A non-magnetic Kondo effect in UAsSe ferromagnet?

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A non-magnetic Kondo effect in UAsSe ferromagnet?

T. Cichorek[a,b,*], Z. Henkie[a], P. Gegenwart[b], M. Lang[b], A. Wojakowski[a], M. Dischner[b], F. Steglich[b]

[a]*Institute of Low Temperature and Structure Research, Polish Academy of Sciences, Wroclaw, Poland*
[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, γ (20–40 mJ mol^{-1} K^{-2}), and the Debye temperature, Θ_D (252–310 K), have been observed for crystals with different T_C values. Θ_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 compositions. Fig. 1 shows the *ab*-plane resistivity normalized to its value at room temperature, $R(T)/R(300\,\mathrm{K})$, for the single crystals with different T_C values. The low-T upturn (quantified, e.g., by the $\mathrm{RR} = R(4.2\,\mathrm{K})/R_{min}$ ratio, where R_{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 $R(T_C)/R(300\,\mathrm{K}) \approx 1.3$ 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 magnetoresistivity 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 Θ_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).

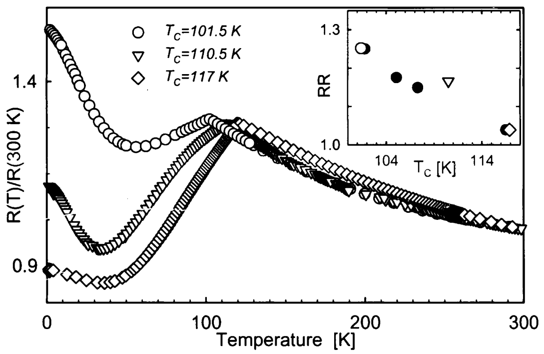

Fig. 1. The temperature dependence of the *ab*-plane electrical resistivity normalized to its room-temperature value for three UAsSe single crystals. Inset: the RR = $R(4.2\,\mathrm{K})/R_{\mathrm{min}}$ ratio as a function of the Curie temperature for several single crystals.

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 $T_C \geqslant 111$ 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. $\gamma = \gamma(T_C)$ shows a minimum close to $T_C = 111$ K. Naively, one would expect a reduction of γ upon going to samples with reduced lattice parameters, i.e., those with enhanced hybridization strength. Therefore, the γ enhancement for the specimens with T_C in excess of 111 K remains unclear at present. Further on, the increase of γ 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_2Se_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

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

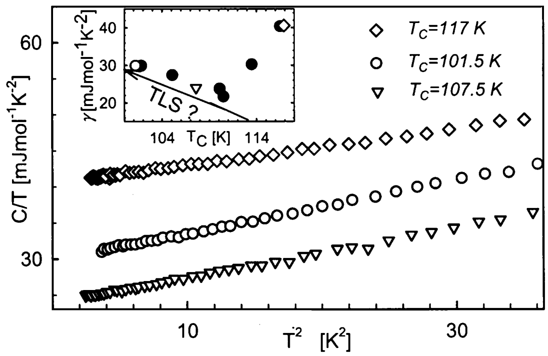

Fig. 2. The low-T specific heat as C/T vs. T for selected UAsSe single crystals. Note that the different slope for the crystal with $T_C = 117$ K which corresponds to a different Debye temperature. Inset: the Sommerfeld coefficient as a function of T_C for several UAsSe single crystals. The solid arrow indicates the potential change of γ due to the TLS.

where m is the electron band mass, n the electronic density, e the electron charge, c_{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 = $R(4.2\,\mathrm{K})/R_{\mathrm{min}}$ accompanied by the T_C enhancement could be related to a decrease of c_{TLS}. Since the tunneling systems are expected to contribute also to the low-T γ, a reduction of c_{TLS} might explain the observed decrease of γ 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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