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**Magnetic and transport properties of UBi<sub>2</sub> and USb<sub>2</sub> single crystals**

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## Magnetic and transport properties of $\text{UBi}_2$ and $\text{USb}_2$ single crystals

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### ABSTRACT

Thermoelectric power,  $S(T)$ , of  $\text{USb}_2$  and  $\text{UBi}_2$  that are tetragonal, uniaxial antiferromagnets below  $T_N = 202$  K and 180.8 K, respectively, have been examined between 0.4 K and 300 K. The  $S(T)$  dependencies, up to now known above 70 K for  $\text{USb}_2$  and unknown for  $\text{UBi}_2$ , are along the  $a$ -axis positive for both compounds in the whole examined temperature range. The  $S(T)$  data for the  $c$ -axis (the easy magnetisation axis) are positive near the room temperature for  $\text{USb}_2$  and  $\text{UBi}_2$  and becomes negative below 120 K and 170 K, respectively, with two very deep minima in  $S(T)$  dependence for  $\text{USb}_2$ . In the latter compound the Fermi surface, known from literature, is composed of the only cylindrical sheets that are slightly corrugated and parallel to the  $c$ -axis.  $\text{UBi}_2$ , the Fermi surface of which is composed of one spherical and two cylindrical sheets shows the corresponding minima to be less pronounced than those in  $\text{USb}_2$ . Having in disposal the highest purity single crystals in comparison to those for which the resistivity,  $\rho(T)$ , has been reported in literature up to now, the  $\rho(T)$  anisotropy could be re-examined for these two systems. Magnon and phonon contributions to their total electrical resistivity have been determined and critical fluctuation behaviour of the resistivity near  $T_N$  for both dipnictides has also been analysed. Although the magnetic susceptibilities of  $\text{UBi}_2$  and  $\text{USb}_2$  reveal a similarity, their transport properties are significantly different due to the difference in the Fermi surface topology.

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*Keywords:* Uranium dipnictides; Magnetic susceptibility; Resistivity; Thermoelectric power

## § 1. INTRODUCTION

The  $\text{UBi}_2$  and  $\text{USb}_2$  compounds belong to group of the uranium dipnictides  $\text{UX}_2$  ( $X=\text{P}, \text{As}, \text{Sb}, \text{Bi}$ ) - they were, and are still intensively investigated due to their unusual transport properties. The magnetic properties of  $\text{UBi}_2$  and  $\text{USb}_2$  are described in Refs. [1] – [5], whereas the limited transport properties are presented in Refs. [6, 7, 8]. The crystal structure and the Fermi surface [9] of both compounds are well defined. Therefore the aim of this work is to show how their features are reflected in the electrical transport coefficients.

$\text{UBi}_2$  and  $\text{USb}_2$  crystallise in a tetragonal structure belonging to the anti- $\text{Cu}_2\text{Sb}$ -type with either  $\text{P4/nmm}$  or  $\text{D}_{4h}^7$  space group. The unit cell contains two formula units and the ratio of unit cell parameters  $c/a$  equals to 2.004 and 2.044 for  $\text{UBi}_2$  and  $\text{USb}_2$ , respectively. The neutron-diffraction study [3] showed that below the Néel temperature they order antiferromagnetically, along the  $c$ -axis, with alternating ferromagnetic sheets in the sequences  $(\uparrow\downarrow\uparrow)$  for  $\text{USb}_2$  and  $(\uparrow\downarrow)$  for  $\text{UBi}_2$ . Such an ordering results in the magnetic unit cell of  $\text{USb}_2$ , which is twice the chemical unit cell along the  $c$ -axis, while the magnetic and chemical unit cells of  $\text{UBi}_2$  are of the same size. Hence the magnetic Brillouin zone for  $\text{UBi}_2$  being fairly flat is the same as that of the chemical one. The ratio of the length  $k_c$  along [001] to the length  $k_a$  along [100] in the magnetic Brillouin zone of  $\text{USb}_2$  and  $\text{UBi}_2$  is 0.24 and 0.5, respectively.

The de Haas-van Alphen and Shubnikov-de Haas experiments have allowed for constructing the Fermi surface for  $\text{UBi}_2$  and  $\text{USb}_2$  in the ordered state [9]. It consists of one spherical ( $\alpha$ ) and two cylindrical ( $\beta$ ) sheets in  $\text{UBi}_2$  and entirely cylindrical sheets in  $\text{USb}_2$ . The magnetic Brillouin zone in  $\text{USb}_2$  consists of one of each cylindrical sheets ( $\alpha$ ) and ( $\delta$ ), and two of each of cylindrical sheets ( $\epsilon$ ) and ( $\gamma$ ). The Fermi surfaces of  $\text{UBi}_2$  are the same in the paramagnetic and antiferromagnetic states. In the case of  $\text{USb}_2$  the paramagnetic Fermi surfaces are related to those in  $\text{UBi}_2$ , because the Brillouin zone in  $\text{USb}_2$  is almost the same as that in  $\text{UBi}_2$  and it is occupied by similar number of electrons. Thus, in the paramagnetic state

the spherical sheet ( $\alpha$ ) in  $\text{UBi}_2$  corresponds to the corrugated cylindrical one in  $\text{USb}_2$  (see figures 24c and 25a in Ref. [9]) and the cylindrical sheet ( $\beta$ ) in  $\text{UBi}_2$  corresponds to the long ellipsoidal Fermi surfaces in  $\text{USb}_2$  (see figure 24d in Ref. [9]). The latter crosses over the Brillouin zone boundary, forming another corrugated cylindrical and a small-pocket ellipsoidal Fermi surfaces.

The paramagnetic to antiferromagnetic transition in  $\text{USb}_2$  increases the number of the Fermi surface sheets and enhances their quasi-two dimensionality, as discussed in Ref. [9].

An angle-resolved photoemission study in single crystal of  $\text{USb}_2$  at 15 K [10] showed the dispersion of extremely narrow bands situated near the Fermi level. The natural linewidth is less than 10 meV. In the normal emission spectrum there is also a dispersion observed of about 10 meV, which indicates a 3D character in electronic structure of  $\text{USb}_2$  in contrast to the earlier evidence [9]. These resonant photoemission measurements confirm the  $5f$ -electron hybridisation with the conduction electrons.

This work presents results of measurements of the magnetic susceptibility  $\chi(T)$ , electrical resistivity  $\rho(T)$ , and thermoelectric power  $S(T)$  down to 0.35 K, for  $\text{UBi}_2$  and  $\text{USb}_2$  single crystals. Their Fermi surfaces distinctly differ from each other.

