

# Dependence of Exchange Coupling on Interfacial Conditions in SmCo<sub>5</sub>/Co System: A First-Principles Study

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We have performed first-principles calculations to study the interfacial exchange coupling in a SmCo<sub>5</sub>/Co multilayer model system. The hard phase *hcp* SmCo<sub>5</sub> and the soft phase *hcp* Co (or Co<sub>1-x</sub>Fe<sub>x</sub>) stacking along (10 $\bar{1}$ 0) direction are structurally well matched. The atomic structure, including the alignment and the separation between layers, were optimized first. Then the non-collinear magnetic structures were calculated to explore the exchange coupling dependence on the variation of the atomic composition across the interface. We found that the inter-phase exchange coupling strength is strongly dependent on the interface condition between the hard and soft phase by comparing the exchange coupling strengths in different interface conditions. The findings were further confirmed by the calculated site-to-site exchange parameters across the interface.

**Keywords:** First-Principle Calculation, Permanent Magnets, Noncollinear Magnetic Calculation, Ferromagnetic Multilayer, Interphase Exchange-Coupling, Cobalt Alloys.

## 1. INTRODUCTION

Since Kneller and Hawig's pioneering work,<sup>1</sup> exchange-coupled permanent magnets containing soft and hard magnetic phases (the exchange-spring magnets) have been extensively studied to achieve high maximum energy product values.<sup>2</sup> The maximum energy product values are expected to be enhanced in these hard/soft composite systems, by combining high saturation magnetization of a soft phase and large anisotropy of a hard phase. In this regard, the hard magnetic materials such as Sm-Co and the soft magnetic materials such as Fe-Co are particularly attractive because of their peculiar properties. Among the commonly used magnetic materials, for instance, SmCo<sub>5</sub> has the largest anisotropy energy ( $1.7 \times 10^7$  J/m<sup>3</sup>) with high Curie temperature, while Co, Fe and their alloys have very high Curie temperatures with large saturation magnetizations.<sup>3</sup> Indeed, high maximum energy product values have been reported<sup>4</sup> recently in exchange-coupled systems with SmCo<sub>5</sub> as hard phase and Fe as soft phase. According to early models by Kneller and Hawig,<sup>1</sup> an ideal hard/soft phase multilayer achieves maximum energy product at the optimum thickness of the soft phase which is equal to two domain wall thickness in the hard phase (it is about 7 nm for SmCo<sub>5</sub>). However,

many recent experimental and theoretical studies show the important effect of the soft phase parameters and interface conditions.<sup>5-9</sup> Thus it is important to understand the influence of these factors in the inter-phase exchange coupling, in order to achieve better energy products. These issues can be tackled in the scope of first-principles electronic structure calculations based on Density Functional Theory (DFT) as demonstrated in recent work.<sup>10,11</sup>

In the present work, we focused on the effects of the interface conditions on the inter-phase exchange coupling in layered SmCo<sub>5</sub>/Co system using first-principles methods. We show that exchange coupling on the interface between the hard and soft phase can be modified by addition of a third element (iron). In this study we consider the nanocomposite exchange-spring multilayer with the size of the hard and soft layer smaller than the thickness of a usual domain wall, so that the exchange-coupling between the two phases will be in effect. A single domain case is considered for both the hard and the soft phase in the present modeling interface. The exchange coupling strength between the soft and hard phases was described in two complimentary ways. First, we model a simulated demagnetization process of the magnetic systems using noncollinear magnetic structure calculations in a superlattice model. In this method we rotate the direction of local magnetic moments at the center of soft layer from

