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TRANSVERSAL ELECTRICAL PROPERTIES OF THE UNIAXIAL ANTIFERROMAGNETS UAs_2 AND USb_2

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The paper presents the results of electrical resistivity measurements carried out on single crystal samples of antiferromagnetic UAs_2 and USb_2 in the temperature range 4.2–300°K in an electric field perpendicular to the *c*-axis, as well as on powder samples in the temperature range 77–500°K. It was found that these compounds exhibit a high anisotropy of the electrical resistivity. The temperature derivative $d\varrho/dT$ of the transversal resistivity has a sharp peak at about T_N. Below 60°K the spin disorder resistivity of UAs_2 is governed by spin wave scattering with a spin-wave activation energy of about 51°K, and above 70°K up to T_N it is proportional to T^2. For USb_2 this resistivity is proportional to $T^{5/2}$.

1. *Introduction*

UX_2 compounds where X = (P, As, Sb, Bi) show antiferromagnetic ordering of the magnetic moments of uranium atoms [1, 2, 3]. As has been shown recently [4], UP_2 exhibits strong anisotropic transport properties with predominant spin disorder scattering. In this paper the investigation of the electrical properties has been extended to UAs_2 and USb_2.

Hitherto, the electrical resistivity has been measured on compact samples of USb_2 in the paramagnetic state (20–600°C) only [5]. Compact samples of USb_2 were obtained by hot-pressing of the powders. The electrical properties of UAs_2 were unknown.

The dependence of the inverse magnetic susceptibility against temperature shows that for UAs_2 the Néel point is at 283°K, and for USb_2 at 206°K. The effective magnetic moments of UAs_2 and USb_2 are 2.94 and 3.04 BM, respectively [2].

According to neutron diffraction measurements [6, 7], the magnetic cell of these compounds compared to the chemical one (tetragonal structure of the PbFCl type) is doubled along the *c*-axis, and the lattice displays ferromagnetic layers of uranium atoms

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which are perpendicular to the c-axis and coupled between each other in the sequence $+--+$. The magnetic moments of the uranium atoms of UAs_2 and USb_2 are 1.61 ± 0.01 and 1.88 ± 0.03 BM, respectively, their direction being parallel to the c-axis.

2. *Experimental*

The electric measurements were performed using monocrystalline samples obtained by the chemical transport method [8], as well as compressed powder samples prepared according to the method described earlier [4]. The dimensions of the powder samples were $15 \times 5 \times 2$ mm^3, their density being 82% and 90% of the theoretical one, respectively for UAs_2 and USb_2. The X-ray analysis of the samples has revealed the existence of the UAs_2 or the USb_2 phase only.

The measurements of the temperature dependence of the resistivity (ϱ) were performed by the conventional four probe *dc* method. The electric contacts with the samples were made by silver dots pressed by springs or by using the Hg-In-Tl alloy. The resistivity was calculated for 100% density according to [9].

3. *Results and discussion*

The results of the resistivity measurements of UAs_2 and USb_2 in the direction perpendicular to the c-axis ($\varrho_{\perp}(T)$ — transversal component) are presented in Fig. 1, in the temperature range from 4.2°K to room temperature. The electrical resistivity for this direction at 300°K calculated as the mean value obtained for four samples is 160 μΩ cm for UAs_2 and 176 μΩ cm for USb_2. The experimental error for these measurements is

