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# Heavy fermion $Ce_3Co_4Sn_{13}$ compound under pressure

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The non-magnetic heavy fermion compound $Ce_3Co_4Sn_{13}$ was studied under pressure. We report single crystalline measurements of electrical resistivity as a function of temperature $\rho(T)$ under pressure. Some characteristic features related to a structural transition ($T_S$), crystalline field effects ($T_{CEF}$), and a low temperature maximum ($T_{max}$), possibly connected simultaneously to the onset of Kondo lattice coherence and short range magnetic correlations, were identified in the $\rho(T)$ data. A pressure-temperature phase diagram with $T_S$ and $T_{max}$ was constructed by mapping these features. Like for most Ce-based heavy fermion compounds, $T_{max}$ moves to higher temperatures with pressure, indicating that it is related to the Kondo energy scale, due to the increase of hybridization induced by pressure. On the other hand, $T_S$, associated to a superlattice distortion and probably combined with a charge density wave transition, decreases as a function of pressure. However, differently from the $Sr_{3-x}Ca_xIr_4Sn_{13}$ system, where a superlattice quantum phase transition is observed [L. E. Klintberg *et al.*, Phys. Rev. Lett. **109**, 237 008 (2012)], in $Ce_3Co_4Sn_{13}$ $T_S \sim 154$ K, at ambient pressure ($P = 0$), seems to stabilize at around 143 K for $P \geq 19$ kilobars. We also investigated $\rho(T)$ in external magnetic fields, at $P = 0$. Negative magnetoresistance and increase of $T_{max}$ are observed, suggesting suppression of low temperature short range magnetic correlations. [http://dx.doi.org/10.1063/1.4913304]

## I. INTRODUCTION

The intermetallic $R_3M_4Sn_{13}$ (3-4-13) series of compounds, where R is a rare-earth or alkaline-earth ion and M is a transition metal, are very interesting materials that can present fascinating physical properties such as antiferromagnetism, superconductivity, and strong electron correlation effects.[1] The series crystallize in the cubic $Pm\bar{3}n$ space group symmetry,[2] similar to that of the thermoelectric skutterudites compounds. A small distortion on the cubo-octahedral rare-earth site results in a local tetragonal symmetry.[3] The magnetism in most of these materials comes from the 4*f* electrons total angular momentum, given by Hund's rule.[4] The interplay between crystalline electrical field (CEF) effects and Ruderman-Kittel-Kasuya-Yosida (RKKY) magnetic interaction promotes different magnetic ground states. Examples of antiferromagnetic compounds are $Gd_3Co_4Sn_{13}$ ($T_N = 14.5$ K)[5] and $Eu_3Ir_4Sn_{13}$ ($T_N = 10$ K),[6,7] which also display a structural transition ($T_S$) at ~55 K.[8] On the other hand, $Pr_3Ir_4Sn_{13}$ and $Nd_3Ir_4Sn_{13}$ show no magnetic ordering down to 1.8 K.[6] In addition, some superconductors can be found when 4*f* magnetism is not present such as in $Ca_3Rh_4Sn_{13}$ ($T_c \sim 8.2$ K), $Yb_3Rh_4Sn_{13}$ ($T_c \sim 8$ K),[1] and $Sr_3Ir_4Sn_{13}$ ($T_c = 5$ K).[9] The latter material additionally undergoes a superlattice structural transition at around 147 K, which doubles its lattice parameter in respect to the cubic $Pm\bar{3}n$ structure. Interestingly, by chemical doping $Ca^{2+}$ ions into $Sr_3Ir_4Sn_{13}$ and applying an external pressure one can suppress the superlattice transition temperature to zero, suggesting a superlattice quantum phase transition.[9]

Furthermore, besides the CEF and RKKY energy scales, compounds containing Ce as the rare-earth ion usually exhibit Kondo effect. The Kondo interaction, which couples the Ce $4f^1$ electron with the conduction-electron-sea, enhances the electron effective mass and affects the electronic scattering, giving rise to heavy fermion physics. $Ce_3Rh_4Sn_{13}$ is an example of non-magnetic 3-4-13 with high $\gamma$ Sommerfeld coefficient values.[10,11] $Ce_3Ir_4Sn_{13}$ is also a heavy fermion [$\gamma = 0.67$ J/(mol-Ce-K$^2$)][6] with possibly some short range ferromagnetic correlations and a low temperature structural phase transition at 2 K, observed in specific heat and electrical resistivity measurements.[12,13]

