# Magnetic properties of RAuSn compounds

S. Baran[a], J. Leciejewicz[b], M. Ślaski[c], P. Hofmann[d], A. Szytuła[a],*

[a]*Institute of Physics, Jagellonian University, Reymonta 4, 30-059 Kraków, Poland*
[b]*Institute of Nuclear Chemistry and Technology, Dorodna 1, 03-169 Warszawa, Poland*
[c]*School of Physics and Space Research, University of Birmingham, Birmingham, England*
[d]*BENSC, Hahn-Meitner Institute, Berlin-Wannsee, Germany*

## Abstract

Neutron diffraction and magnetometric data show that RAuSn compounds (R=Gd–Ho) with the hexagonal LiGaGe-type crystal structure order antiferromagnetically at low temperatures. Their magnetic structures are described by the wave vector **k**=[1/2,0,0]; the magnetic moments are normal to the hexagonal axis in TbAuSn and DyAuSn and parallel to it in HoAuSn. ErAuSn is cubic (MgAgAs-type of crystal structure). The long range magnetic ordering is not observed at $T$=1.6 K.   © 1998 Elsevier Science S.A.

*Keywords:* Rare earth compounds; Antiferromagnetism; Magnetic order

## 1. Introduction

Lanthanide ternary stannides with the composition RAuSn crystallize in the hexagonal structure [1] except for ErAuSn which crystallizes in the cubic unit cell. The magnetic measurements indicate that CeAuSn ($T_N$=4 K [2]) and GdAuSn ($T_N$=35 K [3]) are antiferromagnets. Mössbauer effect measurements on [155]Gd and [119]Sn performed for GdAuSn compound suggest the presence of a long range magnetic order at low temperatures [4]. In this work the results of the magnetometric and neutron diffraction studies of the ternary RAuSn (R=Gd–Ho) compounds are reported.

## 2. Experiments

Polycrystalline samples of RAuSn compounds were obtained by arc melting under purified argon atmosphere of high purity constituent elements in the stoichiometric ratio 1:1:1. The samples contained in the evacuated silica tubes were annealed at 800°C during 4 days.

X-ray powder data were collected at room temperature using DRON 3 diffractometer (Cu $K_\alpha$ radiation). They show that all samples except for ErAuSn have the hexagonal structure.

Magnetic properties were measured using a SQUID (in low magnetic field) and vibrating magnetometer (in high magnetic field up to 120 kOe) in the temperature range 4.2–300 K.

The neutron diffraction experiment for RAuSn (R=Tb–Er) compounds was performed at BENSC Hahn-Meitner-Institute (Berlin) with E6 diffractometer ($\lambda$=2.382 Å) in the temperature range 1.6–17 K. The neutron diffraction data were analysed using the Rietveld technique of the FULLPROF program [5].

## 3. Results

The temperature dependence of the magnetic susceptibility of RAuSn compounds, where R is Gd–Er, holds at low temperatures the maximum typical of the change of magnetic structure from an antiferro- to paramagnetic phase for R=Gd–Ho (see Fig. 1). The values of the Néel temperatures are listed in Table 1. Above 50 K the magnetic susceptibilities of all RAuSn compounds obey the Curie–Weiss law (see Fig. 2) with the negative values of paramagnetic Curie temperatures $\theta_p$ and the values of the effective magnetic moments $\mu_{eff}$ which are close to the free $R^{3+}$ ion values (see Table 1).

Fig. 3 displays the magnetization curves recorded at 4.2 K in the external field up to 120 kOe. They smoothly rise as a function of the external magnetic field. For HoAuSn the presence of two step metamagnetic transitions induced by the external field is observed.

