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Pressure effect in the Kondo semimetal $CeRu_4Sn_6$ with nontrivial topology*

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Kondo semimetal $CeRu_4Sn_6$ is attracting renewed attention due to the theoretically predicted nontrivial topology in its electronic band structure. We report hydrostatic and chemical pressure effects on the transport properties of single- and poly-crystalline samples. The electrical resistivity $\rho(T)$ is gradually enhanced by applying pressure over a wide temperature range from room temperature down to 25 mK. Two thermal activation gaps estimated from high- and low-temperature windows are found to increase with pressure. A flat $\rho(T)$ observed at the lowest temperatures below 300 mK appears to be robust against both pressure and field. This feature as well as the increase of the energy gaps calls for more intensive investigations with respect to electron correlations and band topology.

Keywords: $CeRu_4Sn_6$, Weyl semimetal, heavy fermion, hydrostatic pressure

PACS: 71.27.+a, 75.30.Mb, 72.15.–v **DOI:** 10.1088/1674-1056/27/9/097103

1. Introduction

Kondo insulators are a class of Kondo lattice compounds in which the localized, mostly f-electron band hybridizes with the dispersive conduction band, resulting in a small energy gap of a few meV in both spin and charge excitation channels at the Fermi level.[1] These materials have been attracting interest due to their highly renormalized energy gap by electronic correlations, giving rise to unusual physical properties as observed in optical conductivity,[1] thermoelectric transports,[2] etc. Prototypical examples of Kondo insulators are SmB_6,[3] $Ce_3Bi_4Pt_3$,[4] and so on. In some cases, the energy gap is highly anisotropic or only a pseudogap, such as in the title compound and CeNiSn,[5] which are more appropriate to be classified as Kondo semimetals. Compared to their metallic counterparts known as heavy-fermion metals, investigations on Kondo insulators/semimetals are highly insufficient due to their rareness.

$CeRu_4Sn_6$ is a relatively new member in the Kondo insulators/semimetals family. It crystallizes in a body-centered tetragonal, non-centrosymmetric lattice with space group $I\bar{4}2m$. A monotonic increase of the electrical resistivity $\rho(T)$ upon cooling from room temperature (RT) down to at least $T \approx 20$ K was first observed on a polycrystalline sample by Das and Sampathkumaran.[6] By contrast, its nonmagnetic homolog $LaRu_4Sn_6$ without f electron is a normal metal. Recent band structure calculations by a combination of density functional theory (DFT) and dynamical mean field theory[7,8] or Gutzwiller method[9] have shown a finite direct hybridization gap between Ce-4f and Ru-4d conduction bands, with however considerable residual electronic density of states at the Fermi level.

Subsequent investigations on magnetic, transport, and thermodynamic properties have revealed further details on this compound.[10,11] The magnetic susceptibility $\chi(T)$ is of Curie–Weiss behavior due to localized Ce^{3+} moments down to at least $T = 100$ K, followed by an unusual upturn at lower temperatures.[11] The $\chi(T)$ maximum, which signifies the opening of a spin gap in typical Kondo insulators,[1] has not been observed in $CeRu_4Sn_6$. This is most probably due to the substantial residual in-gap states. Such inference is supported by the specific heat $C(T)$ measurements, where the value of C/T reaches as high as 600 mJ/mol·K^2 when approaching absolute zero.[11] This is a large value even for a typical heavy-fermion metal,[12] relative to the high Kondo temperature $T_K \approx 170$ K as determined from x-ray absorption spectroscopy.[13]

Recently, there appears a revived interest in Kondo insulators/semimetals because of their nontrivial band topology originated from the inherently strong spin–orbit coupling, as observed in SmB_6[14] and CeNiSn.[15] Along this line, $CeRu_4Sn_6$ was also found to fulfill the criteria for nontrivial topology due to the band inversion between 4f and 5d bands;[9,13] because of the lack of inversion center, two types of Weyl nodes in the quasiparticle band structure near the Fermi level were theoretically predicted.[9] In agreement with the notion of Kondo

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semimetal, topologically trivial bulk states at the Fermi level were also predicted due to a negative indirect Kondo gap. This makes it difficult for transport probes to detect the excitations of Weyl fermions. As the quasiparticle bands in heavy-fermion systems are sensitive to external conditions like pressure and field, how the physical properties evolve with external parameters is of great interest in view of the band topology in correlated systems.

