the ordinary Hall coefficient R_0 gives 0.1 e/f.u. for $U_{0.9}Th_{0.1}Sb$, which can be interpreted as resulting from the 0.1 tetravalent Th ions per cation. Considering this proportionality and the fact that the application of the field in this hard antiferromagnet will induce practically no magnetization at 4 K, we set, as a first approximation, $R_H(\mathrm{USb}) = R_0(\mathrm{USb})$, giving 0.02 e/f.u. for USb. This low value gains support from the specific-heat data. For LaSb, $\gamma(0)$ amounts to 0.8 mJ/K^2 mol and the free-electron concentration is 0.12 e/f.u. (Ref. 29). Assuming the same effective mass for USb, we compute, for the d conduction electrons in the latter material, $\gamma(0) = 0.4$ mJ/K^2 mol. This is not too far from the value of (0.2 ± 0.1) mJ/K^2 mol found by Rudigier *et al.*,[12] and it indicates that, at least for very low temperatures, the $5f$ electrons do not contribute to the γ term.

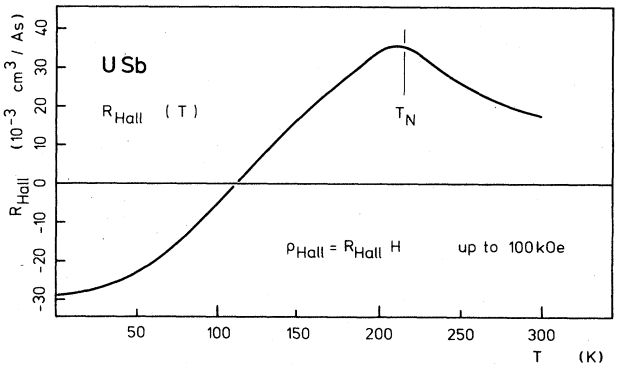

FIG. 8. Temperature dependence of the total Hall coefficient of USb. Up to 100 kOe, only minor changes occur close to T_N.

DISCUSSION OF THE RESISTIVITY

The temperature dependence of the resistivity of magnetic systems has received wide theoretical attention, and many different power and exponential laws have been predicted.[30] For electron scattering on spin waves possessing a quadratic dispersion and an energy gap Δ, it has been found that [31,32]

$$\rho_{\mathrm{magnon}} = c_m T^2 e^{-\Delta/k_B T} . \tag{7}$$

Adding a linear phonon contribution and a residual resistivity term, we can fit the resistivity curves of all uranium monochalcogenides over a remarkably large temperature range. In US, $\Delta = 0$, which we relate[33] to the absence of sharp magnon branches in inelastic-neutron-scattering experiments.[10] For USe, no inelastic neutron experiments have been published so far, but for UTe a sharp magnon branch rising quadratically from its $q = 0$ value of 165 K has been reported. The rather poor agreement with our best-fit value of 95 K should not be overinterpreted due to the variation of Δ with the assumed c_{ph}. In addition, the postulated temperature interval for the fit is large, and therefore the higher-temperature spin fluctuations will tend to suppress the gap in the magnon dispersion curve which has been determined at 4 K.

For a linear magnon dispersion, as is predicted in antiferromagnets, and as, in fact, has been observed in USb,[9] electron-magnon scattering is expected to contribute a term

$$\rho_{\mathrm{magnon}} \propto T^4 . \tag{8}$$

Our resistivity measurements show such a proportionality up to 45 K, but as in the ferromagnetic case [Eq. (7)], a factor $e^{-\Delta/k_B T}$ should be included in Eq. (8) if the magnon dispersion exhibits an energy gap. Because in the vicinity of $q = 0$ the magnon energy of an antiferromagnet possessing a gap is proportional to q^2 rather than q, one may also expect a contribution in the form of Eq. (7).

The total magnetic resistivity can be expressed as[34,35]

$$\rho_m = \frac{3\pi}{2}\,\frac{m^* I_{fd}^2 S(S+1)}{e^2\hbar\epsilon_F N}\,, \tag{9}$$

