

Article

Exchange Interactions and Curie Temperature of Ce-Substituted SmCo_5

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Abstract: A partial substitution such as Ce in SmCo_5 could be a brilliant way to improve the magnetic performance, because it will introduce strain in the structure and breaks the lattice symmetry in a way that enhances the contribution of the Co atoms to magnetocrystalline anisotropy. However, Ce substitutions, which are benefit to improve the magnetocrystalline anisotropy, are detrimental to enhance the Curie temperature (T_C). With the requirements of wide operating temperature range of magnetic devices, it is important to quantitatively explore the relationship between the T_C and ferromagnetic exchange energy. In this paper we show, based on mean-field approximation, artificial tensile strain in SmCo_5 induced by substitution leads to enhanced effective ferromagnetic exchange energy and T_C , even though Ce atom itself reduces T_C .

Keywords: mean-field theory; magnetism; exchange energy; curie temperature

1. Introduction

Owing to the large magnetocrystalline anisotropy, high Curie temperature (T_C) and saturation magnetization (M_s), Sm–Co compounds have drawn attention for the high-performance magnet [1–12]. The net performance of the magnet crucially depends on the inter-atomic interactions, which in turn depend on the local atomic arrangements. In order to improve the intrinsic magnetic properties of Sm–Co system, further approaches [5,7,9,12–19] have been attempted in the past few decades.

A promising scheme is the partial substitution of Co with other transition metals (TM), such as Cu, Ni, Fe, or Zr in $\text{Sm}(\text{Co}, \text{TM})_5$ [15,19–22]. The partial TM substitution enhances magnetic anisotropy energy (MAE) due to slight modifications of the atomic structure and the breaking of the lattice symmetry. Substitution of the Co in the system might make it more economically viable, however, due to the reduced exchange coupling parameters, both T_C and the M_s of the $\text{Sm}(\text{Co}, \text{TM})_5$ compounds decrease with increasing TM content. [15,19,20].

Importantly, any substitution of Co with another TM reduces the effective ferromagnetic exchange coupling and consequently reduces T_C . This is highly undesirable because the most important application field of Sm–Co-based magnets is that of high temperature. Hence, it is vital that we find a way to enhance the magnetic performance in Sm–Co magnets without substituting away the Co, but by carefully modifying the local atomic arrangements, by introducing strain. Strain is a particularly important issue for permanent magnets, because their synthesis typically involves sintering, and thermal processes inevitably introduce strain on fine-grained materials [23,24]. Strain effects are also associated directly to partial element substitution, such as e.g., of Sm with another RE metal.

Ce is a particularly promising candidate because it is the most abundant and second lightest among all RE metals, and CeCo_5 is isomorphous with SmCo_5 [25,26]. It is well-known that the magnetic material parameters of CeCo_5 are substantially weaker than those of SmCo_5 [27], i.e., reduced

T_C of 660 K instead of 1000 K, reduced MAE of 5.3 MJ/m^3 instead of 17.2 MJ/m^3 , and reduced M_s of 0.95 T instead of 1.15 T. Therefore, most researches focused on 3d transition metal substitution instead of Ce substitution, at least so far. Nevertheless, our recent simulation work indicates that a partial substitution of Sm by Ce can have drastic effects on the magnetic performance, because it will introduce strain in the structure and breaks the lattice symmetry in a way that enhances the contribution of the Co atoms to magnetocrystalline anisotropy (MCA) [28]. Furthermore, we have also experimentally confirmed that a partial Ce substitution can enhance the magnetic performance of the system [29]. However, Ce substitutions, which are benefit to improve the MCA with slight modifications of the local atomic arrangements, are detrimental to enhance the Curie temperature. With the requirements of wide operating temperature range of magnetic devices, it is very important to quantitatively explore the relationship between the Curie temperature T_C and ferromagnetic exchange energy.

In this paper we will report, based on mean-field approximation (MFA), that Ce substitution-induced tensile strain in SmCo_5 leads to enhanced effective ferromagnetic exchange energy and T_C . However, the Ce atom itself reduces T_C .

2. Results and Discussion

2.1. Crystal Structure

The intermetallic magnetic compounds SmCo_5 and CeCo_5 crystallize into the hexagonal CaCu_5 structure, where each Sm (Ce) occupies a $1a$ site and Co occupies $2c$ and $3g$ sites in the lattice that Co_3 and SmCo_2 sub-layers are alternatively along the c -axis [30]. To understand the Ce substitution effect on crystal structure and magnetic properties, we performed calculations of the $\text{Sm}_{1-x}\text{Ce}_x\text{Co}_5$ system, where we considered seven different compositions: $x = 0, 0.125, 0.25, 0.375, 0.5, 0.75$, and 1. Configurations shown in Figure 1 are the most stable structures obtained by total energy minimization in the density functional theory calculations [28].