In this work, we have succeeded in growing single crystals of $CeRu_4Sn_6$ from Pb flux and studied their hydrostatic pressure effects. As a complement, chemical pressure effects on polycrystalline samples were also studied. Gapped behaviors in $\rho(T)$ were found in two temperature ranges that are separated by a hump at around 10 K. The corresponding energy gaps were found to expand with pressure, clearly pointing to their correlated nature. Following the opening of the smaller energy gap below $T \approx 2$ K, a flat $\rho(T)$ appears at the lowest temperatures below about 0.3 K. Interestingly, this behavior was found to be robust against both pressure and field. With the larger energy gap properly captured by theoretical calculations, an understanding of the much smaller gap and the flat resistivity toward zero temperature remains a challenge.

2. Experiment

Single crystals of $CeRu_4Sn_6$ were grown from molten flux of Pb. High-purity starting materials of Ce, Ru, Sn, and Pb were mixed in a molar ratio of 1 : 4 : 6 : 80 and loaded into an alumina crucible, which was further sealed under vacuum in a quartz tube. The quartz tube was then heated in a furnace from RT up to 1150 °C over 20 h, and dwelled at this temperature for 24 h before being cooled down to 600 °C at a rate of 2 °C/h. The tube was then inverted, and quickly put into a centrifuge to remove the excess flux.

The obtained single crystals are small but well faceted, with the longest dimension of roughly 0.5 mm, see inset of Fig. 1(a). Energy dispersive x-ray spectroscopy and x-ray diffraction from certain facet of the as-grown crystal (Fig. 1(a)) confirmed the correct composition and good crystallinity. We note that a few published works on single crystalline $CeRu_4Sn_6$ have employed samples grown from a floating zone furnace.[16,17] Polycrystalline samples of $CeRu_4(Sn_{1-x}Ge_x)_6$ were also prepared by arc melting, with Sn partially substituted by Ge in order to achieve a chemical pressure. The foreign peaks as revealed in Fig. 1(b), which have been known for $CeRu_4Sn_6$,[6,10] indicate a slight secondary phase of $Ce_3Ru_4Sn_{13}$. Since $CeRu_4Ge_6$ does not form, Ge therefore replaces Sn only partially. Here the substitution limit was set to $x = 0.15$ because the secondary phase substantially increased upon further substitution. As expected, the lattice constants a and c decrease with x, as shown in Fig. 1(c), resulting in a chemical pressure. The polycrystalline samples allow for detailed thermoelectric investigations, which are not possible with small single crystals grown from flux.

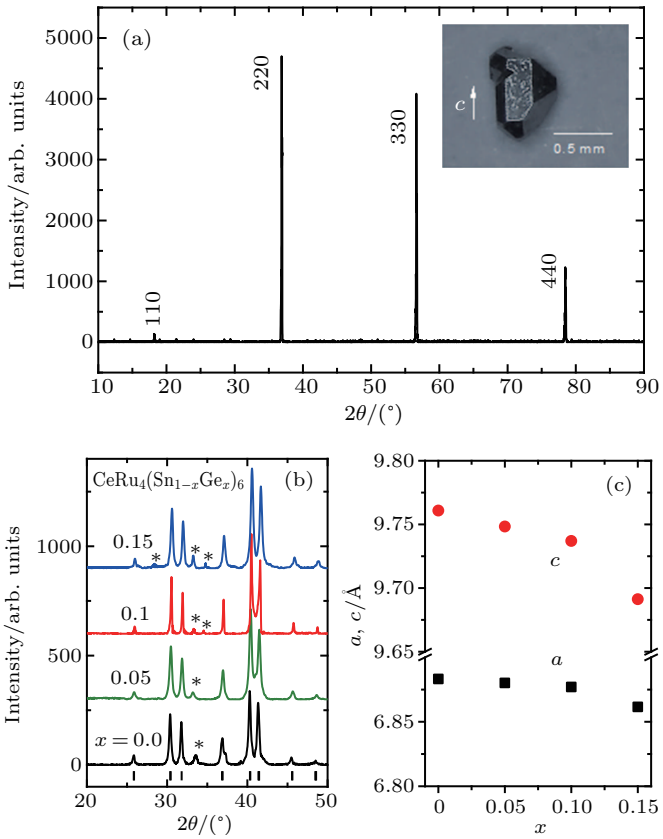


Fig. 1. (color online) (a) X-ray diffraction from one as-grown facet of the single crystalline $CeRu_4Sn_6$. Inset shows the photograph of a typical single crystal. (b) X-ray diffraction of the polycrystalline $CeRu_4(Sn_{1-x}Ge_x)_6$ with different x values. Here x is the nominal substitution concentration. "$*$" marks the peaks of secondary phases. (c) Lattice constants a and c as a function of x.

