MAGNETIC ANISOTROPY AND EXCHANGE INTERACTIONS IN $R_{n+1}Co_{3n+5}B_{2n}$ COMPOUNDS*

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The individual Co site contributions to the magnetocrystalline anisotropy in RCo_5, RCo_4B and $R_3Co_{11}B_4$ compounds have been studied. An analysis is given of the room temperature anisotropy constants of these compounds by using a model of Streever. The values of the rare earth–transition metal exchange coupling J_{RCo}/k_B derived by mean field analysis of the Curie temperatures are -11.2 K, -15 K and -20.9 K, respectively. In general, a decrease in J_{RCo}/k_B with increasing Co concentration is observed.

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1. Introduction

The $R_{n+1}Co_{3n+5}B_{2n}$ type compounds formed between rare earth (R), cobalt and boron are an important and physically interesting class of materials. Compounds in this series are known to exist for $n = 0$ (RCo_5), $n = 1$ (RCo_4B), $n = 2$ ($R_3Co_{11}B_4$), $n = 3$ ($R_2Co_7B_3$) and for $n \to \infty$ (RCo_3B_2). These structures are based on the well-known $CaCu_5$ structure. The unit cells are formed by alternative stacking of one layer of RCo_5 and n layers of RCo_3B_2 unit cells. They all have the hexagonal symmetry and belong to the space group of $P6/mmm$ [1]. These compounds exhibit a strong axial anisotropy for R = Sm [2]. Smit et al. [3] determined the crystal field parameters by measuring Mössbauer spectrum of $Gd_{n+1}Co_{3n+5}B_{2n}$ compounds. The strength of the individual contributions to the magnetic anisotropy and magnetic moments of the non-equivalent Co sites in these compounds was studied using nuclear magnetic resonance (NMR) [4, 5] and neutron diffraction [6].

In the present investigation, the results of magnetocrystalline anisotropy and exchange interactions study of the RCo_5, RCo_4B and $R_3Co_{11}B_4$ compounds are presented. A model proposed by Streever [4] is used to analyse the room temperature anisotropy constants of these compounds.

2. Results and discussion

The bulk anisotropy of rare earth–transition metal compounds arises from two contributions, the $4f$ sublattice and $3d$ sublattice anisotropies.

At high temperature, because the R single ion anisotropy is rather small, the easy magnetization direction of RCo$_5$ compounds is dominated by magnetocrystalline anisotropy of Co sublattice. Streever [4] indicated that the spin–orbit magnetic moment of Co atoms at the (2c) crystallographic site contributes significantly to the anisotropy of RCo$_5$ compounds. The uniaxial easy magnetization direction of RCo$_5$ arises from the Co atoms in the (2c) site wheras the Co atoms in the (3g) site exert relatively small reverse contribution [4]. Streever suggested that the anisotropy of the Co sublattice in RCo$_5$ compounds could be affected by the spin–orbit coupling. The spin–orbit coupling can make an important contribution to the Co anisotropy in the RCo$_5$ compounds due to large orbital moments. A stabilization energy or local anisotropy energy per atom E_s was defined by Streever as the difference of the spin–orbit coupling energies in directions parallel and perpendicular to the hexagonal c-axis. In the literature [4], Streever also calculated the anisotropy E_a of the SmCo$_5$ compound.

In the SmCo$_5$ compound, the E_s values of the Co atoms in the Co$_1$(2c) and Co$_2$(3g) sites may be taken approximately as 25 and -10 cm^{-1}/atom, respectively. These values were supported by the Co NMR studies in RCo$_5$ compounds [4]. The NMR studies showed that the Co atoms at (2c) sites have a large positive anisotropy contribution and those at (3g) sites have a smaller negative contribution. The former favors an easy c-axis alignment of the Co moment and the latter favors an easy plane alignment of the Co moment. The total anisotropy energy per unit cell of SmCo$_5$ can be calculated by the following equation which is equal to 20 cm^{-1}/unit cell:

$$E_a(SmCo_5) = 2E_s(2c) + 3E_s(3g). \qquad (1)$$

The structure of Sm$_{n+1}$Co$_{3n+5}$B$_{2n}$ compounds may be obtained from SmCo$_5$ by partial substitution of two Co atoms by B atom pairs in the SmCo$_5$ lattice. As the B atom pairs preferentially replace the two Co atoms in the (2c) sites in SmCo$_5$ forming SmCo$_3$B$_2$ elementary cell, their unit cells are formed by alternative stacking of a layer of SmCo$_5$ and n layers of SmCo$_3$B$_2$ elementary cells. The (3g) Co atom sites of SmCo$_5$ are replaced by the (6i$_1$) and (6i$_2$) sites in the SmCo$_4$B and Sm$_3$Co$_{11}$B$_4$ compounds. The anisotropy fields at room temperature of these compounds are 400, 175 and 400 kOe, respectively, and the anisotropy constants K_1 are 4.98×10^7, 0.72×10^7 and 3.39×10^7 erg/cm^3, respectively. These values expressed in wave numbers are 3.86, 0.80 and 4.95 cm^{-1}/unit cell.

In the lattice of SmCo$_4$B the Co atoms occupy two kinds of crystallographic sites (2c) and (6i$_1$). One unit cell includes two Co atoms at the (2c) site and six Co atoms at (6i$_1$) site. As the (2c) site of SmCo$_4$B is similar to the (2c) site of SmCo$_5$ and the circumstances of the neighboring atoms around the (2c) sites of both compounds are not changed, we can assume that the stabilization energy E_s of the (2c) site atom in SmCo$_4$B has a value of 25 cm^{-1}/atom, the same as in SmCo$_5$. The anisotropy energy per unit cell of SmCo$_4$B can be written in the form

$$E_s(SmCo_4B) = 2E_s(2c) + 6E_s(6i_1). \qquad (2)$$

So the stabilization energy E_s of the (6i$_1$) site Co atom in SmCo$_4$B has a value of -7.69 cm^{-1}/atom, which can be obtained by using the experimental value of 3.86 cm^{-1}/unit cell.

