University of Nebraska - Lincoln
DigitalCommons@University of Nebraska - Lincoln

Faculty Publications: Materials Research Science and Engineering Center | Materials Research Science and Engineering Center

9-1-2003

Exchange Interactions and Curie Temperature of Y–Co Compounds

Arti Kashyap
University of Nebraska - Lincoln, akashyap2@unl.edu

Ralph Skomski
University of Nebraska-Lincoln, rskomski2@unl.edu

Renat F. Sabiryanov
University of Nebraska - Omaha, rsabirianov@mail.unomaha.edu

Sitaram Jaswal
University of Nebraska, sjaswal1@unl.edu

David J. Sellmyer
University of Nebraska-Lincoln, dsellmyer@unl.edu

Follow this and additional works at: http://digitalcommons.unl.edu/mrsecfacpubs

Part of the Materials Science and Engineering Commons

Kashyap, Arti; Skomski, Ralph; Sabiryanov, Renat F.; Jaswal, Sitaram; and Sellmyer, David J., "Exchange Interactions and Curie Temperature of Y–Co Compounds" (2003). *Faculty Publications: Materials Research Science and Engineering Center.* Paper 25.
http://digitalcommons.unl.edu/mrsecfacpubs/25

This Article is brought to you for free and open access by the Materials Research Science and Engineering Center at DigitalCommons@University of Nebraska - Lincoln. It has been accepted for inclusion in Faculty Publications: Materials Research Science and Engineering Center by an authorized administrator of DigitalCommons@University of Nebraska - Lincoln.

Exchange Interactions and Curie Temperature of Y–Co Compounds

A. Kashyap, R. Skomski, R. F. Sabiryanov, S. S. Jaswal, and D. J. Sellmyer

***Abstract*—The Curie temperature of rare-earth Co intermetallics is investigated by self-consistent spin-polarized electronic-structure calculations on Y–Co compounds. The total exchange interaction of a given site with all other sites (J_o) is calculated by the linear-muffin-tin-orbital method using the local force theorem and employed to obtain mean-field Curie-temperature estimates. The theoretical predictions for YCo_3, YCo_5, and Y_2Co_{17} are in fair agreement with the experimental Curie temperatures. For YCo_5, the variation of exchange interactions (J_{ij}) with distance is analyzed, and it is discussed how the presence of nonequivalent cobalt sites affects the Curie temperature of the compounds.**

***Index Terms*—Curie temperature, exchange interactions, magnetic moment, permanent magnets, rare-earth cobalt intermetallics, YCo_5.**

I. Introduction

RARE-EARTH transition-metal intermetallics have long been valued as permanent-magnet materials. Rare-earth cobalt intermetallics combine reasonably high magnetizations with high or very high anisotropies and high Curie temperatures [1]–[3], which makes them suitable for advanced high-temperature permanent magnets [4]. Due to the smallness of the de Gennes factor of samarium, the Curie temperature is largely determined by the interatomic exchange of the cobalt atoms [2], [5], and in fair approximation it is sufficient to consider isostructural intermetallics with a nonmagnetic rare earth, such as yttrium.

It is interesting to study the interatomic exchange from an atomic point of view because of its crucial importance for the Curie temperature, and because it plays an important role in the realization of the leading rare-earth anisotropy contribution [2]. The tools for the first-principle study of the interactions have been developed in the context of Y–Fe compounds [6]. Here, we perform the calculations for Y–Co compounds and investigate how the magnetic properties are affected by the local environment. The calculated results are compared with the available experimental data.

Manuscript received January 6, 2003. This work was supported in part by the DOE, AFSOR, NRI, NSF MRSEC, and CMRA.

A. Kashyap, R. Skomski, S. S. Jaswal, and D. J. Sellmyer are with the Center for Materials Research and Analysis and Department of Physics and Astronomy, University of Nebraska, Lincoln, NE 68588 USA (e-mail: akashyap2@unl.edu).

