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Magnetic and related properties of a novel compound $Ce_3Co_2Sn_7$

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Abstract

A novel compound $Ce_3Co_2Sn_7$ was synthesized and characterized by means of X-ray powder diffraction, magnetic susceptibility and magnetization, specific heat, electrical resistivity and magnetoresistivity measurements performed at temperatures down to 1.8 K and in magnetic fields up to 9 T. The compound was found to crystallize in the orthorhombic $La_3Co_2Sn_7$ structure type (space group *Cmmm*, no. 65) with lattice parameters: $a = 4.556(8)$ Å, $b = 27.345(5)$ Å and $c = 4.562(3)$ Å. Analysis of the physical properties suggests that only two of three atoms in the formula unit possess localized magnetic moments, which order antiferromagnetically at 4.6 K with some field-induced spin-reorientation in the ordered region. Moreover, $Ce_3Co_2Sn_7$ exhibits some features of dense Kondo systems with the characteristic Kondo temperature of about 1.2 K.

Keywords: cerium intermetallics, antiferromagnetic order, Kondo effect

1. Introduction

Ternary intermetallics based on lanthanides and actinides have been one of the most intensively studied groups of compounds in solid state physics. They have attracted much attention mainly due to unusuall physical phenomena observed in those systems at low temperatures.

In the course of our search for new ternary intermetallics exhibiting interesting physical properties we have focused on cerium-based compounds with stoichiometry 3:2:7 crystallizing with the orthorhombic $La_3Co_2Sn_7$ structure

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type (space group *Cmmm*, no. 65), in which cerium atoms occupy two inequivalent crystallographic sites (i.e. the Wyckoff positions 2*d* and 4*i*) [1]. As far only two cerium-based members of this family have been reported in the literature, i.e. $Ce_3Ni_2Ge_7$ [2] and $Ce_3Ni_2Sn_7$ [3]. Low-temperature measurements of their physical properties revealed that both phases order antiferromagnetically at the Néel temperature of 7.5 and 3.8 K, respectively, and exhibit some features of dense Kondo systems [4, 5, 6, 7]. Neutron diffraction experiments showed that in both systems there is no detectable magnetic moment on the cerium ions located at the 2*d* site. Only those located at the 4*i* position bears an ordered magnetic moment of 1.98(2) μ_B in $Ce_3Ni_2Ge_7$ and 1.89(1) μ_B in $Ce_3Ni_2Sn_7$. Their magnetic structure can be described as a stack of ferromagnetic trigonal prisms with a sequence $+-+-$ along the *b*-axis [8, 9].

Only few more isostructural compounds with other lanthanide and actinide elements have been discovered. Among them there are compounds reported as antiferromagnets ($Pr_3Ni_2Sn_7$, $Nd_3Ni_2Sn_7$ [10]), ferromagnets ($Np_3Co_2Si_7$ [11], $U_3Fe_2Ge_7$ [12], $U_3Co_2Ge_7$ [13, 14]), ferrimagnet ($U_3Ru_2Si_7$ [15]), and paramagnets ($U_3Co_2Si_7$ [11], $U_3Fe_2Si_7$ [16], $La_3Ni_2Sn_7$ [10] and $La_3Co_2Sn_7$). Some of them exhibit also heavy fermion behavior.

In this article we present basic structural and physical properties of a new member of the 3:2:7 family, namely $Ce_3Co_2Sn_7$, investigated by means of X-ray powder diffraction, magnetization, specific heat, resistivity and magnetoresistivity measurements. We show that most probably also in that system the cerium ions occupying different crystallographic sites have different electron configuration.

2. Materials and methods

Polycrystalline samples of $Ce_3Co_2Sn_7$ and $La_3Co_2Sn_7$ were synthesized by conventional arc melting stoichiometric amounts of the constituents (with purity at least 99.9%) under protective titanium-gettered argon atmosphere. The buttons were turned over and remelted several times to ensure good homogeneity. The overall weight losses after the melting were less than 0.5%. The samples were subsequently wrapped with molybdenum foil and annealed in evacuated silica tube at 850°C for two weeks. Quality of the products was verified by means of X-ray powder diffraction using X´pert Pro PANalytical diffractometer with Cu Kα radiation. The experimental patterns were

analysed using the Rietveld method implemented in the Fullprof software [17].

