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Localized states within the gap of $Ce_3Au_3Sb_4$

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Abstract

The temperature dependence of the specific heat and of the resistivity under pressure has been measured for single crystals of the semiconductor $Ce_3Au_3Sb_4$. The transport data follow an exponential activation and variable range hopping at low T, consistent with weak disorder and localization, while C/T has a $-\ln T$ dependence with large entropy. Thus the properties of $Ce_3Au_3Sb_4$ are very different from those of ordinary Kondo insulators.

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Kondo insulators, e.g. SmB_6, CeNiSn, $Ce_3Bi_4Pt_3$, and $Ce_3Pt_3Sb_4$ [1–5], are rare earth compounds with a small gap that originates from the hybridization between f-electrons and conduction electrons [6]. $Ce_3Au_3Sb_4$, on the other hand, has well localized f-moments; the resistivity reveals an insulating ground state [5,7,8]; and the specific heat coefficient, C/T, is rather large [5,7] and depends as $-\ln T$ at low temperatures. The large C/T is not a common feature of Kondo insulators and appears to be unique to $Ce_3Au_3Sb_4$, raising debates on the origin of the gap for this compound [8–14].

To explore the nature of the ground state and the in-gap states in this material, we studied stoichiometric single crystals. The density of in-gap states and their behavior are very different from those of ordinary Kondo insulators. The data are explained by assuming three bands, namely, a conduction and a valence band separated by a gap (as in a ordinary semiconductor), and hybridized with an almost dispersionless f-band in the gap [9–11].

Single crystal $Ce_3Au_3Sb_4$ was grown out of Sb and Au flux and characterized by powder and single crystal X-ray diffraction. The specific heat was measured with the relaxation method using a Quantum Design PPMS. The resistivity under hydrostatic pressure was measured using a Be/Cu piston-cylinder clamp device and a profiled toroidal anvil clamp device. The pressure was determined from the superconducting temperature change in Pb.

Fig. 1 displays the large specific heat coefficient, C/T, showing a logarithmic divergence, and the inset shows the corresponding entropy vs. T. The peak at 8 K is associated with the first crystalline field excitation at 25 K based on a fit to a Schottky anomaly due to two doublets. The entropy suggests a twofold degenerate ground state. The entropy at 5 K is about $0.6\,R\ln 2$ and reaches up to $0.8\,R\ln 2$ in a magnetic field of 3 T. The reduced entropy from $R\ln 2$ at 5 K may indicate that 10–15% of the Ce ions are non-magnetic. We note that isostructural $La_3Au_3Sb_4$ also is a small gap semiconductor, but its low temperature C/T is very small ($0.4\,\mathrm{mJ/mol\,K^2}$), as expected for semiconductor.

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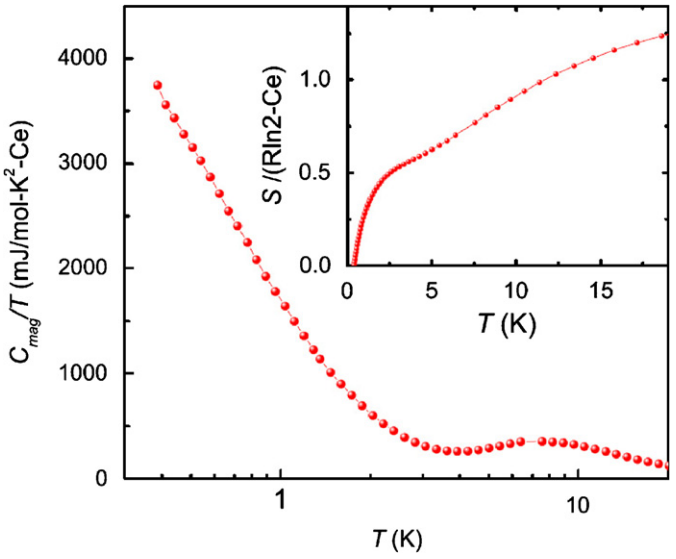

Fig. 1. Specific heat divided by T as a function of T. Inset: The corresponding magnetic entropy.

Thus, the specific heat result of $Ce_3Au_3Sb_4$ suggests a large f-electron density of states pinning the Fermi level within the band gap.

