Magnetic properties of hard/soft composites: $SmCo_5/Co_{1-x}Fe_x$

R. F. Sabiryanov and S. S. Jaswal
Department of Physics and Astronomy and Center for Materials Research and Analysis, University of Nebraska, Lincoln, Nebraska 68588-0111
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First-principles calculations are carried out to study the magnetic hardening of $(SmCo_5)/(Co_{1-x}Fe_x)$ multilayers. The multilayer stacking along the c axis of the hexagonal hard phase $(SmCo_5)$ and the (111) direction of the fcc soft phase $(Co_{1-x}Fe_x)$ is well matched structurally. The self-consistent spin-polarized electronic structure results are used to calculate the magnetic moments and the exchange interaction parameters. The average magnetic moments of the soft (Co_3Fe) and hard phases are $1.83\mu_B$ and $1.2\mu_B$ per atom, respectively. A continuum model of the periodically layered hard/soft composite predicts the optimum thickness of the soft phase to be approximately 13 nm independent of the thickness of the hard phase. Calculated exchange parameters predict the Curie temperature of the hard/soft system to be between the values for each phase (1000–1388 K) depending on the relative thicknesses of the two phases. The optimum theoretical limit to the energy product of the composite is ~65 MGOe, which is almost twice the value for the hard phase.
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I. INTRODUCTION

Substantial efforts are being made to improve the properties of permanent-magnet materials, i.e., to increase the energy product $[(BH)_{\max}]$ and the Curie temperature (T_c). A recent innovation in this area is the possibility of exchange coupling of the hard and soft magnetic phases proposed by Kneller and Hawig.[1] The basic idea here is to combine the large saturation magnetization (M_0) of the soft phase with the large magnetic anisotropy of the hard phase to produce a composite with superior hard-magnet properties.[2] Our recent calculations[3] predict such an improvement for FePt/Fe multilayers and the experimental results[4] also look promising.

$SmCo_5$ has the largest anisotropy field (240–440 kOe) among the rare earth–transition metal (RE-TM) compounds and also has a high Curie temperature (1000 K). However, it has a relatively low theoretical value of the energy product $[(BH)_{\max}^{\text{theor}}=1/4(\mu_0 M_0)^2=33\text{ MGOe}]$, an upper bound to $(BH)_{\max}$. An iron-based bcc $Fe_{65}Co_{35}$ alloy has the largest magnetization $(M=2.43\mu_B)$ making it the best candidate as a soft phase. Since $SmCo_5$ has a strong mismatch in lattice constants to those of bcc Fe, it is quite possible that the use of bcc Fe as a soft phase can stabilize structures different from $SmCo_5$ ($SmCo_3$, Sm_2Co_7, $SmCo_5$, and Sm_2Co_{17} have similar formation energies). The fcc (close-packed) Fe phase has much weaker exchange interactions and therefore is not useful as a soft phase in an exchange spring magnet.[5] Among the fcc phases Co has the highest Curie temperature (1388 K) and large magnetization. An addition of 10–15 % of iron to Co stabilizes the fcc phase and increases the magnetization to 1.8 T, which corresponds to $(BH)_{\max}^{\text{theor}}$ of 82 MGOe. Also the lattice mismatch is only about 1% when the (111) direction of fcc Co is aligned with the c axis of $SmCo_5$ as shown in Fig. 1. Thus $SmCo_5$ and $Co_{1-x}Fe_x$ are excellent candidates as hard and soft phases, respectively, for an exchange spring composite.

We present here first-principles calculations of the magnetization, exchange interactions, and T_c of $SmCO_5/Co_{1-x}Fe_x$ composites represented by a periodic layered structure shown in Fig. 1. These results along with the experimental anisotropy of the hard phase are used in a micromagnetic analysis due to Skomski and Coey[2] to find the optimal thicknesses of the hard and soft phases.

