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NON-MAGNETIC KONDO-LIKE SCATTERING IN UPS AND UAsSe FERROMAGNETS*

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UPS is the second uranium-based pnictochalcogenide where electron-assisted tunneling presumably is realized. Although the unusual properties of UPS and UAsSe originate from the structural disorder, a clear-cut uniformity in their electrical resistivity is observed. The additional resistivity due to non-magnetic Kondo-like scattering was found as large as 140 $\mu\Omega$ cm at low temperatures. The estimated concentration of the fast TLS centers varies between 10^{19} and 10^{20}cm^{-3}.

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Non-magnetic interactions between conduction electrons and two-level system (TLS) may cause behaviors analogous to the ordinary (magnetic) Kondo effect (see [1] for a review). Recently, it has been shown that uncommon transport properties of the structurally disordered UAsSe ferromagnet may be consistently interpreted in terms of the TLS Kondo model. This concerns, *e.g.*, the disorder-dependent low-temperature upturn in the electrical resistance, $R(T)$ [2], as well as changes in the thermoelectric power introduced by tiny variations of Se excess [3]. Similar $R(T)$ behavior has been observed in the diamagnetic homologue ThAsSe [4]. Since the properties of actinide arsenoselenides are caused by interactions of conduction electrons with the fast TLS centers situated in the anionic sublattice, similar behaviors are expected in UPS crystallizing in the same PbFCl-type structure. In the very first experiment on UPS ($T_{\rm C}$ = 118 K), no $R(T)$ anomaly in the ferromagnetic state was observed [5]. However, recent studies on other UPS

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single crystals with slightly reduced T_C values revealed the low-T upturn of a different size [6]. Similarly to UAsSe, the $R(T)$ dependencies for all the investigated UPS specimens are remarkably alike in the paramagnetic state. In the following, uniformity in the resistivity of both uranium pnictochalcogenides is discussed.

Fig. 1 shows the *ab*-plane electrical resistance for three UPS single crystals with different amount of disorder. The results are normalized to the value at $T_{\max}$, where $T_{\max}$ is the temperature at which a maximum of the resistance, $R_{\max}$, occurs. For comparison, the most different $R(T)$ data of UAsSe are presented. The size of the low-T anomaly monotonically increases with Se excess [7]. In both systems, an increase of the upturn is accompanied by a slight reduction of the Curie temperature. Although this is a secondary effect in our TLS-based approach, it allows us, in a simply way, to characterize the samples investigated. To date, no UAsSe sample without the low-T upturn was obtained.

As it becomes evident from Fig. 1, a variation of the low-T anomaly in UPS compares very well with the UAsSe data. Obviously, the same type of interactions, presumably electron-assisted tunneling, are involved in the unusual scattering in both systems. Thus, to some extent, the free-of-anomaly UPS sample can be considered as a reference one to UAsSe,

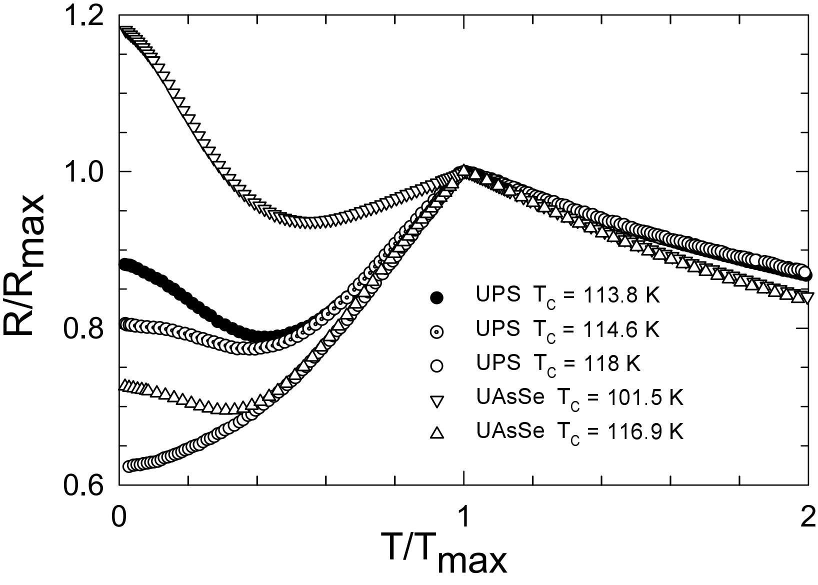

Fig. 1. Normalized *ab*-plane resistance for UPS and UAsSe ferromagnets. Note that $T_{\max}$ is very close to T_C, which was estimated from the maximum of the first derivative of the resistance. The data for the UPS single crystal with $T_C = 118\,\mathrm{K}$ have been taken from [5].

too. Above T_{max}, there are no significant differences between samples of the same system. This fact unambiguously points out that the high-T $R(T)$ dependence is independent of the amount of disorder. We speculate that those parameters having an effect on the anomalous curvature, must have very similar values. Some distinctions between UPS and UAsSe systems might originate in their intrinsic characteristics, being independent of the tiny variations in the anionic composition.