The insulating behavior of the resistivity is seen in Fig. 2. The resistivity ratio between 0.4 and 300 K is more than four orders of magnitude. The saturation of the resistivity below 10 K may be due to states within the gap. The activation behavior appears above 10 K with an activation gap of 90 K for this sample and becomes larger as higher pressure is applied. Since $Ce_3Pt_3Sb_4$ with smaller lattice constant has a larger gap [15], it is not surprising to see that the insulating gap increases under pressure. At low temperature the resistivity is fitted better with variable range hopping, $\rho = \rho_0 \exp(T_0/T)^{1/4}, T_0 \propto [N(E_F)\xi^3]^{-1}$, which indicates the existence of states with disorder in the gap. Here $N(E_F)$ is the density of states at the Fermi level and ξ is the localization length [16]. The fit gives a T_0 of 23.6 K, which decreases to 0.05 K at 53.3 kbar. The value of the resistivity at 1 K also decreases from 110 Ω cm at 1bar to 40 Ωcm at 53.3 kbar. The large reduction of T_0 indicates that the in-gap states are delocalized with pressure although the gap increases. A large effect can already be achieved with modest pressures.

In-gap states have also been observed in the Kondo insulator $Ce_3Bi_4Pt_3$ (isostructural to $Ce_3Au_3Sb_4$) under high pressure [17] with opposite tendency for T_0 (increasing with P), i.e., the in-gap states get localized under pressure. Considering that the gaps in both systems increase under pressure, this opposite trend indicates a different nature and/or origin of the gap and the in-gap states of $Ce_3Au_3Sb_4$ from those of normal Kondo insulators. Moreover, considering the large specific heat and its $-\ln(T)$ divergence in C/T at low temperatures appears in a similar T-range where also variable range hopping is observed, the in-gap states may be relevant to the hopping mechanism which then involves localized

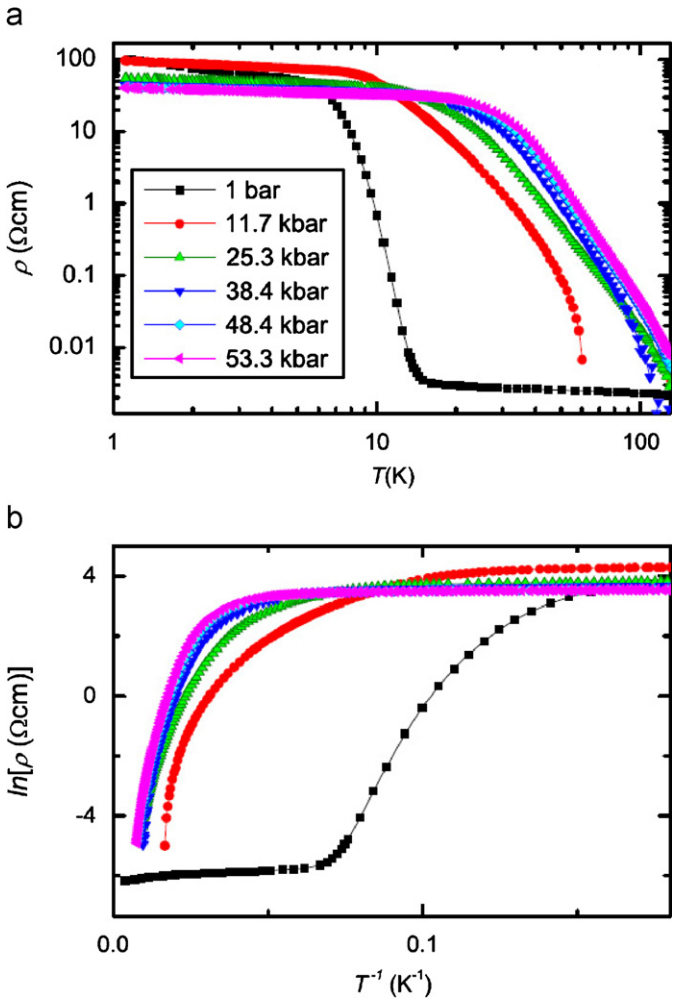

Fig. 2. (a) Resistivity vs. T for various pressures. (b) The same data as $\ln\rho$ vs. T^{-1} to show activation behavior. (See (a) for legend.)

f-electrons. This suggests that the f-band lies inside the gap and is hybridized with the conduction and valence bands.

In summary, we presented specific heat and pressure dependent resistivity data to discuss the unique behavior of $Ce_3Au_3Sb_4$. The existence of in-gap states, the large specific heat at low T and the decrease of T_0 as pressure increases provide strong indications that the gap formation mechanism in this system is different from that of normal Kondo insulators and the localized f-electron states exist inside the gap.

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