Magnetic properties were studied using a Quantum Design MPMS-5 SQUID magnetometer in temperature range 1.8–400 K and in external magnetic fields up to 5 T. Specific heat was measured using a standard time-relaxation method implemented in a Quantum Design PPMS platform at temperatures ranging from 1.8 K to 300 K. Electrical resistivity and magnetoresistivity measurements were carried out on the latter platform using the conventional four-probe technique on bar-shaped samples with spot-welded contacts in temperature range 1.8–300 K and in magnetic fields up to 9 T.

3. Results and discussion

3.1. Crystal structure

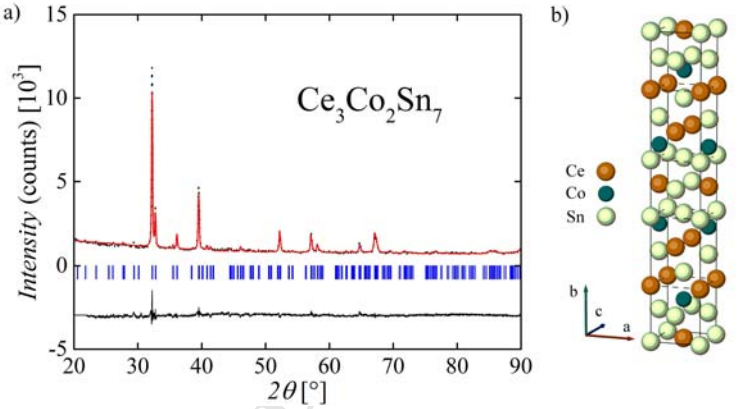


Figure 1: a) X-ray powder diffraction pattern obtained for $Ce_3Co_2Sn_7$ (circles) together with refined positions of the Bragg reflections (vertical ticks), calculated profile (red solid line) and difference plot (black solid line below); b) crystal structure of $Ce_3Co_2Sn_7$ compound.

Analysis of the X-ray powder diffraction pattern obtained for pollycrystalline $Ce_3Co_2Sn_7$ (Fig. 1) revealed that the compound crystallizes in the orthorhombic $La_3Co_2Sn_7$ structure type (space group $Cmmm$, no. 65 [1]). The refined lattice parameters are: $a = 4.556(8)$ Å, $b = 27.345(5)$ Å and $c = 4.562(3)$ Å. No Bragg reflections from any secondary phase were found in the experimental pattern. The obtained values of reliability factors R_p and R_{wp} were 5.02 and 6.12 respectively. Lattice parameters obtained for

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$La_3Co_2Sn_7$ are $a = 4.6068(2)$ Å, $b = 27.6979(1)$ Å, $c = 4.6112(2)$ Å, being in good agreement with those reported previously [1].

The dimensions of the unit cell of $Ce_3Co_2Sn_7$ can be compared to those reported for the Ni-based counterpart, i.e. $Ce_3Ni_2Sn_7$ ($a = 4.5650(2)$ Å, $b = 27.3041(9)$ Å, $c = 4.5690(2)$ Å [4]). As expected, substitution of Ni by Co (atomic radii of which are nearly equal) has very small influence on the unit-cell size.

