MAGNETIC PROPERTIES AND BAND STRUCTURE CALCULATIONS OF $YCo_{2-x}M_x$ WITH M=Cr, Ti

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Abstract: The $YCo_{2-x}M_x$ systems with M = Cr, Ti form solid solutions having $MgCu_2$ -type structure in the composition range $x \leq 0.4$ for M = Cr and $x \leq 0.2$ for M = Ti. Magnetic measurements were performed in the temperature range 4 K – 700 K and fields up to 7 T. At low temperatures, the magnetic susceptibilities, for compounds with Ti, follow a linear T^2 dependence. At temperatures higher than a characteristic value T*, a Curie-Weiss type behavior is evidenced. Similar behavior, at high temperatures, was evidenced in $YCo_{2-x}Cr_x$ system. The experimental data were analysed in spin fluctuation model. Band structure calculations were also performed. The magnetic susceptibilities, at 4 K of $YCo_{2-x}Ti_x$, as well as their temperature coefficients, in the low temperature range, are reasonable described by using the computed density of states.

1. Introduction

The physical properties of the RM_2 intermetallic compounds, where R is a rare earth or yttrium and M a 3d transition metal, were extensively investigated [1]. The RCo_2 compounds crystallize in a cubic C15 type Laves phase structure. In this structure the R and Co atoms, respectively occupy one type of site only. Cubic Laves-phase structures are formed with M = Mn, Fe, Co and Ni, while the compounds YCr_2 and YTi_2 with this structure does not exist [2,3].
The magnetic behavior of YCo_2 exchange enhanced paramagnet is more complicated than that described by a classical Pauli type paramagnet [4-6]. At low temperatures, (T < 10 K), the magnetic susceptibilities, χ, follow a T^2 linear dependence, while at temperatures higher than a characteristic value T*, a Curie - Weiss type behavior was evidenced. The effective cobalt moment is close to that of Co^{2+} ion, considering the spin contribution only. Similar behavior was reported in RCo_2 compounds with R = Lu, Sc, Hf [4] A metamagnetic transition may be achieved for YCo_2 and $LuCo_2$ in high magnetic fields (above 60 T). The aim of the present work is to investigate the composition range in which $YCo_{2-x}M_x$ with M = Cr or Ti form solid solutions and the influence of M atoms on their magnetic properties.

2. Experimental and computing methods

Samples were prepared in purified argon atmosphere in an induction furnace. A small excess of yttrium was used in order to avoid the loss of the weight due to the evaporation of yttrium and consequently the formation of magnetically ordered phases. The samples were thermally treated in vacuum for one weak at 950 °C. The structure of the compounds was studied by X - ray, on powders, using Cu K_α radiation, at room temperature. The X - ray analysis showed the presence of one phase only, having $MgCu_2$-type structure, in composition range $x \leq 0.4$ for M = Cr and $x \leq 0.2$ for M = Ti. The lattice parameters are given in Table. 1. Magnetic measurements were performed in the temperature range 4 K - 700 K and external fields up to 7 T. For each temperature, the magnetic susceptibility has been determined from magnetization isotherms, according to Honda-

Arrott plot [7]: $\chi = \chi_p + cM_S'/H$, by extrapolation to $H^{-1} \rightarrow 0$. By c is denoted a presumed impurity content and M_S' is their saturation magnetization. By this method any possible alteration of χ values, as result of the presence of small quantities of magnetic ordered phases is avoided. Band structure calculations were made using the ab-initio tight binding linear muffin-tin orbital method in the atomic sphere approximation (TB-LMTO-ASA). In the frame of the local density approximation (LDA) the total electronic potential is the sum of the external, Coulomb and exchange correlation potentials [8-9]. The functional form of the exchange - correlation energy used in the present work, was the free electron gas parameterization of von Barth and Hedin [10]. Relativistic corrections are included without the spin-orbit coupling. Supercell calculation were performed with the dimension of supercell of 8 times a unitary cell.