There exist two (2c) site Co atoms and six (6i$_1$) site Co atoms and three (6i$_2$) site Co atoms in each $Sm_3Co_{11}B_4$ unit cell. The difference between the (6i$_1$) site and the (6i$_2$) site is that the neighboring atom planes of the former contain Sm and Co atoms or Sm and B atoms whereas those of the latter contain Sm and B atoms only. Assuming the stabilization energy of (2c) site E_s in $Sm_3Co_{11}B_4$ to be 25 cm^{-1}/atom and that (6i$_1$) site E_s −7.69 cm^{-1}/atom, and using the expression

$$E_a(Sm_3Co_{11}B_4) = 2E_s(2c) + 6E_s(6i_1) + 3E_s(6i_2), \tag{3}$$

the stabilization energy of (6i$_2$) site E_s may be obtained as −1.02 cm^{-1}/atom.

The anisotropic properties of Co atom at different sites in the $Sm_{n+1}Co_{3n+5}B_{2n}$ compounds are shown in Table.

TABLE

Anisotropic properties of different Co atom sites in the $Sm_{n+1}Co_{3n+5}B_{2n}$ compounds.

Compound	E_s [cm^{-1}/atom]				E_a [cm^{-1}/u.c.]
	2c	3g	6i$_1$	6i$_2$	
$SmCo_5$	25	−10			20
$SmCo_4B$	25		−7.69		3.86
$Sm_3Co_{11}B_4$	25		−7.69	−1.02	0.8

In the rare earth–transition metal intermetallics, it is generally accepted that there are three types of interactions, namely, the R–R interactions between the magnetic moments within the R-sublattice, the T–T interactions between the magnetic moments of the T-sublattice, and R–T intersublattice interactions. In rare earth intermetallic system since the $4f$ states are localized, the interaction between the $4f$ moment and the band magnetism can occur only through the local exchange interaction on the rare earth atom, that is between the $4f$ and $5d$ states. However, it is $3d$–$5d$ hybridization that, firstly, produces significant $5d$ conduction electron charge and spin density at the R sites and, secondly, is responsible for the coupling between the $5d$ and $3d$ spin directions, and therefore the $4f$ and $3d$ spin directions.

An important parameter describing the magnetic properties of these compounds is the intersublattice exchange coupling J_{RCo}. In the mean-field approximation for $Gd_{n+1}Co_{3n+5}B_{2n}$ compounds the expression for J_{GdCo} reads [7]

$$(J_{GdCo}/k_B)^2 = 9(T_C - T'_C)T_C/4Z_{RCo}Z_{CoR}S_{Co}(S_{Co} + 1)G, \tag{4}$$

where T_C and T'_C represent the Curie temperatures of rare earth–transition metal compounds in which R is magnetic (Gd) or R is nonmagnetic (Y), respectively; S_{Co} is the actual Co spin quantum number and G represents the de Gennes factor $(g_R - 1)^2 J(J+1)$. For an estimate of J_{GdCo}, the number of Co nearest neighbors of R atoms Z_{RCo} is taken 18 for RCo_5 and 15 for RCo_4B and $Gd_3Co_{11}B_4$. The number of R neighbors of T atoms, Z_{CoR}, amounts to 3.5–4 in these compounds. The values of T_C, T'_C and S_{Co} are taken from [8, 9]. The value of the J_{GdCo} equals −11.5 K, −15 K and −20.9 K for RCo_5, RCo_4B and $R_3Co_{11}B_4$, respectively.

3. Conclusions

The results lead to the following conclusions:

(a) The stabilization energies of the (2c) site and the ($6i_1$) site in $Sm_{n+1}Co_{3n+5}B_{2n}$ series are similar to those of the (2c) and (3g) sites of $SmCo_5$. The stabilization energy of the ($6i_2$) sites in $Sm_3Co_{11}B_4$ is much smaller than that of the (3g) site in $SmCo_5$.

(b) The smaller anisotropy in $Sm_{n+1}Co_{2n+5}B_{2n}$ compounds is attributed to the replacement of B atoms for the (2c) site atoms in $SmCo_5$, which is primarily responsible for the large positive anisotropy contribution.
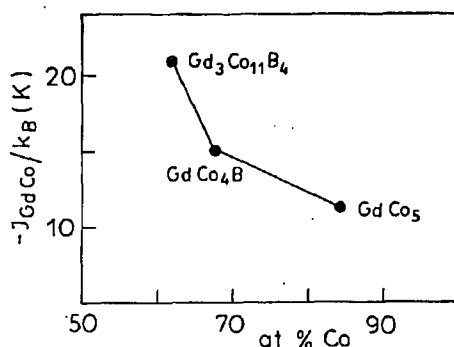

Fig. 1. Dependence of the magnetic coupling constant J_{RCo} in $R_{n+1}Co_{3n+5}B_{2n}$ compounds on the Co content.

(c) A decrease in J_{RCo} with increasing Co concentration is observed (Fig. 1). According to Brooks et al. [10] the $3d$–$5d$ hybridization strongly governs the interatomic $3d$–$5d$ interaction. Therefore, the decrease in J_{RCo} with increasing Co concentraction in the $R_{n+1}Co_{3n+5}B_{2n}$ is likely to be associated with a charge of the hybridization effects.

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