3.2. Magnetic properties

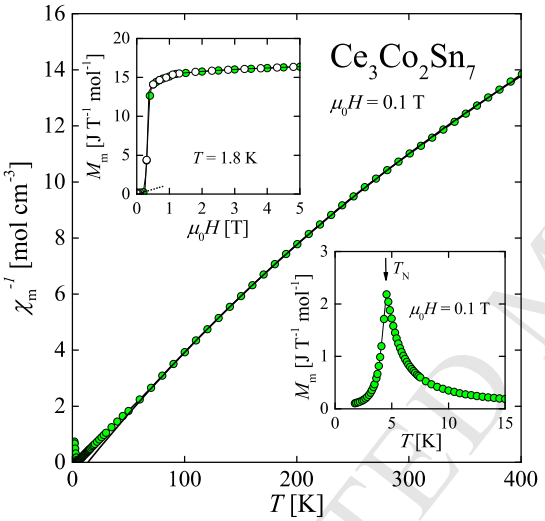


Figure 2: Temperature variation of inverse molar magnetic susceptibility χ_m^{-1} of $Ce_3Co_2Sn_7$ measured in external magnetic field $\mu_0 H$; solid line is a fit of the modified Curie–Weiss law (Eq. 1) to the experimental data. Upper inset displays molar magnetization M_m measured in the ordered region with increasing and decreasing magnetic field (open and closed symbols, respectively); Straight dashed line is added to emphasize the linear character of magnetization below the transition. Lower inset presents $M_m(T)$; solid curves serve as guides for the eye and the arrow marks ordering temperature T_N.

Fig. 2 presents temperature dependence of the inverse molar susceptibility $\chi_m^{-1}(T)$ measured for $Ce_3Co_2Sn_7$. Above about 80 K the $\chi_m^{-1}(T)$ curve is nearly linear and can be described by the modified Curie–Weiss law:

$$\chi_m(T) = \frac{C}{T - \theta_p} + \chi_0, \qquad (1)$$

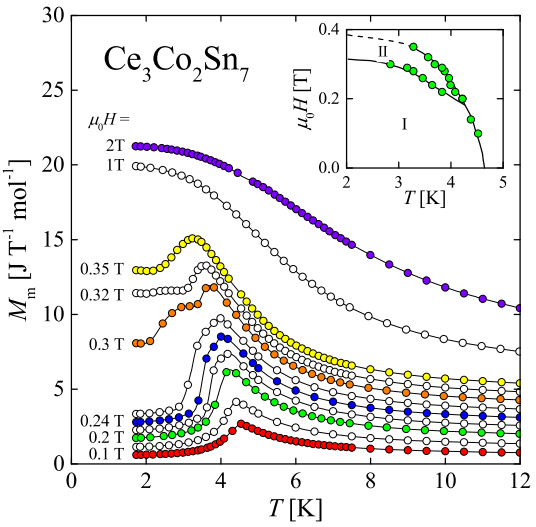


Figure 3: Temperature variation of M_{m} of $Ce_3Co_2Sn_7$ taken in several different magnetic fields (for the sake of clearity all the curves except the lowest one are shifted up by 0.5 J/(T mol)). Inset presents tentative magnetic phase diagram $\mu_0 H(T)$ of the compound; I and II mark the magnetically ordered phases.

where C is the Curie constant, θ_{p} is the paramagnetic Curie–Weiss temperature, and χ_0 stands for a sum of all temperature independent contributions (mainly Pauli paramagnetism of conduction electrons and core diamagnetism). Least-squares fit of Eq. 1 to the experimental data yielded the values: $C = 20.2$ cm^{3}Kmol^{-1}, $\theta_{\mathrm{P}} = 14$ K and $\chi_0 = 6.7 \times 10^{-3}$ cm^{3}mol^{-1}. The Curie constant C is related to the effective magnetic moment μ_{eff} via the expression:

$$C = \frac{n\mu_0 N_{\mathrm{A}} \mu_{\mathrm{eff}}^2}{3k_{\mathrm{B}}} \tag{2}$$

where n is a number of atoms carrying magnetic moment in a formula unit, μ_0 is the magnetic permeability of free space, N_{A} is the Avogadro's number and k_{B} is the Boltzmann's constant. Assuming $n = 3$ one can obtain the average effective magnetic moment $\mu_{\mathrm{eff}} = 2.07(1)\ \mu_{\mathrm{B}}$ which is clearly lower than the theoretical one calculated for a free Ce^{3+} ion, i.e. 2.54 μ_{B}. This finding is similar to the case of $Ce_3Ni_2Sn_7$, where the effective magnetic moment (obtained assuming negligible contribution χ_0) is also smaller than expected (i.e. 2.33 μ_{B} [4]). Neutron diffraction experiments performed on $Ce_3Ni_2Sn_7$ samples revealed that there is no detectable magnetic moment at