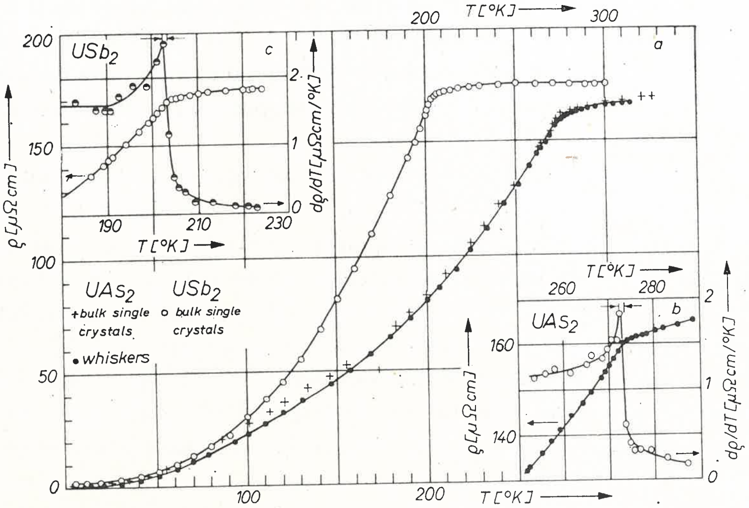

Fig. 1. Resistivity *versus* temperature for UAs_2 and USb_2 single crystals measured perpendicularly to the c-axis (part a), and temperature derivative of the resistivity (parts b and c)

about 5%. The residual resistivity ratio, $\varrho_{300}/\varrho_{4.2}$, varied within 200–260 for UAs_2 and 90–100 for USb_2 samples.

For USb_2 the measurements were performed on crystals grown in the form of regular plates, with the dimension ratio: $l/b > 3$ (l — length, b — width). The c-axis was perpendicular to the plate, and the a-axis parallel to the length. The temperature dependence of the resistivity measured for various samples was practically the same. The results of measurements for one of these samples are presented in Fig. 1.

The resistivity measurements for UAs_2 were performed using whiskers or bulk samples polished from bigger crystals. The X-ray diffraction control of the bulk samples showed that the declination of the samples' lateral surfaces from the a-axis directions was 2°–3°.

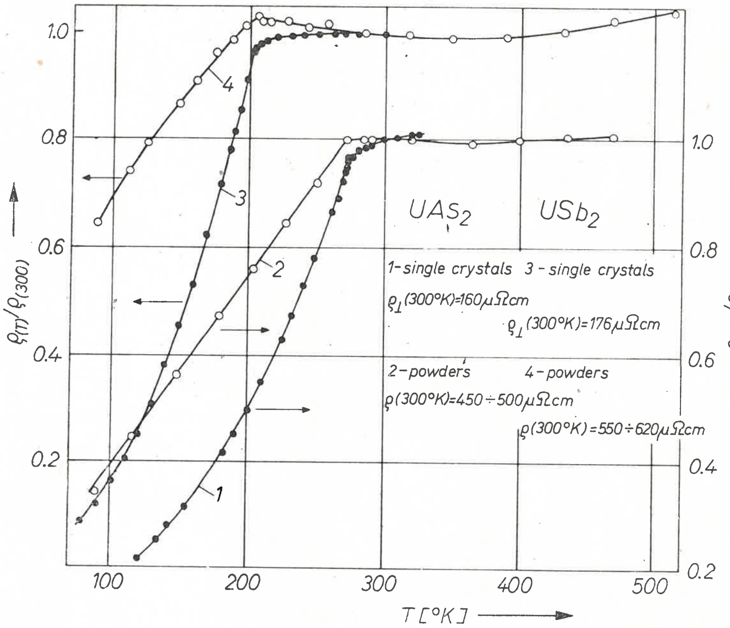

Fig. 2. Temperature dependence of resistivity of powder samples of UAs_2 and USb_2 (curves *2* and *4*) and monocrystals (curves *1* and *3*) measured perpendicularly to the c-axis

The temperature dependence of the resistivity, normalized at the Curie point, is presented for the bulk sample (crosses) and for the whisker of UAs_2 with diameter $1.1 \cdot 10^{-3}$ cm (full circles) in Fig. 1. Some discrepancies of the $\varrho(T)$ values can be noticed there. If the resistivity tensor for UAs_2 were highly anisotropic, then the observed discrepancy between the ϱ *versus* T curves for these samples might be caused by the longitudinal component $\varrho_{||}(T)$ which contributes besides the transversal component $\varrho_\perp(T)$ to the measured value of the resistivity of the bulk sample. For this reason additional measurements of the temperature dependence of the resistivity of powdered samples were performed.