We compare these data to those partially known from the literature and discuss the differences in the observed behaviours of  $\text{USb}_2$  and  $\text{UBi}_2$ . These observed differences are related to the variation in the Fermi surface topology of these two compounds.

## § 2. EXPERIMENTAL DETAILS

$\text{USb}_2$  and  $\text{UBi}_2$  single crystals were grown by the so called „molten metal solution” method [8]. They were grown in the form of plates with the dimensions of about  $6 \times 5 \times 3 \text{ mm}^3$  along the  $a$ -,  $b$ -, and  $c$ - axes, respectively. For electrical resistivity and thermoelectric power examinations the specimens were cut out to the dimensions of about  $4 \times 0.5 \times 0.5 \text{ mm}^3$  and  $3 \times 0.3 \times 0.3 \text{ mm}^3$  for

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Deleted: The investigation of hyperfine interactions at the uranium sites in the antiferromagnetic state of uranium dipnictides was done using  $^{238}\text{U}$  Mössbauer spectroscopy [11]. In  $\text{UBi}_2$  the nuclear quadrupole interaction at  $^{238}\text{U}$  is almost zero, whereas in  $\text{USb}_2$  the sign of these interactions is negative. This suggests that the nuclear quadrupole interactions in both compounds are correlated with their magnetic structures.

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2 carrying out the measurements along the  $a$ - and  $c$ - axes, respectively. The dc magnetic  
3 susceptibility in fields up to 5 T applied along the  $c$ -axis and in the temperature range 1.7 – 400  
4 K was measured with a superconducting quantum interference device magnetometer (Quantum  
5 Design). The electrical resistivity at temperatures of 0.35 – 300 K was measured by a four  
6 point ac method. For the thermoelectric power measurements a modified set-up described in  
7 Ref. [11] has been used.  
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### 13 § 3. RESULTS

#### 14 3.1. Magnetic susceptibility

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Magnetic susceptibility of a  $\text{UBi}_2$  crystal, measured along the  $c$ -axis, shows the temperature dependence being typical for an antiferromagnet with a peak at  $T_N = 180.8$  K (figure 1, see also Ref. [12]). In the paramagnetic state the modified Curie–Weiss law is fulfilled:  $\chi(T) = \frac{N\mu_{eff}^2}{3k_B(T + \Theta_p)} + \chi_0$ , where  $N$  denotes the number of atoms in the unit cell,  $\mu_{eff}$  is the effective magnetic moment,  $k_B$  is the Boltzmann constant,  $\Theta_p$  is the paramagnetic Curie temperature, and  $\chi_0$  is the temperature independent paramagnetic contribution. The parameters are as follows:  $\mu_{eff} = 3.36 \mu_B$ ,  $\Theta_p = -17$  K, and  $\chi_0 = 2.7 \times 10^{-3}$  emu/mol. Obtained from the fit of above equation to experimental data the  $\mu_{eff}$  value appears to be very close to that of  $3.4 \mu_B$ , reported in Ref. [2]. The determined  $\mu_{eff}$  is however smaller from that of  $3.62 \mu_B$  or  $3.58 \mu_B$  expected for the free  $\text{U}^{+3}$  or  $\text{U}^{+4}$  ions, respectively.

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[Insert figure 1 about here]

In the case of  $\text{USb}_2$ , the  $\chi(T)$  dependence, measured along the  $c$ -axis, displays a sharp maximum yielding the Néel temperature of 202 K (figure 1, see also Ref. [13]). In this case, the modified Curie-Weiss law is obeyed in paramagnetic region for  $\Theta_p = -59$  K and  $\mu_{eff} = 3.32 \mu_B$ .

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### 3.2. Electrical resistivity

The electrical resistivity of  $\text{UBi}_2$  single crystals were carried out, in the  $a$  and  $c$  directions, for two different samples having different purity. They were (figure 2), obtained in different syntheses. The resistivity ratio  $\rho_{300\text{ K}}/\rho_{4.2\text{ K}} = \text{RR}$  measured along the  $a$ -axis for the samples obtained in the first (1s) and second (2s) syntheses were 500 and 1110, respectively. The room temperature resistivity,  $\rho_{300\text{ K}}$ , for both the (1s) and (2s) samples in the  $a$  direction was of 0.231 m $\Omega$  cm while in the  $c$  direction it was 3.88 m $\Omega$  cm. In the paramagnetic state the resistivity was found to be almost temperature independent, while that below  $T_N$  decreases rapidly in both directions with decreasing temperature.

[Insert figure 2 about here]

As seen in figure 2, distinct anisotropy in the temperature dependence of the resistivity is observed. Below  $T_N$ , the anisotropy ratio  $(\rho_c - \rho_0)/(\rho_a - \rho_0)$  increases with decreasing temperature down to about 27 K. It achieves a value of about 2.7 times larger than that in the paramagnetic range (figure 3), and then it decreases at lower temperatures.

The temperature dependence of the anisotropy ratio  $(\rho_c - \rho_0)/(\rho_a - \rho_0)$  differs from the results presented in Ref. [9], where the anisotropy ratio defined as  $\rho_c/\rho_a$  achieves a maximum value of about 800 at the temperature of circa 80 K. In the present work the  $(\rho_c - \rho_0)/(\rho_a - \rho_0)$  ratio reaches at 27 K a maximum value of about 50 while the  $\rho_c/\rho_a$  ratio is not larger than about 100 (figure 3).

[Insert figure 3 about here]

This discrepancy in the anisotropy ratio probably arises from the difference in the sample purity. As mentioned above, the RR values for our samples in the  $a$  direction are 500 (1s) and 1110 (2s) compared to  $\text{RR} = 22$  for the sample investigated by the authors of Ref. [9]. The RR values for the  $c$  direction in both cases were of the same order (i.e. 130 and 215 for our samples and 160 for the sample of Ref. [9]). Also the discrepancy in absolute values of the resistivity

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appears; for example, the room temperature resistivity of our samples along the  $a$ - and  $c$ -axes are of 0.23 mΩ cm and 3.9 mΩ cm, while that for the samples from Ref. [9] these values are considerably different and amount to 60 mΩ cm and  $3.4 \times 10^4$  mΩ cm, respectively. The samples differ mainly in the magnitudes of the residual resistivity,  $\rho_0$ .

