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Bulk and local properties of intermetallic GdAgSn compound

K Łątka[1], A W Pacyna[2]

[1] M. Smoluchowski Institute of Physics, Jagiellonian University, Prof. Łojasiewicza 11, 30-348 Kraków, Poland
[2] Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences, Radzikowskiego 152, 31-342 Kraków, Poland

uflatka@cyf-kr.edu.pl

Abstract. The results of magnetic susceptibility as well as electric resistivity measurements complemented with ^{155}Gd Mössbauer spectroscopy investigations carried out within wide temperature range for GdAgSn compound are discussed.

1. Introduction

The ternary equiatomic stannides (R, A)TSn (R or A = rare earth or actinide element, respectively; T = 3*d*, 4*d* or 5*d* transition element) have intensively been studied with respect to their largely varying magnetic and electrical properties [1-3]. Among them the title compound reveals interesting magnetic behaviour as it was shown by the AC susceptibility and Mössbauer measurements at low temperatures [4, 5]. It has been found that on cooling down this compound orders first anitiferromagnetically at T_N=34.3(2) K, while at T_f = 15.3 K undergoes spin glass-like transition (SG).

2. Experimental

Our polycrystalline GdAgSn sample was synthesized by arc melting the constituent elements with purity better than 99.9% in argon atmosphere and consecutive annealing procedure for better homogeneity as previously described [4]. The X-ray powder diffraction pattern was recorded at room temperature with a Siemens D-501 diffractometer using the Ni-filtered CuK_α and the results has already been published elsewhere [4, 5]. Magnetic susceptibility and magnetization measurements were carried out with a 7225 Lake Shore AC Susceptometer/DC Magnetometer operating in AC mode on powdered samples in the temperature range 4.2 K – 200 K and in the external magnetic fields up to 60 kOe. The electrical resistivity (ρ) measurements were performed by means of Physical Property Measurement System (Quantum Design, PPMS-9T) on a bulk probe using a steady-current, standard four point technique and applying an excitation current of 80 mA in zero external magnetic field. Our specimen had of approximate dimensions 1x1x5 mm^3. (86.5 keV (I_g = 3/2, *E1*, I_e = 5/2 transition) ^{155}Gd Mössbauer spectra were collected in a transmission geometry cryostat using a conventional constant-acceleration spectrometer of the Kankeleit type. The 20 mCi ^{155}Eu:$SmPd_3$ source was kept at 4.2 K to increase the efficiency of the resonance emission. The temperature of the absorber was varied between 4.2 and 50 K with a stabilisation better than 0.05 K. In contrast to the fitting method applied in our previous investigations (where Lorentzian approximation was used [5]), for the proper description of the recorded ^{155}Gd Mössbauer spectra a least-squares fitting procedure which includes the diagonalization of the full hyperfine Hamiltonian within the transmission integral approximation was used.

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3. Results and discussion

X-ray diffraction investigations made on the polycrystalline GdAgSn sample revealed the existence of only single phase and confirmed that this compound crystallizes in the hexagonal $CaIn_2$-type of structure with space group $P6_3/mmc$ and the derived lattice parameters are following: a = 470.9(1) pm and c = 742.5(3) pm [2]. The temperature dependence for the absolute value of the magnetic AC susceptibility $\chi_{AC} = [(\chi')^2 + (\chi'')^2]^{1/2}$ and its reciprocal are displayed in figure 1.

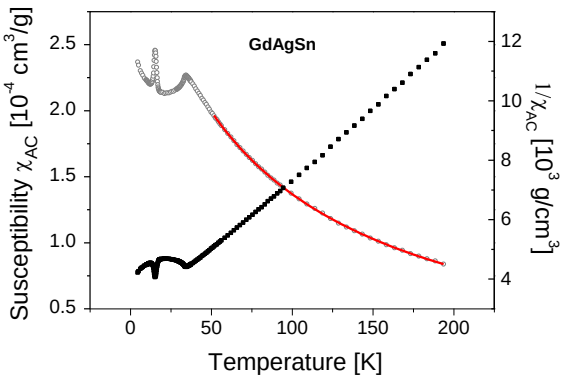

Figure 1. Temperature dependence for the absolute value of the magnetic ac susceptibility $\chi_{AC} = [(\chi')^2 + (\chi'')^2]^{1/2}$ (left-hand scale) and its reciprocal (right-hand scale) recorded for GdAgSn in zero external magnetic field at the internal frequency f = 375 Hz and the applied oscillating field strength H_{AC} = 5 Oe. The solid line is a result of the fitting procedure in the paramagnetic range, for details see the text.