The resistivity of powdered samples at 300°K, calculated for 100% density, varied from 450 to 500 μΩ cm for UAs_2 and from 550 to 620 μΩ cm for USb_2. The values given in this paper for the latter compound agree with the value of 600 μΩ cm reported by

Warren and Price [5]. At room temperature the resistivity of the powder samples of UAs_2 and USb_2 is about three times larger then that of the transversal components of the single crystals (Fig. 2). In addition, the temperature dependence of the resistivity of powder samples of these compounds (Fig. 2, curves *2* and *4*) differs from that of $\varrho_{\perp}(T)$ (Fig. 2, curves *1* and *3*). As has been reported earlier for UP_2 [4], similar differences between the resistivity $\varrho(T)$ of powder samples and the transversal component $\varrho_{\perp}(T)$ for the single crystal have been ascribed to the strong anisotropy of the transport properties; namely, at room temperature the resistivity along the *c*-axis ($\varrho_{\parallel}(T)$) was about three times larger than $\varrho_{\perp}(T)$. One might expect, therefore, that for the UAs_2 samples a strong anisotropy is responsible for the discrepancy of $\varrho(T)$ for the bulk sample and the whiskers, due to the deviation the lateral surfaces from the *a*-axis. Because the crystals were very thin along the *c*-axis, it was imposible to measure the component of the resistivity in this direction.

The spin disorder resistivity was obtained from $\varrho_{\perp}(T)$ measurements by the use of Matthiessens rule:

$$\varrho_{\perp}(T) = \varrho_{\text{phonon}}(T)+\varrho_{\text{spin}\,\perp}(T)+\varrho_{\text{residual}\,\perp} \tag{1}$$

where the symbols have the usual meaning.

The $\varrho_{\text{residual}}$ of the individual samples, taken as the resistivity value at 4.2°K, is 0.4, *i.e.*, 1.1% of the resistivity value at room temperature.

The temperature derivative, $d\varrho/dT$, of the electrical resistivity near T_N varies in UAs_2 and USb_2 (Fig. 1, part b and c) like the magnetic specific heat of UP_2 [10] (specific heats for UAs_2 and USb_2 are unknown) and shows the same positive divergence. As shown by Fisher and Langer [11] for ferromagnets and Marcelja [12] for antiferromagnets, this behaviour is caused by the dominant contribution of the short-range spin fluctuations to the spin disorder resistivity of the metal.

At the highest applied temperatures, $d\varrho/dT$ is still temperature dependent which makes the determination of ϱ_{phonon} impossible. However, as $d\varrho/dT \to 0$ with increasing temperature, one can suppose that the phonon contribution to the total resistivity is small and can be neglected in evaluating ϱ_{spin}.

The temperature dependence of the spin disorder resistivity has been considered many times (see [13, 14]). For this reason, we have compared our results for UAs_2 and USb_2 only with Kasuya's theory [15]:

$$\varrho_{\text{spin}}(T)/\varrho_{\text{spin}\,\infty} = 1-\frac{M(T)\,(M(T)+1)}{M(0)\,(M(0)+1)} \tag{2}$$

where $M(T)$ is the sublattice magnetization at temperature T. The magnetic moment obtained from neutron diffraction measurements [6, 7] has been used as the $M(0)$ value.

The magnetic entropy change of UP_2 [10] $\Delta S = 1.31$ cal mol^{-1} deg^{-1} is very close to the value which one might expect for a doublet as the ground state of the localized electrons.

On the basis of these data, we assume that in UAs_2 and USb_2 a pseudo-doublet with the splitting gap Δ is the ground state of the $5f^2$ configuration electrons of the paramagnetic ions. In the molecular field $H_m = \lambda M(T)$, the energy gap between the two levels is

$$\varepsilon = g\mu_B \lambda M(T) + \Delta \tag{3}$$

and the sublattice magnetization $M(T)$ is

$$M(T) = \tfrac{1}{2}\, g\mu_B \frac{1-e^{-\varepsilon/kT}}{1+e^{-\varepsilon/kT}} \tag{4}$$

where λ is the molecular field parameter and the $M(0)$ value is $\frac{1}{2}\, g\mu_B$. For the numerical calculations the following formula is convenient:

$$\frac{M(T)}{M(0)} = \frac{1-e^{-\alpha}}{1+e^{-\alpha}} \quad \text{where } \alpha = A\frac{T_N}{T}\frac{M(T)}{M(0)} + B\frac{T_N}{T}. \tag{5}$$