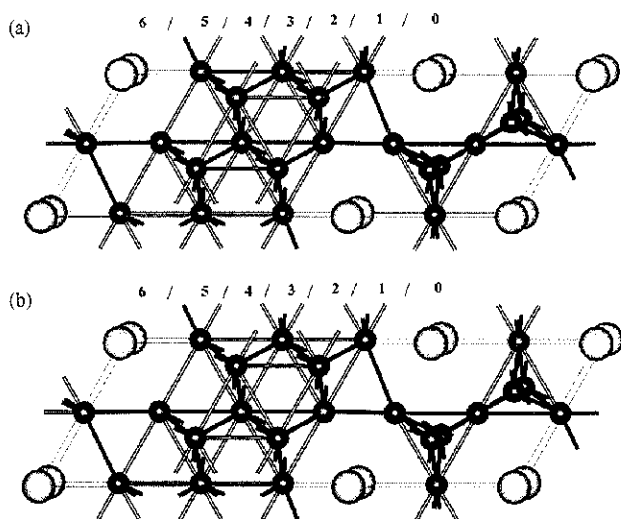
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ferromagnetic to a finite angle  $\theta$ . We vary this angle as a parameter to extract the strength of the interlayer exchange coupling. Second, we calculate pair exchange parameters across interface using a perturbative method.

## 2. MODEL AND METHODOLOGY

The superlattice model is adopted to construct the interface structure for our simulation. A supercell consists of  $\text{SmCo}_5$  and 5 layers of Co stacking along  $(10\bar{1}0)$  direction as shown in Figure 1(a).  $\text{SmCo}_5$  and *hcp* Co lattice constants have a mismatch of only 0.2% along this direction. There are totally 18 atoms in the supercell. The self-consistent spin-polarized electronic structure calculations with periodic boundary conditions were performed to optimize the atomic structure and conduct the noncollinear magnetic calculations.

We use the projector augmented wave (PAW) method implemented in Vienna *ab initio* simulation package (VASP) to obtain an optimized crystal structure of the system.<sup>12–15</sup> Then, we apply the linear-muffin-tin-orbital (LMTO) method<sup>16,17</sup> in atomic sphere approximation and in near orthogonal representation to perform non-collinear magnetic configurations.<sup>18</sup> PAW method calculations reproduce excellently the lattice constants of crystal  $\text{SmCo}_5$ , Co and Fe. We use the energy cut-off of 335 eV for the plane wave expansion of the PAWs. All the structural relaxations are performed until the Hellman-Feynman forces on the relaxed atoms become less than 1 meV/cell. The optimized separation between the soft and hard phases is 2.5 Å.

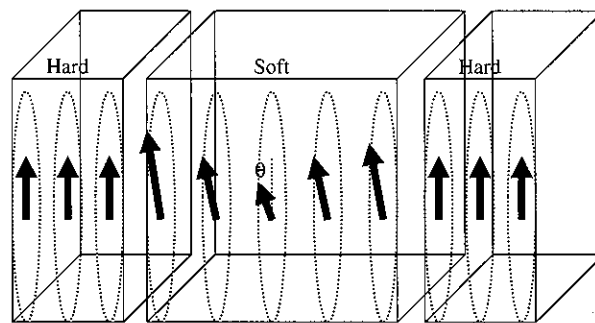


**Fig. 1.** Atomic configurations of the two-phase model systems. The soft and hard phases are aligned along  $(10\bar{1}0)$  direction. The green large, blue small and orange small balls represent Sm, Co and Fe atoms, respectively. (a) the “original” system  $\text{SmCo}_5/\text{Co}$ :  $\text{SmCo}_5$  as hard phase and pure *hcp* Co as soft phase; (b)  $\text{SmCo}_5/\text{CoFe}$ : Co atoms in one sublattice in the soft phase replaced with Fe atoms. The label on top of each panel indicates the planes parallel to the interface. The periodic boundary condition has been used in the graph.