The electrical resistivity $\rho(T,B)$ was measured with a four-probe technique down to $T = 2$ K in the physical properties measurement system (PPMS, Quantum Design), and down to $T = 25$ mK by using a ^{3}He–^{4}He dilution refrigerator in a magnetic field up to 14 T. A self-clamped piston-cylinder pressure cell made of nonmagnetic CuBe and NiCrAl was employed to produce hydrostatic pressures, with glycerol as the pressure-transmitting medium. The generated pressure was monitored by the superconducting transition temperature of a small piece of Sn mounted together with the sample. Thermoelectric properties were measured in PPMS by using the arc-melted polycrystalline samples.

3. Results and discussion

Figure 2 shows the temperature-dependent resistivity $\rho(T)$ measured in various external pressures up to $P =$ 22.9 kbar for a single crystal, with the electrical current applied along the c axis. According to previous work at ambient pressure,[16] the c axis is more conductive than the a axis and its resistivity is approximately one half of that along the a axis.

Several prominent features can be immediately identified from Fig. 2: (i) an overall enhancement of the $\rho(T)$ values with increasing pressure; (ii) in all pressures, the value of $\rho(T)$ increases upon cooling in two separated temperature windows, from RT down to ~ 20 K and from ~ 2 K down to ~ 0.3 K; (iii) $\rho(T)$ tends to be flat at the lowest temperatures below ~ 0.3 K in all pressures.

Band structure calculations predicted a direct hybridization gap, albeit considerable residual states at the Fermi level.[7,9,13] This explains the semiconductor-like behavior at high temperatures. The upturn below $T \approx 2$ K is already visible in ambient pressure (Fig. 4(b)) and has also been observed in arc-melted polycrystalline sample,[10] suggestive of an intrinsic in-gap structure of this compound. The continuous enhancement of $\rho(T)$ with pressure is essentially different from the resistivity of a metallic Kondo system of similar T_K, where the resistivity decreases with pressure due to enhanced phase coherence among Kondo scattering, as observed in $CePd_3$.[18]

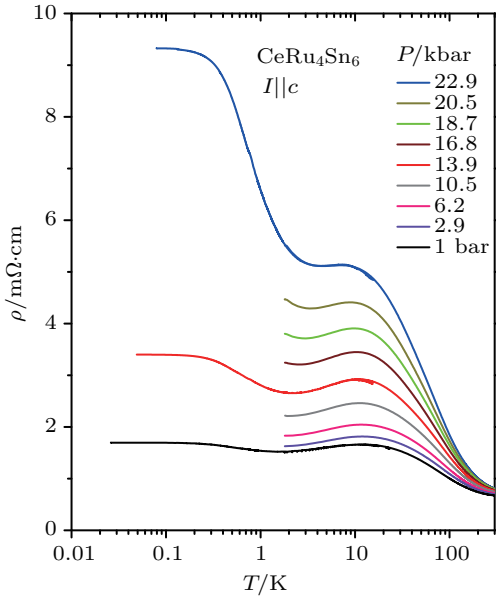


Fig. 2. (color online) Electrical resistivity $\rho(T)$ measured with current applied along c axis at various pressures from $P = 1$ bar to 22.9 kbar.

Following the previous interpretation of the high-T resistivity as thermally-activated behavior,[16] we use the Arrhenius equation, $\rho(T) = \rho_0 \exp[E_g/(2k_B T)]$, to describe the resistivity at $T > 120$ K. The energy gap obtained in this temperature range is denoted as E_{g1} and shown in Fig. 3(a) as a function of pressure. Selected curves are shown as $\ln\rho$ vs. $1/T$ in the inset, whose slope measures the value of E_{g1}. Note that $E_{g1} = 130$ K at ambient pressure is consistent with the band structure calculations[9] as well as the estimation reported in Ref. [16], but is more than two times larger than that of polycrystalline samples.[6,10,11,19] Similarly, we apply the Arrhenius equation to the low-T semiconducting behavior at $T < 2$ K, see the quasi-linear $\ln\rho$ vs. $1/T$ (denoted by dashed lines) in Fig. 3(b) inset. The estimated energy gap E_{g2} is only on the order of 0.1 K and increases by a factor of 8 in the highest pressure $P = 22.9$ kbar (Fig. 3(b)). The small energy gap, however, cannot be captured by the calculated band structures currently available. Similarly, two distinct gapped regimes have also been reported for SmB_6,[20] which are considered as a consequence of crystal-field splitting of the 4f states. Different multiplets of the 4f states may hybridize with the conduction band at different energies, leading to multiple Kondo gaps. We also note that there exists a report of resistivity measurements under hydrostatic pressures down to $T = 1.8$ K on polycrystalline $CeRu_4Sn_6$ (Ref. [21]). There, a nonmonotonic change of the energy gap was reported, together with an unexpected shoulder in $\rho(T)$ curves that is absent in the current work on single crystals.