The parameters A and B are proportional to λ and Δ, respectively. We have chosen T_N to be 273 ± 0.5°K for UAs_2 and 203 ± 0.5°K for USb_2, corresponding to the temperature at which $d\varrho/dT$ has a peak. These temperatures differ by 10° and 3°K, respectively, from those determined from magnetic measurements for the powder samples [2].

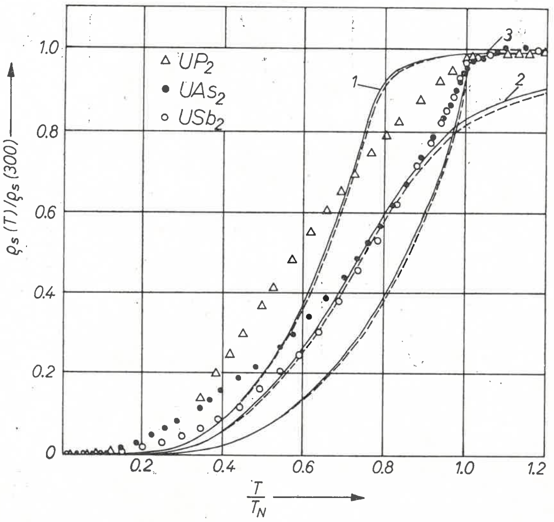

Fig. 3. Normalized spin disorder resistivity as a function of T/T_N. The solid curves are calculated from the theory of Kasuya [15] for various molecular field parameters and splitting gaps of the pseudo-doublet. The spin disorder resistivity data for UP_2 are taken from paper [4]

In Fig. 3 the curves calculated for UP_2 (full line) and for UAs_2 (dashed line) from the formulae (2) and (5) for various parameters A and B are compared with the experimental data. The values of the above parameters for the three curves are: *1)* $A = 1.5$, $B = 0.015$; *2)* $A = 1.5$, $B = 0.15$; *3)* $A = 2$, $B = 0.0015$. The agreement is only qualitative. However, as shown in Fig. 4 for USb_2 the dependence $\varrho_{\text{spin}} \sim T^{5/2}$ is fulfilled. The measurements for UAs_2 can be adequatly described by the relation $\varrho_{\text{spin}} \sim T^2$ for the temperature range from 70°K up to T_N, and by $\varrho_{\text{spin}} \sim T^2 \exp(-E/kT)$ below 60°K, with the value $E/k = 51$°K (Fig. 4, part b).

Several theories were developed to describe the behaviour of the electrical resistivity of ferromagnets and antiferromagnets near their transition temperatures. Our results

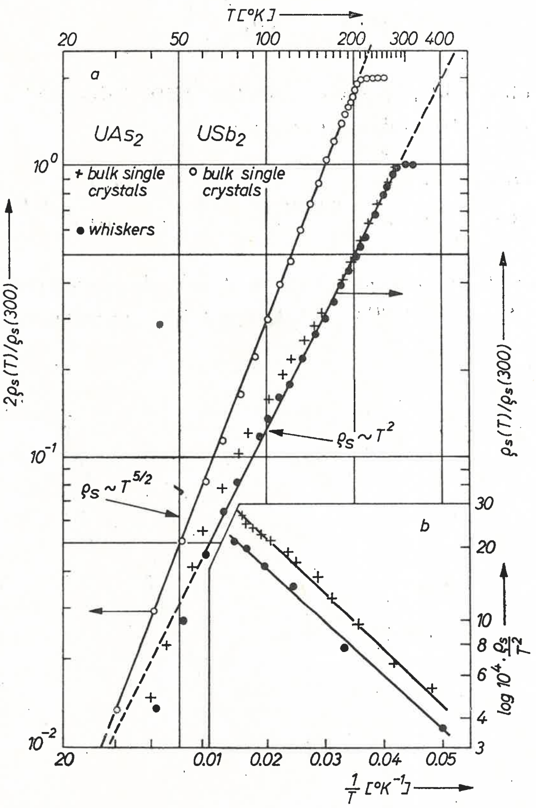

Fig. 4. Spin disorder resistivity perpendicular to c-axis *versus* temperature

obtained for the uniaxial antiferromagnetic uranium compounds UAs_2 and USb_2 (spin disorder resistivity perpendicular to the spin axis) show a qualitative agreement with Fisher and Langer [11] or Marčelja's [12] theories.