## 3. RESULTS AND DISCUSSION

The optimized structure obtained in PAW calculations is used as an input for non-collinear magnetic order calculations using LMTO method.<sup>18</sup> Our model mimics a domain wall which forms in the demagnetization process. Due to the computational restrictions we consider the directions of the magnetic moments of the atoms in the hard phase were fixed to the easy magnetization axis direction and five layers of soft phase with magnetic moments rotated from its FM order as illustrated in Figure 2. The magnetic moments of atoms in the middle layer of the soft phase were fixed to turn a given angle  $\theta$  relative to the direction of the magnetic moments of the hard phase, while the magnetic moments of other atoms in the soft phase were free to relax. Upon the convergence of the calculations is reached, the total energy is obtained for each given angle  $\theta$ . The total energy difference for the system,  $\delta E(\theta) = E(\theta) - E(\theta = 0^\circ)$ , as a function of the turning angle  $\theta$  is shown in Figure 3. We find that  $\delta E(\theta)$  behaves as a quadratic function of  $\theta$ , manifesting the spring behavior and the exchange coupling between the soft and hard phases in this system. We compare results in the case of soft phase made of pure *hcp* Co and the Fe-doped Co soft phase. The iron doping is expected to strengthen the exchange coupling because FeCo alloy have stronger exchange than pure phases, and, at the same time, an increased magnetization. Furthermore, an iron doping can produce more gradual variation of anisotropy across interface. Two model systems are shown in Figure 1.

The structure of the interface shows a substantial change in the local coordination of Co atoms when going from  $\text{SmCo}_5$  to *hcp* Co phase. Co atoms near the interface lack some of their nearest neighbors as can be seen from Figure 1. Thus, we can expect that the exchange coupling near the interface should be different from the one in either phase. Because of the reduction of number of nearest neighbors across the interface, the interface exchange coupling is reduced. It can be seen from the variation of the layer resolved angle of rotations of atomic moment



**Fig. 2.** A schematic view of noncollinear magnetic orderings in the two-phase systems. The arrows represent the directions of magnetic moments of the atoms in each layer.  $\theta$  is the angle between the directions of magnetic moments of the atoms in the hard phase and those in the middle layer of the soft phase, which are fixed.

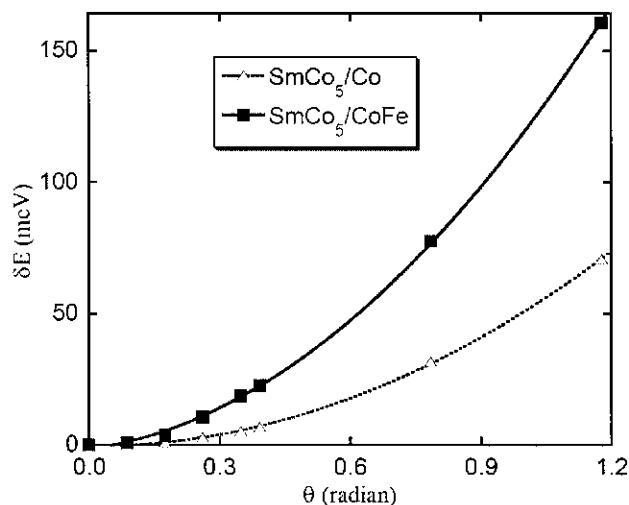


Fig. 3. The calculated total energy differences,  $\Delta E(\theta) = E(\theta) - E(\theta = 0)$  (the marks) and their fitting to a quadratic curve for the two systems considered in Figure 1.

across the soft phase as shown in Figure 4. The middle-layer angle is fixed at  $\theta = 15^\circ$  in this example. Two other layers have their relaxed angles at 10 and 11.5 degrees which is indication that coupling is the strongest between first and second layer, but weakest across the interface. In the system with uniform exchange coupling these angles are expected to be close to 5 and 10 degrees, respectively. Thus, the exchange coupling oscillates in the soft phase as function of distance from the interface. Using one dimensional Heisenberg model we fit the inter-phase exchange coupling parameter per cross-section of the unit cell to be about 16 meV and 25 meV in case of *hcp* Co, and FeCo