the $2d$ site [9] and only two cerium atoms at the $4i$ position bear an ordered magnetic moment. Applying this scenario to $Ce_3Co_2Sn_7$ and assuming $n = 2$, one can get the value of the effective magnetic moment $\mu_{\mathrm{eff}} = 2.53(6)\ \mu_{\mathrm{B}}$. This value almost ideally corresponds to the theoretical one and thus gives an argument for a presence of well localized magnetic moments only at the $4i$ cerium positions in the unit cell similarly to $Ce_3Ni_2Sn_7$ compound. Positive value of the paramagnetic Curie–Weiss temperature θ_{p} in $Ce_3Co_2Sn_7$ points out some contribution of ferromagnetic interactions between the localized magnetic moments of cerium.

Below about 80 K the $\chi_{\mathrm{m}}^{-1}(T)$ curve of $Ce_3Co_2Sn_7$ deviates from the behavior described by Eq. 1 , manifesting reduction of the effective magnetic moment most probably due to thermal depopulation of the crystal field levels and at the lowest temperatures possibly due to Kondo screening. Distinct cusp-shaped anomaly visible at about 4.6 K in T-dependence of the molar magnetization M_{m} manifests antiferromagnetic ordering of the compound (see the lower inset to Fig. 2). Field dependence of M_{m} measured at 1.8 K and displayed in the upper inset in Fig. 2 seems to be in line with the latter finding. As seen, magnetization of the compound initially increases linearly with increasing magnetic field (as expected for antiferromagnets) and subseuently undergoes sharp metamagnetic-like phase trasition in the magnetic field of about 0.35 T. At higher fields M_{m} shows clear tendency to saturation and at 5 T reaches a value of 16.4 $\mathrm{JT^{-1}mol^{-1}}$. Assuming that in 5 T the magnetic moments are all aligned along the direction of applied magnetic field, one can estimate the value of the saturated magnetic moment μ_{sat} as equal to about 1.47(4) μ_{B} per Ce atom (if averaged over two Ce atoms).

In order to shed more light on the low temperature behavior of $Ce_3Co_2Sn_7$, temperature variation of the magnetization was measured in a few different magnetic fields. As can be inferred from Fig. 3, upon increasing magnetic field the antiferromagnetic peak at T_{N} shifts towards lower temperatures and broadens. In magnetic fields higher than about 1 T the $M_{\mathrm{m}}(T)$ curve takes a Brillouin-like shape, characteristic of ferromagnets.

In the intermediate fields presence of a second peak is clearly visible in the ordered region. Evolution of the position of the observed anomalies in $M_{\mathrm{m}}(T)$ is shown on a tentative magnetic phase diagram of $Ce_3Co_2Sn_7$ displayed in the inset to Fig. 3. Exact character of the antiferromagnetic phases I and II visible in the diagram remains however an open question, which needs further experiments (*e.g.* neutron diffraction).

3.3. Specific heat

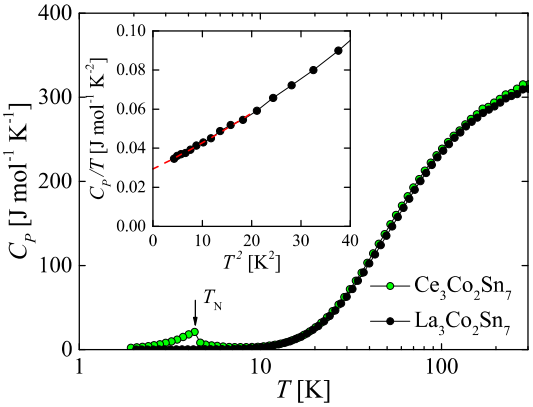


Figure 4: Temperature dependence of specific heat C_P of $Ce_3Co_2Sn_7$ (green circles) and $La_3Co_2Sn_7$ (black circles); the arrow marks the Néel temperature T_N. Inset shows C_P/T vs. T^2 for the La-based compound; red dashed line presents a fit of Eq. 3 to the experimental data.