According to the Matthiessen's rule the temperature dependence of the resistivity for an antiferromagnet can be written as follows:

$$\rho(T) = \rho_0 + a(T/\Theta)^n \int_0^{\Theta/T} \frac{z^n dz}{(e^z - 1)(1 - e^{-z})} + bT(1 + 2T/\Delta) \exp\left(-\frac{\Delta}{T}\right) = \rho_0 + \rho_{\text{ph}} + \rho_{\text{m}}, \quad (1)$$

where  $\rho_{\text{ph}}$  is approximated by the generalised Bloch-Grüneisen equation for electron-phonon scattering with  $n = 3$  (similarly as for another uranium compounds, e.g. for URhGa<sub>5</sub> [14]). In Eq. (1) the  $\rho_{\text{m}}$  denotes the magnon component,  $\Theta$  is the Debye temperature,  $\Delta$  ( $= \varepsilon/k_{\text{B}}$ ) is a gap in the magnons spectrum, and  $a$  and  $b$  are the fitting parameters. Whereas the third part of Eq. (1), which concerns the electron-magnon scattering, is limited to temperatures below  $T_{\text{N}}/3$ , the second part of Eq. (1) is valid in the whole temperature range (dashed line in figure 4).

[Insert figure 4 about here]

In figure 4 the fit of Eq. (1) to the experimental data is displayed by the solid line. The  $\rho_{\text{m}}(T)$  equation was used previously for the fitting of the data in the case of the USb antiferromagnet [11].

The experimental  $\rho(T)$  data of a USb<sub>2</sub> single crystal are displayed in figure 5. The results are similar to those obtained and described earlier in Refs. [7, 8] and [9], however a discrepancy in the anisotropy ratio is observed in comparison to the data of Ref. [9]. In the latter case a maximum value of about 90 is achieved in the  $\rho_c/\rho_a$  ratio at a temperature of circa 60 K, while for our sample this maximum value is found to be higher even than 190 and occurs at the temperature of 23 K (figure 3).

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Deleted: As mentioned above, USb<sub>2</sub> exhibits an antiferromagnetic ordering below  $T_{\text{N}} = 202$  K.

In the paramagnetic state, near the critical temperature,  $\rho(T)$  in the  $c$  direction in the limit of  $0 \leq t \leq 0.04$  ( $t = (T - T_N)/T_N$ ) can be expressed as that in Ref. [15]:

$$\rho(T) = A t^{1-\alpha} + B|t| + C, \quad (2)$$

where  $\alpha$  is the critical exponent,  $A$  is the critical amplitude of the resistivity;  $B$  and  $C$  are the fitting parameters (first part of the solid curve 1 in figure 5). The exponent  $(1 - \alpha)$  from Eq. (2) becomes  $\frac{1}{2}$  in the limit  $0.04 \leq t \leq 0.12$  (second part). As seen from figure 5 (third part of the curve 1), formula (2) with  $\alpha = 0.326$  gives a good fit to the experimental data at temperatures above 225 K. For the  $a$ -axis the resistivity between  $T_N$  and 260 K increases and then decreases slightly with increasing temperature to room temperatures.

[Insert figure 5 about here]

Below 70 K, for the  $\rho(T)$  data taken in a similar way like in  $\text{UBi}_2$  along the  $a$ -axis a sufficient approximation is given by Eq. (1) (curve 2 in the inset of figure 5). A good fit in the antiferromagnetic range is given by the equation:  $\rho(T) = \rho_0 + \rho_{\text{ph}} + aT^{5/2}$  (dash-dotted line) [7]. For the  $c$  direction below  $T_N/3$  Eq. (1) gives rather a poor fit.

The behaviours of critical resistivity for the  $\text{UBi}_2$  and  $\text{USb}_2$  single crystals in the  $a$  and  $c$  directions are presented in figure 6 as a function of the reduced temperature  $t = (T - T_N)/T_N$ . Using the classification from Ref. [16], the critical resistivity of a  $\text{UBi}_2$  crystal in the  $a$  direction behaves like that for an antiferromagnetic semiconductor (figure 6a), while that along the  $c$ -axis as for an antiferromagnetic metal (figure 6b). This rule concerns  $\text{USb}_2$  as well (figures 6c and 6d). The solid lines in figure 6 represent Eq. (2) fitted to the experimental data of the  $\text{UBi}_2$  and  $\text{USb}_2$  resistivities within the temperature limit  $-0.08 \leq t \leq 0.08$  (for  $\text{UBi}_2$  along the  $c$ -axis (figure 6b) this limit is equal to  $-0.04 \leq t \leq 0.08$ ). The exponent  $(1 - \alpha)$  becomes  $\frac{1}{2}$  at boundary marked by  $t_G$ .

[Insert figure 6 about here]

### 3.3. Thermoelectric power

The  $\text{UBi}_2$  single crystals reveal a large anisotropy in the temperature dependence of the thermoelectric power  $S(T)$  (see figure 7). Above the Néel temperature,  $S(T)$  decreases and increases for the  $a$  and  $c$  directions, respectively. Below  $T_N$ , a few extremes in  $S(T)$  are observed: for the  $a$  direction, after the change in slope at  $T_N$ , the two maxima in the temperature dependence of the thermoelectric power occur, the first one:  ${}^1aS_{\max} = 30.6 \mu\text{V/K}$  at  ${}^1aT_{\max} = 130 \text{ K}$  and the second maximum  ${}^2aS_{\max} = 2.5 \mu\text{V/K}$  at  ${}^2aT_{\max} = 3.5 \text{ K}$ .

[Insert figure 7 about here]

For the  $c$ -axis two minima in  $S(T)$  exist: i.e.  ${}^1cS_{\min} = -5.9 \mu\text{V/K}$  and  ${}^2cS_{\min} = -1.0 \mu\text{V/K}$  at  ${}^1cT_{\min}$  and  ${}^2cT_{\min}$  of 103 K and 13.5 K, respectively. The  $S(T)$  curve for  $\text{UBi}_2$  along the  $c$  direction changes its sign thrice, namely at temperatures: 33.5 K, 38 K, and 166 K.

The  $S(T)$  dependencies for a  $\text{USb}_2$  single crystal along the  $a$  and  $c$  directions are presented in figure 8. They are similar to the results presented in Refs. [8, 17] along the  $a$  direction in the temperature range 50 – 300 K and are similar to the results of Ref. [17] in the  $c$  direction in the temperature range 75 – 300 K. However in the latter case they differ in absolute values of  $S$  at lower temperatures. In general, the thermoelectric power for  $\text{USb}_2$  differs from that of  $\text{UBi}_2$ , particularly in that taken along the  $a$ -axis. However, a maximum of  ${}^aS_{\max} = 14.2 \mu\text{V/K}$  in the antiferromagnetic range exists, but it is single and appears at  ${}^aT_{\max} = 25 \text{ K}$ . This maximum can be explained if we assume that the total thermoelectric power is a sum of the two components: the diffusion one,  $S_d$ , that increases linearly with temperature and the magnon/phonon-drag one,  $S_g$ , which is peaked. The phonon-drag thermoelectric power depends on temperature as  $T^3$  before the maximum of the peak and as  $1/T$  on the right hand side of  $S_{\max}$  [18]. The solid curve 1 (the inset on the left hand side in figure 8) presents the equation:  $S(T) = S_d + S_g = aT + b/T$  fitted to the experimental data of  $S(T)$  within the temperature range 25 – 90 K. The  $S_d$  (straight line 2) was calculated from the above equation,

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whereas the  $S_g$  (curve 3) was estimated as the difference between the experimental data and diffusion contribution. The maximum of  $S_g$  appears at  $T^* = 7.5$  K.