The neutron diffraction data obtained at the temperatures

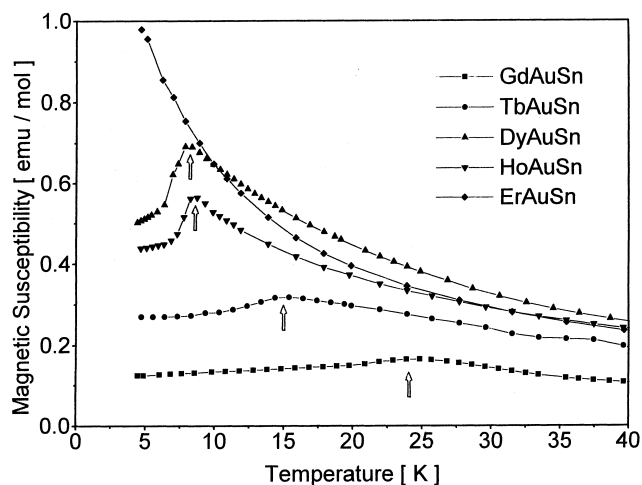*Corresponding author. Tel.: +48 12 336377/546; fax: +48 12 337086; e-mail: szytula@if.uj.edu.pl

Fig. 1. Temperature dependence of the magnetic susceptibilities of RAuSn compounds (R=Gd–Er) at low temperatures.
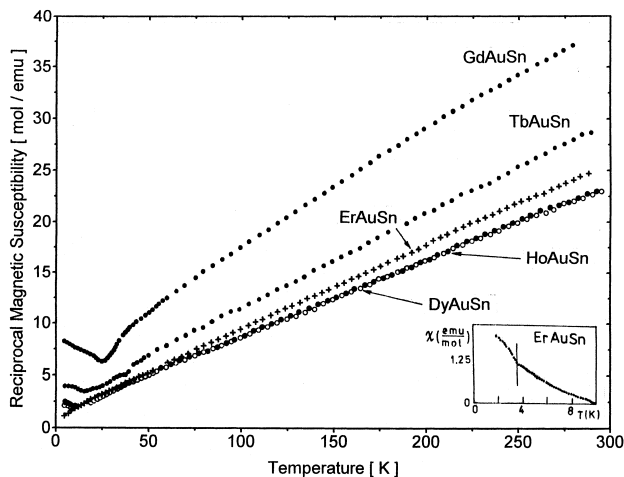


Fig. 2. Reciprocal magnetic susceptibilities $\chi^{-1}$ (measured in the external field $H=10$ Oe) vs temperature curves for RAuSn compounds (R=Gd–Er).

above the respective Néel points indicate that RAuSn compounds crystallize in the hexagonal LiGaGe-type (space group $P6_3mc$) for R=Gd–Ho and the cubic MgAgAs-type (space group $F\bar{4}3m$) for R=Er.

Neutron diffractograms recorded at 1.6 K for RAuSn compounds (except for ErAuSn) disclose the presence of additional peaks of a magnetic origin (see Fig. 4) which are indexed with the wave vector $\mathbf{k}=(1/2,0,0)$, or with an orthorhombic unit cell: $a_o=\sqrt{3}a_h$, $b_o=a_h$, $c_o=c_h$ ('o' and 'h' refer to the orthorhombic and hexagonal cells, respectively).

In the orthorhombic unit cell the magnetic moments localized on the lanthanide ions occupy the sites with the following coordinates: $S_1(0,0,1/4)$; $S_2(0,0,3/4)$; $S_3(1/2,1/2,1/4)$; $S_4(1/2,1/2,3/4)$.

The group theory considerations [6] point to the three possible antiferromagnetic ordering schemes:

$$A = S_1 + S_2 - S_3 - S_4; C = S_1 - S_2 + S_3 - S_4; G = S_1 - S_2 - S_3 + S_4.$$

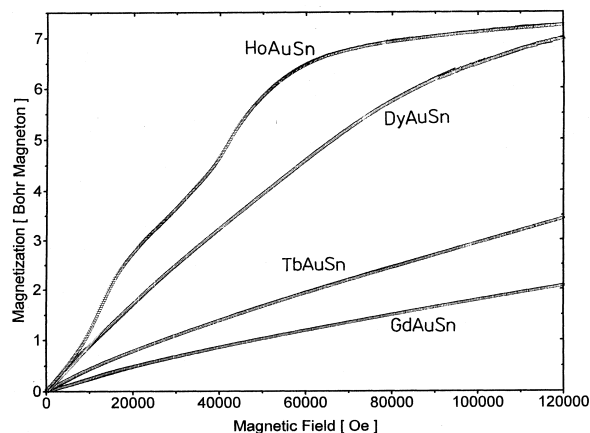The best agreement with the experimental data gives the A