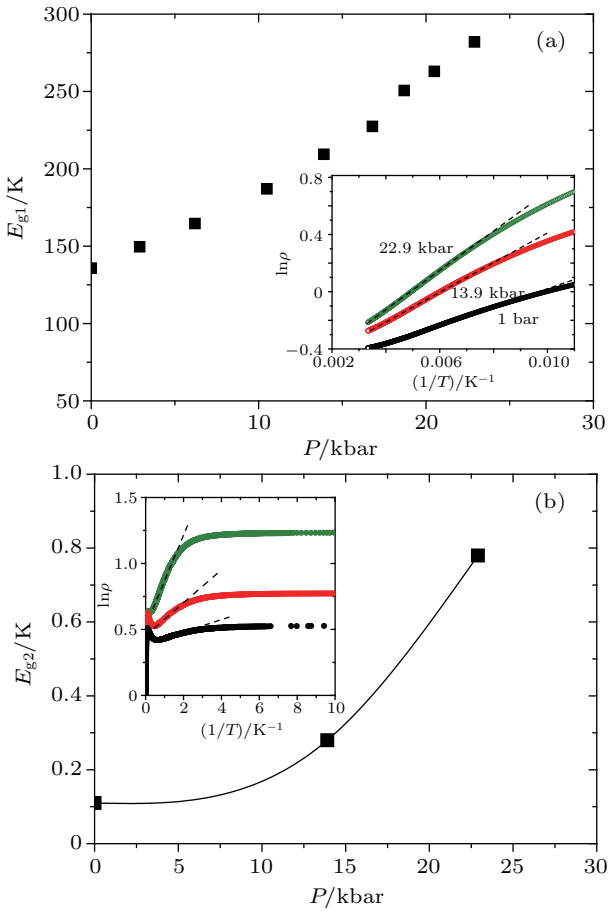


Fig. 3. (color online) (a) Energy gap E_{g1} estimated from the high-T resistivity curves, shown as a function of pressure for the single crystalline $CeRu_4Sn_6$. Inset shows $\ln\rho$ vs. $1/T$ for selected pressures, where a linear change follows the Arrhenius equation. (b) Energy gap E_{g2} estimated from the low-T window, 0.3 K$< T <$ 2 K. Inset highlights the $\ln\rho$ vs. $1/T$ curves in this temperature window.

The $\rho(T)$ upturn below 2 K as well as the flat resistivity at $T < 0.3$ K are robust against pressure. These further corroborate that the enhancement of the overall resistivity with pressure is due to an increase of the energy gaps, rather than a reduction of the in-gap states. Increase of hybridization gap

with pressure is not unusual and has been observed in, e.g., $Ce_3Bi_4Pt_3$.[4] There, the hybridization gap was found to be well traced by the single-ion Kondo temperature, the latter being enhanced by pressure through strengthened hybridization. This is a remarkable feature of Kondo insulators distinct from ordinary band insulators without significant electron correlations.

The transverse magnetoresistance (MR) measured at $T = 0.08$ K as a function of field is shown in Fig. 4(a) for three selected pressures, i.e., $P = 1$ bar, 13.9 kbar, and 22.9 kbar. At ambient pressure, the value of MR(B) is negative at low fields and turns upward after assuming a minimum. With increasing pressure, the position of the MR(B) minimum shifts from 3 T (1 bar) to 5.5 T (22.9 kbar), meanwhile the MR values are pushed to be even negative. Apparently, the pressure drives $CeRu_4Sn_6$ to be more insulating on one hand and more negative in its MR response on the other. It is obvious that there exist two competing mechanisms of MR in $CeRu_4Sn_6$ at low temperatures; one produces negative MR and the other produces positive MR. The latter can be ascribed to normal orbital magnetoresistance as observed in most nonmagnetic conductors. As to the negative MR, incoherent Kondo scattering is a frequently invoked origin in Kondo lattice. This, however, is unapplicable to $CeRu_4Sn_6$ because the hybridization gap is derived from phase coherent Kondo resonance in analogy to the coherent Fermi-liquid state in heavy fermion metals.[22] Alternatively, a feasible mechanism of negative MR generic to Kondo insulators is the closing of Kondo gap in magnetic field due to the Zeeman splitting effect of the renormalized bands, as has been observed in CeNiSn[23] and YbB_{12}.[24] Suppression of the smaller energy gap has already been indicated by the resistivity of polycrystalline $CeRu_4Sn_6$.[10]

