



Crystal structure and magnetic properties of $R_2Co_3Al_9$ compounds ($R=Y, Pr, Gd, Tb, Dy, Ho, Er, Tm$)

Ch. Routsis*, J.K. Yakinthos

Democritos University of Thrace, Electrical and Computer Engineering Department, Physics Laboratory, 67100 Xanthi, Greece

Abstract

The crystal structure and the magnetic properties of the ternary $R_2Co_3Al_9$ ($R=Y, Pr, Gd, Tb, Dy, Ho, Er, Tm$) alluminides have been studied from 2 to 300 K under magnetic fields up to 5.5 T. $Y_2Co_3Al_9$ is a Pauli paramagnet while $Pr_2Co_3Al_9$ orders antiferromagnetically at low temperature. $Gd_2Co_3Al_9$ presents two characteristic anomalies around 100 and 20 K. This also occurs with $Tb_2Co_3Al_9$, around 15 and 100 K. $Dy_2Co_3Al_9$, $Ho_2Co_3Al_9$, and $Er_2Co_3Al_9$ order ferromagnetically with low Curie temperatures and $Tm_2Co_3Al_9$ remains paramagnetic down to 2 K. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Crystallographic and magnetic properties of the rare earth cobalt gallides have been the subject of our systematic investigations [1–6]. $RCo_{5-x}Ga_x$ ($x=1,2$) compounds (CaCu₅ type structure) show ferromagnetic behavior (light rare earth), while those with the heavy rare earth are ferrimagnetic [1–5]. R_2CoGa_3 compounds ($R=Gd, Tb, Dy, Ho, Er$ and Y) crystallize in the hexagonal $P6_3/mmc$ space group [7] and order ferromagnetically with relatively low Curie temperatures (3 K (Er) < T_c < 50 K (Gd)) [6]. Finally $RCoGa_4$ compounds ($R=Ce, Pr, Nd$) crystallize in the orthorhombic $Cmcm$ space group [8] and present rather paramagnetic behavior down to 4.6 K [2]. As a continuation of the above study, we report in this paper the crystal structure of $R_2Co_3Ga_9$ (actually $R_{0.66}CoGa_3$) and $R_2Co_3Al_9$ compounds and the magnetic properties of $R_2Co_3Al_9$ compounds.

2. Experimental

All of the samples reported here, were prepared from high purity elements by arc melting under an argon atmosphere and then annealed at 900°C for 10 days to ensure homogeneity. X-ray analysis showed that the compounds $R_2Co_3Al_9$, with $R=Y, Gd, Tb, Dy, Ho, Er$ and Tm were single-phase. Using the Lazy Pulverix program,

the lattice parameters were determined on the basis of the orthorhombic $Cmcm$ space group and are given in Table 1.

The atom positions are similar to that of the isotypic compound $Y_2Co_3Al_9$ [9], the first alluminide identified with the $Y_2Co_3Ga_9$ structure type [10].

The as cast and after annealing $Pr_2Co_3Al_9$ crystallize in the orthorhombic $LaCoAl_4$ type of structure (space group $Pnma$) [11] and $Nd_2Co_3Al_9$ was identified as two-phase mixture.

Powder X-ray diffraction patterns for all $R_2Co_3Ga_9$ compounds, do not confirm the crystal structure suggested by Yu.N. Grin et al. [10]. The crystal structure of the $CeCoAl_4$ and $PrCoAl_4$ (orthorhombic structure, space group $Pnma$) looks as more likely [12,13]. Therefore, in order to clarify the space group and atom positions, more investigation is desirable for the $R_2Co_3Ga_9$ compounds.

The magnetic measurements were performed by a SQUID Magnetometer in the temperature range from 2 to 300 K, for fields up to 5.5 Tesla.

Table 1
Crystallographic data for the $R_2Co_3Al_9$ compounds

R	Lattice parameters (Å)		
	a	b	c
Gd	12.757(2)	7.570(5)	9.450(2)
Tb	12.745(3)	7.510(7)	9.3709(3)
Dy	12.740(3)	7.522(8)	9.411(4)
Ho	12.746(4)	7.511(5)	9.380(3)
Er	12.738(3)	7.509(7)	9.378(4)
Y	12.740(5)	7.523(8)	9.411(3)

*Corresponding author.