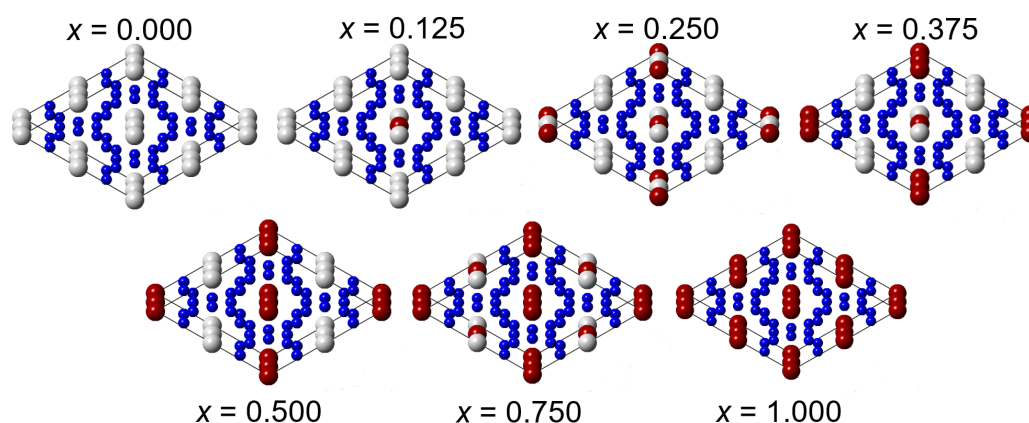


Figure 1. Illustration of the crystal structure of $\text{Sm}_{1-x}\text{Ce}_x\text{Co}_5$ for $x = 0, 0.125, 0.25, 0.375, 0.5, 0.75$, and 1. Gray, red, and blue spheres correspond to Sm, Ce, and Co atoms, respectively.

The atomic volume decreases linearly from 14.12 \AA^3 in SmCo_5 to 13.84 \AA^3 in CeCo_5 with increasing x , whereas the strain increases correspondingly monotonically with increasing x (see Figure 2). Result that decreased volume by Ce concentration well corresponds with ref. [31] which presented that CeCo_5 compound stands out as a anomalous in the lattice constants. Due to lanthanide contraction, smooth decrease in atomic volume can be expected through the series from lanthanum to terbium in the periodic table, however, there is a pronounced deviation for CeCo_5 . In our related research [28], we found substitution-induced tensile strain on $\text{Sm}_{1-x}\text{Ce}_x\text{Co}_5$ has drastic effects on the magnetic state. While the decreased magnetic moment for CeCo_5 have been interpreted as evidence for a mixed-valence

behavior of the Ce ion [32,33], the effective occupation number of the 4*f* orbitals can be determined by induced strain rate.

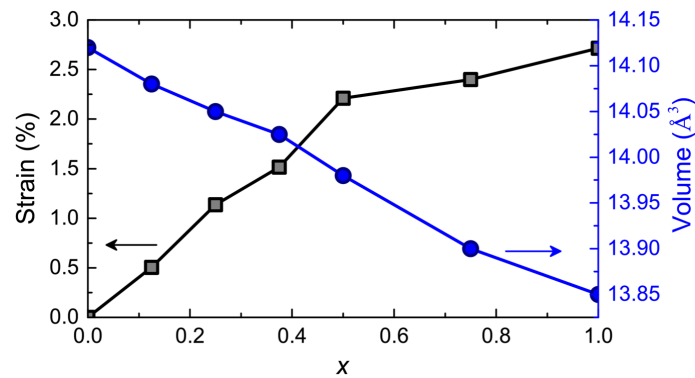


Figure 2. Modification of the effective strain (square) compared to the SmCo₅ cell, and the atomic volume (circle) in Sm_{1-x}Ce_xCo₅, as a function of Ce substitution *x*.

2.2. Exchange Interactions

Mean-field Hamiltonian for the 3-sublattice system could be written as

$$H_{MFA} = -J_{c-c}m_c \sum_i^{N_c} S_{c,i} - 2J_{c-g}m_c \sum_i^{N_g+N_c} S_{g,i} - J_{g-g}m_g \sum_i^{N_g} S_{g,i} - 2J_{g-a}m_g \sum_i^{N_g+N_a} \sigma_{a,i} - 2J_{c-a}m_c \sum_i^{N_c+N_a} \sigma_{a,i}, \quad (1)$$

where J_{ij} , m_i , and S_i are exchange coupling between sites i and j , magnetization, and classical spin vector at crystal site i , respectively. Labels of c and g correspond to 2*c* and 3*g* sites of Co atom, while a represents 1*a* site of Sm.