R. F. Sabiryanov is with the Center for Materials Research and Analysis and Department of Physics, University of Nebraska, Omaha, NE 68182-0266 USA (e-mail: rsabirianov@mail.unomaha.edu).

Digital Object Identifier 10.1109/TMAG.2003.815744

II. Method of Calculation

For rhombohedral YCo_3, hexagonal YCo_5, and hexagonal Y_2Co_{17}, self-consistent, semirelativistic, spin-polarized electronic structure calculations are performed using the linear muffin-tin orbitals (LMTO) method within the atomic sphere approximation [7]. A minimal basis set consisting of s, p, and d orbitals is used for both type of atoms and Barth–Hedin exchange-correlation potential is used. The spin up and spin down local densities of states (DOS) and magnetic moments are calculated using the linearized tetrahedron method. Due to the considerable different sizes of Y and Co atoms it is necessary to assume different Wigner–Seitz (WS) cell radii. The ratio of WS radii is kept at 1.35 for all compounds, as suggested by Coehoorn [8], and the crystallographic data for Y–Co compounds are taken from Pearson's Handbook [9]. The interatomic exchange interactions (J_{ij}) are calculated in the Heisenberg approximation.

A method to calculate J_{ij}, based on the local approximation to spin density functional theory has been developed by Liechtenstein *et al.* [10], [11]. Using spherical charge and spin densities and a local force theorem, expression for J_{ij} is

$$J_{ij} = \frac{1}{4\pi}\sum_{LL'} \mathrm{Im}\int_{-\infty}^{\varepsilon_F} d\varepsilon \Delta_l^i(\varepsilon) T_{LL'}^{ij\uparrow}(\varepsilon)\Delta_{l'}^j(\varepsilon) T_{LL'}^{ij\downarrow}(\varepsilon). \quad (1)$$

Here, $T_{LL'}^{ij\sigma}(\varepsilon)$ is the scattering path operator in the site (i, j) representation for different spin projections ($\sigma = \uparrow, \downarrow$), and $\Delta_l^i(\varepsilon) = t_{i\uparrow}^{-1} - t_{i\downarrow}^{-1}$ is the difference of the inverse single-site scattering matrices. The total exchange of a given site with all sites ($J_o = \sum_{i\neq 0} J_{oi}$) can also be calculated from the relation

$$J_o = \frac{1}{4\pi}\sum_{LL'} \mathrm{Im}\int_{-\infty}^{\varepsilon_F} d\varepsilon \{\Delta_l^0(\varepsilon)[T_{LL'}^{00\uparrow}(\varepsilon) - T_{LL'}^{00\downarrow}(\varepsilon)] + \Delta_l^0(\varepsilon) T_{LL'}^{00\uparrow}\Delta_{l'}^0(\varepsilon) T_{LL'}^{00\downarrow}(\varepsilon)\}. \quad (2)$$

The parameter J_o reflects the energy change due to small-angle rotation of the moment at one site. In contrast to the J_{ij}, it is given by the site-diagonal scattering matrix (or Green function), where $i = j$.

III. Results and Discussions

An approximate mean-field approach to calculate the Curie temperature is to use the relation $T_c = 2\langle J_0\rangle/3k_B$ where J_0 is the exchange interaction averaged over all inequivalent sites. Here, we use this approximation to calculate the Curie temperature and discuss corrections due to the involvement of nonequivalent sites.