Similarly like for  $UBi_2$ ,  $S(T)$  in the  $c$  direction changes the sign in the antiferromagnetic ordering and passes through two minima:  ${}^1cS_{\min} = -18.4 \mu\text{V/K}$  and  ${}^2cS_{\min} = -19.2 \mu\text{V/K}$  at  ${}^1cT_{\min} = 78.5$  K and  ${}^2cT_{\min} = 6.4$  K. The two latter minima are deeper than those in the  $UBi_2$  case and occur at lower temperatures.

[Insert figure 8 about here]

The lowest part of the  $S(T)$  data are well fitted by the equation:  $S(T) = a'T + b'T^3$  being well known for the case of two components, i.e. for the diffusion and phonon-drag components in the thermoelectric power (the inset on the right hand side in figure 8). In the ordered state one should consider rather the magnon/phonon-drag component.

#### § 4. DISCUSSIONS AND CONCLUSIONS

The temperature dependencies of the magnetic susceptibilities for  $UBi_2$  and  $USb_2$  single crystals reveal a similarity displaying a sharp maxima leading to the Néel temperatures of 180.8 K and 202 K, respectively. Below  $T_N$ , at about  $T = 21$  K and 50 K, in the  $\chi(T)$  curves of  $UBi_2$  and  $USb_2$  there are observed small minima (for  $UBi_2$  see the inset of figure 1), which may result from the crystal-field effects. In  $UBi_2$  this valley is not as deep as that observed for  $USb_2$  or also for  $UNiSb_2$  and  $UPdSb_2$  there measured on the polycrystalline samples [19]. In the ordered state, when the crystal field interaction is comparable with the exchange field interaction, the anomalies in the  $\chi(T)$  dependencies can occur. A crystal-field splitting (ground state is a doublet) of order of the exchange interaction yields an anomalous temperature variation of the magnetic susceptibility as was shown for  $CeSb$  and  $CeBi$  [20].

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The electrical resistivity of  $UBi_2$  measured along the  $c$  direction (figure 2) differs considerably from that of  $USb_2$  (figure 5). A large hump in the resistivity curve observed in  $USb_2$  just below  $T_N$  in the  $c$  direction can be easily explained by the fact that the corrugated

1  
2 cylindrical Fermi surface in the paramagnetic state is transformed into the two cylindrical  
3 sheets in magnetically ordered state, whilst the long ellipsoidal Fermi surface changes into the  
4 two cylindrical ones [9]. In  $\text{UBi}_2$  the Fermi surface is not rebuilt during the transition from the  
5 paramagnetic to antiferromagnetic state and such a large hump in the resistivity is not  
6 observed, but only a small hump in  $\rho(T)$  occurs (figure 6b).  
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10 The genesis of six times greater resistivity anisotropy for  $\text{USb}_2$  than that for  $\text{UBi}_2$  (figure 3) is  
11 in the shape of the Fermi surface. The group velocity  $v_g$  for the electronic transport defined as  
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13  $\hbar^{-1}(\partial E/\partial k)$  is zero at the band extrema and zone faces (see e.g. Ref. [21]). Vector  $\mathbf{v}_g = \hbar^{-1}$   
14  $(\partial E(\mathbf{k})/\partial \mathbf{k})$  is in the direction normal to the energy surface. This condition cannot be fulfilled  
15 along the  $c$  direction for the cylindrical surface. This means that in the ordered state of  $\text{USb}_2$ ,  
16 for the  $c$  direction the group velocity of electrons should be zero. In practice, for  $\text{USb}_2$  the  
17 cylindrical Fermi surfaces are corrugated [9] and a small component  $v_g$  along the  $c$ -axis exists.  
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19 In a real  $\text{USb}_2$  crystal, at low temperatures the resistivity (in relation to the electron-magnon  
20 and electron-phonon interactions) in the  $c$  direction is almost 300 times larger than that in the  $a$   
21 direction (figure 3).  
22

23 Because of a hybridisation effect with the conduction band, the  $5f$ -electrons, which are  
24 responsible for existing the magnetic moment in the uranium atoms, also contribute to the  
25 electronic transport properties of  $\text{UBi}_2$  and  $\text{USb}_2$ . They are considered to be partially localised  
26 and partially itinerant. It is known from the  $^{238}\text{U}$  Mössbauer spectroscopy results [22] and  
27 measured large cyclotron masses, that the  $5f$ -electrons in  $\text{UBi}_2$  are more hybridised with the  
28 conduction electrons than those in  $\text{USb}_2$  despite the larger  $d_{\text{U-U}}$  distance for the former  
29 compound.  
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31 The electrical resistivity in both crystals:  $\text{UBi}_2$  and  $\text{USb}_2$  is satisfactorily described, below  
32  $T_N/3$ , by Eq. (1), although a better fitting is obtained for a  $\text{UBi}_2$  crystal probably due to  
33 existence of the spherical Fermi surface. In  $\text{USb}_2$ , where only cylindrical Fermi surfaces exist,  
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1 the best description for the ordered state is given by  $\rho \sim T^{5/2}$  [7], but the mechanism yielding  
2 such a proportionality is as yet unknown.  
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5 Near the critical temperature in the  $a$  direction, the resistivity in  $\text{USb}_2$  differs from that  
6 observed in  $\text{UBi}_2$  which is firstly indicated by the temperature dependence of the derivative  
7  $d\rho/dT$ . Contrary to  $\text{UBi}_2$ , the  $d\rho/dT$  curve of  $\text{USb}_2$  exhibits a peak at 202 K (the smaller inset in  
8 figure 5). Secondly, the critical exponent  $\alpha$  in  $\text{UBi}_2$  is negative. In  $\text{UBi}_2$  the temperature  
9 dependence of  $\rho$  below  $T_N$  is convex while in the case of  $\text{USb}_2$  concave (figures 6a and 6c),  
10 probably due to the different values in the energy gap on the Brillouin zone boundary.  
11 However, it should be noticed that in  $\text{USb}_2$  the ellipsoidal ( $\beta$ ) sheet crosses over the Brillouin  
12 zone boundary, while in  $\text{UBi}_2$  such a case does not occur.  
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15 A metallic shape of  $\rho(T)$  in the critical region, characteristic for an antiferromagnet (in  
16 agreement with classification from Ref. [16]), is revealed for both the  $\text{UBi}_2$  and  $\text{USb}_2$  crystals  
17 only in the  $c$  direction. In view of the shape of the  $\rho(T)$  curve along the  $a$ - axis these two  
18 systems may be classified as antiferromagnetic semiconductors. This discrepancy in the  
19 classification may be caused by the fact that the authors of Ref. [16] assumed the spherical  
20 Fermi surface. In Ref. [23] it is shown that the critical behaviour of the resistivity is very  
21 sensitive to the shape of the Fermi surfaces and the position of the point of instability,  $Q$ , in the  
22 Brillouin zone.  
23

