



Bulk and local properties of DyRhSn

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ABSTRACT

Electric, magnetic and ¹¹⁹Sn Mössbauer spectroscopy measurements were carried out on the hexagonal DyRhSn, where all atoms occupy single crystallographic sites. This stannide undergoes a transition from a paramagnetic to an antiferromagnetic state at $T_N = 7.5(1)$ K. The obtained results are compared with previously published magnetic and neutron diffraction data.

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1. Introduction

The RERhSn stannides crystallize in the hexagonal ZrNiAl-type structure (space group $P\bar{6}2m$) [1]. Recently, neutron diffraction studies revealed that the magnetic structure of DyRhSn corresponds to a propagation vector $\mathbf{k} = [1/2, 0, 1/2]$ [2]. It was shown that the best description of its magnetic structure below $T_N = 6$ K can be obtained with a model of a triangular arrangement of the magnetic moments within the basal plane.

In this work polycrystalline DyRhSn was studied using powder X-ray diffraction, AC susceptometry, DC magnetometry, electrical resistivity measurements as well as ¹¹⁹Sn Mössbauer spectroscopy, and the obtained results are discussed in comparison with those obtained by neutron methods [2]. Especially, ¹¹⁹Sn Mössbauer spectroscopic measurements have been applied to deliver deeper insight into the local electronic and magnetic properties of DyRhSn.

2. Results and discussion

2.1. Bulk properties

The preparation of the DyRhSn sample was made by arc melting and a consecutive annealing procedure as previously described [1]. The hexagonal ZrNiAl-type structure was confirmed with lattice parameters $a = 753.1(4)$ and $c = 377.1(3)$ pm being in a good agreement with recent neutron diffraction data [2]. Detailed magnetic measurements were performed by means of a Cahn RG automatic electrobalance in a temperature range 6.8–285 K at a field $H_0 = 450$ Oe on a bulk sample as well as using a 7225 Lake Shore AC susceptometer/DC magnetometer. Above 15 K, the recorded susceptibility obeys fairly well a modified Curie–Weiss law in the form $\chi_\sigma = \chi_0 + C/(T - \Theta_p)$, with the temperature independent factor $\chi_0 = -9.75 \times 10^{-6}$ cm³/g, the Curie constant $C = 4.093 \times 10^{-2}$ K cm³/g, and the paramagnetic Curie temperature $\Theta_p = -2.8$ K (see Fig. 1). The small but negative value of Θ_p is indicative for antiferromagnetic correlations. The effective magnetic moment was derived from the formula $\mu_{\text{eff}} = p_{\text{eff}} \mu_B$, where $p_{\text{eff}} = (3k_B/N_A)^{1/2} (MC)^{1/2} / \mu_B$ (here k_B is the Boltzmann constant, N_A is the Avogadro number, and M is the molar mass expressed

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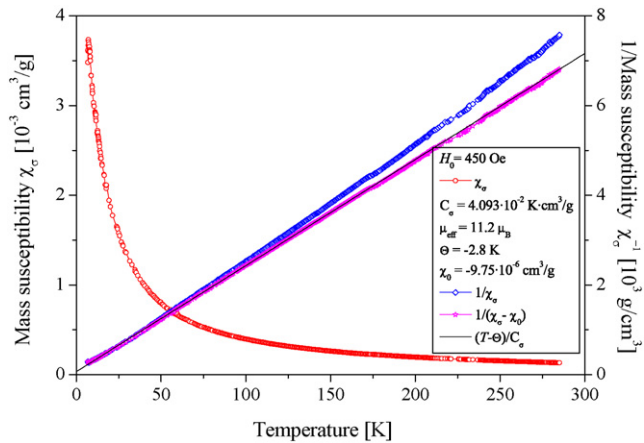


Fig. 1. Temperature dependence of the magnetic susceptibility (left-hand scale) and inverse susceptibility (right-hand scale) as measured with a Cahn RG automatic electrobalance for DyRhSn in an external magnetic field $H_0 = 450$ Oe. In the inset, the magnetic parameters obtained from the fit according to a modified Curie–Weiss law are presented, as explained in the text.