Table I summarizes the theoretical predictions and compares them with experimental data. For all compounds, it lists the WS

0018-9464/03$17.00 © 2003 IEEE

TABLE I
THE WS CELL RADII (IN ATOMIC UNITS), DOS AT THE FERMI LEVEL (IN STATES/Ry), MAGNETIC MOMENTS (IN μ_B), EXCHANGE PARAMETERS (IN meV) AND CURIE TEMPERATURE (IN K) FOR Y–Co COMPOUNDS

	YCo_3	YCo_5	Y_2Co_{17}
WS Cell Radii			
Y	3.5022	3.5426	3.5487
Co	2.5598	2.6242	2.6287
DOS			
Down	270.80	129.36	994.75
Up	93.23	15.38	95.52
Magnetic Moment			
Total (Theory)	4.81	6.97	25.89
Total (Expt.)	4.95	7.52	27.50
Co1	0.67	1.40	1.72
Co2	0.82	1.52	1.50
Co3	0.49		1.54
Co4			1.57
Exchange J_0			
Co1	16.86	123.91	186.42
Co2	22.17	132.15	159.77
Co3	10.74		140.63
Co4			163.64
Exchange$\langle J_0 \rangle$	13.96	128.85	171.83
Curie Temperature			
Theory			
Expt.	108	998	1330
	305	987	1186

radii, the calculated ↑ and ↓ DOS at the Fermi level, theoretical and experimental magnetic moments, exchange constants, and theoretical and experimental Curie temperatures. The experimental data are taken from [12].

We see that the agreement for the magnetic moment is quite reasonable. The inclusion of spin-orbit interaction would further improve the total magnetic moment but here we are mainly interested in the nature of exchange interactions in these compounds. The orbital moment of the cobalt atoms is reported to be about 0.25 μ_B in YCo_5 [13].

Fig. 1 shows the distance dependence of the exchange interactions for two different Co sites in YCo_5. The exchange interaction is very strong at small interatomic distances but *de facto* short-range, its magnitude rapidly decaying with interatomic distance. The same trend is found in YCo_3 and in Y_2Co_{17}. By contrast, in Y–Fe compounds the exchange interactions are weaker, have a somewhat longer range, and exhibit more pronounced RKKY-type oscillations [6]. This reflects the well-established instability of the ferromagnetism of dense-packed Fe intermetallics [2], [5].

Average exchange interactions for all the three compounds studied are given in Table I. With the increase of Co concentration, the average exchange interactions for Co atoms increase considerably, thereby increasing the Curie temperature. This is in agreement with experiment and with mean-field type model predictions. By contrast, in Y–Fe compounds band-structure effects tend to yield a decrease of T_c with increasing transition metal concentration.

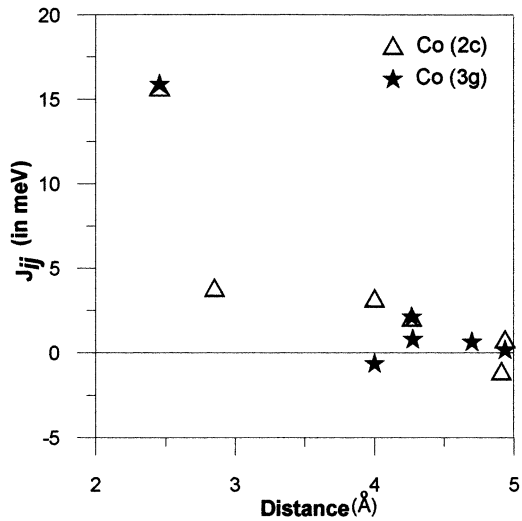

Fig. 1. Distance dependence of the interatomic exchange for the two nonequivalent Co sites in YCo_5. The data include 35 neighbors for both the Co sites.

The average exchange interaction for YCo_5 is 128.85 meV, corresponding to a Curie-temperature estimate of 998 K. This is very close to the experimental value of 987 K. For Y_2Co_{17}, the Curie temperature is slightly overestimated. By comparison, for Y–Fe compounds the method overestimates the Curie temperature by 25%. For YCo_3, the agreement is worse. The J_0 values for YCo_3 are small and very disparate in magnitude, indicating that the mean-field approximation is poor in this case. The main reason is that the comparatively low Co content causes the alloy to remain close to the onset of ferromagnetism. This leads to long-range spin fluctuations and to corrections to the Curie temperature [16], [17].