Fig. 4 presents molar specific heat C_P of $Ce_3Co_2Sn_7$ and $La_3Co_2Sn_7$. $C_P(T)$ of $La_3Co_2Sn_7$ is characteristic of non-magnetic metals and can be described at low temperatures by the formula:

$$C_P(T) = \gamma T + \beta T^3 + \delta T^5 \tag{3}$$

where the first term is the low-temperature approximation of the electron contribution to the total specific heat of metals with the Sommerfeld coefficient γ, and the latter two terms are the first two terms of the polynomial approximation of the Debye formula for low-temperature phonon specific heat. Least-squares fit of Eq. 3 to the experimental data yielded $\gamma = 29$ mJmol^{-1}K^{-2}, $\beta = 1.2$ mJmol^{-1} K^{-4} and $\delta = 9.8$ μJmol^{-1} K^{-6} (see small open circles in the inset in Fig. 4). Using the second value, one can estimate the Debye temperature of $La_3Co_2Sn_7$ $\theta_D = 267$ K. The λ-shaped peak at $T_N = 4.6$ K dominates the low temperature part of $C_P(T)$ of $Ce_3Co_2Sn_7$. It corroborates bulk character of the magnetic phase transition evidenced in magnetic property measurements.

Assuming that the specific heat of isostructural $La_3Co_2Sn_7$ is a good approximation of a non-magnetic part of C_P of $Ce_3Co_2Sn_7$, one can estimate magnetic contribution to the specific heat ΔC_P of the latter compound as:

$$\Delta C_P(T) = C_P(Ce_3Co_2Sn_7) - C_P(La_3Co_2Sn_7). \tag{4}$$

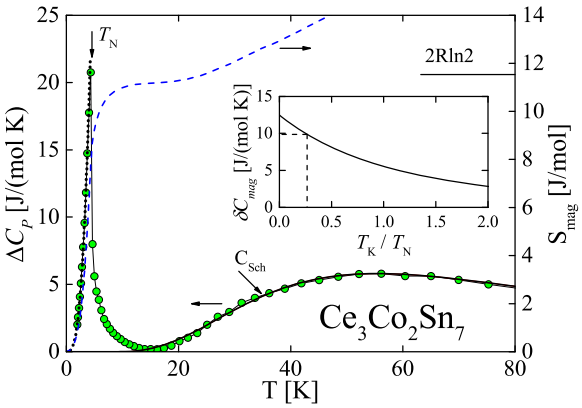


Figure 5: The non-lattice contribution ΔC_P to the specific heat of $Ce_3Co_2Sn_7$ (green circles, thin solid curve serves as guides for the eye; left axis) and calculatetd magnetic entropy S_{magn} (blue dashed line; right axis). The thick solid line is the fit of Eq. 5 to the experimental data in paramagnetic region. The inset presents graphical solution of Eq. 7 (for details see the text).

$\Delta C_P(T)$ obtained in such a manner is presented in Fig. 5. As seen, at elevated temperatures broad maximum in $\Delta C_P(T)$ is well visible. It can be attributed to the Schottky effect due to the crystal field effect, being in agreement with magnetic susceptibility behavior. The Schottky contribution to the specific heat can be described by the formula (see e.g. Ref. [18]):

$$C_{\mathrm{Sch}}(T) = \frac{R}{T^2}\left[\frac{\sum_i g_i E_i^2 e^{-E_i/T}}{\sum_i g_i e^{-E_i/T}} - \left(\frac{\sum_i g_i E_i e^{-E_i/T}}{\sum_i g_i e^{-E_i/T}}\right)^2\right], \qquad (5)$$

where R is the gas constant, E_i are energies of the crystal field levels in units of temperature, and g_i are degeneracies of these levels. For the $^2F_{5/2}$ term (resulting from the $4f^1$ configuration of the Ce^{3+} ions) a doublet–doublet–doublet scheme of the crystal field splitting is appropriate, i.e. $i = 0, 1, 2$ and $g_0 = g_1 = g_1 = 2$. Least-squares fit of the experimental data to Eq. 5 (see the bold black line in Fig. 5) yielded the relative energies $E_1 = 115$ K and $E_2 = 187$ K (E_0 is by definition equal to 0 K).