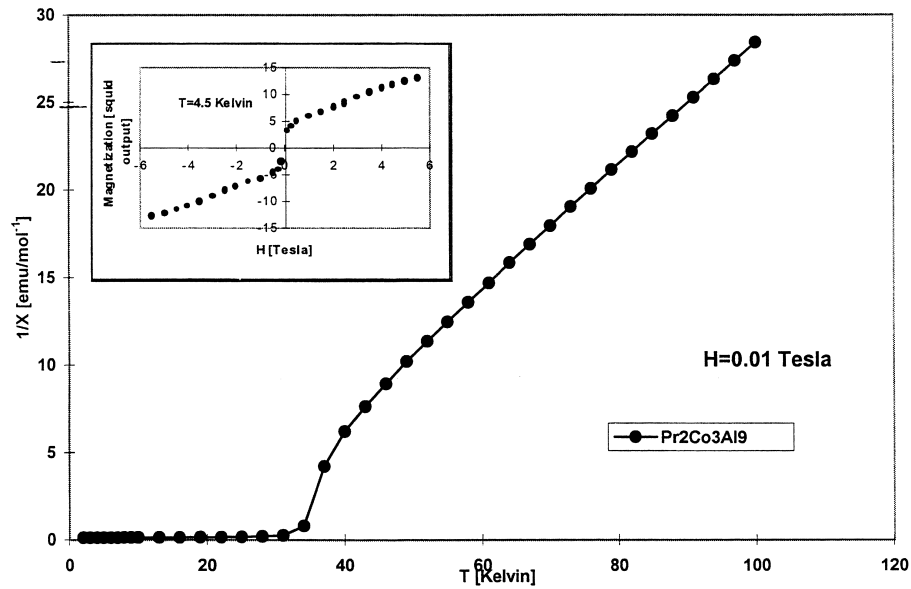


Fig. 1. Inverse magnetic susceptibility for $\text{Pr}_2\text{Co}_3\text{Al}_9$. Insert field dependence of the magnetization at 4.5 K.

3. Results

The $\text{Y}_2\text{Co}_3\text{Al}_9$ compound is a weak Pauli paramagnet with a small susceptibility that is only slightly temperature dependent. In these compounds, the cobalt atoms do not carry any magnetic moment.

$\text{Pr}_2\text{Co}_3\text{Al}_9$ compound orders antiferromagnetically at $T_N=37$ K (Fig. 1). The result is in agreement with similar results obtained on approximately the same composition PrCoAl_4 compound [13]. Curie–Weiss behaviour is reasonably well followed above 40 K. Inset of Fig. 1 presents hysteresis cycle for this compound.

In the case of the Gd sample, we observed two well defined magnetic transitions, one at 25 K and the other around 130 K (Fig. 2). The field dependence of the magnetization at 4.5 K is shown in the inset of Fig. 2.

A jump is observed at about 15 K in the magnetic susceptibility curve for $\text{Tb}_2\text{Co}_3\text{Al}_9$ compound associated with a phase transition (Fig. 3). A second change in the slope occurs at about 100 K. Similar compound Tb_2CoGa_3 orders ferromagnetically with relatively low T_c temperature (27 K) [14]. The anomaly at low temperature in the $\chi(T)$ curve could be associated with the ordering temperature. There is no hysteresis in the magnetization curve, the

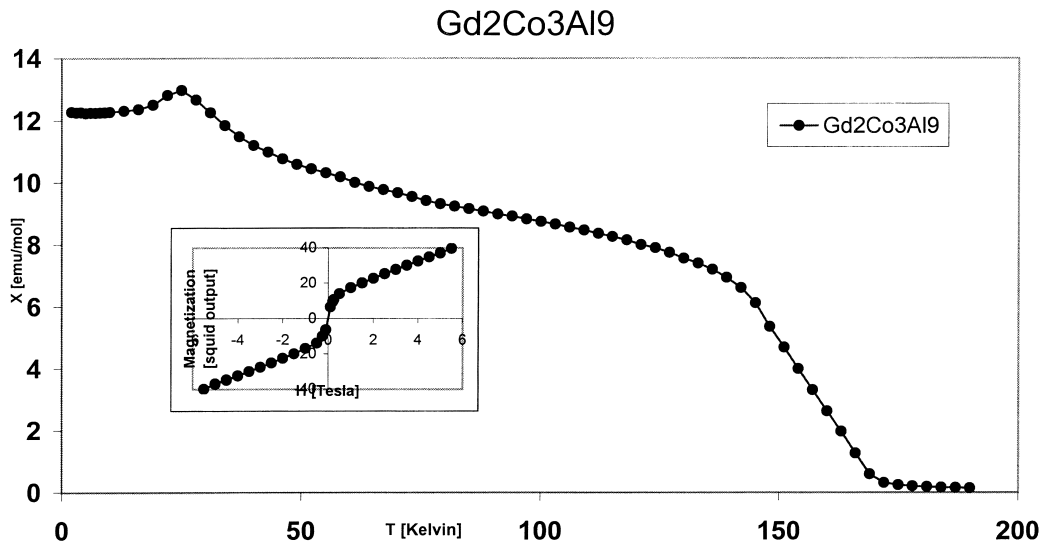


Fig. 2. Magnetic susceptibility for $\text{Gd}_2\text{Co}_3\text{Al}_9$. Insert field dependence of the magnetization at 4.5 K.

