

ThAsSe Diamagnet: Evidence for a Kondo Effect Derived from Two-Level Systems

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Within the broad spectrum of properties of amorphous and disordered solids the phenomena related to quantum tunneling take a special position. This is due to the fact that these processes may determine the low-energy physics of matter with the same kind of disorder. The most popular realization of tunneling center is believed to be an atom that tunnels between two minima of the double-well potential (Fig. 1). An ensemble of them, called two-level systems (TLS), is typically characterized by a broad distribution of such parameters like, e.g., the

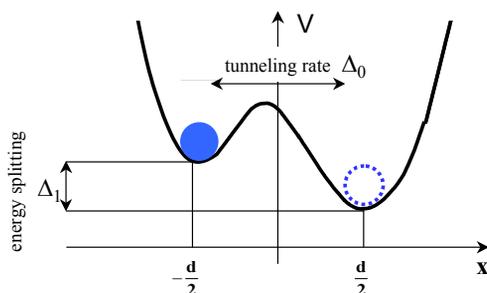


Fig. 1: Schematic representation of an atomic two-level system.

energy splitting or/and the tunneling rate. One of the facets of the TLS is the appearance of an exotic Kondo effect; the Hamiltonian of the TLS interacting with a degenerate Fermi gas may be mapped to the Hamiltonian of the spin-1/2 Kondo problem [1]. In the case of the TLS, however, a logarithmic singularity in the electrical resistance, $R(T)$, has an origin in non-magnetic interaction of the itinerant electrons with the tunneling centers. As a consequence, due to the electron-assisted tunneling, a moveable particle cannot be localized in one of the minima. This resembles the quenching of a magnetic-impurity moment via the conduction electrons in the spin Kondo problem. At strong-coupling fixed point of the TLS model, the spin of the electrons acts as a “flavor” variable and reflects the channel index in the two-channel Kondo effect. Thus, in this particular case non-Fermi liquid behavior is expected [1]. Studies on a bulk TLS material, performed by different experimental techniques,

might verify some of the aspects of the TLS Kondo model and open a new route to the non-Fermi liquid (NFL) problem.

Although point-contact spectroscopy, telegraph noise or scanning tunneling microscopy measurements suggest a strong coupling of the tunneling centers to the conduction electrons on a mesoscopic scale, so far, exist no convincing examples for electron-assisted tunneling on a microscopic scale [1,2]. Our interest in ThAsSe, whose layered-type crystal structure consists only of non-magnetic atoms (Fig. 2), originates from recent studies on its U-based homologue [3,4]. It has been shown that uncommon transport properties of the structurally disordered UAsSe ferromagnet may be consistently interpreted in terms of a dynamic disorder. This concerns, e.g., a disorder-dependent low-temperature upturn in $R(T)$ far below the ferromagnetic transition at around 110 K [3], as well as changes in the thermoelectric power introduced by tiny variations of Se excess [4]. The presence of the structural disorder in the anionic sublattice of UAsSe, favored by the similar atomic radii of As and Se, has been suggested by means of X-ray and scanning electron microscopy studies as well as ^{77}Se

Fig. 2: Crystallographic unit cell of ThAsSe with exposed $[\text{ThAs}_4\text{Se}_5]$ polyhedron formed by the anion nearest neighbor of thorium.

NMR measurements [3,4]. Thus, a similar situation is expected for ThAsSe.

All the ThAsSe single crystals investigated (plate-like of the thickness less than 1 mm and the mass of a few mg) display an anomalous low- T increase of $R(T)$ whose magnitude and temperature dependence is strongly sample dependent. However, for several specimens a distinct $\log T$ behavior over one decade in temperature has been found, as showed in Fig. 3 ($R(300\text{ K}) \approx 200\ \mu\Omega\text{cm}$ [5]). In addition, the influence of the hydrostatic pressure up to 1.88 GPa on the low- T $R(T)$ data, normalized to the value at 2 K is plotted [6]. While applied pressure does not alter the $\log T$ behavior, the hump-like anomaly observed at $T = 65\text{ K}$ (ambient pressure) is completely suppressed at $p = 1.88\text{ GPa}$. The way, in which p influences this anomaly, i.e., by shifting it to lower temperatures accompanied by a uniform reduction of its size, hints at its relation to the electronic structure. Since similar energy scales for both singularities, their different response to pressure indicates that they are of different origin. In particular, the low- T upturn in $R(T)$ cannot be caused by thermally activated hopping, because the pressure would change the tiny energy difference between the localized states and hence the magnitude of the hopping term.