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25 The large- $q$  critical spin fluctuations grow in an antiferromagnet when the temperature  
26 decreases down through  $T_N$  because the antiferromagnetic order is associated with the large-  
27 wave-vector  $Q$  [15]. In a metallic antiferromagnet where  $Q \approx k_F$  this growth yields an increase  
28 in the resistance when the temperature is decreased through  $T_N$ . This large momentum-transfer  
29 is consistent with the conservation of energy and momentum. The temperature dependence of  
30 the resistivity of an antiferromagnetic metal, predicted in the vicinity of the critical temperature  
31 [15], is confirmed by our results especially for those obtained in  $\text{UBi}_2$  along the  $c$  direction  
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(figure 6b) and also for USb<sub>2</sub> (figure 6d), where, as mentioned above, a large hump in the resistivity behaviour occurs.

In conclusion, the resistivity near the critical temperature for both the UBi<sub>2</sub> and USb<sub>2</sub> crystals observed along the *a* and *c* directions varies in a manner as predicted for the energy-like behaviour [15, 16]: i.e.  $\rho \sim \pm |t|^{1-\alpha}$  and then it becomes proportional to  $\pm |t|^{1/2}$  close to the Ginsburg temperature,  $t_G$ , at the vicinity of a transition from the critical to mean-field behaviour.

Looking at the temperature dependencies of the thermoelectric power (figure 8) and of the resistivity (figure 5) for USb<sub>2</sub>, both measured for the *c* direction, one sees that they have the extremes almost at the same temperature  $T(\rho_{\max}) \approx {}^1cT_{\min}$ . This can be explained if we take into account the fact, that in a two band model of a compensated metal the contribution of the particular band's thermoelectric power to the total thermoelectric power is weighted by the contribution of the particular band's conductivity to the total conductivity. The second minimum in the  $S(T)$  dependence occurs at the temperature  ${}^2cT_{\min}$ , which is close to the temperature of a maximum anisotropy ratio in the resistivity (the solid line in figure 3). This also confirms the influence of the conductivity on the thermoelectric power behaviour. Similarly for UBi<sub>2</sub> (figure 7),  ${}^2cT_{\min}$  is close to the temperature of a maximum anisotropy ratio of the resistivity (dashed line in figure 3).

Undoubtedly, the shape of the Fermi surface is reflected in the temperature dependence of the thermoelectric power, e.g. for USb<sub>2</sub>, where only the cylindrical sheets exist along the *c*-axis (figure 8), the minima in  $S(T)$  are more strongly pronounced than those in the case of UBi<sub>2</sub> (figure 7), where besides the cylindrical sheets, a spherical one was also detected. This means, that in the case when only the cylindrical Fermi surfaces exist (USb<sub>2</sub>), the electron transport along the *c* direction due to the temperature gradient is also limited, like in the case of the

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2 presence of the electrical field gradient. Additionally, in relation to the Umklapp processes, the  
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4 electrons may drift in opposite direction to the temperature gradient.

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6 For good quality crystals, one cannot omit the magnon/phonon-drag contribution to the total  
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8 thermoelectric power at low temperatures. This contribution is visible on the  $S(T)$  dependence  
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10 especially along the  $a$ -axis for both the  $\text{UBi}_2$  and  $\text{USb}_2$  single crystals. In  $\text{UBi}_2$  (figure 7) the  
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12 small maximum  ${}^{2a}S_{\max}$  observed at  ${}^{2a}T_{\max}$  along the  $a$ -axis certainly arises from the  
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14 magnon/phonon-drag effect. This thesis was confirmed in our measurements of the thermal  
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16 conductivity, which will be published elsewhere, and the thermoelectric power data of two  
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18  $\text{UBi}_2$  samples having a different purity. For the sample with a higher maximum of the thermal  
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20 conductivity, a higher maximum in the thermoelectric power at  ${}^{2a}T_{\max}$  was observed. It is well  
21  
22 known, that the magnon/phonon-drag effect often occurs and is large when crystals are of a  
23  
24 high purity. Similarly for  $\text{USb}_2$  (figure 8), in the  $a$  direction  ${}^a S_{\max}$  at  ${}^a T_{\max}$  should arise from the  
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26 magnon/phonon-drag effect as well (see the left hand inset in figure 8). Obviously, this effect  
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28 should exist also along the  $c$ -axis. Our calculations showed that the temperature  $T^*$  at the  
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30 vicinity of which the maximum value of  $S_g$  is observed, for the  $a$ -axis, is close to the  ${}^{2c}T_{\min}$ .

31 This suggests that the  ${}^{2c}S_{\min}$  also originate from the magnon/phonon-drag effect.

32 Generally, the electron-magnon/phonon scattering (including normal as well as the Umklapp  
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34 processes) will result in a negative magnon/phonon-drag thermoelectric power if the  $\mathbf{q}$  vector  
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36 of the absorbed magnon/phonon crosses an occupied region of the Brillouin zone, while  $S_g$  will  
37  
38 be positive if the  $\mathbf{q}$  vector crosses an unoccupied region of the Brillouin zone [18].

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40 Thus, the charge transport in relation to a particular sheet of the Fermi surfaces may contribute  
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42 with different magnitude and sign to the total thermoelectric power, including both the  
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44 contributions: i.e. the diffusion and magnon/phonon-drag ones. Thus this gives such extremes,  
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46 especially observed in  $\text{USb}_2$  (figure 8) and in  $\text{UBi}_2$  (figure 7) along the  $c$  direction.  
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**Deleted:** As mentioned above, the total thermoelectric power is the sum of diffusion and magnon/phonon-drag contributions. The former part can be dominant and in the  $c$  direction the latter part can be poorly visible. However in the case, when the magnon/phonon-drag is enhanced an extreme in the  $S(T)$  curve is possible to occur.