Sublattice magnetization is

$$m_i = \frac{2S_i + 1}{2S_i} \coth\left(\frac{2S_i + 1}{2S_i} \frac{1}{T} \sum_{ij} z_{ij} J_{i-j} m_j\right) - \frac{1}{2S_i} \coth\left(\frac{1}{2S_i} \frac{1}{T} \sum_{ij} z_{ij} J_{i-j} m_j\right) \quad (2)$$

where z_{ij} is the number of neighboring sites in j -th sublattice to i -th sublattice.

While the inter-atomic ferromagnetic exchange interactions can be obtained as the difference between ferromagnetic and antiferromagnetic spin configuration, i.e., $J = (E_{\uparrow\uparrow} - E_{\uparrow\downarrow})/2$ by using density functional theory calculations [28], the effective exchange energies of Co and Sm are calculated as

$$J_{\text{eff}}^{\text{Co}} = (z_{gg} + z_{gc} + z_{cc} + z_{cg})J_{\text{Co-Co}} \quad (3)$$

$$J_{\text{eff}}^{\text{R}} = (z_{ac} + z_{ca} + z_{ag} + z_{ga})J_{\text{Co-R}},$$

respectively.

Due to the smallness of the de Gennes factor of Sm, the Curie temperature is dominantly determined by the interatomic exchange interaction of the Co atoms [9,34]. Therefore the calculations on the exchange interaction are focused on the Co-Co couplings. To understand the role of Ce substitution-induced strain and Ce substitution itself, magnetic moment, inter-site exchange coupling (J_{ij}) and effective exchange energy (J_{eff}) are calculated as a function of the strain in SmCo₅ (Figure 3) and Ce concentration in Sm_{1-x}Ce_xCo₅ (Figure 4).

In Figure 3a, the magnetic moments at each crystallographic site decrease nearly linearly with increasing tensile strain. The reason for the decrease of the magnetic moments is the decreasing overlap of the *d*-orbitals as the *c*-axis grows with strain. Further, we calculate the inter-site exchange coupling and overall effective exchange coupling parameters. The intra-plane interaction strengths $J_{\text{Co}(2c)\text{-Co}(2c)}$ and $J_{\text{Co}(3g)\text{-Co}(3g)}$ increase while the inter-plane interaction strength $J_{\text{Co}(2c)\text{-Co}(3g)}$ decreases

with increasing tensile strain (Figure 3b). This illustrates the dependence of the ferromagnetic exchange on the inter-atomic distance: as the c - c and g - g distances decrease the corresponding exchange interaction increases, whereas as the c - g distance increases with increasing tensile strain the exchange interaction decreases. Then we obtain effective exchange interactions J_{eff} , which are the sum of the exchange coupling constants within a sphere of radius $R = 5a$. From these calculations we observe that the total ferromagnetic exchange interaction increases monotonically with increasing strain (see Figure 3c), in contrast to the magnetic moments.

