

The magnetic behaviour of cobalt in $Y(\text{Co}_x\text{Al}_{1-x})_3$ compounds

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The $Y(\text{Co}_x\text{Al}_{1-x})_3$ system crystallizes in BaPb_3 -type structure for $x \leq 0.4$ and PuNi_3 -type for $x \geq 0.6$. The magnetic measurements were performed in temperature range, 4.2-800 K and external magnetic fields up to 10 T. The compounds with $x \geq 0.6$ are ferromagnetically ordered. For $x \leq 0.4$ no magnetic order, down to 4.2 K is shown. For all compositions, the thermal variations of magnetic susceptibilities obey Curie-Weiss type dependencies. Cobalt magnetic behaviour is analyzed in spin fluctuation model.

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1. Introduction

The magnetic properties of rare-earth R – transition metal (T) compounds is an interesting subject of research, both from basic and technical point of view. As a function of alloying partner and composition, the transition metals can cover a wide range of behaviour from nonmagnetic state to well defined magnetism [1]. The YCo_3 compound crystallizes in a PuNi_3 -type structure having $R\bar{3}m$ -space group [2]. In this structure, yttrium atoms are located in two and cobalt atoms in three non-equivalent sites. YAl_3 crystallizes either in Ni_3Sn -type structure having $P6_3/mmc$ space group or in BaPb_3 -type, having $R\bar{3}m$ type lattice [3,4]. The YCo_3 compound has been shown to be a typical itinerant electron metamagnet. The compound is ferromagnetically ordered. The cobalt magnetic moments are dependent on lattice sites. A mean cobalt moment of $0.41 \mu_B/\text{atom}$ was determined by magnetization studies at 4.2 K [1]. The neutron diffraction studies show values of cobalt moments of 0.55(3), 0.79(4) and $0.04(1) \mu_B$ [5]. The Co magnetic instability is the origin of the two metamagnetic transitions, at 60 T respectively 82 T, which were evidenced in magnetization curves under ultrahigh external magnetic fields. At 60 T the ground state of the Co subsystem goes from a low magnetic state to an intermediate one and than at the second transition, to a strong magnetic state [6]. This transitions can also be observed by employing the large molecular field due to magnetically ordered 4f moments in the magnetic RCo_3 compounds [7-10]. These metamagnetic transitions are believed to occur at different crystallographic Co sites and are considered to come from a special shape of density of state near the Fermi level. YAl_3 shows a Pauli-type paramagnetism [4]. The magnetic moment of cobalt can be changed by decreasing the 3d electron concentration.

The $Y(\text{Co}_x\text{Al}_{1-x})_3$ compounds in the rich cobalt region ($x \geq 0.9$) were studied by means of magnetization

in external magnetic fields up to 7 T, AC susceptibility and nuclear magnetic resonance [11]. They have conclude that the system goes from a low magnetic state ($x = 1$) to a very weak itinerant ferromagnet ($x = 0.1$). In order to a better understanding of the magnetic behaviour of $Y(\text{Co}_x\text{Al}_{1-x})_3$ compounds we analyze in this paper the effect of Co substitution by Al on the magnetic properties over a large temperature and composition range and in higher magnetic fields.

2. Experimental

The samples were prepared by arc-melting the high purity constituent elements (better than 99.9%) in a purified argon atmosphere. In order to obtain a good homogeneity, the ingots were remelted several times. A small excess of yttrium was added to compensate their loss caused by evaporation. The samples were thermally treated for one week at 900°C and then rapidly cooled to room temperature. The X-ray analysis shows that the $Y(\text{Co}_x\text{Al}_{1-x})_3$ compounds crystallize in BaPb_3 -type structure for compositions $x \leq 0.4$, while for $x \geq 0.6$ the crystal structure is of PuNi_3 -type. Two phases were evidenced in the intermediate composition range. The lattice parameters are given in Table 1.