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2 In summary, the  $\chi(T)$  results are similar for both compounds (figure 1) so that the  
3 magnetic properties are alike, but the transport properties at low temperatures show interesting  
4 differences particularly if measured in the  $c$  direction. These differences can be explained in  
5 terms of the details of the two different Fermi surfaces dictated by the different magnetic  
6 structures found in these dipnictides.  
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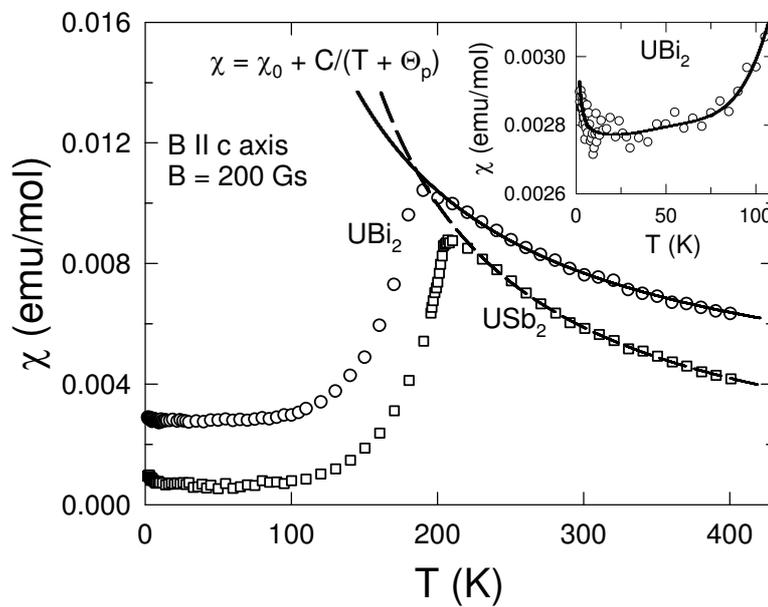
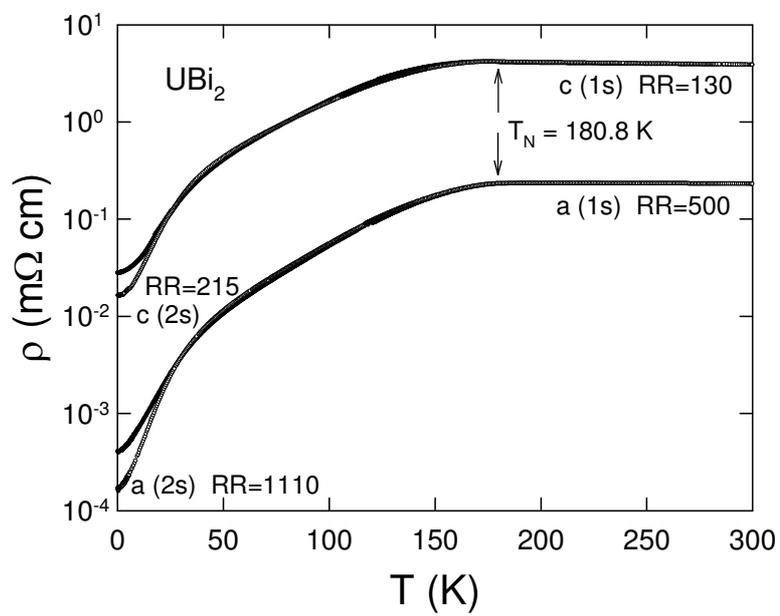


Figure 1



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Figure 2

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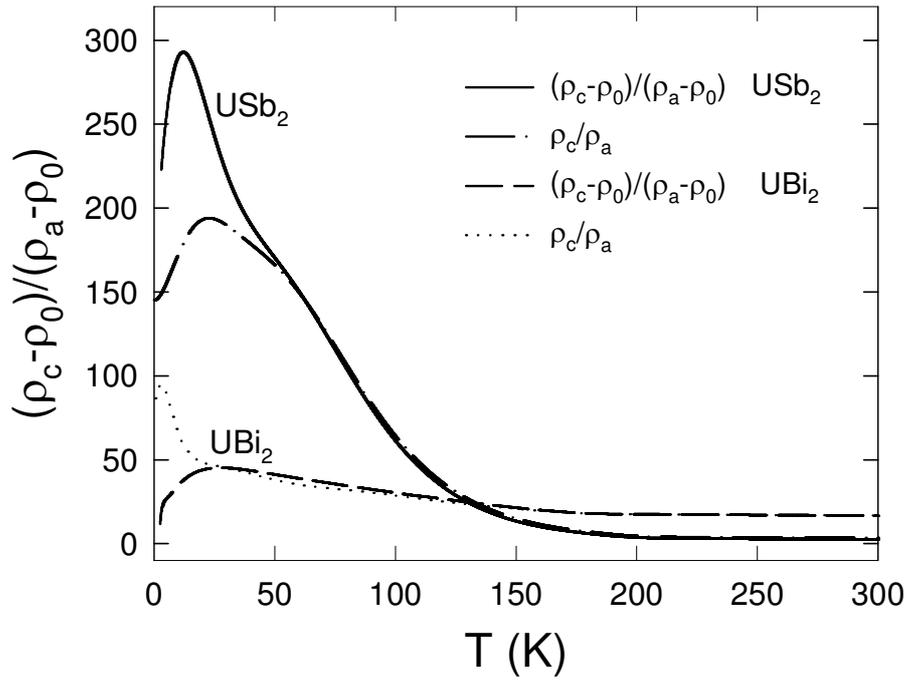