Fig. 3. Magnetization vs magnetic field strength curves recorded at $T=4.2$ K.

mode with the magnetic moment normal to c-axis for TbAuSn and DyAuSn and parallel to c-axis for HoAuSn. The values of the magnetic moment are smaller that the $R^{3+}$ free ion values (see Table 1).
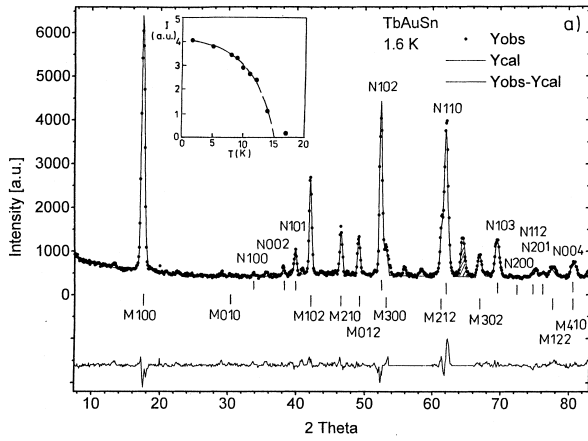
Table 1
Magnetic data for RAuSn compounds

| R | $T_N(K)$ | | $\theta_p(K)$ | $\mu_{eff}$ ($\mu_B$) | | $\mu(\mu_B)$ | | | $H_{Cr}(kOe)$ |
|---|---|---|---|---|---|---|---|---|---|
| | $M^a$ | ND | | Exp. | Theor. | Exp. | | Theor. | |
| | | | | | | $M^b$ | ND | | |
| Gd | 24 | – | −46.7 | 8.42 | 7.94 | 2.1 | – | 7 | |
| Tb | 15 | 15 | −21.4 | 9.31 | 9.72 | 3.5 | 5.7 | 9 | |
| Dy | 8 | – | −10.2 | 10.33 | 10.65 | 7.0 | * | 10 | |
| Ho | 9 | 9 | −14.3 | 10.35 | 10.61 | 7.3 | 6.6 | 10 | 10, 35 |
| Er | – | – | −11.1 | 9.83 | 9.85 | – | – | 9 | |

$M^a$ - SQUID measurements results.
$M^b$ - magnetic measurements at $T=4.2$ K and $H=120$ kOe.
* - Neutron diffraction data for DyAuSn give the very small value of the Dy magnetic moment as equal to 3.9(2) $\mu_B$.

Fig. 4. Observed and calculated neutron diffraction patterns for TbAuSn at $T = 1.6$ K. The open squares represent the observed points while the solid lines - the calculated profile and the difference between observed and calculated data (below). The upper row of marks corresponds to the reflections related to the crystal structure of the hexagonal LiGaGe-type. The lower row corresponds to the magnetic structure described in the orthorhombic unit cell. The shaded peaks arise from an impurity. The insert shows the temperature dependence of magnetic peak intensity.



Fig. 5. The Néel $T_N$ and paramagnetic $\theta_p$ temperatures of RTSn (R - heavy rare earth metal, T = Cu, Ag, Au) compounds as compared with the de Gennes function. The solid lines represent the de Gennes functions for RCuSn, RAgSn and RAuSn as normalized with respect to GdCuSn, GdAgSn and GdAuSn.

The temperature variation of the strongest magnetic peak intensities gives the Néel temperature which is in fair agreement with the data derived from magnetic measurements (see Table 1).

Only one broad and weak magnetic peak has been observed on the diffractogram of ErAuSn taken at 1.6 K. Thus it is not possible to determine the magnetic structure of this compound.