in grams). The experimental value $\mu_{\text{eff}} = 11.2\mu_B$ is comparable to the theoretical free-ion value $\mu_{\text{eff}} = g\mu_B[J(J+1)]^{1/2} = 10.65\mu_B$ for the free Dy^{3+} ion. The temperature dependence of the AC susceptibility (Fig. 2) reveals a behavior typical for a phase transition from a paramagnetic to an antiferromagnetic state (PM \leftrightarrow AFM) at the Néel temperature $T_N = 7.5(1)$ K, which is a little higher than that reported previously [2]. Under an applied external magnetic field at temperatures below T_N , the magnetization of DyRhSn presents a typical metamagnetic behaviour with a characteristic spin-flop transition. Electrical resistivity, ρ , measurements were carried out using a conventional DC four probe technique in the

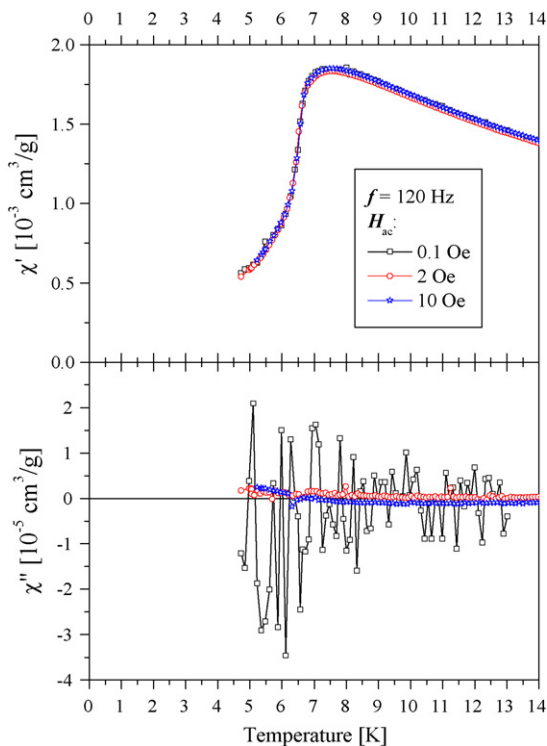


Fig. 2. DyRhSn AC susceptibilities χ' and χ'' recorded simultaneously, using a 7225 Lake Shore AC susceptometer/DC magnetometer, as a function of temperature with different amplitudes of oscillating fields H_{ac} at an internal frequency $f = 120$ Hz.

temperature range 1.9–100 K. They fully confirm a phase transition below 7.5 K.

2.2. ^{119}Sn Mössbauer spectroscopy

Mössbauer spectra of DyRhSn were obtained by means of the 23.875 keV resonance transition of ^{119}Sn . Experiments were made with a spectrometer of conventional design and a liquid helium cryostat. The velocity scale was calibrated with a $^{57}\text{Co}(\text{Rh})$ source and a metallic iron foil at room temperature. The $\text{Ca}^{119\text{m}}\text{SnO}_3$ source was kept close to 4.2 K while the temperature of the absorber was varied between 4.2 K and room temperature. Special care was taken to optimize the absorber thickness that is equal to 30.5 mg/cm². The γ -rays were detected using a 3-cm thick NaI(Tl) scintillation counter. A palladium foil of 0.05-mm thickness was used as a critical absorber for tin X-rays to reduce the background intensity of the recorded spectra.

Since the tin position in the ZrNiAl-type structure of DyRhSn has low site symmetry ($m2m$), a full hyperfine interaction Hamiltonian was applied to analyze the magnitudes of the hyperfine parameters. A transmission integral formula was used to describe the resonance lineshape. The following parameters were least-squares fitted to the resonance spectra: the isomer shift δ_{IS} , the quadrupole coupling constant $\Delta E_Q = eQV_{zz}$, the θ and φ polar angles which define the direction of the magnetic hyperfine field H_{hf} with respect to the main axis of the electric field gradient (EFG) tensor and the asymmetry parameter $\eta = (V_{xx} - V_{yy})/V_{zz}$ ($0 \leq \eta \leq 1$).

In accordance with the non-cubic point symmetry of the tin site ($m2m$), the ^{119}Sn Mössbauer spectrum recorded at $T = 293$ K, i.e. in the paramagnetic state, is perfectly well described by a single quadrupole component and the following hyperfine parameters were derived from the fitting procedure: the effective quadrupole interaction constant $|\Delta E_Q^{\text{eff}}| = |eQV_{zz}|(1 + 1/3\eta^2)^{1/2} = 1.23(1)$ mm/s, the isomer shift $\delta_{\text{IS}} = 1.72(1)$ mm/s, and the experimental absorption line widths $\Gamma = \Gamma_s + \Gamma_a = 0.89(1)$ mm/s. These data are consistent with the room temperature data given by Dwight [3].