To shed further light on the low-temperature resistivity, in Figs. 4(b) and 4(c), we show $\rho(T)$ measured in different fields at the ambient and the highest pressures. Surprisingly, the flat resistivity at the lowest temperatures is robust against field as well, in addition to its robustness in pressure (Fig. 2). As is already clear from Fig. 4(a), the low-T saturating resistivity in Figs. 4(b) and 4(c) is not monotonically suppressed by the field. Its re-enhancement in higher fields can be, at least partially, ascribed to the enhanced positive orbital MR. Given the robustness of the low-temperature flat resistivity, one is tempted to consider the topologically protected electronic structure as one underlying origin. Whether this inference is correct appears to be interesting in future investigations.

We now turn to the Ge substituted polycrystalline samples $CeRu_4(Sn_{1-x}Ge_x)_6$. Here, our motivation is to study the thermoelectric properties, which are not accessible by the single crystals due to their small dimensions. As shown in Fig. 5(a), the $\rho(T)$ curves measured for these samples reveal a clear effect of chemical pressure. Their values are enhanced gradually by substitution with an increasing trend of the energy gap, see the upper inset in Fig. 5. These features are qualitatively consistent with the hydrostatic pressure effects shown in Fig. 2. Similar to the observations in the single crystals, a hump at around 10 K is also visible in Fig. 5(a), except for the case of $x = 0.05$, where an extrinsic drop of $\rho(T)$ at lower temperatures has been observed. On the other hand, the thermal conductivity $\kappa(T)$ continuously decreases with increasing x due to the enhanced chemical disorders (the lower inset in Fig. 5).

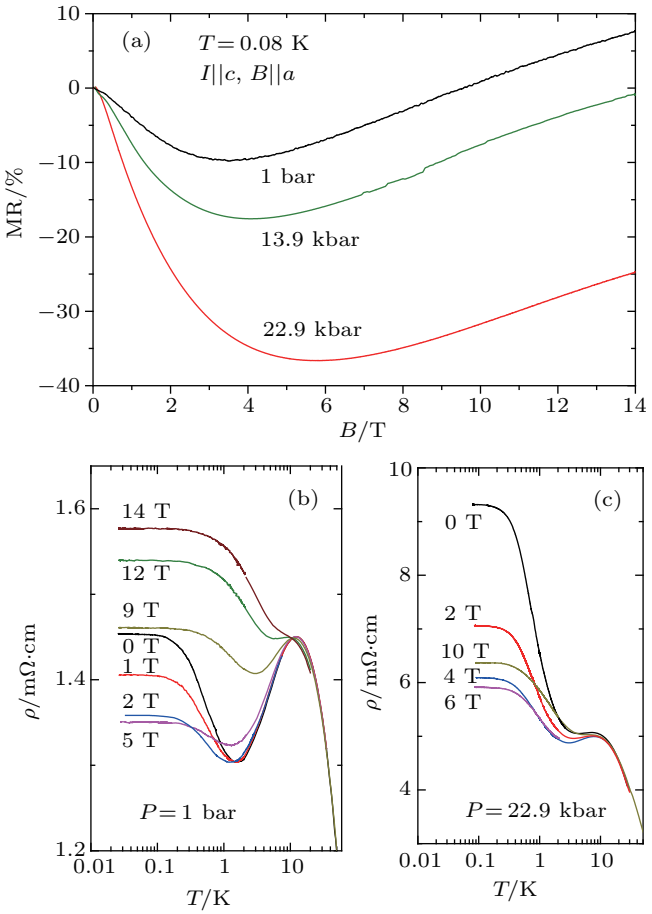


Fig. 4. (color online) (a) Isothermal transverse magnetoresistance, $MR = (\rho_B - \rho_{0T})/\rho_{0T}$, measured as a function of field at $T = 0.08$ K, at three selected pressures. Panels (b) and (c) display the low-temperature $\rho(T)$ measured in different fields at ambient pressure and at $P = 22.9$ kbar, respectively. Note that the low-temperature flat resistivity is robust against both pressure and field.

