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IS THE LOW TEMPERATURE THERMOELECTRIC POWER BEHAVIOUR AN EVIDENCE FOR THE MULTI-CHANNEL KONDO EFFECT IN $UAs_{1-x}Se_{1+x}$?[*]

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We have investigated the anisotropy and off-stoichiometry effects on the thermoelectric power $S(T)$ of uranium dipnictides — $UP_{1.7}As_{0.3}$ and UAs_2 and arsenoselenide — $UAs_{1-x}Se_{1+x}$. These two groups of compounds have tetragonal PbFCl-type structure and order at low temperatures anti- or ferromagnetically, respectively. The $S(T)$ curves are strongly anisotropic for all the examined systems. The a-axis thermopower of the ferromagnetic $UAs_{1-x}Se_{1+x}$ system shows a spectacular dependence of the shape of $S(T)$ curve on x. This behaviour is discussed in the frame of the multi-channel Kondo model.

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UAsSe is an anisotropic, uniaxial ferromagnet ($T_C \sim 110$ K) [1] with a narrow $5f$-electron band at the Fermi energy [2]. Its resistivity, $\rho(T)$, shows an unusual Kondo-like behaviour scaled by the Kondo temperature $T_K = 50$ K [1]. This behaviour was ascribed to a Two-Level System (TLS) Kondo mechanism [3–5]. The original TLS Kondo model can not explain such a high T_K value (see [6,7]). However, as realised recently, electron-hole symmetry breaking, present in all realistic band-structure-based density of states, can account for an increase in T_K substantially. This becomes very promising to resolve the long standing problem of the high T_K values [7,8] observed in certain systems. Since the thermoelectric power also depends

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on the electron–hole asymmetry at the Fermi level, it should be an excellent tool for the TLS Kondo system examination. This motivates us to perform study on an effect of off-stoichiometry ($x \neq 0$) on the thermoelectric power of the $UAs_{1-x}Se_{1+x}$ system as well as the study of $S(T)$ for the uranium dipnictides $UP_{1.7}As_{0.3}$ and UAs_2 as references. The resistivity of the latter compounds show no sign of the Kondo-like behaviour [5, 9].

All the examined systems crystallise in the tetragonal, PbFCl-type structure with the magnetic moments arranged parallel to the c-axis. Note that in the case of the dipnictides, which at low temperature order antiferromagnetically, the magnetic unit cell is doubled along the c-axis (see Ref. [10]). This should be taken into account when comparing their properties to those of arsenoselenides.

We have determined the $S(T)$ and $\rho(T)$ dependences of the relevant single crystals which were grown by a chemical vapour transport method [5]. Composition of the $UAs_{1-x}Se_{1+x}$ ferromagnet was estimated from the $x(T_C)$ dependence given in Ref. [4]. Fig. 1 presents the $S(T)$ behaviour for $UP_{1.7}As_{0.3}$. Its specific feature is a broad peak in ${}^cS(T)$ growing below about 150 K and a small negative peak in ${}^aS(T)$ reaching a minimum at 20 K. There is also a slope change in ${}^aS(T)$ at $T_N = 221$ K. The behaviour of $S(T)$ for $UP_{1.7}As_{0.3}$ differs slightly from that for UAs_2. The single peak of ${}^cS(T)$ seen for the former compound corresponds to two peaks in UAs_2, while ${}^aS(T)$ tends roughly linearly to zero with lowering temperature, with-

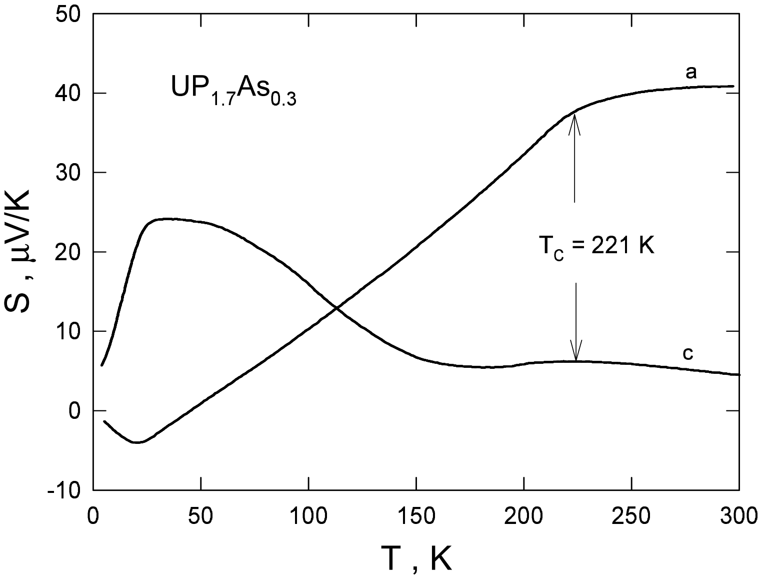

Fig. 1. Thermoelectric power anisotropy of $UP_{1.7}As_{0.3}$.