where I_{fd} is the f-d exchange integral, N is the number of formula units per unit volume, and S is the spin of the $5f$ state. de Gennes[36] has replaced the term $S(S+1)$ by $(g-1)^2J(J+1)$ to include possible orbital contributions to the magnetic moment. Later, Coqblin and Schrieffer,[37] stating that the exchange Hamiltonian $H=-2\Gamma(g-1)\vec{s}\cdot\vec{J}$ leading to the above term in ρ_m would not give rise to a Kondo effect in compounds and dilute alloys containing Ce^{3+}, derived the factor $4J(J+1)/(2J+1)^2$. Because the same argument of a negative $g-1$ as for the $4f^1(^2F_{5/2})$ ground state of Ce^{3+} also holds for the f^3 configuration of U^{3+}, which in Russel-Saunder coupling has $J=\frac{9}{2}$ and $g=\frac{8}{11}$, all three expressions are used to evaluate I_{fd} from the resistivity measurements. Experimentally, ρ_m has been determined for US and USe by extrapolating the high-temperature resistivity linearly to $T=0$ and subtracting the residual resistivity. For UTe, UP, UAs, and USb, the determination of ρ_m is less straightforward due to the negative $d\rho(T)/dT$ over the entire paramagnetic temperature range of our measurements. As an order of magnitude we have included in Table I for the latter materials, values which are obtained by subtracting, from the maximum resistivity, the residual resistivity and an estimated phonon contribution. The amazing result is that the so-determined ρ_m of the three uranium monochalcogenides can be accounted for by the same exchange parameter I_{fd}. Setting $\epsilon_F=1$ eV and $m^*=m$, the exchange integral I_{fd} takes the values 0.19, 0.27, and 0.37 eV for the Kondo, de Gennes, and Coqblin expressions, respectively. The first value agrees perfectly with the 0.2 eV which we derived previously for the Kondo system $\mathrm{U}_{0.14}\mathrm{Th}_{0.86}\mathrm{Sb}$ using the same Kondo expression and the same parameters m^* and ϵ_F.[23] However, recent magneto-optical measurements on $\mathrm{UAs}_x\mathrm{Se}_{1-x}$ (Ref. 38) and $\mathrm{USb}_{0.9}\mathrm{Te}_{0.1}$ (Ref. 39) have given evidence for an f moment which is dominated by an orbital part, and we favor an interpretation using the total angular momentum. In Ref. 39 the largest exchange-induced red shift of the $f\rightarrow d$ transition in any uranium compound is reported to be (0.23 ± 0.03) eV. This red shift ΔE is related to the f-d exchange by $2\Delta E=2I_{fd}\vec{s}\cdot\vec{S}$, giving, for I_{fd}, a value of 0.3 eV, in reasonable agreement with our estimates from ρ_m.

For the uranium monopnictides, ρ_m is substantially larger than for the corresponding uranium chalcogenides and increases strongly with the anion mass (Table I). Because the free-electron concentration is certainly smaller in the pnictides than in the chalcogenides, this trend suggests an interpretation for the pnictides in which ϵ_F is the free parameter, while I_{fd} is taken to be constant within the pnictide series and the same values as for the chalcogenides are assumed. With this choice, ϵ_F decreases from 1 eV in the chalcogenides to 0.75 eV in UP, 0.6 eV in UAs, and 0.2 eV in USb. If we scale the latter value with the Fermi energy of 1 eV and one conduction electron per molecule for UTe using the expression $n\propto\epsilon_F^{3/2}$ for parabolic bands, we derive $n(\mathrm{USb})=0.09$ e/f.u. This is certainly an upper limit considering that estimates for m^* are closer to $2m$ than to m.

As indicated by the fit in Fig. 1, the decrease of the resistivity of UTe for $T\gg T_C$ is logarithmic, as is typical for Kondo systems.[35] To ascertain this temperature behavior we have undertaken efforts to extend the resistivity measurements to higher temperatures. A first measurement up to 750 K on UTe shows a continuation of the logarithmic temperature dependence of the resistivity up to 750 K. Thus UTe exhibits the typical resistivity features of dense Kondo systems such as CeB_6,[40] CeAl_2 and CeAl_3,[41] and intermetallic intermediate-valence materials such as CePd_3.[42] This relationship is further sustained by the very recent observation of a negative elastic shear constant c_{12} (Refs. 10 and 43) as has also been found in CeB_6 (Ref. 40) and TmSe (Ref. 44). The remarkable difference from the cerium compounds is the ferromagnetic order of UTe. Theories for the electric resistivity of Kondo lattices have mostly concentrated on the explanation of a possible antiferromagnetic ordering, and the possibility of ferromagnetic order is sometimes only briefly mentioned. In a model in which the Kondo interaction is transformed into a fictitious "s-f" hybridization and the transport properties are calculated after the introduction of thermal fluctuations of the hybridization, the type of magnetic order is found to depend on the number of conduction electrons per formula unit, n.[45] Ferromagnetic order is predicted for small n, while for n close to 1, antiferromagnetic order is obtained. The temperature of the resistivity maximum is related to the Kondo temperature by $T_{\max}\propto(1-n)^{1/3}T_K$.