Figure 3

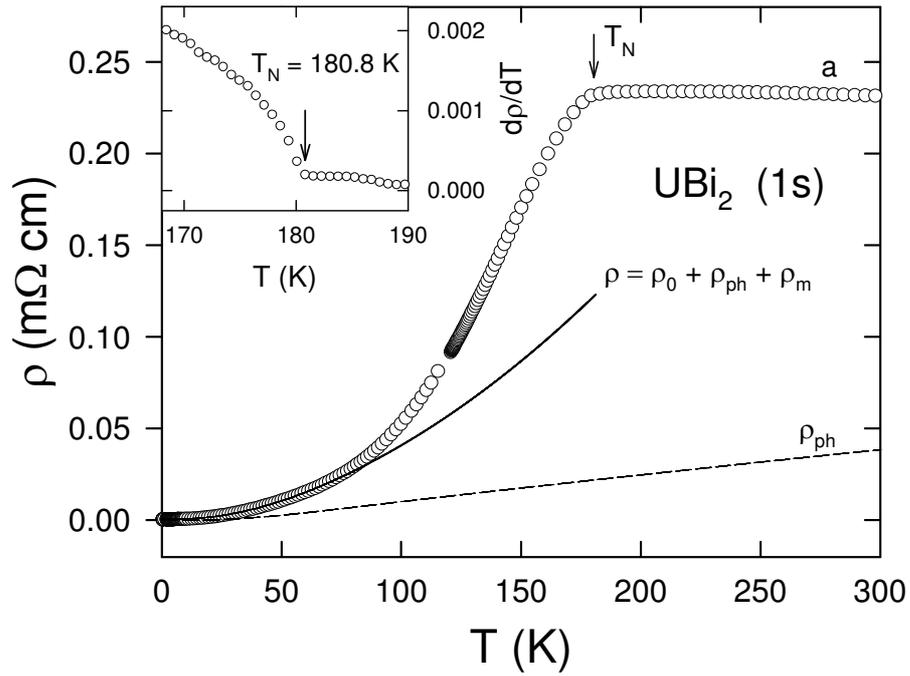


Figure 4

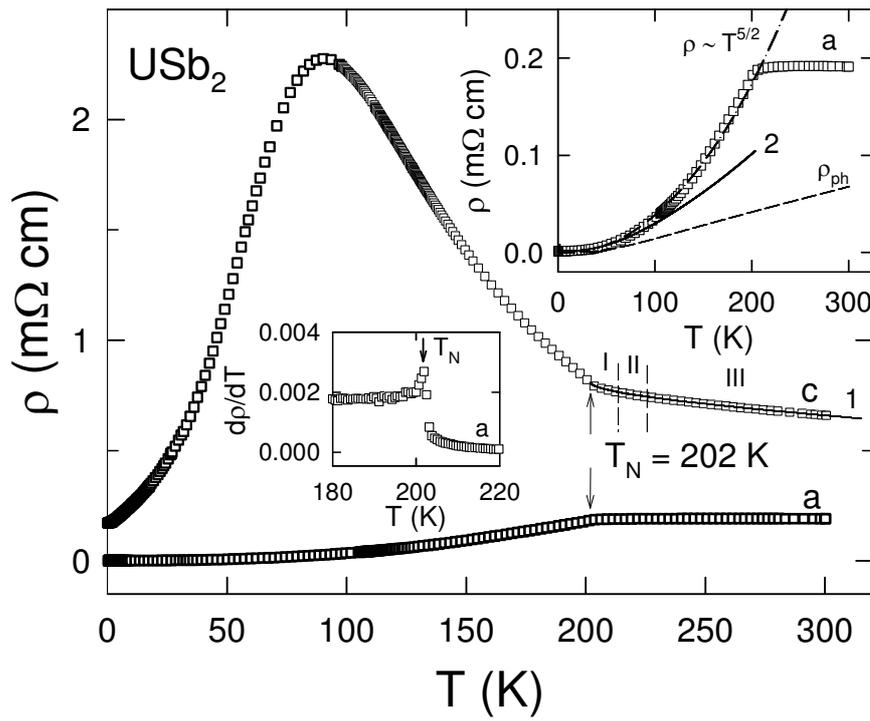


Figure 5

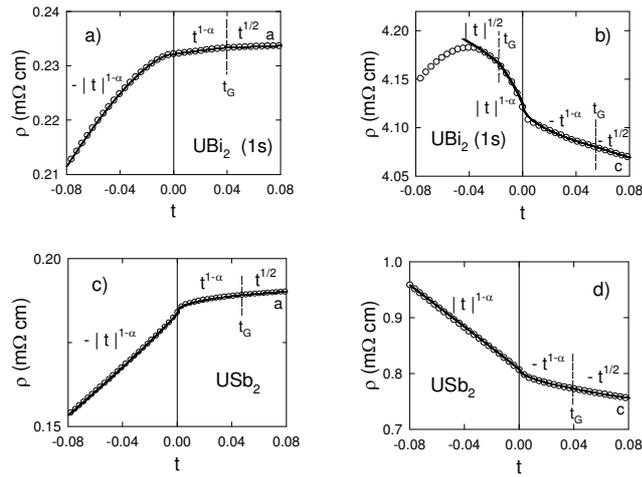


Figure 6

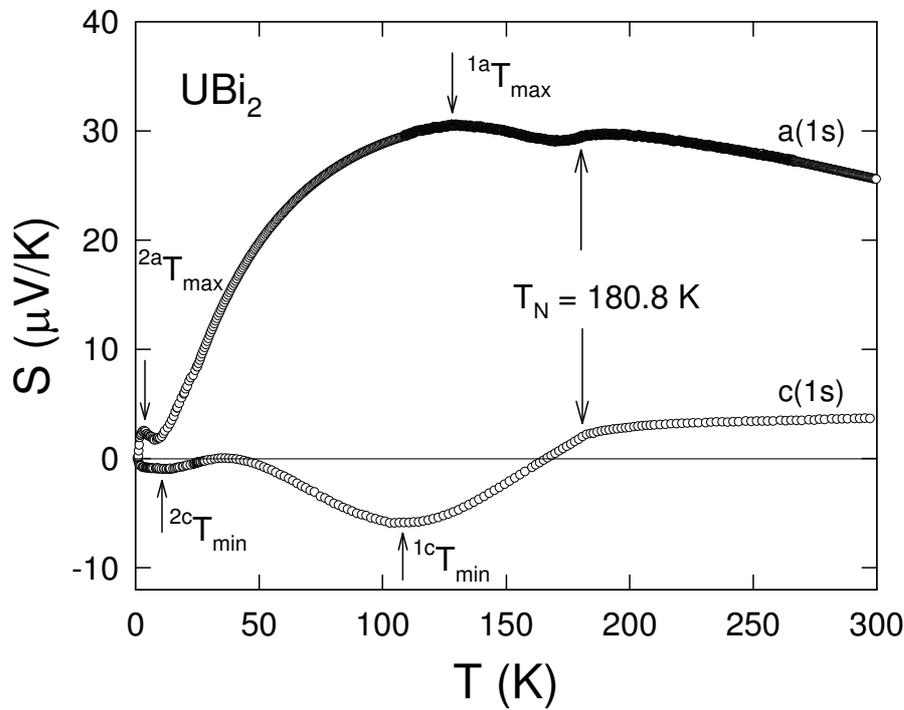
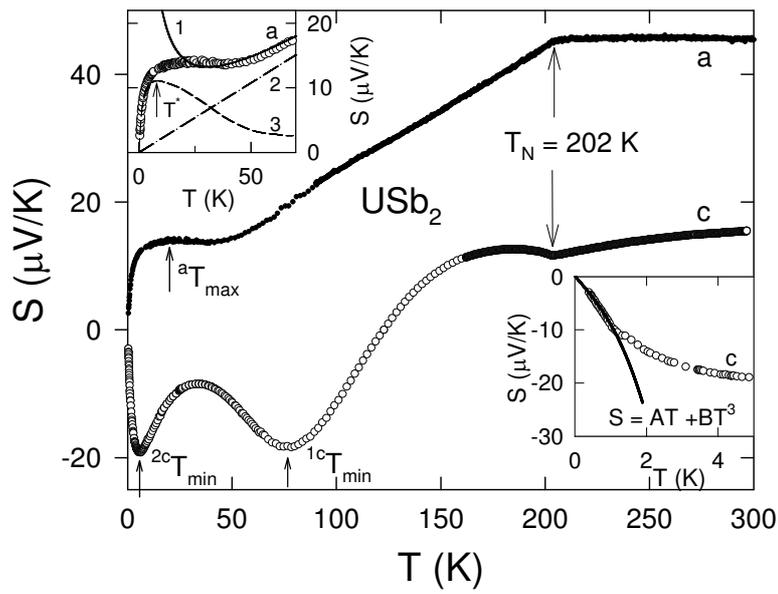