To fit the magnetically split spectrum registered at 4.2 K (Fig. 3) in a relatively simple way several simplifications were made. Firstly, during the fitting procedure the effective quadrupole interaction constant, ΔE_Q^{eff} , was fixed to the value obtained in the paramagnetic state while the asymmetry parameter η , and the polar angles θ , φ were varied independently. In addition, the resonance linewidth Γ was fixed to the value obtained from the spectrum taken at 293 K. Secondly, similarly to the TbRhSn case [4, and references therein] it was assumed that the V_{yy} axis is parallel to the c -axis and then as a consequence the V_{zz} have to be within the ab -basal plane. Additionally, taking into account that with each of the three tin positions in the unit cell are associated three different principal systems of axes of the EFG tensor, which are equivalent after rotation around the c -axis by 120°, one can expect that a magnetically split spectrum can be composed of three subspectra (see discussion in Ref. [5]) having the same intensities, magnitudes of H_{hf} , ΔE_Q , δ_{IS} , and η parameters but different polar angles if all Dy magnetic moments are lying in the basal plane and are equal. Indeed, under the above mentioned conditions, the best fit ($\chi^2 = 1.65$) can be obtained for the magnetically split spectrum measured at 4.2 K giving the following hyperfine parameters: $H_{\text{hf}} = 9.5(1)$ kOe, $\Delta E_Q = 1.23$ mm/s = const. (as kept equal to the room temperature value), $\delta_{\text{IS}} = 1.74(1)$ mm/s, $\varphi = 0^\circ = \text{const.}$ (this corresponds to the assumption that V_{yy} is along the c -axis), $\eta = 0.57$ (2), and θ close to 60°. These results are consistent with the model used in Ref. [6] for description of the closely related TbRhSn system, where

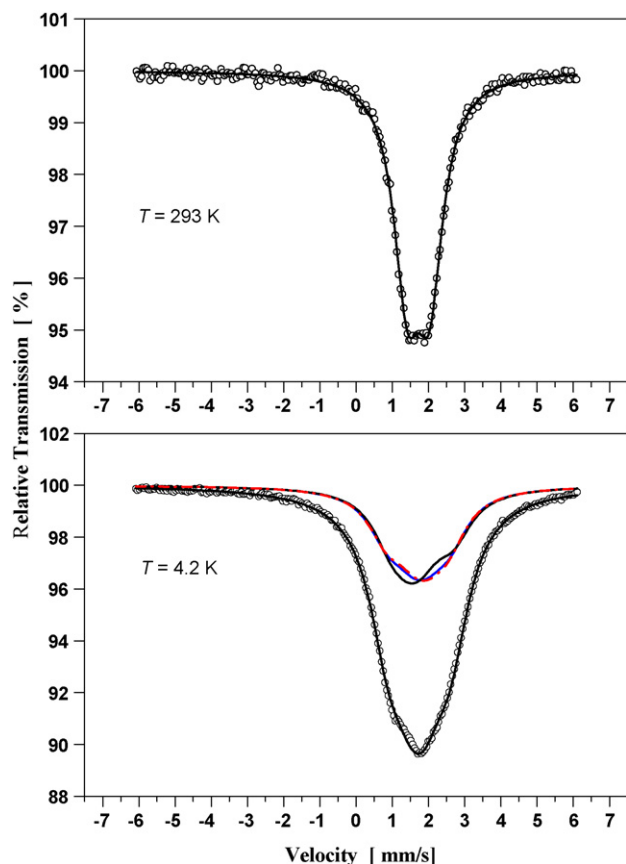


Fig. 3. ^{119}Sn resonance spectra for DyRhSn recorded at $T=293\text{ K}$, i.e. in the paramagnetic region (at the top), and at $T=4.2\text{ K}$ (at the bottom). The continuous line represents the least-squares fits to the experimental points.

also all magnetic Tb moments have equal magnitudes and lay in the **ab**-plane forming 60° , 180° , and 300° angles with the **a**-axis (i.e. in accordance with symmetry consideration giving following angles: $\theta = 60^\circ$, $\theta + 120^\circ = 180^\circ$, and $\theta + 240^\circ = 300^\circ$), as well as recent neutron results for DyRhSn [2].

The fact that the spectra observed in the paramagnetic range can be fitted with pure quadrupole interaction using only one component indicates also that our sample does not exhibit any tin impurity and that the tin atoms occupy the single positions being not distributed over RE or Rh sites in agreement with X-ray diffraction results.

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