II. METHOD

The self-consistent spin-polarized electronic structure calculations are carried out using the linear-muffin-tin-orbital (LMTO) method in atomic sphere approximation and in near orthogonal representation.[6]

The electronic structure results are used to calculate the

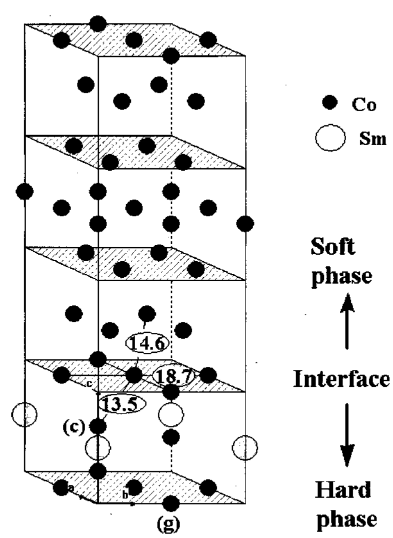

FIG. 1. Crystal structure near the interface of hexagonal $SmCo_5$ and fcc Co with the c axis of $SmCo_5$ and the (111) direction of Co aligned normal to the interface. The interface pair-exchange parameters (meV) are shown in the figure.

magnetization and the exchange parameters J_{ij} in the Heisenberg Hamiltonian. The expression for J_{ij} derived in Ref. 7 is as follows:

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

Here $T_{LL'}^{ij\sigma}$ 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 interaction of a given site 0 with all the other sites i,

$$J_0=\sum_{i\neq 0}J_{0i}, \tag{2}$$

can also be calculated from the relation[7]

$$J_0=-\frac{1}{4\pi}\sum_{LL'}\ \mathrm{Im}\int_{-\infty}^{\varepsilon_F}d\varepsilon\Delta_l^0(\varepsilon)(T_{LL'}^{00\uparrow}(\varepsilon)-T_{LL'}^{00\downarrow}(\varepsilon))+\Delta_l^0(\varepsilon)T_{LL'}^{00\uparrow}(\varepsilon)\Delta_{l'}^0(\varepsilon)T_{LL'}^{00\downarrow}(\varepsilon). \tag{3}$$

The procedure to calculate $T_{LL'}^{ij\sigma}$ in the LMTO formalism has been developed by Gunnarsson, Jepsen, and Andersen.[8] The exchange parameters are calculated for nearest and next-nearest neighbors.

Skomski and Coey[2] minimize the magnetic energy of a periodical layered structure in the presence of a nucleation field to arrive at the following micromagnetic relation among the various parameters of hard and soft phases:

$$\sqrt{(2K_h-M_hH_N)/2A_h}\tanh\left(\frac{\lambda_h}{2}\sqrt{(2K_h-M_hH_N)/2A_h}\right)=\frac{A_s}{A_h}\sqrt{M_sH_N/2A_s}\tan\left(\frac{\lambda_s}{2}\sqrt{M_sH_N/2A_s}\right), \tag{4}$$

where M_h and M_s are the saturation magnetizations in hard and soft phases, respectively, K_h is the anisotropy constant of the hard phase (anisotropy of the soft phase is neglected), A_h and A_s are the exchange stiffness constants for hard and soft phases, respectively, H_N is the nucleation field, and λ_s and λ_h are the soft and hard layer thicknesses, respectively. The exchange stiffness constants A of the continuum model are related to the exchange parameters of the discrete model as follows. Stiffness tensor of spin waves $D_{\alpha\beta}$ is defined as follows:[9]

$$D_{\alpha\beta}=\sum_{ij}J_{ij}(\mathbf{R}_i-\mathbf{R}_j)_\alpha(\mathbf{R}_i-\mathbf{R}_j)_\beta, \tag{5}$$

where $\mathbf{R}_i$ is the position of the ith atom. The exchange stiffness constant for the crystals with inversion symmetry is connected with D through the relation

$$A_{\alpha\beta}=\frac{D_{\alpha\beta}}{V}, \tag{6}$$

where V is the volume of the unit cell.

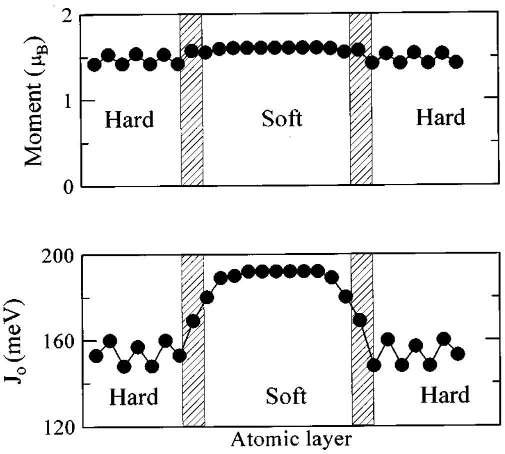

FIG. 2. Magnetic moments and total exchange parameters of various Co atoms in a hard/soft multilayer.