Figure 7



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Figure 8

Figure captions

Figure 1. Magnetic susceptibility  $\chi$  versus temperature of the  $\text{UBi}_2$  (circles) and  $\text{USb}_2$  (squares) single crystals measured in a magnetic field of 200 Gs parallel to the  $c$ -axis. Solid and dashed lines represent  $\chi(T) = \chi_0 + C/(T + \theta_p)$  with  $\chi_0 = 2.7 \times 10^{-3}$  emu/mol, Curie constant  $C = 1.41$  emu K/mol, and  $\theta_p = -17$  K for  $\text{UBi}_2$  and  $\chi_0 = 1.42 \times 10^{-4}$  emu/mol,  $C = 1.38$  emu K/mol, and  $\theta_p = -59$  K for  $\text{USb}_2$ . The inset displays the low temperature  $\chi(T)$  data for  $\text{UBi}_2$  (solid line is a guide for the eyes).

Figure 2. Electrical resistivity versus temperature for two  $\text{UBi}_2$  single crystals with different purity: 1s (RR = 500 and 130) and 2s (RR = 1110 and 215) measured for the  $a$  and  $c$  crystallographic directions, respectively.

Figure 3. Temperature dependence of the anisotropy ratio  $(\rho_c - \rho_0)/(\rho_a - \rho_0)$  for  $\text{UBi}_2$  and  $\text{USb}_2$ .

Figure 4. Electrical resistivity of a  $\text{UBi}_2$  single crystal versus temperature for the 1s sample along the  $a$ -axis. The solid line represents Eq. (1) with:  $\rho_0 = 4.05 \times 10^{-4}$  m $\Omega$  cm,  $a = 3.403 \times 10^{-2}$  m $\Omega$  cm,  $b = 5.73 \times 10^{-6}$  m $\Omega$  cm,  $\Theta =$

267 K and  $\Delta = 3.7$  K. The dashed line represents the  $\rho_{\text{ph}}(T)$  function (the second term in Eq. (1)). The inset shows the temperature dependence of the derivative  $d\rho(T)/dT$  in units of  $\text{m}\Omega \text{ cm K}^{-1}$ .

Figure 5. Electrical resistivity of a  $\text{USb}_2$  single crystal versus temperature along the  $a$  and  $c$  directions. The squares denote the experimental data. The solid line 1 represents Eq. (2) for the  $c$ -axis in the I part of the paramagnetic region with the following parameters:  $\alpha = 0.452$ ,  $A = -0.198 \text{ m}\Omega \text{ cm}$ ,  $B = 5.506 \times 10^{-5} \text{ m}\Omega \text{ cm}$ , and  $C = 0.807 \text{ m}\Omega \text{ cm}$ . These parameters in the II part are:  $\alpha = 0.5$ ,  $A = -0.206 \text{ m}\Omega \text{ cm}$ ,  $B = 0.012 \text{ m}\Omega \text{ cm}$ , and  $C = 0.813 \text{ m}\Omega \text{ cm}$ , while in the III part:  $\alpha = 0.326$ ,  $A = -0.223 \text{ m}\Omega \text{ cm}$ ,  $B = 1.553 \times 10^{-4} \text{ m}\Omega \text{ cm}$ , and  $C = 0.796 \text{ m}\Omega \text{ cm}$ . The inset shows the  $\rho(T)$  data along the  $a$ -axis; the solid line 2 presents Eq. (1) with  $\rho_0 = 1.178 \times 10^{-3} \text{ m}\Omega \text{ cm}$ ,  $a = 8.935 \times 10^{-2} \text{ m}\Omega \text{ cm}$ ,  $b = 2.025 \times 10^{-6} \text{ m}\Omega \text{ cm}$ ,  $\Theta = 386 \text{ K}$ , and  $\Delta = 2.73 \text{ K}$ ; the dashed line represents  $\rho_{\text{ph}}(T)$  (the second term in Eq. (1)); and the dash-dotted line displays the formula:  $\rho(T) = \rho_0 + \rho_{\text{ph}} + aT^{5/2}$ , where  $a = 1.424 \times 10^{-7} \text{ m}\Omega \text{ cm K}^{-5/2}$ . In the inset the derivative  $d\rho(T)/dT$  is given in  $\text{m}\Omega \text{ cm K}^{-1}$  unit.

Figure 6. Electrical resistivity versus reduced temperature ( $t = (T - T_N)/T_N$ ) along the  $a$  and  $c$  directions for a  $\text{UBi}_2$  crystal a), b) and a  $\text{USb}_2$  crystal c), d).

Figure 7. Thermoelectric power of the  $\text{UBi}_2$  single crystal (1s) versus temperature measured along the  $a$  and  $c$  directions. The superscripts 1a and 2a at  $T_{\text{max}}$ , and 1c and 2c at  $T_{\text{min}}$  denote: the first and second maximum along the  $a$ -axis, and the first and second minimum along the  $c$ -axis, respectively.

Figure 8. Thermoelectric power versus temperature for  $\text{USb}_2$  single crystal along the  $a$  and  $c$  directions. Curve 1 in the inset on the left hand side displays the equation:  $S(T) = aT + b/T$  with the fitting parameters  $a$  and  $b$  equal to  $0.216 \mu\text{V}/\text{K}^2$  and  $203 \mu\text{V}$ , respectively, determined along the  $a$ -axis. The straight line 2 and curve 3 present the diffusion and magnon/phonon-drag contributions to the total thermoelectric power, respectively. The inset on the right hand side presents the low temperature part of  $S(T)$  for the  $c$ -axis. The solid line displays the equation:  $S(T) = a'T + b'T^3$ , where  $a' = -7.80 \mu\text{V}/\text{K}^2$  and  $b' = -1.36 \mu\text{V}/\text{K}^4$ .