It should be noted however, that due to interplay of the Kondo and crystal field effects our analysis is rather qualitative. As it was shown by Desgranges and Rasul [19, 20, 21], the crystal field levels in dense Kondo systems cannot be described properly using exclusively the Schottky formula. And the deconvolution of the experimental data into a Kondo and a Schottky-

type peak in some cases is hard or even impossible to perform. Therefore the estimated energies of the splitting may differ from those of pure crystal field.

The low-temperature anomaly in $\Delta C_P(T)$ can be in turn described assuming that it is the result from presence of antiferromagnetic magnons. Taking dispersion relation $\omega = \sqrt{\Delta^2 + Dk^2}$ at temperatures smaller than the spin-wave gap Δ, magnetic contribution to the specific heat in the ordered region follows the formula [22]:

$$C_{\mathrm{magn}}(T) = \gamma^* T + c\Delta^{7/2} T^{1/2} e^{-\Delta/T} \left[1 + \frac{39}{20}\left(\frac{T}{\Delta}\right) + \frac{51}{32}\left(\frac{T}{\Delta}\right)^2 \right], \qquad (6)$$

in which the first term describes additional conduction-electron contribution, and the coefficient c is related to the spin-wave stiffness D by $c \propto 1/D^{3/2}$. Least-squares fit of Eq. 6 to the experimental data below T_{N} yielded $\gamma^* = 334$ mJK^{-2}mol^{-1}, $c = 24$ mJK^{-5}mol^{-1} and $\Delta = 5.8$ K (see the black dotted line in Fig. 5).

Moderately enhanced value of γ^* might point out at presence of strong electronic correlations in $Ce_3Co_2Sn_7$. According to the phenomenological model developed by Bredl *et al.* for antiferromagnetically ordered $CeAl_2$ [23] the specific heat jump δC_{magn} in such systems is related to the Kondo temperature T_{K}. In antiferromagnetically ordered Ce-based compounds it can be described by the formula [24]:

$$\delta C_{\mathrm{magn}} = \frac{6k_B}{\psi'''(\frac{1}{2}+\zeta)} \left[\psi'(\frac{1}{2}+\zeta) + \zeta\psi''(\frac{1}{2}+\zeta) \right]^2, \qquad (7)$$

where $\zeta = (T_{\mathrm{K}} \,/\, T_{\mathrm{N}})/2\,\pi$ and ψ', ψ" and ψ"' are first, second and third derivative of the digamma function. The inset to Fig. 5 presents δC_{magn} plotted as a function of $T_{\mathrm{K}}/T_{\mathrm{N}}$ using Eq. 7. Since the jump δC_{magn} in $Ce_3Co_2Sn_7$ is of about 20 Jmol^{-1} K^{-1} for two atoms carrying localized magnetic moments, one can estimate T_{K} of the system as equal to about 1.2 K (see the dashed line in the inset to Fig. 5).

Temperature dependence of the magnetic entropy S_{magn} defined as:

$$S_{\mathrm{magn}}(T) = \int_0^T \frac{\Delta C_P}{T} dT \qquad (8)$$

is plotted in the main panel of Fig. 5. As seen, S_{magn} reaches at T_{N} only about 75% of the value $2R\ln 2$, expected for two atoms with a doublet as a ground

state, and saturates at about 97% of $2R\ln 2$ at about 13 K. Such reduction of the entropy – characteristic of Kondo systems – is another manifestation of the presence of strong electron correlations in the studied system.