Furthermore, the low- T upturn in $R(T)$ is not due to the spin Kondo effect (Fig. 4). While the $\log T$ signature in $R(T)$ is not altered by $B \leq 13.5\text{ T}$ (the inset (b) of Fig.4), a pronounced saturation of the dc magnetic susceptibility already below about 8 K

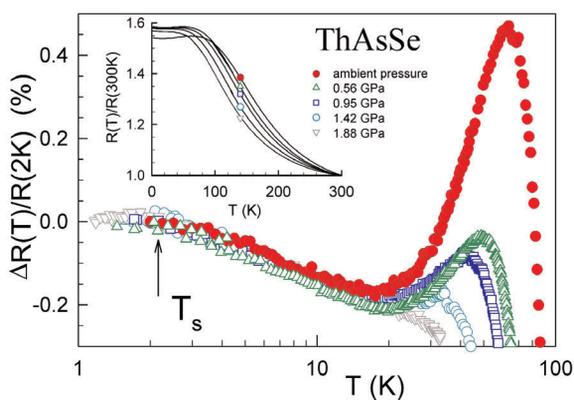


Fig.3: Effect of the hydrostatic pressure on the relative change of the low- T resistance normalized to the corresponding 2 K value for the single crystal of ThAsSe. Inset shows the ab -plane $R(T)/R(300\text{ K})$ dependence at $p \leq 1.88\text{ GPa}$ in the entire temperature range. The arrow indicates the temperature of saturation.

in $B = 5\text{ T}$ is found (the inset (a) of Fig.4). It proves that the applied magnetic field, being sufficient to align magnetic moments of impurities (less than 0.01 % of U^{+4}), does not change the anomalous scattering cross-section.

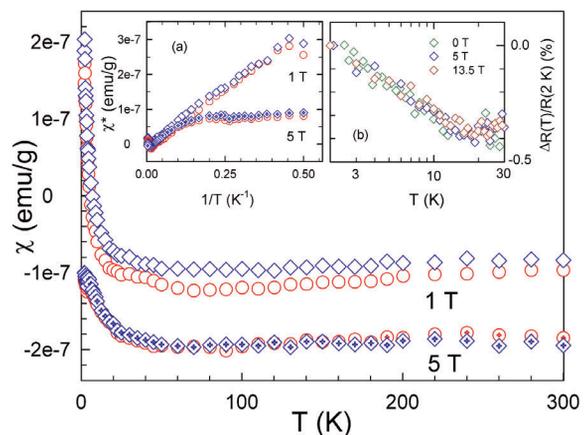


Fig. 4: The dc magnetic susceptibility for ThAsSe measured in magnetic fields of 1 T and 5 T applied perpendicular (circles) and parallel (diamonds) to the c axis. Inset (a): The same results plotted as χ^* vs. $1/T$, where χ^* denotes the low- T susceptibility data, from which average value of the diamagnetic component was subtracted. Inset (b): Irrelevance of the magnetic field up to 13.5 T for the low- T upturn in $R(T)$. All measurements were done on the same specimen.

A very weak (0.05 % per Tesla) and linear response of the resistance on $B \leq 17\text{ T}$ brings into question a scenario based on the weak localization as well. Especially that this, in general, two-dimensional effect usually disappears in a few times smaller fields [6]. This is in a striking contradiction to the behavior observed for ThAsSe. Finally, there is no experimental evidence for the 2D electron gas in ThAsSe. For this reason, an interpretation of the $\log T$ behavior in ThAsSe based on the electron-electron interaction may be seriously questioned. Most importantly, a saturation of the resistance below $T_S \sim 2\text{ K}$ (see Fig. 3) is in clear disagreement with the theoretical models for the electron-electron interaction, which predict a $-\log T$ dependence in the limit $T \rightarrow 0$ [7].