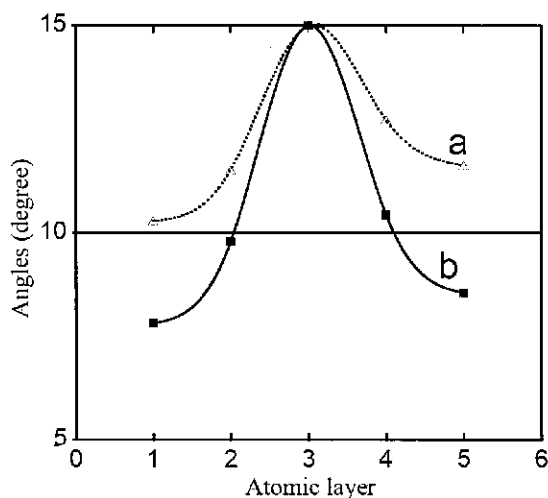


Fig. 4. The angle distributions for the soft phase atomic layers parallel to the interface plane (refer to Fig. 1). Layer 0 and layer 6 are the fixed hard phase layers in the superlattice model. Layer 3 is the middle layer of the soft phase, whose atomic magnetic moments are turned at a fixed value ( $15^\circ$  here) away from those in the hard phase layers. All the atomic magnetic moment orientation in layers 1, 2, 4, and 5 are obtained self-consistently.

alloy, respectively. The exchange between Co plane at the interface and next layer of Co in the soft phase is order of magnitude larger. Such large difference is due to a large increase in number of bonds across planes (by factor of 6) and the increase in the pair exchange parameters (by factor of 1.5).

We find that the effect of doping on the exchange coupling is significant. Figure 1(b) illustrates our model where Co atoms in the *hcp*-Co film are replaced by Fe atoms in the second layer. By introducing Fe atom in this extreme model, Fe atoms were counted as 40% in the soft phase. In this system,  $\Delta E(\theta)$  is also a quadratic function of  $\theta$  (Fig. 3). However, the curve of this system is much steeper than that of pure Co, indicating that the exchange coupling in system with Fe-doped soft phase is stronger than the exchange coupling in system with pure Co as soft phase. Comparing relaxed angles of the Fe-doped interface we observe smaller angle of rotation in the interface layer. This indicates that the interface exchange coupling between the hard and soft phases is stronger in Fe-doped system and it is stronger with respect to the coupling of Fe to the next interlayer coupling in soft phase. Thus, Fe-doping not only strengthens the exchange in FeCo alloy it makes exchange coupling variation smoother across the system.

To further understand the phenomena, we have calculated the site-to-site exchange interaction parameters  $J_{ij}$  between sites  $i$  and  $j$  using the obtained electronic structure. The expression for the exchange parameters  $J_{ij}$  in a Heisenberg Hamiltonian is given in Ref. [19]:

$$J_{ij} = \frac{1}{4\pi} \sum_{LL'} \text{Im} \int_{-\infty}^{\epsilon_F} d\epsilon \Delta_i^i(\epsilon) T_{LL'}^{ij\uparrow}(\epsilon) \Delta_i^j(\epsilon) T_{LL'}^{ij\downarrow}(\epsilon)$$

Here  $T_{LL'}^{ij\sigma}(\epsilon)$  is the scattering path operator in the site ( $i, j$ ) representation for different spin projections ( $\sigma = \uparrow, \downarrow$ ), and  $\Delta_i^i(\epsilon) = t_{i\uparrow}^{-1} - t_{i\downarrow}^{-1}$  is the difference of the inverse single-site scattering matrices. Since  $J_{ij}$  decreases fairly rapidly as a function of the distance, the calculation is limited to the few nearest neighboring pairs only. The  $J_{ij}$ 's for pairs across the hard/soft interface are averaged over the atomic pairs between the two layers and the results for the two interface models are  $-8.8$  meV for SmCo<sub>5</sub>/Co and  $-31.6$  meV for SmCo<sub>5</sub>/CoFe. The minus sign of  $J_{ij}$  indicates that the systems are in the ferromagnetic state. It is clear that the site-to-site exchange parameters of the interface atomic pairs in system SmCo<sub>5</sub>/CoFe are larger (absolute values) than those of the corresponding pairs in system SmCo<sub>5</sub>/Co. This also supports that the inter-phase exchange coupling in system SmCo<sub>5</sub>/CoFe is stronger than that in system SmCo<sub>5</sub>/Co, in agreement with the present noncollinear magnetic order simulation as discussed above.