3.4. Electron transport properties

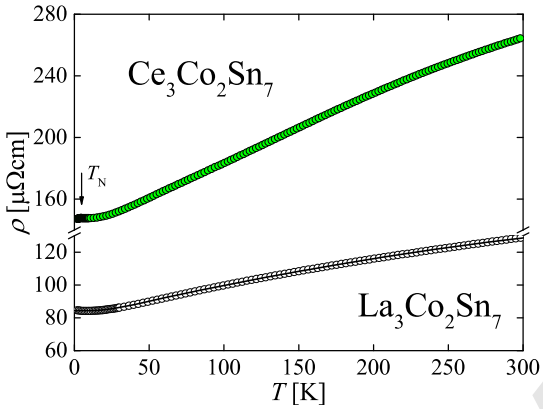


Figure 6: Temperature variation of the electrical resistivity of $Ce_3Co_2Sn_7$ and $La_3Co_2Sn_7$. Solid line is a fit of Eq. 9 to the experimental data; the arrow marks ordering temperature T_N.

Temperature dependences of the electrical resistivity ρ of $Ce_3Co_2Sn_7$ and $La_3Co_2Sn_7$ are presented in Fig. 6. $\rho(T)$ of the latter non-magnetic compound can be described using the commonly known Bloch–Grüneisen–Mott relation:

$$\rho(T) = \rho_0 + 4R\Theta_R \left(\frac{T}{\Theta_R}\right)^5 \int_0^{\frac{\Theta_R}{T}} \frac{x^5 dx}{(e^x-1)(1-e^{-x})} - KT^3, \qquad (9)$$

where ρ_0 is the residual resistivity, the second term represents electron-phonon scattering, and the third term is a contribution due to Mott's s-d interband electron scattering. The least-squares fit of Eq. 9 to the experimental data (see the solid line in Fig. 6) yielded the values of the parameters $\rho_0 = 84\ \mu\Omega$cm, $R = 0.17\ \mu\Omega\text{cmK}^{-1}$, $\Theta_R = 139$ K and $K = 2.7 \times 10^{-7}\ \mu\Omega\text{cmK}^{-3}$, which are from a range characteristic of polycrystalline specimens of intermetallics. The characteristic temperature θ_R is lower then the Debye temperature estimated from the specific heat, which is acceptable because of slightly different assumptions of the theoretical models developed for $C_P(T)$ and $\rho(T)$.

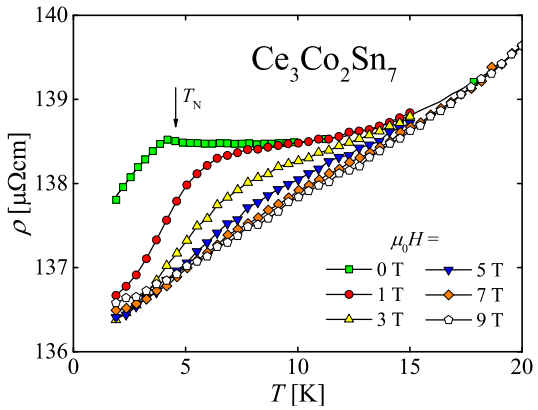


Figure 7: Low-temperature behavior of resistivity of $Ce_3Co_2Sn_7$ measured in various magnetic fields; the arrow marks the Néel temperature.

Temperature variation of the resistivity of $Ce_3Co_2Sn_7$ shows at first glance quite similar, metallic behavior (Fig. 6). Below 10 K it exhibits however slight increase with decreasing temperature followed by a sudden drop just below T_N, confirming the bulk antiferromagnetic ordering of the compound. The small negative slope of $\rho(T)$ above T_N could be ascribed to the Kondo effect. Unfortunately, our attempts to extract and reliably analyse magnetic part of the resistivity of $Ce_3Co_2Sn_7$ failed due to large difference in absolute values of ρ of the La- and Ce-based compounds, caused most probably by significantly larger number of internal cracks in the latter one.