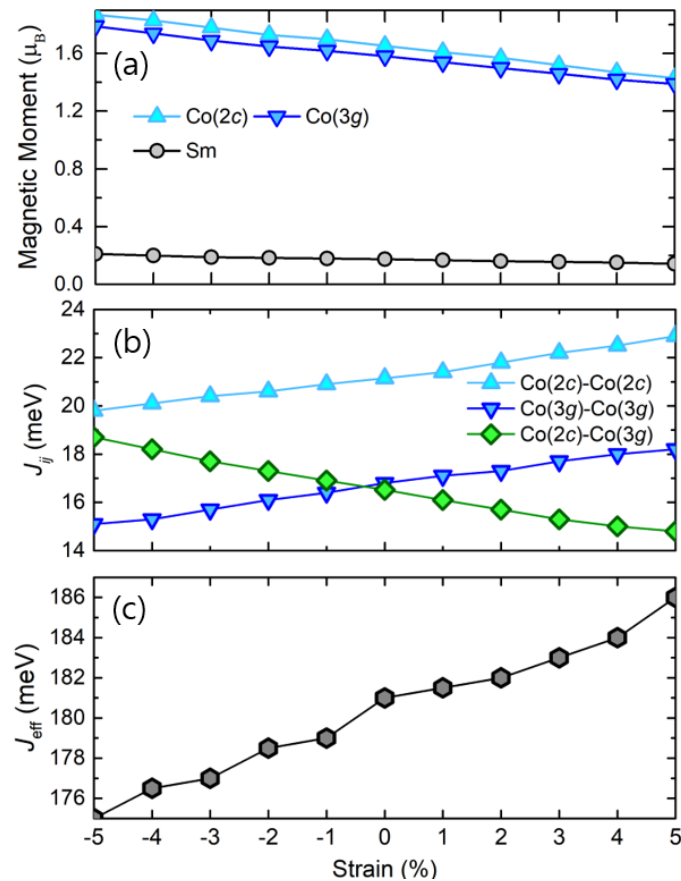


Figure 3. Intrinsic magnetic properties as a function of strain in SmCo₅: (a) atomic magnetic moments of Co atoms at sites 2c (triangles) and 3g (inverse-triangles) and Sm atoms (circles); (b) exchange coupling between Co sites i and j ; and (c) effective total ferromagnetic exchange energy.

For Ce substituted Sm_{1-x}Ce_xCo₅ without strain, the calculated magnetic moments of the Co, Ce, and Sm atoms decrease monotonically with increasing Ce-substitution x , which correlates to the decreasing atomic volume, following Vergard's law [35] (see Figure 4a). Also, the calculated $J_{\text{Co-Co}}$ and J_{eff} decrease strongly with increasing x , as seen in Figure 4b,c.

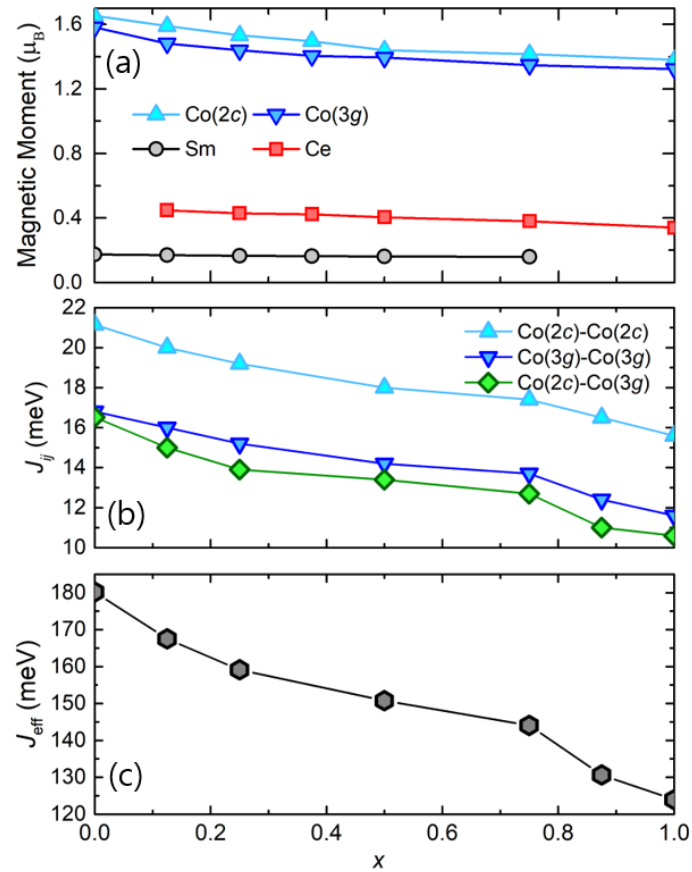


Figure 4. Magnetic properties as a function of Ce-substitution of Sm: (a) atomic magnetic moments of Co atoms at sites 2c (triangles) and 3g (inverse-triangles) and Sm atoms (circles); (b) exchange coupling between Co sites i and j ; and (c) effective total ferromagnetic exchange energy.

2.3. Calculations of Curie Temperature

The corresponding Curie temperatures are also derived from the calculated exchange parameters based on the MFA.

The MFA is based on the notion of single-spin excitations, and the Hamiltonian is

$$\hat{H}_i = -\vec{\mu}_i \cdot \vec{H}_i = -g\mu_B \vec{S}_i \cdot \vec{H}_i \quad (4)$$

where g and μ_B are the Lande factor and Bohr magneton, respectively. The molecular field could be thus defined as

$$\hat{H}_i = -\frac{1}{g\mu_B} \sum_j J_{ij} \langle \vec{S}_j \rangle \quad (5)$$

with

$$\langle S_{iz} \rangle = S_i B_{S_i}(x_i), \quad x_i = \frac{g\mu_B H_i}{k_B T} \quad (6)$$

where $B_{S_i}(x_i)$ is the Brillouin function, k_B is the Boltzmann constant, and T is the temperature in K. When T is very high, such as $k_B T \gg \mu_B H_i$, the $B_{S_i}(x_i)$ and $\langle S_{iz} \rangle$ are rewritten as

$$B_s(x) = \frac{(S+1)x}{3}, \quad \langle S_{iz} \rangle = \frac{S_i(S_i)\mu_B H_i}{3k_B T}. \quad (7)$$