The temperature dependence of the spin disorder resistivity for the UX_2 compounds (where X = P, As, Sb) shows a still better fit with Kasuya's theory and the simple molecular field model with the increasing atomic number of the metaloid (Fig. 3). One might expect a better agreement between experimental results and theory by taking into account the influence of the crystal field on the central U^{4+} ion as well as the anisotropy constant determined experimentally. This might also be helpful in explaining the differences in the temperature dependence of ϱ_{spin}.

Due to the electron-spinwave scattering [16, 17], in simple ferromagnets the relationship $\varrho_{\text{spin}} \sim T^2$ should be fulfilled. According to Mackintosh [18], in ferromagnets with high magnetocrystalline anisotropy the spin resistivity should involve a term depending on the spin wave excitation energy E, the contribution of which is $\varrho_{\mathrm{spin}} \sim T^2 \exp(-E/kT)$. However, the scattering of electrons by disturbances in the spin system can be analysed in terms of the spin-wave spectrum at low temperatures ($T < \frac{1}{3} T_N$), the relation $\varrho_{\mathrm{spin}} \sim T^2$ being fulfilled above this range. The origin of the $T^{5/2}$ term is not yet clear.

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REFERENCES

[1] W. Trzebiatowski, R. Troć, *Bull. Acad. Polon. Sci., Ser. Sci. Chim.*, **11**, 661 (1963).
[2] W. Trzebiatowski, A. Sępichowska, A. Zygmunt, *Bull. Acad. Polon. Sci., Ser. Sci. Chim.*, **12**, 687 (1964).
[3] W. Trzebiatowski, A. Zygmunt, *Bull. Acad. Polon. Sci., Ser. Sci., Chim.*, **14**, 495 (1966).
[4] Z. Henkie, W. Trzebiatowski, *Phys. Status Solidi*, **35**, 827 (1969).
[5] J. H. Warren, C. E. Price, *Canad. met. Quart.*, **3**, 245 (1964).
[6] A Oleś, *J. Phys. Radium*, **26**, 561 (1965).
[7] J. Leciejewicz, R. Troć, A. Murasik, A. Zygmunt, *Phys. Status Solidi*, **22**, 517 (1967); Erratum, *Phys. Status Solidi*, **24**, 763 (1967).
[8] Z. Henkie, *Roczniki Chemii*, **42**, 363 (1968), in Polish.
[9] H. J. Juretschke, R. Steinitz, *J. Phys. Chem. Solids*, **4**, 118 (1958).
[10] B. Staliński, Z. Biegański, R. Troć, *Bull. Acad. Polon. Sci., Ser. Sci. Chim.*, **15**, 257 (1967).
[11] M. E. Fischer, J. S. Langer, *Phys. Rev. Letters*, **20**, 665 (1968).
[12] S. Marĉelja, *J. Phys. C.*, **4**, L 26 (1971).
[13] G. Koszai, L. Maksimov, *Fiz. Metallov Metallovedenie*, **27**, 976 (1969), in Russian.
[14] C. W. Searle, S. T. Wang, *Canad. J. Phys.*, **48**, 2023 (1970).
[15] T. Kasuya, *Progr. Theor. Phys.*, **16**, 58 (1956).
[16] J. Mannari, *Progr. Theor. Phys.*, **22**, 335 (1959).
[17] T. Kasuya, *Progr. Theor. Phys.*, **22**, 227 (1959).
[18] A. R. Mackintosh, *Phys. Letters*, **4**, 140 (1963).