Considering the other possible doping sites, we take into account the possibilities of both the Fe atoms diffusion and being Fe-doped into the hard phase SmCo<sub>5</sub>.<sup>20</sup> One Fe

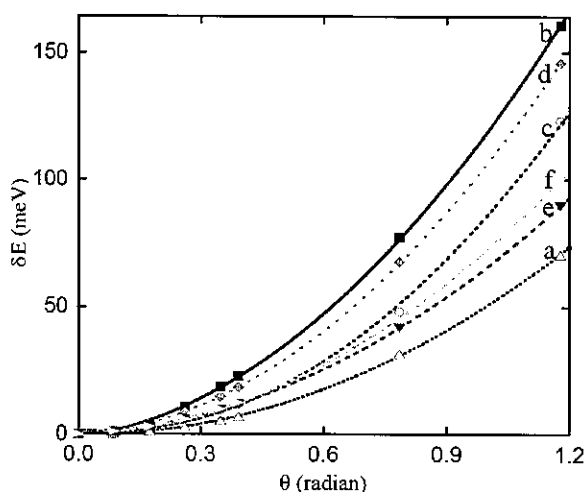


Fig. 5. The calculated total energy differences,  $\delta E(\theta) = E(\theta) - E(\theta = 0)$  (the marks), together with their fittings for the four Fe-doped hard phase ( $\text{SmCo}_5$ ) systems. For the comparison purpose, the two curves from Figure 3 are also plotted as the upper (curve b) and the lower (curve a) bounds. The curves c and d are for an Fe substitution of a Co in the first and second atomic layer from the interface while the soft phase is  $\text{CoFe}$ . The curves e and f are for an Fe substitution of a Co in the first and second atomic layer from the interface while the soft phase is pure  $\text{hcp Co}$ .

substitution of a Co atom in the first layer and the second layer from the interface in the  $\text{SmCo}_5$  part has been studied separately in the two soft phase models discussed earlier (see Fig. 1).  $\delta E(\theta)$  versus  $\theta$  curves of these systems from the noncollinear magnetic order calculations are all shown in Figure 5, with the two curves in Figure 3 shown again for comparison purpose. The curve of  $\text{SmCo}_5/\text{CoFe}$  model serves as upper bound while the  $\text{SmCo}_5/\text{Co}$  model serves as the lower bound. For the soft  $\text{CoFe}$  model (Fig. 1(b)), an Fe in the second layer and in the first layer in the hard phase causes degradation of the exchange coupling, showing by the curvatures of these curves which lie right below the upper bound curve (Curves c and d in Fig. 5). With the pure Co as soft phase, however, Fe doping to the first and second layer of the hard phase strengthens the exchange coupling since the two corresponding curves (Curves e and f in Fig. 5) are above their reference, i.e., the lower bound. In both soft phase cases, Fe in the second layer of the hard phase has a stronger exchange coupling than that of Fe in the first layer of the hard phase. On the other hand, comparing the curves in Figure 5 for pair systems (a) and (b), (c) and (e), and (d) and (f), where the hard phase is the same for each pair, we can conclude that with a same hard phase, doping of Fe in soft phase will improve the exchange coupling. Overall, system (b), in which  $\text{SmCo}_5$  is hard phase and Fe doped Co is soft phase, has the strongest exchange coupling among the systems we considered.