Table I reveals a considerable dispersion of the J_{ij}. This makes it necessary to distinguish between nonequivalent sites. Note that even on a mean field level, site-resolved exchange interactions yield Curie-temperature corrections.

The basic idea is to write down separate mean-field equations for the nonequivalent sites, as discussed in other contexts [2], [5], [14], [15], [18]. Linearization of the Brillouin functions then leads to an eigenvalue problem, and the largest eigenvalue is equal to the exact mean-field Curie temperature. The calculation, which will be published elsewhere, involves intersublattice exchange constants derived from (1).

The anisotropic exchange parameters listed in Table I yield a pronounced anisotropy of the exchange stiffness, making it necessary to distinguish between $A_{||}$ for magnetization gradients parallel to the c-axis and $A_{\perp}$ for magnetization gradients in the basal plane. This yields a variety of micromagnetic phenomena, such as a dependence of the Bloch wall width on the orientation of the wall and a directional dependence of domain-wall energies. For example, in YCo_5 the intra-sublattice interaction of the Co 2c atoms corresponds to only about 127 K as compared with the 2c–3g intersublattice exchange of about 770 K.[1] However,

[1]The bond anisotropy considered in this paper is nonrelativistic, that is, it does not involve spin-orbit coupling and is independent of the magnetization direction. It must be distinguished from the much smaller exchange anisotropy, which involves spin-orbit coupling and yields a small direction-dependent contribution of typically less than 1% to the spontaneous magnetization.

in the present case the strong intersublattice exchange ensures a stable Co 2c magnetization, and the Curie-temperature reduction is very small, about 5%. Note that exchange anisotropy can also be calculated by performing calculations of the total magnetic energy [19], but the present approach makes it possible to assign the net exchange anisotropy to individual sites and to trace the origin of the exchange anisotropy. From a formal point of view, this will be achieved by a Fourier-transformation method, in analogy to the use of the Lindhard function [20] to discuss RKKY interactions in k-space.

From the point of view of future research, this study establishes a new approach toward the understanding and development of multisublattice permanent magnets. First, the exchange parameters $\mathrm{J_{ij}}$ enable a direct access to many-sublattice mean-field analysis [21], which provides reliable finite-temperature predictions of magnetic properties. In the past, intersublattice interactions were approximated in terms of parameters of the type $\mathrm{J_{TT}}$, $\mathrm{J_{RT}}$, and $\mathrm{J_{RR}}$ [5], [21], [22], ignoring the transition-metal dispersion shown in Fig. 1 and in Table I. Second, as mentioned above, the bond anisotropy of the $\mathrm{J_{ij}}$ leads to far-reaching but largely unexplored anisotropic micromagnetic phenomena. Third, a site-resolved analysis will make it possible to gauge how specific substitutions will affect the finite-temperature magnetization and anisotropy of high-temperature permanent magnets.

In conclusion, we have investigated how the Curie temperature $\mathrm{T_c}$ of rare-earth cobalt intermetallics is realized by interatomic exchange. Electronic-structure calculations on Y–Co compounds have been used to analyze the leading transition-metal contribution to $\mathrm{T_c}$. Compared with Y–Fe compounds, the interatomic exchange is stronger but exhibits a more rapid decay with increasing distance and less pronounced oscillations. Further research is necessary to explore the consequences of the existence of more two or more transition-metal sublattices, particularly with respect to the temperature dependence of the rare-earth and transition-metal sublattice anisotropies.

REFERENCES

[1] M. Richter, "Band structure theory of magnetism in 3d–4f compounds," *J. Phys. D., Appl. Phys.*, vol. 31, pp. 1017–1048, May 1998.

[2] R. Skomski and J. M. D. Coey, *Permanent Magnetism*. Bristol, U.K.: Inst. Physics, 1999.

[3] K. Kumar, "$\mathrm{RETM_5}$ and $\mathrm{RE_2TM_{17}}$ permanent magnets development," *J. Appl. Phys.*, vol. 63, pp. R13–R57, Mar. 1988.