Nevertheless, measurements of the resistivity of $Ce_3Co_2Sn_7$ in various magnetic fields (Fig. 7) seem to support our hypothesis on the presence of the Kondo effect. In particular, the characteristic small increase of ρ with decreasing T is fully suppressed by external magnetic field, as expected for the Kondo systems.

Fig. 8 presents field dependence of the transverse magnetoresistivity defined as:

$$\frac{\Delta\rho}{\rho_0}(\mu_0 H) \;=\; \frac{\rho(\mu_0 H, T) \;-\; \rho(0, T)}{\rho(0, T)}, \tag{10}$$

measured at various temperatures in the paramagnetic region. As can be seen, the magnetoresistivity curves $\frac{\Delta\rho}{\rho_0}(\mu_0 H)$ have sigmoid shape, characteristic of Kondo systems. Moreover, they can be superimposed onto each other

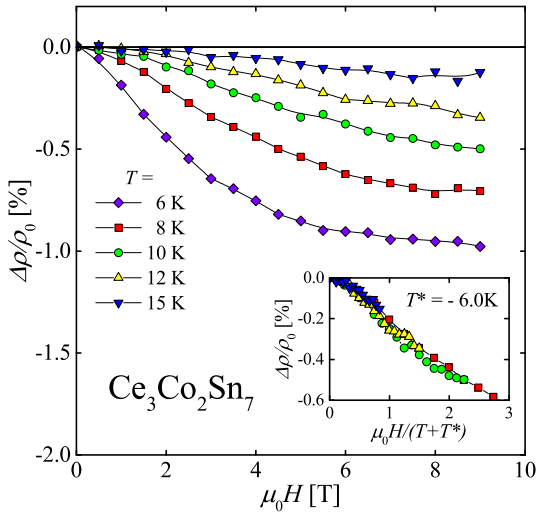


Figure 8: Magnetic field dependence of transverse magnetoresistivity of $Ce_3Co_2Sn_7$ at various temperatures. Inset presents Schlottmann's scaling of the curves.

(see the inset to Fig. 8) using Schlottmann's scaling [26]:

$$\frac{\Delta\rho}{\rho_0} = f\left(\frac{\mu_0 H}{T+T^*}\right), \tag{11}$$

where T^* is a characteristic temperature. The scaling performed for $Ce_3Co_2Sn_7$ yielded T^* = -6.0 K. The negative value of T^* cannot be interpreted as the Kondo temperature – it suggests rather existence of strong ferromagnetic correlations in $Ce_3Co_2Sn_7$. Such behavior was already observed in such dense Kondo systems as UBe_{13} [27], YbPtSn [28], $CeNiGe_2$ [29] and $CeNiGe_3$ [30]. The presence of the ferromagnetic correlations in the antiferromagnetically ordered $Ce_3Co_2Sn_7$ is higly probable. Neutron diffraction studies of isostructural $Ce_3Ni_2Sn_7$ revealed that within the layers of trigonal prism blocks magnetic moments localised on cerium atoms (Ce2) are arranged ferromagnetically and the whole compound consists of antiferromagnetically ordered ferromagnetic layers separated by other blocks of atoms bearing no magnetic moment (please refer to [9] for more details).

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4. Concluding remarks

A novel compound $Ce_3Co_2Sn_7$ crystallizes in the orthorhombic $La_3Co_2Sn_7$-type structure, in which three cerium atoms from the formula unit occupy two inequivalent crystallographic positions in the unit cell. Analysis of its magnetic properties revealed that most probably only two of them possess localized magnetic moments, while the third one remains non-magnetic. The magnetic moments order antiferromagnetically below $T_N = 4.6$ K, what manifests itself as distinct anomalies in all the physical properties studied. Moreover specific heat and electrical resistivity of $Ce_3Co_2Sn_7$ show some features characteristic of the Kondo effect, suggesting presence of strong electron correlations in the studied compound.

Acknowledgements

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- New compound Ce3Co2Sn7 was synthesized and their physical properties were studied.
- Localized magnetic moments of cerium order antiferromagnetically at 4.6 K.
- Strong electronic correlations are manifested in thermodynamic and electron transport properties of the compound.

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