A quite convincing comparison with theory can be made for our resistivity data on USb. In a theory of the Anderson lattice, Coqblin *et al.*[46] calculate the electric resistivity as a function of the ratio of the temperature and the half-width of the conduction band W for the total number of electrons $(f+d)$ N ranging from 1 to 1.05. With a set of parameters for the Coulomb correlation energy, the hybridization width and the f binding energy intended to describe intermediate-valence TmSe, these authors find, for $N>1$, an antiferromagnetic and semimetallic ground state and a temperature dependence of ρ which is in qualitative agreement with our results for USb. Comparing the parameters used with those observed in USb, we find the correct order of magnitude for (i) $T_N/T_{\max}\approx2$, (ii) $k_BT_{\max}/W\approx0.008$, giving, for the hybridizing d-band half-width, $W=1.3$ eV, and (iii) $U=0.5W$. Thus USb appears to be a good example for the description of a dense intermediate-valence system within the Anderson-lattice Hamiltonian.[46] In USb the total number of f and d electrons (taking, for the f^3 configuration, $n_f=1$ and, for the f^2 configuration, $n_f=0$) is very close to 1, and the hybridization of the narrow f-like band with d and p states produces a pseudogap. In the antiferromagnetic phase the Fermi level lies within this pseudogap, leading to the observed strong increase of the resistivity below T_N and to the low $\gamma(0)$ value.

UTe, on the other hand, orders ferromagnetically, so that a possible gap can only be located at ϵ_F for one spin direction, excluding semiconducting or semimetallic behavior. From the available data we then prefer to classify UTe as a dense Kondo system. Experiments on magnetically dilute materials are in preparation to investigate the dependence of the magnetic ordering and the Kondo temperature on the exchange and the bandwidth.

ACKNOWLEDGMENTS

We are indebted to P. Wachter for his support of this work. The collaboration of J. W. Allen and G. Travaglini in the early stage of these investigations and the technical assistance of P. Dekumbis and K. Mattenberger are gratefully acknowledged. This work was financially supported by the Swiss National Science Foundation and the Swiss National Energy Foundation.

---

[1] J. Grunzweig-Genossar, M. Kuznietz, and F. Friedman, Phys. Rev. **173**, 562 (1968).

[2] Y. Baer, Physica **102B**, 104 (1980).

[3] M. Erbudak and J. Keller, Z. Phys. B **32**, 281 (1979).

[4] R. Baptist, M. Belakhovsky, M. S. S. Brooks, R. Pinchaux, Y. Baer, and O. Vogt, Physica **102B**, 63 (1980).

[5] B. Reihl, N. Mårtensson, and O. Vogt, J. Appl. Phys. **53**, 2008 (1982).

[6] Y. Baer, in *Handbook of the Physics and Chemistry of the Actinides,* edited by A. J. Freeman and G. Lander (North-Holland, Amsterdam, 1984).

[7] J. Schoenes, Phys. Rep. **66**, 187 (1980).

[8] W. Reim, J. Schoenes, and O. Vogt, J. Appl. Phys. **55**, 1853 (1984).

[9] G. H. Lander and W. G. Stirling, Phys. Rev. B **21**, 436 (1980).

[10] T. M. Holden, W. J. L. Buyers, E. C. Svensson, J. A. Jackman, A. F. Murray, O. Vogt, and P. de V. DuPlessis, J. Appl. Phys. **53**, 1967 (1982).

[11] E. F. Westrum, Jr., R. R. Walters, H. E. Flotow, and D. W. Osborne, J. Chem. Phys. **48**, 155 (1968).

[12] H. Rudigier, Ch. Fierz, H. R. Ott, and O. Vogt, Solid State Commun. **47**, 803 (1983) and H. Rudigier, Ph.D. thesis, Eidgenössiche Technische Hochschule, Zürich, 1984.

[13] R. Troć and H. Mädge, Treizièmes Journées des Actinides, Elat, Israel, 1983 (unpublished).

[14] J. Schoenes, G. Travaglini, O. Vogt, and P. Wachter, Physica **102B**, 328 (1980).

[15] R. Troć and L. Krawczyk, J. Less-Common Met. **90**, L45 (1983).