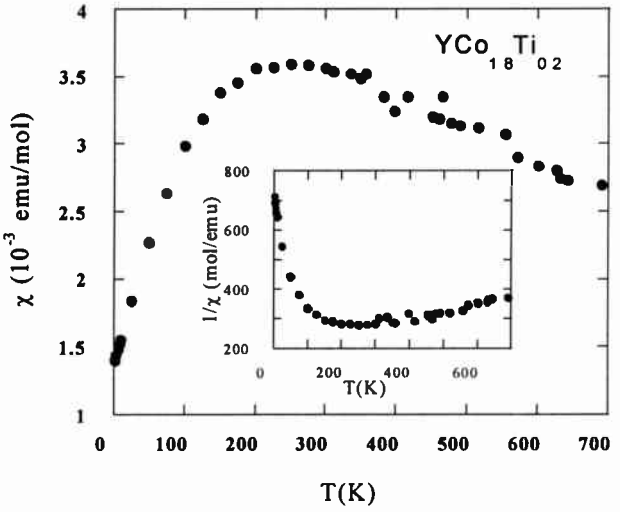

Fig. 1. Temperature dependence of magnetic susceptibilities (reciprocal susceptibilities inset) for the compound $YCo_{1.8}Ti_{0.2}$.

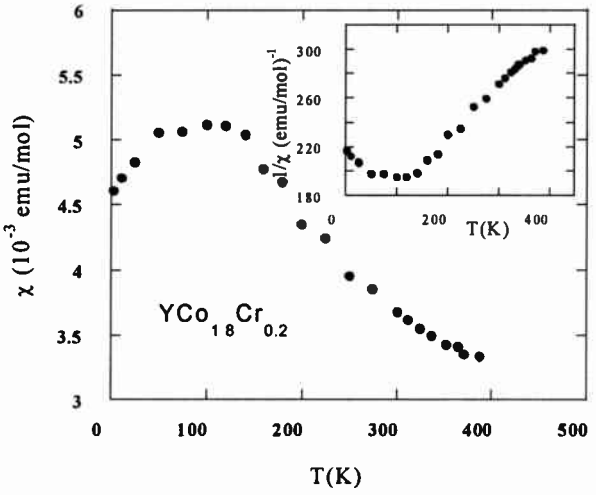

Fig. 2. Temperature dependence of magnetic susceptibilities (reciprocal susceptibilities inset) for the compound $YCo_{1.8}Cr_{0.2}$.

3. Experimental and computed data

The temperature dependences of the magnetic susceptibilities, χ, for some samples are plotted in Figs. 1 and 2. In inserts, the χ^{-1} versus T values are given. The susceptibilities increase up to a temperature T_{max}, and then decrease. Above a characteristic temperature T^*, a Curie-Weiss type behavior is shown. The low temperature magnetic susceptibilities of $YCo_{2-x}Ti_x$ (Fig. 3) first decrease as compared to the value determined for YCo_2 and then increase. In case of chromium substituted samples the low temperature magnetic susceptibilities are higher than reported for YCo_2. As example, for $YCo_{1.6}Cr_{0.4}$ the χ values are nearly two times greater. Probably, that at low temperatures there are small antiferromagnetic exchanges between Cr atoms, which lead to the increase of the susceptibility. Since for an antiferomagnet the M versus H values follow a linear dependence, the

contributions to χ values arising from the above can not be separated from the paramagnetic ones. Thus in $YCo_{2-x}Cr_x$ system reliable data can be obtained only in the high temperature region, where samples are in the paramagnetic state. In any case, if exists, the antiferomagnetic exchange interactions involving chromium are small.

At high temperatures, above a characteristic value, T^*, a Curie-Weiss type behavior was evidenced. From the Curie constants we determined the, μ_{eff} (M), effective moment per transition metal atom as well as μ_{eff} (Co) supposing that Ti and Cr do not contribute to the Curie constants. The above values are listed in Table 1.

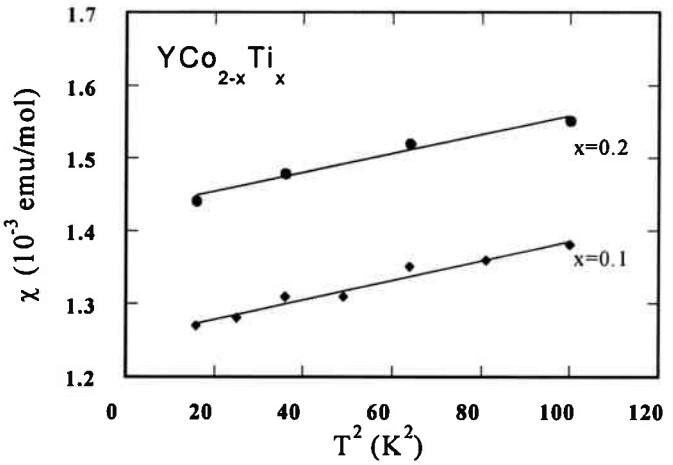

Fig. 3. T^2 dependence of magnetic susceptibilities for the $YCo_{2-x}Ti_x$ series at T<10 K.

