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Magnetic and related properties of a novel compound $Ce_3Co_2Sn_7$

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ABSTRACT

A novel compound $Ce_7Co_3Sn_2$ 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.

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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 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 [4–7]. 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

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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

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 $La_3Co_2Sn_7$ 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

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:

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

where C is the Curie constant, θ_p is the paramagnetic Curie–Weiss

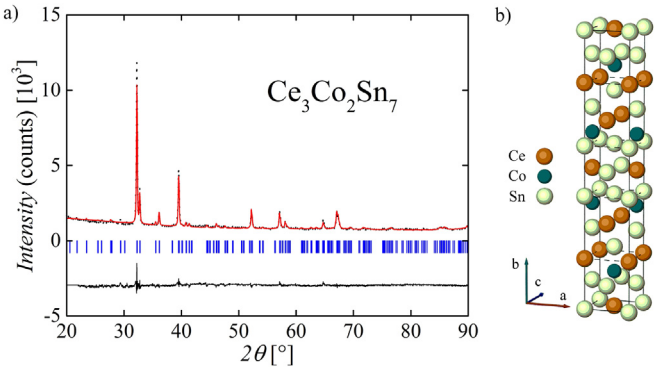

Fig. 1. a) X-ray powder diffraction pattern obtained for $Ce_3Co_2Sn_7$ (circles) together with refined positions of the Bragg reflections (vertical ticks), calculated profile (red solid line) and difference plot (black solid line below); b) crystal structure of $Ce_3Co_2Sn_7$ compound. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

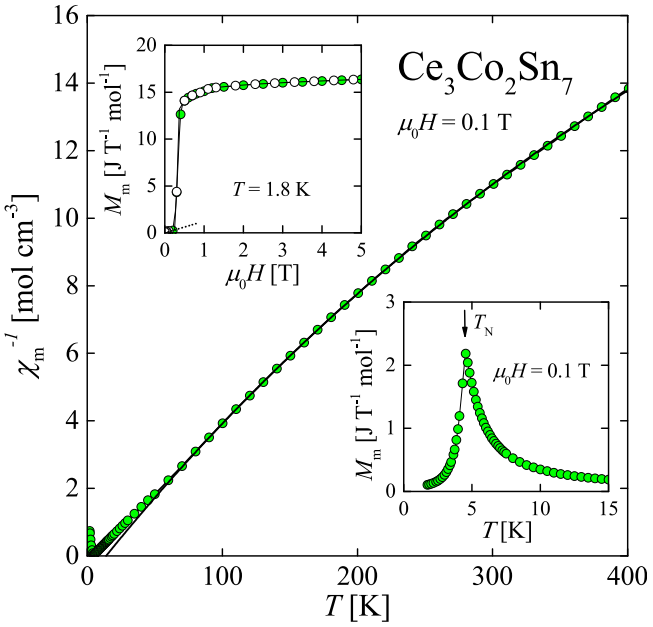

Fig. 2. Temperature variation of inverse molar magnetic susceptibility χ_m^{-1} of $Ce_3Co_2Sn_7$ measured in external magnetic field $\mu_0 H$; solid line is a fit of the modified Curie–Weiss law (Eq. (1)) to the experimental data. Upper inset displays molar magnetization M_m measured in the ordered 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.

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}, $\theta_P = 14$ 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:

$$C = \frac{n\mu_0 N_A \mu_{eff}^2}{3k_B} \quad (2)$$

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 $\mu_{eff} = 2.07(1)\ \mu_B$ 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 2*d* site [9] and only two cerium atoms at the 4*i* 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 $\mu_{eff} = 2.53(6)\ \mu_B$. 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 4*i* 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 $\chi_m^{-1}(T)$ 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

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