In this work, we study the $Ce_3Co_4Sn_{13}$ heavy fermion compound under pressure. This material shows no evidence for long range magnetic order, down to 0.3 K, and presents a large increase in the specific heat at low temperatures yielding a high $\gamma \sim 4$ J/(mol-Ce-K$^2$) Sommerfeld coefficient, with a Kondo temperature estimated to be $T_K = 1.2$ K.[14–17] The magnetic moment is approximately 2.58 $\mu_B$ per $Ce^{3+}$ ion, close to the value of 2.54 $\mu_B$ for the free ion.[14] At low temperatures, magnetization data fit well for an isolated doublet ground state,[15] which arises from the lift of the six-fold

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degenerated $J = 5/2$ state of the free ion by a tetragonal CEF. Similar to $Sr_3Ir_4Sn_{13}$, a superlattice structural transition at around 150 K was observed.[3] Also, a possible connection of this transition with a charge density wave (CDW) transition was reported,[18] providing a rare example of a CDW order in a 3D material. Here, we present our electrical resistivity data as a function of temperature for $Ce_3Co_4Sn_{13}$ single crystals measured under pressure. Interesting features attributed to the presence of structural transition, CEF effects and a low temperature maximum were observed. A pressure-temperature phase diagram with the structural transition and the low temperature maximum was obtained by mapping these features. In addition, the behavior of the low temperature maximum under magnetic fields, at ambient pressure, was investigated.

## II. EXPERIMENT DETAILS

$Ce_3Co_4Sn_{13}$ single crystals were grown using the Sn-flux method, where elemental Ce:Co:Sn are weighted in the ratio 1:1.3:20, yielding samples with dimensions around $0.5 \times 0.5 \times 0.5\,\mathrm{cm}^3$. Samples were heated at 200 °C/h to 1100 °C for 2 h, then cooled to 650 °C at a rate of 5 °C/h, where the excess of flux was spun. Powder X-ray diffraction data showed no presence of spurious phases. Samples were previously screened for Sn inclusions. Unaligned single crystals were used in electrical resistivity measurements. A clamp-type Cu-Be pressure cell, with Fluorinert FC70 + FC77 as pressure transmitting medium, adapted to an AC resistance measurement system in a four-contact configuration, was used in a He3/He4 dilution refrigerator, which can reach temperatures around 50 mK. Room and low temperature manometers were manganin and lead, respectively. The temperature was determined by a calibrated Cernox sensor. Ambient pressure experiments under magnetic fields were performed in a Quantum Design PPMS with He3 option.

## III. RESULTS AND DISCUSSION

The electrical resistivity as a function of temperature $\rho(T)$ for a $Ce_3Co_4Sn_{13}$ single crystal, at ambient pressure, is shown in Fig. 1, and are consistent with previous measurements.[15] The room temperature value of the resistivity varied between 450 and 125 $\mu\Omega$ cm for different samples. Some characteristic features related to a $T_S$, crystalline field effects ($T_{CEF}$), and a low temperature maximum ($T_{max}$) were identified in the $\rho(T)$ data. Like in $Sr_3Ir_4Sn_{13}$, an increase of the resistivity is seen just below $T_S$, which might be due to a decrease in the density of states at the Fermi level.[9]

For $20 < T < 150$ K, a hump-like feature at $T_{CEF} \sim 35$ K can be attributed to CEF effects, which due to the tetragonal site symmetry of $Ce^{3+}$ ions in the $Ce_3Co_4Sn_{13}$ structure,[3] splits the six-fold degenerated $J = 5/2$ multiplet into three Kramers doublets, as in $Ce_3Ir_4Sn_{13}$.[13] An increase of the $\rho(T)$ data for $T \lesssim 20$ K is observed and a maximum at $T_{max}$ is observed, possibly connected simultaneously to the onset of Kondo lattice coherence[19,20] and short range magnetic correlations, seen in low temperature specific heat.[16] The $\rho(T)$ behavior at ambient pressure is quite different in polycrystalline samples, where the resistivity decreases for $T < T_S$.[11,18]