4. Discussion

The experimental data for $YCo_{2-x}Ti_x$ as well as for $YCo_{2-x}Cr_x$ in the high temperature range may be analysed in self-consistent theory of spin fluctuations [11]. In this model, the wave number dependent susceptibility, χ_q, of a nearly ferromagnetic alloy has a large enhancement due to electron-electron interaction for small q values. The temperature dependence of χ_q is insignificant only when q values are small. The average amplitude of local spin fluctuation $\langle S^2_{loc}\rangle = 3k_B T \Sigma_q \chi_q$ is a temperature dependent quantity which increases until reaches an upper limit determined from charge neutrality condition, at a temperature T^*. If $\langle S^2_{loc}\rangle$ is saturated, the charge neutrality condition leads to an effective moment characteristic for the given electron configuration. In this case the moment is localized in q space.

As in pure metals, we suppose that in high temperature range Ti and Cr do not contribute to the Curie constants. The effective cobalt moments thus determined are close to that of Co^{2+} involving only spin contribution (~3.87μ_B).

In the low temperature range the susceptibilities for $YCo_{2-x}Ti_x$ follow a relation of the form $\chi=\chi_0(1+AT^2)$, where

$$A = \frac{\pi^2}{6}\left(2\frac{\eta''}{\eta} - 1.2\frac{\eta'^2}{\eta^2}\right)_{E_F} s^2 \qquad (1)$$

We denoted by η, η' and η'' the state density at the Fermi level, the first and second of their derivatives and s is the exchange enhancement factor.

From the computed band structure we determined the A values according to the relation (1). The values thus obtained are in reasonable agreement with experimental data - Table 1.

The magnetic susceptibilities of $YCo_{2-x}Ti_x$ first decrease and then increase when increasing titanium content. This can be correlated with the band structure calculations. The Fermi level shifts

at the beginning in a region with low density of states and then the state density at Fermi level increase. Thus, the susceptibilities at 0 K computed from band structure $\chi=\mu_o\mu_B^2\eta(E_F)$ follow the same trend as experimentally observed. There are differences between computed and experimentally determined values – Table 1. These can be attributed to the fact that in computing $\eta(E_F)$ we used the room temperature cell parameters and in addition the Co 3d correlations were not considered.

Table 1. Lattice parameters, experimental and theoretical susceptibilities at low temperatures, effective magnetic moments per transition metal and cobalt atoms, and characteristic temperatures.

Com - pound	x	a (nm)	χ_{exp} (10^{-3} emu/ mol)	χ_{th} (10^{-3} emu/ mol)	T_{max} (K)	T^* (K)	A(exp) (10^{-3} K^{-2})	A(th) (10^{-3} K^{-2})	μ_{eff}(M) (μ_B/ atom)	μ_{eff}(Co) (μ_B/ atom)
YCo_2		0.7216	1.93	2.25	230	450				3.86
$YCo_{2-x}Cr_x$	0.1	0.7234	4.256		80	180			3.65	3.75
	0.125			2.236						
	0.2	0.7238	4.663		95	200			3.6	3.8
	0.25			2.515						
	0.4	0.7235	4.179		80	250			3.53	3.95
$YCo_{2-x}Ti_x$	0.1	0.7214	1.271		275	450	1.068		3.31	3.4
	0.125			1.796				0.9961		
	0.2	0.7214	1.442		250	450	0.908		3.69	3.9
	0.25			2.046				0.8945		

Finally, we conclude that the magnetic behavior of $YCo_{2-x}M_x$ (M=Ti, Cr) can be described in spin fluctuation model. A reasonable agreement is obtained between the experimental data and computed values from band structure calculations.

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