The temperatures observed here for both maxima are in perfect agreement with those previously published [4, 5] i.e. a phase transition from a paramagnetic to an antiferromagnetic state (PM→AFM) takes place at the Néel temperature T_N = 34.3(2) K while the second one at T_f = 15.3(2) is that which have been ascribed to the spin-glass transition. Above 52 K (see Fig. 1), the recorded susceptibility obeys fairly well a modified Curie-Weiss law in the form: $\chi_{AC} = \chi_0 + C/(T - \theta_p)$ with the temperature independent factor $\chi_0 = -1.21 \cdot 10^{-6}$ cm^3/g, the Curie constant $C = 2.092 \cdot 10^{-2}$ Kcm^3/g, and the paramagnetic Curie temperature $\theta_p = -53.7$ K. The negative value of θ_p is indicative for antiferromagnetic correlations. The effective magnetic moment was derived from the formula $\mu_{eff} = 2.83(MC)^{1/2}$ where M is the molar mass. The experimental value μ_{eff} = 8.01 μ_B is somewhat higher but agrees well with the theoretical Russel-Saunders value $\mu_{eff} = g\mu_B[J(J+1)]^{1/2}$ = 7.94 μ_B for the free Gd^{3+} ion. Figure 2 shows the temperature variation of electrical resistivity recorded at zero external magnetic field.

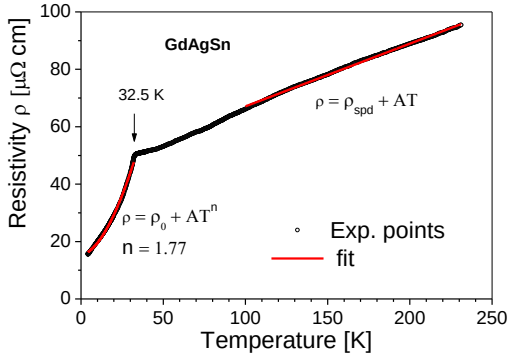

Figure 2. Temperature dependence of electrical resistivity recorded for GdAgSn. The continuous lines present the least-squares fits to the low and high temperature parts of $\rho(T)$ according to the given formulas below each part, respectively.

The linear behavior at high temperatures and the positive temperature coefficient of resistivity ($d\rho/dT>0$) indicate the metallic nature of the compound. The striking feature is a lack of any resistivity anomaly around T_f =15.3 K while a drastic change of resistivity slope is registered at T = 32.5 K, i.e. close to T_N = 34.3. It has to be underlined here, that such an observation is in line with the proposed spin glass-like character of GdAgSn below T_f and its ordered antiferromagnetic character below T_N [4,5]. As a matter of fact, the transport properties do not reveal any transition at spin glass temperature but only a gradual transformation which is associated with the lack of ordered state (see [6]). Below T_N we have found that the best fit to the experimental data in an expanded range (see Fig. 2.) may be approximated by the simple power low in the form: $\rho(T) = \rho_0 + AT^n$ with the ρ_0 = 15.44 μΩcm, A = 0.069 μΩcm/K$^{1.77}$ and n = 1.77. At high temperatures ($T>T_N$) the resistivity $\rho_{mag}(T) \equiv \rho_{spd}(T)$ = constant, where ρ_{spd} is the spin-disorder contribution and the phonon contribution $\rho_{ph}(T)$ is fairly well linear function of T [7]. Thus, in this temperature region the formula: $\rho(T) = \rho_{spd} + AT$ can be fitted. The estimated in such a way values of ρ_{spd} and A are 45.27 μΩcm, and 0.218 μΩcm/K, respectively.
Low temperature ^{155}Gd Mössbauer spectra obtained around and below T_N for GdAgSn can be fitted with the single magnetic hyperfine component rather well, and no drastic change can be registered in the shape of these spectra below T_f in spite of expected field distribution characteristic for spin glass state. The obtained quadrupole interaction constant $\Delta E_Q = eQ_gV_{zz}$ = –1.592(2) mm/s can be easily converted into value of the electric field gradient (EFG) V_{zz} at the gadolinium nuclei by the formula: $V_{zz} = 2.2206 \cdot 10^{21}\ \Delta E_Q$ [mm/s] V/m^2 = –3.535(4)x10^{21} V/m^2. Furthermore, knowledge of the EFG at the Gd site allows an estimation of the quadrupolar term B_2^0 in the Stevens expansion of the crystal field Hamiltonian: $\sum B_n^m O_n^m$. This term is directly related to ΔE_Q(^{155}Gd) by the expression:

$$B_2^0\,[\mathrm{K}] = -\alpha_J * \langle r^2\rangle_{4f} * 90.2 * \Delta E_Q\ (^{155}\mathrm{Gd})$$

Here, α_J is the appropriate Stevens factor, the mean squared radius $\langle r^2\rangle_{4f}$ is expressed in atomic units and ΔE_Q in mm/s. Under assumption that B_2^0 is the leading term in the crystal field Hamiltonian, then when translated for other rare earth ions in the family of isostructural RAgSn compounds, where the rare earths metals have non-zero angular momentums its sign gives information about the magnetocrystalline anisotropy, since then H_2^0 (CEF) = B_2^0 $[3J_z^2 - J(J+1)]$. For a negative B_2^0 the ground state doublet is built up of states with a maximum absolute value of J_z which means that the magnetic moment is pointed in the direction of the z-quantization axis. In contrary, for positive values of B_2^0 only states with the possible smallest $|J_z|$ contribute to the electronic ground state and the magnetic moment is aligned in the basal plane ($\perp z$).

Conclusions

All carried measurements confirm very well the transition observed at T_N, however, the transition at T_f has no noticeably influence on the shape of ^{155}Gd spectra what could be explained by an assumption that the local nearest-neghbour Gd^{3+} spin configuration below T_f is not significantly modified.

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