out sign change. There is also observed a distinct peak in d^aS/dT for UAs_2 at $T_N = 273\,K$. However, such a behaviour is rather typical for a ferromagnet [11]. We just observe it though UAs_2 is antiferromagnet because its magnetic unit cell dimension along the a-axis remains the same as the chemical one. $S(300\,K)$ reaches the values 40.7 and $46.4\mu V/K$ along the a-axis and 4.5 and $3.6\mu V/K$ along the c-axis for $UP_{1.7}As_{0.3}$ and UAs_2, respectively. There is rather weak effect of different pnicogen anion substitution on the thermoelectric power behaviour in uranium dipnictides.

The inset in Fig. 2 displays the $S(T)$ anisotropy for $UAs_{0.979}Se_{1.021}$. Furthermore, the inset shows that there is an anomalous temperature behaviour in the thermoelectric power as compared to that observed for ferromagnetic metal. Below $T_C = 108\,K$ this compound exhibits a broad peak in ${}^aS(T)$ and two peaks for ${}^cS(T)$. However, a similar low-temperature ${}^cS(T)$ behaviour for $UP_{1.7}As_{0.3}$ and UAs_2 shows that ${}^cS(T)$ peaks in compounds of the PbFCl family can be of non-Kondo origin as well. Therefore, we focus our attention on ${}^aS(T)$ for the $UAs_{1-x}Se_{1+x}$ system. After all it is worth stressing that the $S(T)$ curve was found to be strongly anisotropic for all the examined systems.

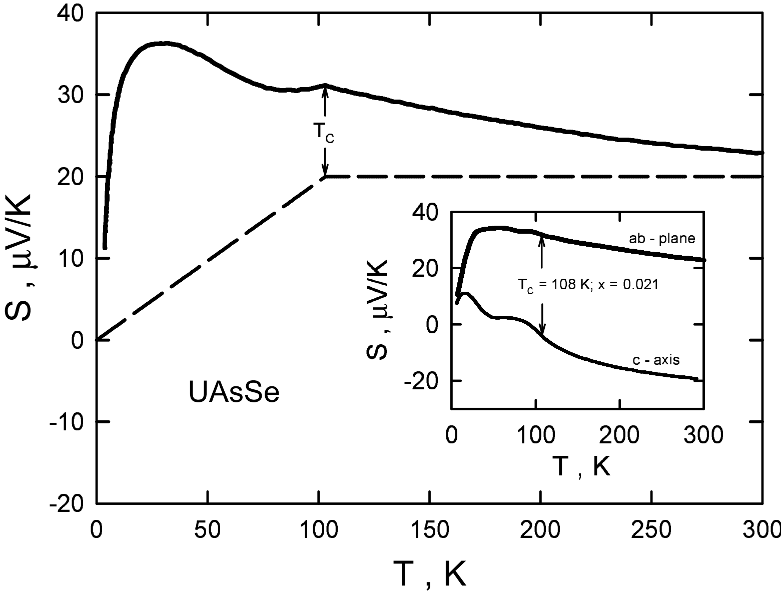

Fig. 2. The calculated $S(T)$ dependence for uranium arsenoselenide (solid line) being a weighted sum of the Kondo and the ferromagnetic contribution (dashed line) as described in the text. Inset: The anisotropy of $S(T)$ for a $UAs_{0.979}Se_{1.021}$ single crystal. Note the similarity between ${}^aS(T)$ and the calculated $S(T)$.