## 4. Discussion

In the RAuSn compounds, as in the RCuSn and RAgSn compounds, the $R^{3+}$–$R^{3+}$ interionic distances are long enough (~3.6 Å along c-axis and ~4.5 Å in the basal plane) to exclude any direct magnetic interactions. One may assume therefore that the magnetic ordering observed at low temperatures is due to the long range magnetic interactions of the RKKY type. If so, the paramagnetic Curie $\theta_p$ and the Néel $T_N$ temperatures should be proportional to the de Gennes function $G = (g-1)^2 J(J+1)$ [7]. Fig. 5 shows these dependencies for RTSn (R - heavy rare earth metal, T = Cu, Ag, Au). The de Gennes scaling is obeyed only partially. The observed difference can result from the crystalline electric field effect [9,10]. The crystal electric field effect hamiltonian for the hexagonal point symmetry is given by the formula

$$H_{CEF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^3 O_4^3 + B_6^0 O_6^0 + B_6^3 O_6^3 + B_6^6 O_6^6$$

where $B_n^m$ are the CEF parameters, $O_n^m$ represent the polynomials of the angular momentum operators [8]. The
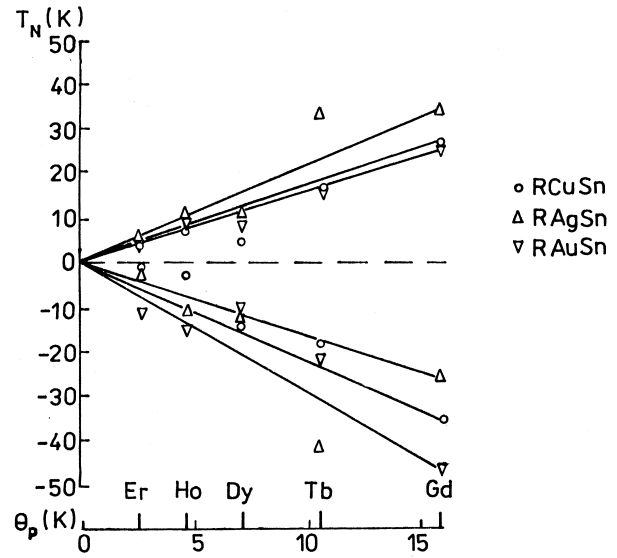
$B_n^m$ parameters were determined only for CeTSn (T = Cu, Ag, Au) compounds. The obtained results show that the $B_2^0$ parameter is predominant [11]. When the $B_2^0$ is positive the magnetic moment is perpendicular to the c-axis, when it is negative the magnetic moment is parallel to the c-axis. The change in the direction of the magnetic moment observed from R = Dy to R = Ho indicates the change in the sign of the $B_2^0$ parameter with an increase in the number of 4f electrons.

The magnitudes of ordered magnetic moments observed at 1.6 K on the heavy rare earth ions are much smaller than the free ion values (see Table 1). One can conclude that this is also caused by the crystal field effects.

## Acknowledgements

## References

[1] A.E. Dwight, in: C.E. Lundin (Ed.), Proceedings of the 12th Rare Earth Research Conference, Denver Research Institute, Denver, 1976, p. 480.

[2] M. Lenkewitz, S. Corsépius, G.R. Stewart, J. Alloys Compd. 241 (1996) 121.

[3] H. Oesterreicher, J. Less-Common. Met. 55 (1997) 131.

[4] D. Bialić, R. Kruk, R. Kmieć, K. Tomala, J. Alloys Compd. 257 (1997) 49.

[5] J. Rodriguez-Carvajal, Physica B 192 (1993) 55.

[6] E.F. Bertaut, Acta Crystall. A 24 (1968) 217.

[7] P.G. de Gennes, J. Phys. Radium 23 (1962) 510 also p. 630.

[8] M.T. Hutchings, Solid State Phys. 16 (1964) 227.

[9] J.E. Gredan, V.U.S. Rao, J. Solid State Chem. 6 (1973) 387.

[10] J.E. Gredan, V.U.S. Rao, J. Solid State Chem. 8 (1973) 386.

[11] D.T. Adroja, B.D. Rainford, A.J. Neville, J. Phys. Condens. Matter 9 (1997) L391.