A non-magnetic Kondo effect in UAsSe ferromagnet?

T. Cichorek\textsuperscript{a,b,*}, Z. Henkie\textsuperscript{a}, P. Gegenwart\textsuperscript{b}, M. Lang\textsuperscript{b}, A. Wojakowski\textsuperscript{a}, M. Dischner\textsuperscript{b}, F. Steglich\textsuperscript{b}

\textsuperscript{a}Institute of Low Temperature and Structure Research, Polish Academy of Sciences, Wroclaw, Poland
\textsuperscript{b}Max-Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany

Abstract

The upturn in the resistivity upon cooling far below the ferromagnetic transition in the metallic system UAsSe is rather unusual. Measurements on several single crystals show that its magnitude monotonically increases with decreasing Curie temperature ($T_C$ between 101.5 and 117 K). On the other hand, the resistivity in the paramagnetic state shows a weak sample dependence only. Distinct differences for both the Sommerfeld coefficient, $\gamma$ (20–40 mJ mol\(^{-1}\) K\(^{-2}\)), and the Debye temperature, $\Theta_D$ (252–310 K), have been observed for crystals with different $T_C$ values. $\Theta_D$ increases only for specimens with $T_C > 111$ K, while $\gamma = \gamma(T_C)$ shows a minimum close to $T_C = 111$ K. No effect in magnetic fields up to 14 T has been observed. The observations suggest a non-magnetic Kondo effect arising from two-level systems.

The UAsSe ferromagnet crystallizes in the tetragonal PbFCl-type structure which can be described as consisting of layers stacked along the c direction (easy axis) with the sequence: As–U–Se–Se–U–As. Neutron-diffraction experiments showed that about 6% of the As sites can be occupied by Se ions and vice versa [1]. In addition, X-ray diffraction measurements reveal very large anisotropic displacement factors for all atoms in UAsSe [2]. UAsSe shows an anomalous upturn of the resistivity in the magnetically ordered state. Recently, distinct differences in the low-$T$ specific heat have been found for off- and nearly-stoichiometric UAsSe single crystals with $T_C = 105$ K, $\gamma = 27.4$ mJ mol\(^{-1}\) K\(^{-2}\), $\Theta_D = 260$ K and $T_C = 117$ K, $\gamma = 40.6$ mJ mol\(^{-1}\) K\(^{-2}\), $\Theta_D = 307$ K, respectively [3].

Here we present the preliminary results of a systematic investigation of the low-$T$ specific heat, $C$, and the electrical resistivity, $R$, of UAsSe. Measurements have been performed on several single crystals obtained from three synthesis processes that differ in their substrate composition. Fig. 1 shows the $ab$-plane resistivity normalized to its value at room temperature, $R(T)/R(300$ K), for the single crystals with different $T_C$ values. The low-$T$ upturn (quantified, e.g., by the RR = $R(4.2$ K)/$R_{\text{min}}$ ratio, where $R_{\text{min}}$ is a minimum of the resistivity in the ferromagnetic state) monotonically increases with decreasing $T_C$ (cf. inset Fig. 1) while the resistivity in the paramagnetic state is only weakly sample dependent. A ratio $R(T_C)/R(300$ K) $\approx 1.3$ seems to be characteristic for the UAsSe system. The increase of the low-$T$ resistivity upon cooling indicates the action of an additional scattering mechanism in the ferromagnetically ordered state. The very small magneto-resistivity at 4.2 K (0.34% T\(^{-1}\)) in fields up to 14 T indicates its non-magnetic origin.

In Fig. 2, we show specific-heat data as $C/T$ vs. $T^2$ between 1.5 and 6 K for three UAsSe single crystals obtained from different synthesis processes. The $T_C$ temperatures of these crystals are representative of each growing runs. Far below $T_C$ the magnon contribution to the specific heat, $C_m \sim T^{3/2}$, is negligible. The fact that the data are field-independent up to 14 T indicates that also the variations in the low-$T$ specific heat have a non-magnetic origin.

The decrease of $T_C$ from 117 to 111 K is accompanied by a decrease of the Debye temperature $\Theta_D$ from 310 to

*Corresponding author. Max-Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 48, D-01187 Dresden, Germany. Fax: +49-351-871-1612.
E-mail address: cichorek@cpfs.mpg.de (T. Cichorek).
252 K, respectively. However, upon going to specimens with $T_c < 111$ K, the Debye temperature remains unchanged within our experimental uncertainty. In addition, the results of a crystallochemical analysis [2] yield a significant change of the unit-cell volume only for specimens with higher $T_c$ values. This suggests that the hybridization between 5f and conduction-electron states in UAsSe is reinforced only for the specimens with $T_c \geq 111$ K while it remains unchanged for those samples with $T_c$ below 111 K.

The inset of Fig. 2 presents the dependence of the Sommerfeld coefficient, $\gamma = C/T$, on $T_c$. $\gamma(T_c)$ shows a minimum close to $T_c = 111$ K. Naively, one would expect a reduction of $\gamma$ upon going to samples with reduced lattice parameters, i.e., those with enhanced hybridization strength. Therefore, the $\gamma$ enhancement for the specimens with $T_c$ in excess of 111 K remains unclear at present. Further on, the increase of $\gamma$ for $T_c$ decreasing below 111 K, i.e., for the specimens from a regime where the hybridization does not change, suggests that another mechanism contributes to the low-$T$ specific heat of UAsSe.

Various energetically almost degenerate positional configurations in disordered system like, e.g., in the semiconducting glass As$_2$Se$_3$ could be the source for a quantum-mechanical tunneling of atoms. In general, these tunneling processes are rare. A much more efficient mechanism is electron-assisted tunneling, in which the scattering of a conduction electron off the tunneling atom induces the transition [4]. Such dynamical scattering centers are considered in the two-level-system (TLS) scenario [5] as a source of a “non-magnetic Kondo effect”.

Taking into account the disorder in the anionic sublattice, we propose the TLS Kondo model as a possible explanation for the unusual low-$T$ physical properties in UAsSe. According to Ref. [5], the additional resistivity due to the TLS can be written as

$$\Delta R(T) \approx \frac{m}{ne^2} \gamma_{\text{TLS}} \Delta \tau^{-1},$$

where $m$ is the electron band mass, $n$ the electronic density, $e$ the electron charge, $\gamma_{\text{TLS}}$ the concentration of the TLS scattering centers and $\Delta \tau^{-1}$ the scattering strength per impurity. Assuming that $m$, $n$ and $\Delta \tau^{-1}$ do not change very much for the various UAsSe crystals then the decrease of $RR = R(4.2 \text{K})/R_{\text{min}}$ accompanied by the $T_c$ enhancement could be related to a decrease of $\gamma_{\text{TLS}}$. Since the tunneling systems are expected to contribute also to the low-$T$ $\gamma$, a reduction of $\gamma_{\text{TLS}}$ might explain the observed decrease of $\gamma$ upon increasing $T_c$ up to 111 K. As mentioned above, the subsequent rise of $\gamma(T_c)$ for $T_c > 111$ K signals another contribution which has yet to be identified.

More detailed studies concerning this subject, especially band-structure calculations as well as investigations of the non-magnetic homologue ThAsSe, are in preparation.

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References