[16] I. H. Warren and C. E. Price, Can. Metall. Q. **3**, 183 (1964).

[17] J. Danan, B. Griveau, J. P. Marcon, J. P. Gatesoupe, C. H. de Novion, in *Rare Earths and Actinides* (IOP, London, 1971), p. 176.

[18] S. Nasu, T. Kurasawa, H. Matsui, T. Tamaki, and M. Okuda, in *Plutonium and Other Actinides,* edited by H. Blank and R. Lindner (North-Holland, Amsterdam, 1976).

[19] R. Troć and Z. Kletowski, Bull. Acad. Pol. Sci. Ser. Sci. Chim. **22**, 621 (1974).

[20] H. Matsui, S. Nakashima, K. Katori, M. Tamaki, and T. Kirihara, J. Nucl. Mater. **110**, 282 (1982).

[21] M. Haessler and C. H. de Novion, J. Phys. C **10**, 589 (1977).

[22] E. Kaldis and B. Fritzler, Prog. Solid State Chem. **14**, 95 (1982).

[23] B. Frick, J. Schoenes, O. Vogt, and J. Allen, Solid State Commun. **42**, 331 (1982).

[24] D. L. Tillwick and P. de V. DuPlessis, J. Magn. Magn. Mater. **3**, 319 (1976).

[25] C. M. Hurd, *The Hall Effect in Metals and Alloys* (Plenum, New York, 1972).

[26] J. Rossat-Mignod, P. Burlet, S. Quézel, O. Vogt, and H. Bartholin, in *Proceedings of the IVth Conference on Crystalline Electric Field Effects in f-electron Magnetism,* edited by R. P. Guertin, W. Suski, and F. Zołnierek (Plenum, New York, 1982), p. 501.

[27] R. V. Colvin, S. Legvold, and F. H. Spedding, Phys. Rev. **120**, 741 (1960).

[28] R. J. Elliott and F. A. Wedgwood, Proc. Phys. Soc. (London) **81**, 846 (1963).

[29] F. Hulliger, in *Handbook on the Physics and Chemistry of Rare Earths,* edited by K. A. Schneider, Jr. and L. Eyring (North-Holland, Amsterdam, 1979).

[30] See, for example, S. V. Vonsovskii, *Magnetism* (Wiley, New York, 1974).

[31] K. Niira, Phys. Rev. **117**, 129 (1960).

[32] A. R. Mackintosh, Phys. Lett. **4**, 140 (1963).

[33] J. Schoenes and O. Vogt, Douzièmes Journées des Actinides, Orsay, 1982 (unpublished), p. 46.

[34] T. Kasuya, Prog. Theor. Phys. **16**, 45 (1956).

[35] J. Kondo, Prog. Theor. Phys. **32**, 37 (1964).

[36] P. G. de Gennes, J. Phys. Radium **23**, 510 (1962).

[37] B. Coqblin and J. R. Schrieffer, Phys. Rev. **185**, 847 (1969).

[38] W. Reim, J. Schoenes, and O. Vogt, Phys. Rev. B **29**, 3252 (1984).

[39] W. Reim, J. Schoenes, and O. Vogt, Helv. Phys. Acta **57**, 226 (1984).

[40] T. Kasuya, K. Takegahara, Y. Aoki, T. Suzuki, S. Kunii, M. Sera, N. Sato, T. Fujita, T. Goto, A. Tamaki, and T. Komatsubara, in *Valence Instabilities,* edited by P. Wachter and H. Boppart (North-Holland, Amsterdam, 1982), p. 359.

[41] K. H. J. Buschow and H. J. van Daal, Solid State Commun. **8**, 363 (1970).

[42] R. D. Hutchens, V. U. S. Rao, J. E. Greedan, W. E. Wallace, and R. S. Craig, J. Appl. Phys. **42**, 1293 (1971).

[43] J. Neuenschwander, H. Boppart, J. Schoenes, E. Voit, O. Vogt, and P. Wachter, Proceedings of the Quatorzièmes Journées des Actinides, edited by J. Schoenes, Davos, 1984 (unpublished), p. 30.

[44] H. Boppart, A. Treindl, P. Wachter, and S. Roth, Solid State Commun. **35**, 483 (1980).

[45] M. Lavagna, C. Lacroix, and M. Cyrot, J. Phys. F **12**, 745 (1982).

[46] B. Coqblin, A. K. Bhattacharjee, and R. Jullien, J. Magn. Magn. Mater. **15-18**, 995 (1980).