In a previous experiment on $R(T)$ for the UAsSe single crystal with $T_{\mathrm{C}} = 102\,\mathrm{K}$, a negative temperature coefficient of the resistance on either side of T_{max} has been attributed to the same Kondo-like component described by the Arai formula with $T_{\mathrm{K}} = 40\,\mathrm{K}$ [8]. However, strongly sample-dependent $R(T)$ at $T < T_{\mathrm{max}}$ brings this interpretation into question. If a negative rise of the resistivity at $T > T_{\mathrm{max}}$ is also related to TLS scattering, rather large T_{K} values are expected. A closer inspection of theoretical results, however, raises questions as whether the TLS scattering is observable at intermediate temperatures at all [1]. For a detailed discussion of the anomalous high-temperature $R(T)$ dependence of the U-based pnictochalcogenides we refer to a forthcoming publication [9].

In the remainder discussion we briefly address observations which could give some quantitative information on the fast TLS centers in UPS and UAsSe. To this aim we subtract the normalized $R(T)$ data of the free-of-anomaly UPS sample from all the others presented in Fig. 1. Note that this procedure ignores possible differences in the phonon and magnetic contributions to the resistivity between various specimens. Further on, we assume the same value of the room-temperature resistivity equal to $250\mu\Omega\,\mathrm{cm}$ for all the samples under discussion. Consequences of the above-mentioned approximations are discussed below. Nevertheless, the results presented in Fig. 2 in absolute units give a rough estimate of the additional resistivity due to the interaction of the TLS with the conduction electrons, $\Delta\rho(T)$ in the UPS and UAsSe ferromagnets.

The $\Delta\rho(T)$ dependencies presented in Fig. 2 appear to resemble the single-ion Kondo contribution to the electrical resistivity characteristic for dilute alloy systems, including the tendency of $\Delta\rho(T)$ to saturate as $T \to 0$. The temperature, below which the $\Delta\rho(T)$ data become finite, is found to increase with the extrapolated value of $\Delta\rho(T \to 0)$. One might be inclined to relate this with an increase of the Kondo temperature. However, a reasonable estimate of T_{K} requires, *e.g.*, acoustic measurements that determine the coupling strength between the TLS and conduction electrons. Note that $\Delta\rho(T)$ of the UAsSe single crystal with $T_{\mathrm{C}} = 101.5\,\mathrm{K}$ does not change smoothly at $T = T_{\mathrm{max}}$ because of differences between the magnetic components of this particular sample and the free-of-anomaly UPS one ($T_{\mathrm{C}} = 118\,\mathrm{K}$), as highlighted by their significantly different T_{C} values.

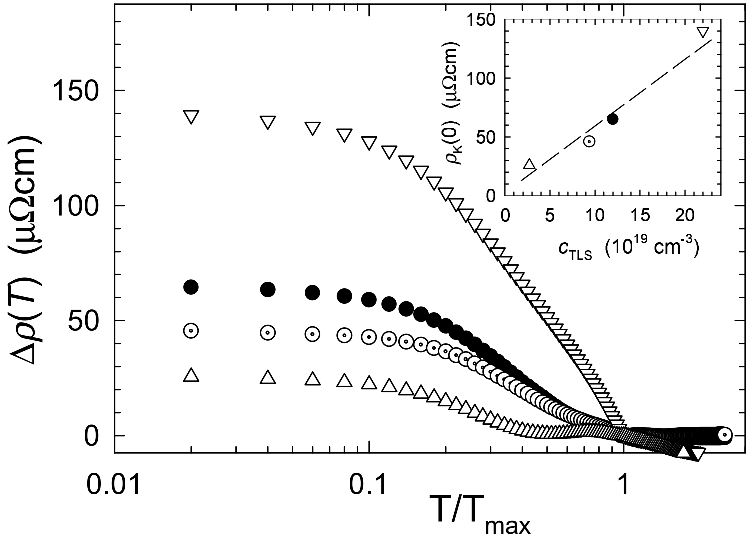

Fig. 2. The additional resistivity due to the TLS for various UPS and UAsSe single crystals. For detailed descriptions of the symbols see Fig. 1. Inset: Relation between $\Delta\rho(T)$ and the concentration of the fast TLS centers in uranium pnictochalcogenides. The dashed line is a guide to the eye only.