[4] J. Zhou, R. Skomski, C. Chen, G. C. Hadjipanayis, and D. J. Sellmyer, "Sm–Co–Cu–Ti high-temperature permanent magnets," *Appl. Phys. Lett.*, vol. 77, pp. 1514–1516, 2000.

[5] J. M. D. Coey, Ed., *Rare-Earth Iron Permanent Magnets*. Oxford: University Press, 1996.

[6] R. F. Sabiryanov and S. S. Jaswal, "Electronic structure and magnetic properties of Y–Fe compounds," *Phys. Rev.*, vol. B57, pp. 7767–7772, Apr. 1998.

[7] O. K. Andersen, "Linear methods in band theory," *Phys. Rev.*, vol. B8, pp. 3060–3083, Oct. 1975.

[8] R. Coehoorn, "First principles band structure calculations for rare earth-transition metal compounds: Magnetization, hyperfine parameters and magnetocrystalline anisotropy," *J. Magn. Magn. Mater.*, vol. 99, pp. 55–70, 1991.

[9] P. Villars and L. D. Calvert, *Pearson's Handbook of Crystallographic Data for Intermetallic Phases*. Metals Park, OH: Amer. Soc. Metals, 1985.

[10] A. I. Liechtenstein, M. I. Katsnelson, V. P. Antropov, and V. A. Gubanov, "Local spin density functional approach to the theory of exchange interactions in ferromagnetic metals and alloys," *J. Magn. Magn. Mater.*, vol. 57, pp. 65–74, 1988.

[11] V. A. Gubanov, A. I. Liechtenstein, and A. V. Postnikov, *Magnetism and the Electronic Structure of Crystals*, M. Cardona, P. Fulde, K. von Klitzing, and H.-J. Queisser, Eds. Berlin, Germany: Springer-Verlag, 1992.

[12] K. H. J. Buschow, "Intermetallic compounds of rare-earth and 3d transition metals," *Rep. Prog. Phys.*, vol. 40, pp. 1179–1256, Oct. 1977.

[13] M. Yamaguchi and S. Asano, "First principles calculations of the 3d magnetocrystalline anisotropy energy of $\mathrm{YCo_5}$, $\mathrm{Y_2Co_7}$, $\mathrm{YCo_3}$, and $\mathrm{Y_2Co_{17}}$," *J. Magn. Magn. Mater.*, vol. 168, pp. 161–168, 1997.

[14] J. S. Smart, *Effective Field Theories of Magnetism*. Philadephia, PA: Saunders, 1966.

[15] J. P. Gavigan and D. Givord, "Intrinsic and extrinsic properties of rare earth-transition metal compounds and permanent magnets," *J. Magn. Magn. Mater.*, vol. 84, pp. 288–298, 1990.

[16] E. P. Wohlfarth, "The Curie temperatures of compounds of the heavy rare earths and yttrium with cobalt," *J. Phys. F: Met. Phys.*, vol. 9, pp. L123–L128, 1979.

[17] P. Mohn, *Magnetism in the Solid State*. Berlin, Germany: Springer-Verlag, 2003.

[18] R. Skomski and D. J. Sellmyer, "Curie temperature of multiphase nanostructures," *J. Appl. Phys.*, vol. 87, pp. 4756–4758, 2000.

[19] K. Belachtchenko, unpublished, 2003.

[20] N. W. Ashcroft and N. D. Mermin, *Solid-State Physics*. Philadelphia, PA: Saunders, 1976.

[21] N. H. Duc, T. D. Hien, D. Givord, J. J. M. Franse, and F. R. de Boer, "Exchange interactions in rare-earth transition-metal compounds," *J. Magn. Magn. Mater.*, vol. 124, pp. 305–311, 1993.

[22] R. Skomski, "Finite-temperature behavior of anisotropic two-sublattice magnets," *J. Appl. Phys.*, vol. 83, pp. 6724–6726, 1998.