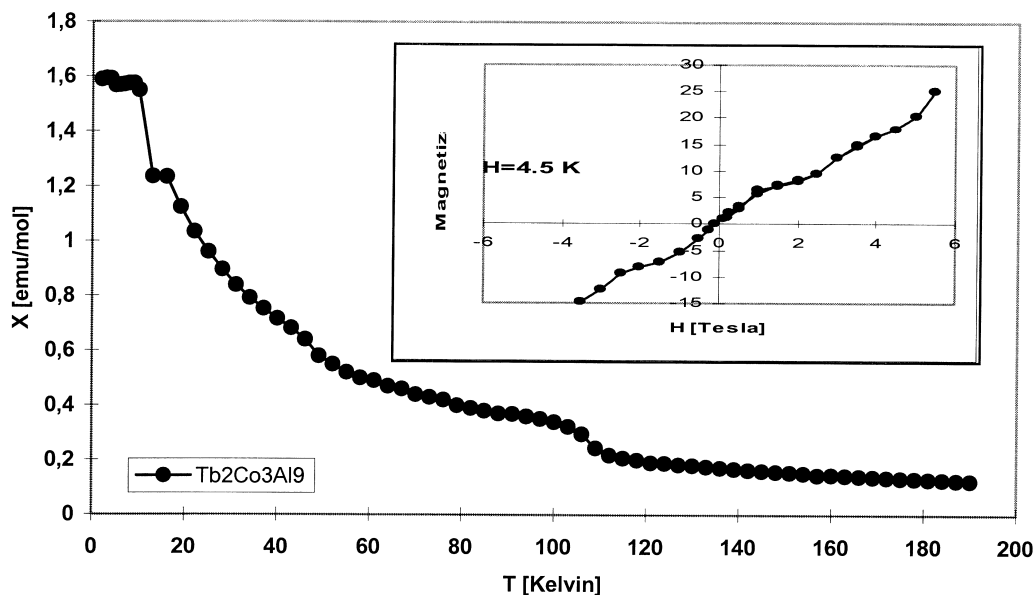


Fig. 3. Magnetic susceptibility for $\text{Tb}_2\text{Co}_3\text{Al}_9$. Insert field dependence of the magnetization at 4.5 K.

measurements with increasing and decreasing field being coincident. This curve exhibits two transitions at 2.5 and 5 T.

$\text{Dy}_2\text{Co}_3\text{Al}_9$, $\text{Ho}_2\text{Co}_3\text{Al}_9$ and $\text{Er}_2\text{Co}_3\text{Al}_9$ compounds order ferromagnetically with low Curie temperatures, 13, 10 and 5 K, respectively (Fig. 4). $\text{Tm}_2\text{Co}_3\text{Al}_9$ remains paramagnetic down to 2 K. The reciprocal susceptibility curves vs. T of the $\text{R}_2\text{Co}_3\text{Al}_9$ compounds ($\text{R}=\text{Dy}$, Ho , Er , Tm) follow Curie–Weiss law above about 30 K (inset of Fig. 4). The paramagnetic Curie temperatures θ_p are positive or near zero.

R_2CoGa_3 ($\text{R}=\text{Dy}$, Ho , Er) compounds present similar behaviour [6].

The nature of the magnetic transitions of the $\text{R}_2\text{Co}_3\text{Al}_9$ compounds will be the subject of neutron diffraction studies planned in the near future.

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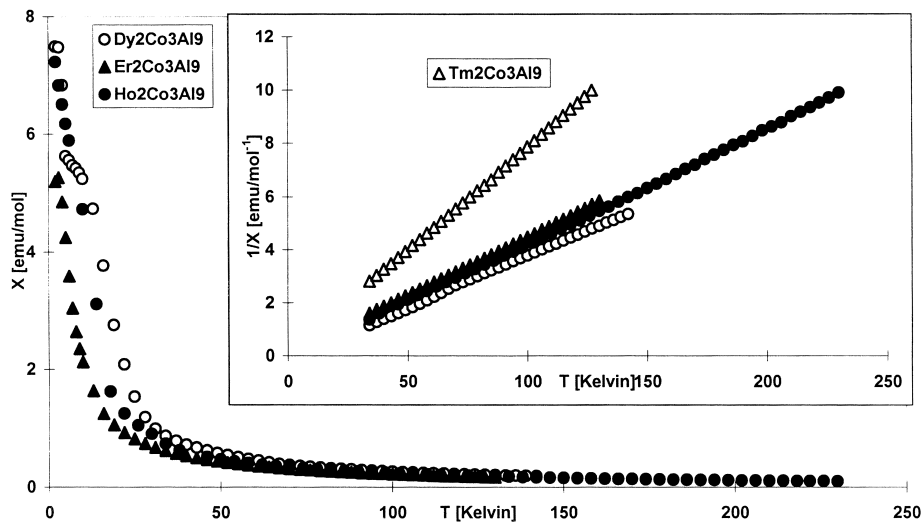


Fig. 4. Magnetic susceptibilities for $\text{R}_2\text{Co}_3\text{Al}_9$ compounds ($\text{R}=\text{Dy}$, Ho , Er) in a 0.01 T field. Insert shows inverse magnetic susceptibilities for $\text{R}=\text{Dy}$, Ho , Er and Tm .

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