In the following we discuss the concentration of the fast TLS centers, $c_{\rm TLS}$, in UPS and UAsSe samples that can be estimated from the simplified formula [11]

$$c_{\rm TLS} = 8.75 \times 10^{19}\, \frac{\rho_{\rm K}(0)}{E_{\rm F}} \frac{m_0}{m^*}\, (n_{22})^2\ [{\rm cm}^{-3}]\,,$$

where $\rho_{\rm K}(0)$ is the TLS Kondo resistivity as $T \to 0$ in $\mu\Omega$cm, $E_{\rm F}$ the Fermi energy in eV, m^*/m_0 the effective mass ratio, and n_{22} denotes the carrier number per 10^{-22} cm^3. From the Hall-effect measurement on the UAsSe sample with $T_{\rm C} = 102$ K [6] , the carrier density amounts to $n_{22} \sim 0.7$ at 300 K. Thus, the Fermi energy appears to be relatively low, $E_{\rm F} \sim 1$ eV. Since U-based pnictochalcogenides display a room-temperature Seebeck coefficient of the order of 20 μV/K in the *ab*-plane [3], the same values of $n_{22} \sim 0.7$ and $E_{\rm F} \sim 1$ eV are assumed for the UPS specimens and for the UAsSe single crystal with $T_{\rm C} = 116.9$ K as well. A specific-heat effective mass $m^* \sim 19.3$, 29.7 and 40.6 was taken for UPS and UAsSe with $T_{\rm C} = 101.5$ K and 116.9 K, respectively [6,10]. From the low-temperature $\Delta\rho(T)$ data we can extrapolate the $\rho_{\rm K}(0) = \Delta\rho(T \to 0)$ values (*cf.* Fig. 2).

The results of our calculation, plotted as $\rho_{\rm K}(0)$ vs| $c_{\rm TLS}$, are summarized in the inset of Fig. 2. The $c_{\rm TLS}$ values for UPS and UAsSe are close to the typical concentration of the TLS in amorphous materials $\sim 10^{18} \div 10^{19}$cm^{-3}.

Two facts may lead to inaccuracies of our estimates, *i.e.* inaccuracies in both the room-temperature resistivity via $\rho_{\rm K}(0)$ and the density of carriers. While with $\rho(300~{\rm K}) = 250~\mu\Omega$cm one slightly underestimates $c_{\rm TLS}$ only in one case, *i.e.* for the UAsSe sample with $T_{\rm C} = 101.5\,{\rm K}$ only, a slightly inappropriate value of $n_{22}(0.7)$ would significantly influence all the results. In fact, considering the experimental value of $\rho(300{\rm K}) = 275~\mu\Omega\,$cm for the UAsSe sample ($T_{\rm C} = 101.5\,{\rm K}$) one gets a small increase of $c_{\rm TLS}$ of less than 20%. For all the other samples, the deviations from the $250\mu\Omega$cm value are still within the margin of error. Since $c_{\rm TLS} \sim (n_{22})^2$, a wrong estimation of n_{22} is much more important. The following observations provide evidence for the $n_{22} \sim 0.7$ being a somewhat overestimated value. Firstly, the carrier density obtained from the magneto–optical Kerr effect is less than $n_{22} \sim 0.45$ [12]. Secondly, according to [8] one can expect a significant reduction of the carrier number with lowering temperature. Thirdly, presumably n_{22} decreases with decreasing concentration of the chalcogenide atoms, which have one p electron more than the nearest-neighbor pnictogens. Thus, the correct calculations require a precise experimental input. However, the re-estimated $c_{\rm TLS}$ values are expected to be smaller than those presented here and, hence, better agreement with the typical concentration of the TLS in amorphous materials should be achieved.

For crystals with $T_{\rm C} = 102\,{\rm K}$, the chemical composition $\rm UAs_{0.94}Se_{1.06}$ has been estimated, *i.e.* $9\times10^{20}{\rm cm}^{-3}$ of Se excess [7]. This could mean that less than one fourth of the excess chalcogenide atoms create the fast TLS centers. An overwhelming majority of the excess atoms behaves as typical, non-magnetic impurities and will enlarge a static disorder only. Indeed, this can explain the slightly larger value $\rho(300\,{\rm K}) = 275~\mu\Omega\,$cm for the UAsSe sample with $T_{\rm C} = 101.5\,{\rm K}$.

In summary, uniformity in the low-T electrical resistivity of UAsSe and UPS was found. This allowed us to estimate the additional resistivity due to the interaction of the TLS with conduction electrons. Presumably, less than 25% of the excess chalcogen atoms are involved in the TLS scattering.

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