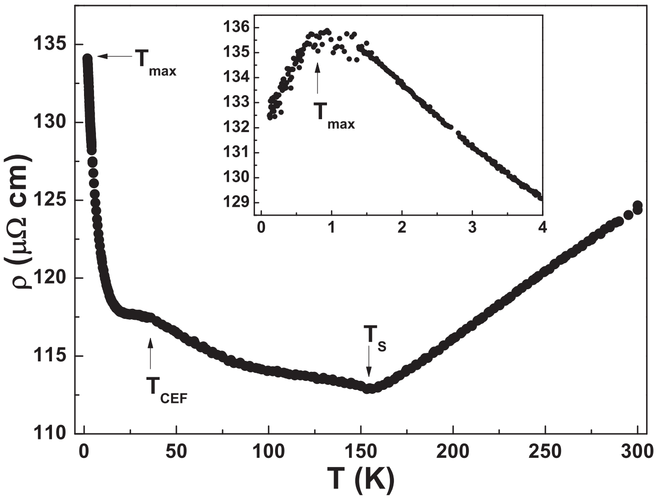

FIG. 1. $Ce_3Co_4Sn_{13}$ electrical resistivity as a function of temperature. The arrows indicate some characteristic features related to a $T_S$, $T_{CEF}$, and a low temperature maximum ($T_{max}$).

Fig. 2 displays the pressure evolution of the temperature dependent electrical resistivity for $Ce_3Co_4Sn_{13}$ single crystals. These curves are representative of the qualitative behavior for all applied pressures. We observe the suppression of the CEF hump with pressure as a result of enhancing the hybridization between the $Ce^{3+}$ 4*f* electrons and the conduction bands, which mixes the broad CEF levels and recovers the behavior for a Kondo single impurity scattering regime.[21]

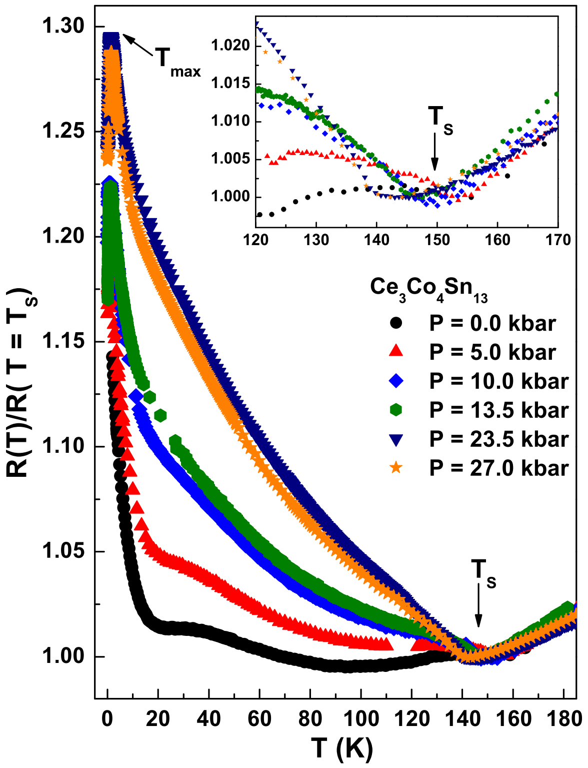

FIG. 2. $Ce_3Co_4Sn_{13}$ representative electrical resistivity curves as a function of temperature at different pressures, normalized at $T_S$. Inset shows, in detail, the pressure evolution of the $T_S$.

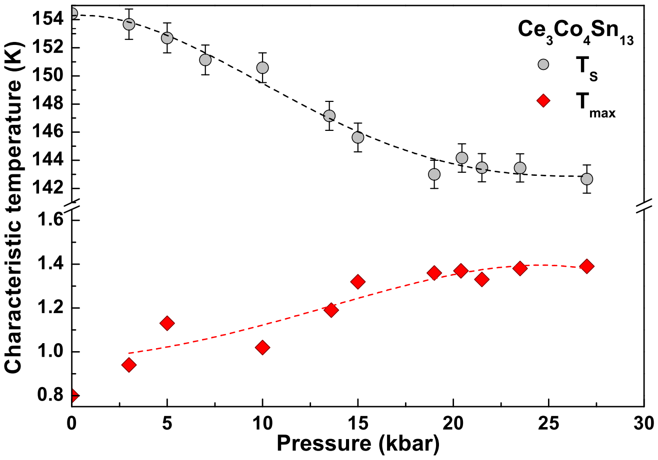

FIG. 3. Pressure-temperature phase diagram for $Ce_3Co_4Sn_{13}$ showing the pressure dependence of a $T_S$ and the low temperature maximum ($T_{max}$) seen in the electrical resistivity data. Dashed lines are guides to the eyes.