In Fig. 5 (a), we show specific-heat data for ThAsSe, as $C(T)/T$ vs. T^2 , obtained between 0.37 K and 5 K. A very small density of states allowed to detect an additional contribution to $C(T)$ below 2 K, as displayed in Fig. 5 (c). Since the

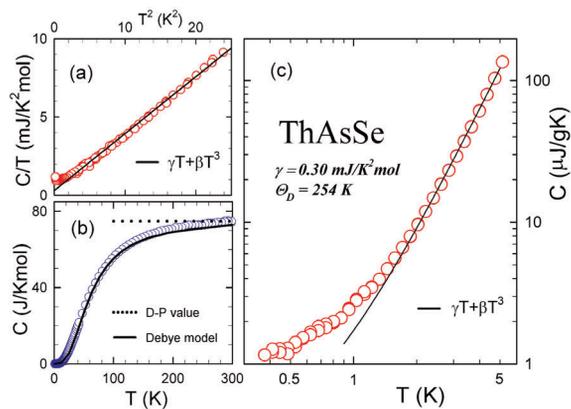


Fig.5: (a) The low- T specific heat, as $C(T)/T$ vs. T^2 , of a diamagnetic ThAsSe between 0.37 K and 5 K. The solid line shows the $\gamma T + \beta T^3$ dependence with $\beta = 3 \times 1944 \Theta_D^3$ in units of $J/K^4 \text{mol}$, where fitting parameters were determined from the temperature range 1.7 K – 5 K. (b) Molar heat capacity for ThAsSe up to 300 K. The solid line is the Debye approximation with $\Theta_D = 254$ K and the dotted line denotes the Dulong and Petit value. (c) The low- T $C(T)$ data on a double-logarithmic scale.

nuclear heat capacity can be safely ignored in ThAsSe, a quasilinear-in- T dependence is undoubtedly due to the TLS. Note that very similar $C(T)$ data were reported for, e.g., vitreous SiO_2 [8].

Anomalous low- T $R(T)$ properties of ThAsSe being unchanged by neither strong magnetic fields nor high hydrostatic pressure are apparently due to the TLS interacting with the conduction electrons. In general, the electron-assisted tunneling may lead to a complex temperature dependence of the resistance [1]. However, in the case of $\Delta < T_K$, i.e., when the Kondo temperature exceeds the TLS splitting, a logarithmic correction to $R(T)$ is predicted. For the Kondo effect with dominant scattering on the asymmetrical TLS, the $\log T$ relation transforms into $\Delta R(T) \sim 1 - aT^2$ at $T \ll T_K$. On the other hand, if $\Delta \ll T_K$ and the electrons are predominantly scattered by the symmetrical TLS, the saturation will be preceded by a $\Delta R(T) \sim 1 - aT^{1/2}$ dependence, i.e., the NFL behavior caused by the two-channel Kondo effect [1]. At present, no signature for $\Delta R(T) \sim 1 - aT^{1/2}$ was found. This points to a dominating scattering on the asymmetrical TLS. However,

since T_S varies significantly (by more than one order of magnitude) with a tiny difference in the As-Se chemical composition the realization of a non-Fermi liquid ground state in ThAsSe might be possible. A search for the non-Fermi liquid properties in ThAsSe and related systems, e.g., ZrAsSe or HfAsSe is the scope of the future work.

In summary, ThAsSe appears to be highly suited to study the TLS Kondo effect because (i) a concentration of the dynamical scattering centers can be controlled by the As-Se chemical composition, (ii) its ground state is not affected by a low-lying phase transition and (iii) high-quality single crystals can be studied, opening the possibility to determine a directional dependence of the electron-assisted tunneling. Additionally, comparative experiments on the diamagnetic ThAsSe and its ferromagnetic U-based homologue reflect the dependence of the interaction between the conduction electrons and the TLS on the character of the former ones.

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