In conclusion, we have carried out first-principles calculations of the exchange coupling across  $\text{SmCo}_5/\text{Co}$  multilayer. Using both the noncollinear magnetic structure simulation and the calculation of the site-to-site exchange parameters across the interfaces, we found that the exchange coupling in  $\text{SmCo}_5/\text{Co}$  is enhanced by introducing Fe in the soft phase. However, the introduction of Fe atom into the hard phase,  $\text{SmCo}_5$ , will have the different effect, depending on the soft phase composition. For a pure  $\text{hcp Co}$  soft phase, it enhances the exchange coupling. But for the  $\text{Co}_{1-x}\text{Fe}_x$  soft phase, it leads to the degradation of the exchange coupling.

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## References and Notes

1. F. Kneller and R. Hawig, *IEEE Trans. Magn.* 27, 3588 (1991).
2. S. D. Bader, *Rev. of Mod. Phys.* 78, 1 (2006).
3. R. Skomski and J. M. D. Coey, *Permanent Magnetism*, Taylor & Francis, Bristol, U.K. (1999), p. 13.
4. J. Zhang, K. Takahashi, R. Gopalan, and K. Hono, *Appl. Phys. Lett.* 86, 122509 (2005).
5. Z. S. Shan, J. P. Liu, V. M. Chakka, H. Zeng, and J. S. Jiang, *IEEE Trans. Magn.* 38, 2907 (2002).
6. Z. J. Guo, J. S. Jiang, J. E. Pearson, S. D. Bader, and J. P. Liu, *Appl. Phys. Lett.* 81, 2029 (2002).
7. G. Asti, M. Solzi, M. Ghidini, and F. M. Neri, *Phys. Rev. B* 69, 174401 (2004).
8. Y. Choi, J. S. Jiang, Y. Ding, R. A. Rosenberg, J. E. Pearson, S. D. Bader, A. Zambano, M. Murakami, I. Takeuchi, Z. L. Wang, and J. P. Liu, *Phys. Rev. B* 75, 104432 (2007).
9. A. J. Zambano, H. Oguchi, I. Takeuchi, Y. Choi, J. S. Jiang, J. P. Liu, S. E. Loffland, D. Josell, and L. A. Bendersky, *Phys. Rev. B* 75, 144429 (2007).
10. R. F. Sabirianov and S. S. Jaswal, *J. Magn. Magn. Mater.* 177–181, 989 (1998).
11. R. F. Sabirianov and S. S. Jaswal, *Phys. Rev. B* 58, 12071 (1998).
12. G. Kresse and J. Hafner, *Phys. Rev. B* 47, R558 (1993).
13. G. Kresse and J. Hafner, *Phys. Rev. B* 49, 14251 (1994).
14. G. Kresse and J. Furthmüller, *Phys. Rev. B* 54, 11169 (1996).
15. P. E. Blöchl, *Phys. Rev. B* 50, 17953 (1994).
16. O. K. Andersen, *Phys. Rev. B* 12, 3060 (1975).
17. O. Gunnarsson, O. Jepsen, and O. K. Andersen, *Phys. Rev. B* 27, 7144 (1983).
18. O. N. Mryasov, R. F. Sabirianov, A. J. Freeman, and S. S. Jaswal, *Phys. Rev. B* 56, 7255 (1997). The ratio of the Wigner-Seitz radii of Sm, Fe and Co in the composite systems was set to be 1.4:1.04:1.00, as the ratio of their corresponding atomic radii. An  $8 \times 3 \times 10$   $k$ -point set were used in the integration over Brillouin zone assuring the convergence of electronic and magnetic results for this systems.
19. A. I. Liechtenstein, M. I. Katsnelson, V. P. Antropov, and V. A. Gubanov, *J. Magn. Magn. Mater.* 67, 65 (1987).
20.  $\text{SmFe}_3$  is unstable phase and tends to produce multiphase composites containing 2–17 and 1–3 phases. Thus, Fe substitute to hard phase is not desirable.

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