A pressure-temperature phase diagram of the relevant temperatures scales for $Ce_3Co_4Sn_{13}$ was obtained from the data displayed in Fig. 2 and is presented in Fig. 3. Like for most Ce-based heavy fermion compounds, $T_{max}$ moves to higher temperatures due to the increase of hybridization induced by pressure, as expected if this characteristic temperature is related to the crossover from incoherent to coherent electronic scattering regime in a Kondo lattice.[19,20] On the other hand, $T_S$, associated to a superlattice distortion[3] and probably combined with a charge density wave transition,[9] decreases as a function of pressure (see inset Fig. 2). However, differently from the $Sr_{3-x}Ca_xIr_4Sn_{13}$ system, where a superlattice quantum phase transition is observed,[9] in $Ce_3Co_4Sn_{13}$, the phase diagram clearly shows that $T_S \sim 154$ K (at ambient pressure, $P = 0$) shifts to lower temperatures reaching a stable value of approximately 143 K for $P \geq 19$ kilobars.

We also investigated the effect of external magnetic fields on the $\rho(T)$ data. Fig. 4 shows the behavior of representative $\rho(T)$ curves for a $Ce_3Co_4Sn_{13}$ single crystal, at ambient pressure, under different magnetic fields. The inset shows that $T_{max}$ moves to higher temperatures, similar as it does under pressure. Low temperature specific heat measurements indicated the suppression of short range magnetic correlations for magnetic fields $H \gtrsim 2.5$ T.[16] This suggests that the magnetic field enhancement of $T_{max}$ and negative magnetoresistance, also observed in Ref. 11, are associated to the decrease of low temperature incoherent short range magnetic scattering contribution to $\rho(T)$. For high magnetic fields, it is expected that $T_{max}$ represent only the onset of Kondo lattice coherence, and for lower temperatures, a $T^2$ Fermi-liquid regime is recovered. $T_S$ does not change for $H \leq 9$ T (not shown).

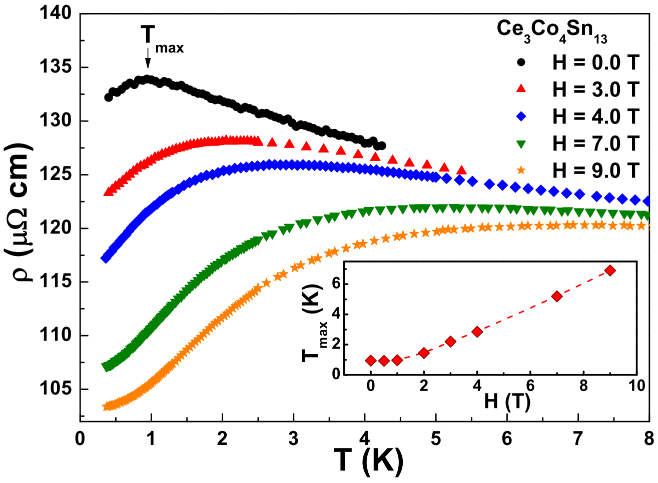

FIG. 4. Representative temperature dependent electrical resistivity curves for different magnetic fields for $Ce_3Co_4Sn_{13}$ at ambient pressure. Inset shows the low temperature maximum ($T_{max}$) evolution under external magnetic fields. Dashed lines are guides to the eyes.

## IV. SUMMARY

In summary, we report temperature dependent electrical resistivity $\rho(T)$ measurements under pressure on single crystals of the heavy fermion $Ce_3Co_4Sn_{13}$ compound. Interesting features were identified in the $\rho(T)$ data and mapped as a function of pressure. We observe that the low temperature maximum moves to higher temperatures with pressure. On the contrary, a structural transition ($T_S \sim 154$ K at ambient pressure) seems to stabilize at around 143 K for $P \geq 19$ kilobars. This behavior is different from the $Sr_{3-x}Ca_xIr_4Sn_{13}$ system, where a superlattice quantum phase transition is observed.[9] We also investigated the effect of external magnetic fields on $\rho(T)$ at $P = 0$. Negative magnetoresistance and enhancement of $T_{max}$ are observed, suggesting suppression of low temperature short range magnetic correlations.

## ACKNOWLEDGMENTS

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