Fig. 2 displays the $S(T)$ calculated as the weighted sum of a ferromagnetic metal $S_{\mathrm{F}}(T)$ and a Kondo $S_{\mathrm{K}}(T)$ contributions

$$S = S_{\mathrm{F}}\left(\frac{\rho_{\mathrm{F}}}{\rho}\right) + S_{\mathrm{K}}\left(\frac{\rho_{\mathrm{K}}}{\rho}\right) .$$

Here $\rho = \rho_{\mathrm{K}} + \rho_{\mathrm{F}}$, where $\rho_{\mathrm{K}}(T)$ is the Kondo resistivity and $\rho_{\mathrm{F}}(T)$ the resistivity of a ferromagnetic metal. The a-axis resistivity components were those determined for UAsSe in Ref. [1]. The $S_{\mathrm{F}}(T)$ was approximated by the temperature dependence shown by broken line in Fig. 2. In turn the $S_{\mathrm{K}}(T)$ was approximated with the dependence given in Ref. [12] for the T_{K}/D_E ratio of 0.0053 ($T_{\mathrm{K}} = 50$ K and D_E is the conduction band width). $S_{\mathrm{K}}(T)$ of Ref. [12] was multiplied by 1/3 to get the calculated $S(T)$ being close to that of $\mathrm{UAs_{0.979}Se_{1.021}}$ at 14 K. At this temperature the $S_{\mathrm{F}}(\rho_{\mathrm{F}}/\rho)$ contribution is negligible. As shown by the inset in Fig. 2, the calculated $S(T)$ dependence is similar to that measured for the a-axis. Therefore, we may think that the anomalous $S(T)$ behaviour observed for $\mathrm{UAs}_{1-x}\mathrm{Se}_{1+x}$ system is due to the Kondo components. Fig. 3 shows that the positive peak-like component observed for $x = 0.021$ weakens when x either decreases or increases. In the latter case, the Kondo component transforms even to the negative peak seen for $x = 0.038$. Such behaviour contrasts with the behaviour of the Kondo resistivity, which decreases with decreasing x rather monotonously.

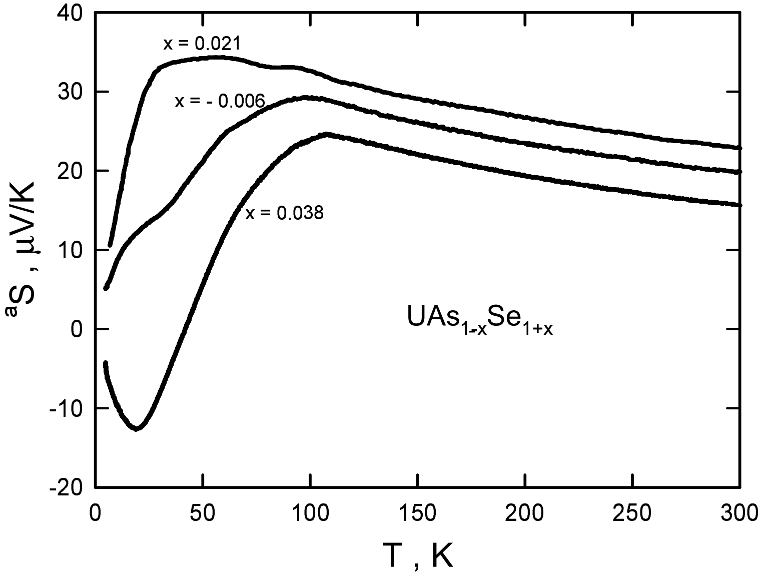

Fig. 3. The a-axis thermoelectric power for various $\mathrm{UAs}_{1-x}\mathrm{Se}_{1+x}$ crystals with x (T_{C}) equal to 0.038 (104 K), 0.021 (108 K) and -0.006 (114 K)

Following the prediction [3, 13] of great similarities of the TLS Kondo system behaviour to that of magnetic Kondo one, we use the theory on n-channel Kondo effects on the thermoelectric power for Ce^{3+} impurities [14] as a guide for our research. This theory predicts that the peak-like thermoelectric power should be positive at low T for $n = 1$, strongly negative for $n = 2$ and weakly negative for $n = 3$. It is expected that TLS centres in the $UAs_{1-x}Se_{1+x}$ system are produced by anion disorder [4] consisting of small part of As lattice positions occupied by Se and *vice versa* [5]. The disorder leads to an anomalous large atomic displacement factor D, which decreases monotonously with decreasing x in the same manner as the Kondo resistivity does [4]. We think that the decrease of D may reflect either the decrease of the TLS centres' density or a change of the two levels splitting energy and hence the change in the n number. While all the TLS centres give positive contribution to the Kondo resistivity, both the sign and size of contributions to the thermoelectric power depend on the n number. We conclude that the observed dependence of the amplitude of the Kondo resistivity on x being different from that of the thermoelectric power, may be related to the multi-channel Kondo effect.

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