Journal of Alloys and Compounds 478 (2009) 34–37

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Instability of the Co magnetic state in Y_2Co_7-based compounds: Effect of alloying for Y- and Co-sites

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ARTICLE INFO

Article history:
Received 22 October 2008
Received in revised form
19 November 2008
Accepted 22 November 2008
Available online 30 November 2008

Keywords:
Rare earth – 3d transition metal intermetallics
Magnetic properties
Itinerant electron metamagnetism
High field
Magnetocrystalline anisotropy

ABSTRACT

High field magnetization process has been studied in the $(Y_{1-x}Zr_x)_2Co_7$, $(Y_{1-x}U_x)_2Co_7$ and $Y_2(Co_{1-x}Al_x)_7$ systems in order to reveal the possibility of the transformation of the Co magnetic ions from the high to the low moment state. The spontaneous magnetic moment, Curie temperature and anisotropy constant have been found to decrease monotonously with increasing x in the single phase interval. The results suggest that the Co moment in present systems is unstable with respect to both electron concentration and exchange interactions. The average Co moment reduces to the value characteristic of the YCo_3 compound. However, no signatures of the formation of the low moment Co state was found in the studied Y_2Co_7-based systems.

1. Introduction

In the R-Co series (R – rare earth) of intermetallic compounds with nonmagnetic trivalent R = Y (Y_2Co_{17}, YCo_5, Y_2Co_7, YCo_3 and YCo_2), the Co moment strongly depends on the atomic ratio of the alloying elements. It is well established that for Y_2Co_{17}, YCo_5 and Y_2Co_7 the high moment state (HMS) with $\mu_{Co} \approx 1.37$–$1.66\,\mu_B$ is realized. On going to lower Co/Y ratio, magnetism of the Co atoms transforms to the low moment state (LMS) with $\mu_{Co} \approx 0.5\,\mu_B$ for YCo_3 and finally the Co becomes paramagnetic for YCo_2. Two successive field-induced metamagnetic transitions from the low magnetic state (LMS) to the high magnetic state (HMS) for YCo_3 were found at 60 T and 82 T critical fields [1]. The value of the magnetization at 50 T just before the transitions is $0.72\,\mu_B$/Co. The magnetization changes from 0.72 to $0.88\,\mu_B$/Co and from 0.88 to $1.23\,\mu_B$/Co by both transitions. Above the second transition field, the magnetization is almost completely saturated. Metamagnetic transition from LMS to HMS is also observed by the substitution of magnetic R for nonmagnetic Y due to the effective 4f–3d exchange interactions. As regards paramagnetic YCo_2, first order metamagnetic transition from paramagnetic to ferromagnetic state with $\mu_{Co} \approx 0.55\,\mu_B$ is observed at the critical field of ~70 T [2].

The observed behavior of the Co moment may be explained within the itinerant model of the Co magnetism taking into account the peculiarities of the band structure [3]. Theoretical calculations of the electronic structure of YCo_3 and Y_2Co_7 by the recursion method have shown that their density of states (DOS) profiles are similar to each other, with a sharp sub-peak above a main peak near the top of the 3d band [4]. A local minimum (a valley) is present just below the sub-peak. The magnetic moment as a function of 3d-electron concentration can be explained qualitatively in terms of the rigid band model with this DOS [3]. This model suggests that YCo_3 is weakly ferromagnetic and the Fermi level of the majority spin band, E_F^+, just below the two metamagnetic transition fields is pinned at the valley. The metamagnetic transition from LMS to HMS occurs when E_F^+ pinned at the valley jumps over the sub-peak on the application of the ultrahigh magnetic field or due to the effective 4f–3d exchange interactions. The origin of the two successive field-induced metamagnetic transitions from the LMS to the HMS for YCo_3 is attributed to the nonequivalent crystallographic sites occupied by Co atoms. (See Fig. 1 where the crystal structures of related Y-Co intermetallics are shown).

Recently, an alternative model of ground magnetic state and magnetic phase transitions in YCo_3 was suggested by Cui et al. [5]. Using a combination of neutron powder diffraction measurements

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doi:10.1016/j.jallcom.2008.11.104