A combination of electronic correlations and small energy gap in Kondo insulators/semimetals has long been pursued for superior thermoelectric properties.[2] The thermopower $S(T)$ of unsubstituted poly- and single-crystalline $CeRu_4Sn_6$ has already been reported.[10,17] As shown in Fig. 5(b), the $S(T)$ curves obtained for Ge substituted polycrystalline samples are qualitatively similar to that of the pure $CeRu_4Sn_6$[10] with, however, gradually enhanced absolute values. The sign change at $T \approx 50$ K is robust against chemical pressure and hinders higher $S(T)$ values to

be achieved. Different to the thermopower of typical Kondo lattice compounds,[17] a positive maximum corresponding to the energy scale of local Kondo effect, i.e., T_K, has not been observed in $CeRu_4(Sn_{1-x}Ge_x)_6$. This reflects the complicated situation of $CeRu_4Sn_6$, where the overall temperature dependence of $S(T)$ is influenced by at least two effects, i.e., the thermal activation through the hybridization gaps and the Kondo scattering of the residual electronic states.

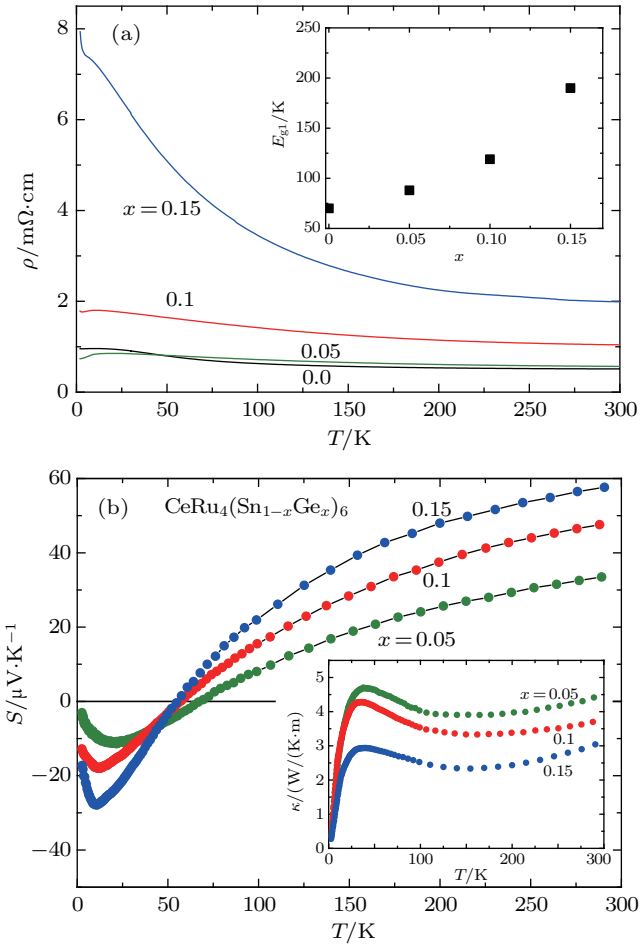


Fig. 5. (color online) (a) Resistivity $\rho(T)$ and (b) thermopower $S(T)$ of the polycrystalline samples $CeRu_4(Sn_{1-x}Ge_x)_6$. Upper inset: the energy gap E_{g1} estimated for $CeRu_4(Sn_{1-x}Ge_x)_6$. Lower inset: thermal conductivity $\kappa(T)$ of the same samples.

4. Conclusion

We have successfully grown single crystals of the Kondo semimetal $CeRu_4Sn_6$ from Pb flux. Besides the larger hybridization gap $E_{g1} \sim 130$ K, a resistivity upturn at below 2 K was also observed, reflecting a tiny energy gap (E_{g2}) that is less than 1 K. An increase of the energy gaps by applying chemical and hydrostatic pressures was observed and is a clear manifestation of enhanced electronic correlations in the hybridized bands. Tunability of the energy gaps will bring about better opportunities to approach the topological properties of this compound. The flat resistivity at the lowest temperatures was found to be a robust feature against both pressure and field. Whether the topologically nontrivial electronic structure plays a role in the low-temperature $\rho(T)$ and how it evolves at even higher pressures appear to be interesting topics for future investigations.

Acknowledgement

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