By using the exchange integral J_{ij} previously obtained, we get

$$\begin{aligned} \langle S_{iz} \rangle &= -\frac{S_i(S_i + 1)}{3k_B T} \sum_j J_{ij} \langle S_{jz} \rangle, \\ T \langle S_{jz} \rangle + \frac{S_i(S_i + 1)}{3k_B} \sum_j J_{ij} \langle S_{jz} \rangle &= 0 \end{aligned} \quad (8)$$

which has nonzero solution only if the determinant

$$\begin{bmatrix} a_{11} - T & \dots & \dots & a_{1n} \\ \dots & \dots & \dots & \dots \\ a_{n1} & a_{n2} & \dots & a_{nn} = T \end{bmatrix} = 0, a_{ij} = \frac{S_i(S_i + 1)}{3k_B} \sum_j J_{ij}. \quad (9)$$

Among solutions in Equation (9), the highest positive T is the desired Curie temperature T_C .

As shown in Figure 5a, T_C increases from 1085 K to 1110 K with increasing tensile strain along c -axis from 0% to 5%. For non-strained SmCo_5 (0%), the calculated T_C (1085 K) is comparable with the experimental values ($T_C = 1020$ K) [8], considering the MFA Curie temperature is generally overestimated by about 10–20% [36]. On the other hand, T_C rapidly reduces with increasing concentration of Ce substitutions, and eventually CeCo_5 exhibit only $T_C = 660$ K. Because for $\text{Sm}_{1-x}\text{Ce}_x\text{Co}_5$ without strain the decrease of the effective ferromagnetic exchange is reinforced by the decrease of the magnetic moments. The appearance of the value of T_C results from the strain dependence of the effective exchange parameters J_{eff} .

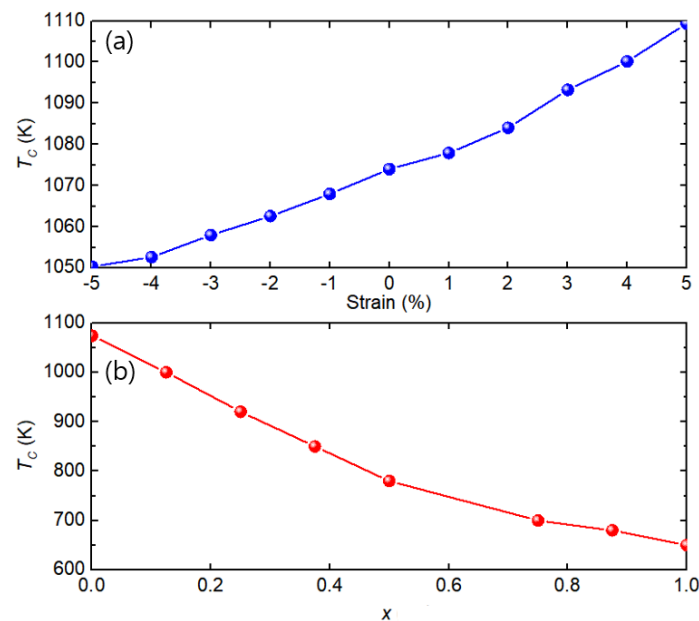


Figure 5. Variation of the Curie temperature with respect to (a) strain and (b) Ce concentration x .

3. Conclusions

Using mean-field theory approximation, we investigated the Ce substitution and strain effect on ferromagnetic exchange energy and corresponding Curie temperature of SmCo_5 compounds. It is found that tensile strain along c -direction improves T_C which is key property for hard magnets. In comparison with an equilibrium state of SmCo_5 , about 40 K higher T_C is observed when 5% tensile strain is applied. The enhancement of T_C can be explained by responses of exchange energy parameters to the lattice strain. Even though the artificial strain due to Ce substitution improves the T_C , Ce itself rather reduces T_C . Therefore, it is necessary to find a substance which can produce a similar strain

on SmCo_5 but does not cause a decrease in T_C by substitution. In addition, our work can propose a guideline since the interest on the role of strain as a tool to control novel quantum functionalities is rapidly growing in related condensed matter systems like ultrathin strongly correlated magnetic ultra thin films [37], cuprates [38] and iron-based high T_C superconductors [39] and charge density wave materials [40].

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Conflicts of Interest: The author declares no conflict of interest.

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