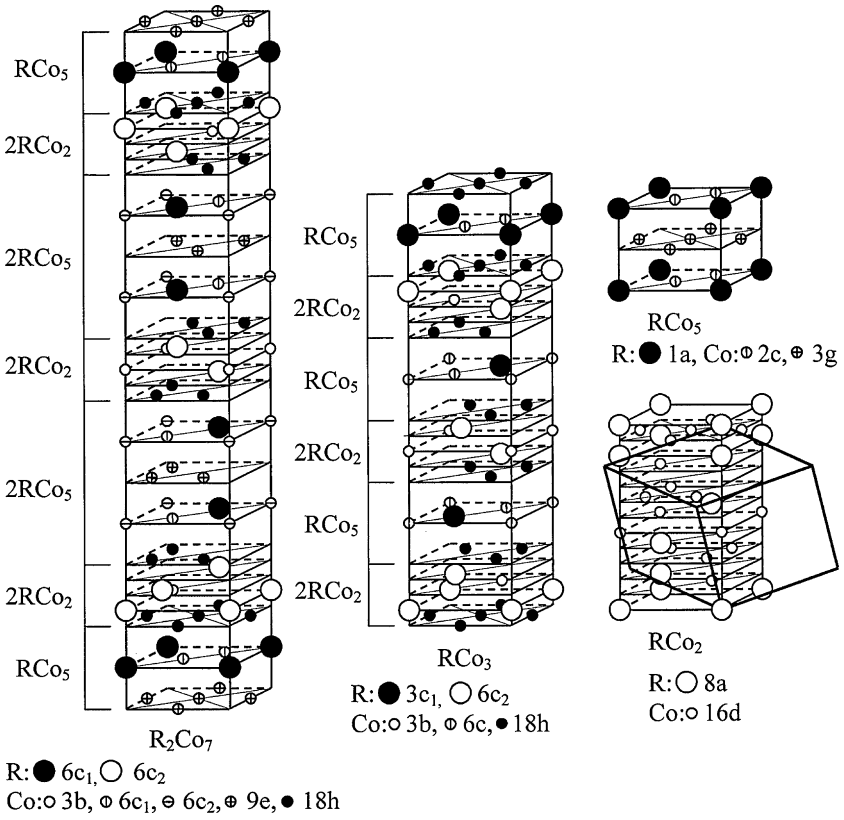

Fig. 1. Crystal structures of the rhombohedral modifications of R_2Co_7 and RCo_3 as constructed from the structural blocks RCo_5 and RCo_2 (shown to the right).

and first-principles full potential augmented plane wave calculations with inclusion of the spin–orbit coupling they concluded that the ground state of YCo_3 is ferrimagnetic with the spin of the Co_2 atoms directed opposite to those of Co_1 and Co_3 atoms. The field-induced magnetic transitions start form the ferrimagnetic phase at low temperatures and moment alignment of Co_2 atoms changes from antiparallel to nonmagnetic for the first transition and then to ferromagnetic during the second transition. Therefore, the changes in magnetization under high fields are mainly due to a spin-flip process on the Co_2 atom.

On the other hand, the Y_2Co_7 compound with slightly higher Y/Co ratio has the ferromagnetic ground state with the Co in HMS in spite of a very similar crystal structure [6]. The transition from the HMS to the LMS can be expected in Y_2Co_7-based systems with alloying for some elements. This transition may be induced by the substitution of tetravalent Zr^{4+} or U having intermediate valence higher than 5 for trivalent Y due to additional supply of electrons to 3d band. Also the concentration metamagnetic transition may be expected by the substitution of nonmagnetic metals for Co due to the weakening of 3d–3d exchange interactions. Studying the magnetic behavior of quasi-binary Y_2Co_7-based compounds may shed light on the ground state and the origin of field-induced magnetic transitions in YCo_3.

In the present study we have performed magnetization measurements on $(Y_{1-x}Zr_x)_2Co_7$, $(Y_{1-x}U_x)_2Co_7$ and $Y_2(Co_{1-x}Al_x)_7$ systems in pulsed high magnetic fields up to 40 T to reveal the mechanism of the transformation of the Co moment from the high to the low moment state in Y-Co intermetallic compounds.

2. Experimental procedure

The compounds $(Y_{1-x}Zr_x)_2Co_7$ ($x \le 0.5$) and $Y_2(Co_{1-x}Al_x)_7$ ($y \le 0.4$) were prepared by induction melting in an argon atmosphere, as well as $(Y_{1-x}U_x)_2Co_7$ ($x \le 0.3$) by arc melting, followed by annealing at 1100 °C for one week. The phase purity and the lattice parameters were determined by a standard X-ray powder diffraction using a DRON diffractometer with Fe K_α radiation. The alloys were found to be single phase (less than 3% of a secondary phase) with the rhombohedral Gd_2Co_7-type of crystal structure for the homogeneity ranges with $x = 0.25$ for $(Y_{1-x}U_x)_2Co_7$, $x = 0.4$ for $(Y_{1-x}Zr_x)_2Co_7$ and $x = 0.2$ for $Y_2(Co_{1-x}Al_x)_7$.

The Curie temperature was determined using ac susceptibility measurements. The high field magnetization curve was measured using an induction method in a pulsed magnetic field up to 40 T with a pulse duration time of about 16 ms. The anisotropy field B_a was determined by observing a singular point appearing in the field dependence of the second derivative of magnetization, d^2M/dB^2. The spontaneous magnetization M_s of the sample was determined from the linear extrapolation of the high field magnetization curve to zero field. Considering the magnetization processes of the single crystalline Y_2Co_7 [7], the anisotropy constant K_2 is negligibly small compared with K_1. Hence, the anisotropy energy calculated from $E_a = M_sB_a/2$ can be defined as K_1.

3. Results and discussion

According to our X-ray diffraction measurements, parent Y_2Co_7 compound has the lattice parameters $a = 4.995$ Å, $c = 36.145$ Å, in agreement with the previously reported data [8]. Concentration dependence of the a and c lattice parameters of all series of compounds are shown in Fig. 2. As it can be expected, the lattice parameters decrease with the Zr and U concentration due to the smaller than Y atomic radius and increase with the Al concentration due to the larger than Co atomic radius. The change of the unit cell volume $\Delta V/V$ for the maximum substituted element concentration is estimated to be −4.1% for Zr ($x = 0.4$), −3.0% for U ($x = 0.25$) and +3.2% for Al ($x = 0.2$). In the YCo_3-based compounds the metamagnetic transition from the LMS to the HMS induced by the application of magnetic field is accompanied by the significant increase of the volume magnetostriction as $\omega(H) = n_{CoCo}\ (\Delta M_{Co})^2$ with the typical value of magnetoelastic coefficient for itinerant metamagnetic compounds $n_{CoCo} = (5.8 \pm 1.3) \times 10^{-3}\ (\mu_B/Co)^{-2}$ [9]. Hence, the decrease of the unit cell volume in $(Y_{1-x}Zr_x)_2Co_7$ and $(Y_{1-x}U_x)_2Co_7$ systems with Zr or U concentration makes favorable transition from the HMS to the LMS. On contrary, for the