Table 1. Lattice constants and magnetic properties of $Y(\text{Co}_x\text{Al}_{1-x})_3$.

x	0.0	0.2	0.4	0.6	0.8	1.0
a(Å)	6.195	6.197	6.201	5.011	5.016	5.020
c(Å)	21.137	21.145	21.171	24.27	24.32	24.41
$M_S(\mu_B/\text{f.u.})$	-	-	-	0.11	1.14	1.25
at T=4.2K						
$M_{\text{eff}}(\text{Co})$	-	1.48	1.88	2.42	2.63	3.30
(μ_B)						

The magnetic measurements were performed in the temperature range 4.2-800 K and external magnetic fields up to 10 T. The spontaneous magnetization, M_S , were determined from the magnetizations isotherms, according to the approach to the saturation law: $M = M_S(1 - b/H) + \chi_0 H$ where we denoted by b the coefficient of magnetic hardness and χ_0 is a field independent susceptibility. Above the Curie points, T_C , in order to avoid any possible alteration of magnetic susceptibility as result of the presence of small quantities of magnetic ordered impurities, the susceptibilities, χ , were determined from Honda-Arrrott plots according to the relation $\chi_m = \chi + dM_s'H^{-1}$ by extrapolation to $H^{-1} \rightarrow 0$, [12]. By d we denoted a presumed impurity content and M_s' is their saturation magnetization.

3. Results and discussion

Some magnetization isotherms, obtained at 4.2 K, are plotted in Fig. 1. Changes in their form are shown in the composition range $0.6 \leq x \leq 0.8$. For high cobalt content a ferromagnetic type ordering is shown while for $x = 0.6$ a mictomagnetic type behaviour is suggested. This behaviour is similar with that observed in $Y(Fe_xAl_{1-x})_3$ system [13]. We note that the spontaneous magnetizations determined for compounds with $x = 0.4$ are very small ($< 4 \times 10^{-3} \mu_B/f.u.$). In order to obtain additional information on this matter, the magnetic properties of $Y(Co_xAl_{1-x})_3$ system were analysed in a large temperature range. The spontaneous magnetization, M_S , and Curie temperatures decrease when the aluminum content increase – Fig. 2. A great variation of M_S values is shown in composition range $0.6 \leq x \leq 0.8$, the spontaneous magnetization per formula unit decreasing by $\sim 0.90 \mu_B$. Such high magnetization decrease can be correlated only with the transition from ferromagnetic to a mictomagnetic type ordering moreover confirmed by a. c susceptibility measurements. For samples with $x \leq 0.4$ no magnetic ordering was observed down to 4.2 K. Probably that in this composition range the greatest part of cobalt atoms are not magnetic and only a very small number of cobalt atoms are coupled by exchange interactions ($< 4\%$). The above behaviour can be correlated with their different local environments as a result of deviation from random substitutions of Al at different Co sites. From the field dependencies of the magnetization curves, at 4.2 K, we conclude that for $x \leq 0.4$ cobalt exhibits an exchange enhanced susceptibility.

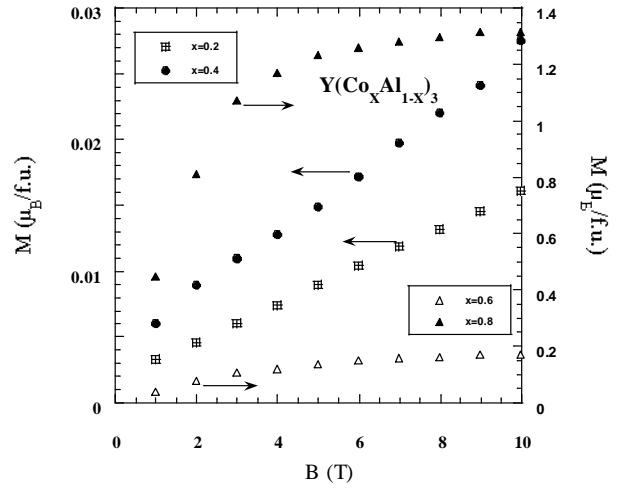


Fig. 1. Magnetization isotherms at 4.2 K.

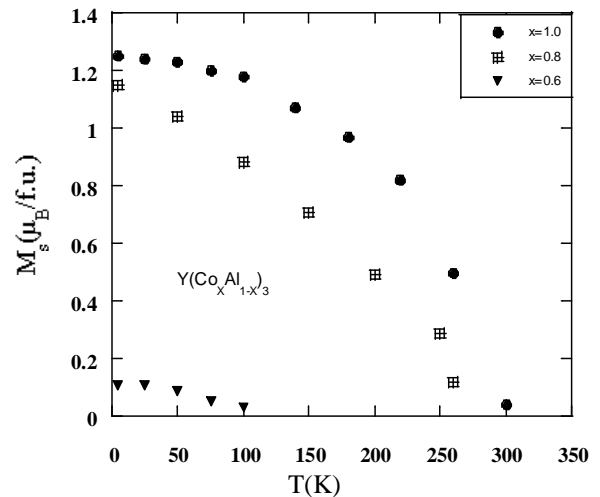


Fig. 2. Thermal variations of spontaneous magnetization.

The thermal variations of magnetic susceptibilities, χ , follow a Curie-Weiss type dependences: $\chi = C(T - \theta)^{-1}$ Fig. 3. We denoted by C the Curie constant and θ is the paramagnetic Curie temperature.

From the Curie constants, the mean effective cobalt moments, $M_{eff}(Co)$, were determined – Table 1. The $M_{eff}(Co)$ values decrease when cobalt atoms are substituted by aluminum. This suggest a gradual change of cobalt electronic configuration. The θ values change from negative for aluminum rich samples to positive values for cobalt rich compositions range.

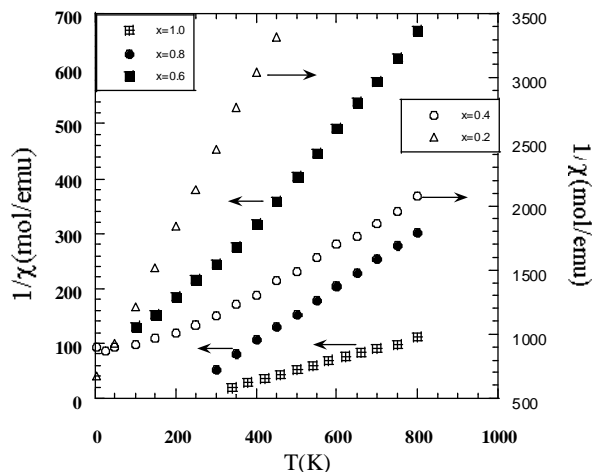


Fig. 3. Thermal variations of reciprocal susceptibilities.

In $Y(\text{Co}_x\text{Al}_{1-x})_3$ system, at 4.2 K, cobalt shows a transition from an exchange enhanced paramagnetism to magnetic state as the aluminum content decreases. In the same time, for all studied samples, the reciprocal susceptibilities follow a Curie-Weiss behaviour. The paramagnetic Curie temperatures are negative for samples where cobalt shows, at 4.2 K, an exchange enhanced magnetic susceptibility and positive for alloys where magnetic ordering is shown. The above features may be described in the self consistent renormalization (SCR) theory of spin fluctuations [14]. The model predicts that the effective moment is independent on the saturation moment at $T = 0$ K, but depends on the band structure around the Fermi energy. For a nearly or weak ferromagnet, as cobalt in the present system, the mean square of local spin fluctuation amplitude, $\langle S_{\text{loc}}^2 \rangle$, increases significantly in a narrow temperature range, leading to a Curie-Weiss type dependence of the magnetic susceptibility. Thus, the system at high temperatures, behaves as having local moments, with $M_{\text{eff}}(\text{Co}) \propto \langle S_{\text{loc}}^2 \rangle$. Changes in the band structure are suggested by the fact that $M_{\text{eff}}(\text{Co})$ varies with composition. This can be correlated with changes in crystal structure as well as to band filling effects, as result of cobalt substitution by aluminum.

4. Conclusions

In compounds with $x \leq 0.4$ mainly longitudinal components of local spin fluctuation (LSF) are present, as suggested by lack of magnetic ordering at $T = 4.2$ K as well as by the negative paramagnetic Curie temperatures. For higher cobalt content both longitudinal and transverse components of LSF are present, the contribution of transverse components increasing when the cobalt content increases.

Finally we conclude that the magnetic behaviour of cobalt in the above system seems to be well described by the spin fluctuation model.

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