III. RESULTS AND DISCUSSION

Calculations were carried out for three supercells with 3, 6, and 12 layers of Co and four units of $SmCo_5$ in each case. The results for magnetization and exchange parameters are almost the same in each case. The calculated magnetic moments and effective exchange parameters J_0 for various Co sites for the largest supercell composite are plotted in Fig. 2. The local magnetic moments in two phases are quite similar as expected. The exchange parameters in the soft phase are slightly larger than those in the hard phase reflecting the difference in their T_c. The exchange parameters at the interface are in between those of the two phases. It is clear from the calculated exchange parameters that T_c for a composite is expected to be in between the values of T_c for $SmCo_5$ (1000 K) and Co (1388 K) depending on the relative amount of the two phases.

These results show that $SmCo_5$/Co are strongly exchange coupled with excellent hard-magnet properties. As mentioned in the introduction, a small amount of Fe is necessary to stabilize Co in the fcc phase. We find that the exchange stiffness parameters for Co-rich $Co_{1-x}Fe_x$ are almost independent of Fe concentration. Therefore, we have chosen fcc Co_3Fe as a representative of the soft phase $Co_{1-x}Fe_x$ in the following micromagnetic calculations.

The micromagnetic equation (4) used to find the optimum thicknesses of the two phases neglects the changes in the magnetic properties at the interface. This is not expected to be a serious limitation for determining the optimum thicknesses of two phases in the present composite because the interface layer exchange coupling parameters are very close to the ones in pure phases as can be seen from Table I and Fig. 1. There is a decrease in the interplane pair-exchange parameters to the side of the hard phase but an increase in the in-plane exchange for the interfacial layer, which results in the value of J_0 to be intermediate between the hard and soft phase values (Fig. 2). The pair exchange parameters for the bulk phases needed in Eq. (5) are listed in Table I. Since the exchange parameters J_{ij} decrease fairly rapidly as a function of the distance in this system, limiting the computation of J_{ij} to first and second neighbors is reasonable. Equations (5) and (6) give the exchange stiffness parameters as follows:

TABLE I. Pair exchange parameters in Co, Co_3Fe, and $SmCo_5$.

Compound	Neighbors Type	Number	Distance (Å)	J_{ij} (meV)
Co	Co(a)-(a)	12	2.519	14.14
	Co(a)-(a)	6	3.563	1.30
Co_3Fe	Co(a)-Co(a)	8	2.519	13.19
	Co(a)-Fe(a)	4	2.519	14.51
	Co(a)-Co(a)	6	3.563	3.13
	Fe(a)-Fe(a)	6	3.563	7.75
$SmCo_5$	Co(c)-(g)	6	2.512	15.22
	Co(c)-(c)	3	2.861	4.46
	Co(c)-(c)	2	4.131	3.65
	Co(c)-(c)	12	4.311	2.11
	Co(g)-(g)	4	2.478	15.65
	Co(g)-(c)	4	2.512	15.22
	Co(g)-(g)	2	4.131	−0.39
	Co(g)-(g)	4	4.291	0.73

$$D_{xx}^{\mathrm{Co_3Fe}}=462\ \ \mathrm{meV\ Å}^2,\quad A=1.3210^{-11}\,\frac{J}{m}=A_s\,,$$

$$D_{xx}^{\mathrm{SmCo_5}}=308\ \ \mathrm{meV\ Å}^2,\quad A_{xx}=0.8810^{-11}\,\frac{J}{m},$$

$$D_{zz}^{\mathrm{SmCo_5}}=379\ \ \mathrm{meV\ Å}^2,\quad A_{zz}=1.0810^{-11}\,\frac{J}{m}=A_h\,.$$

The magnetization values for two phases are

$$M(\mathrm{SmCo_5})=1.05T=M_h\,,$$

$$M(\mathrm{Co_3Fe})=1.8T=M_s\,.$$

We use the value of $K_h=17(MJ/m^3)$ from the experiment.[10] Using the parameters listed above, the micromagnetic Eq. (4) is solved numerically for the optimal thicknesses of hard and soft phases for a given $(BH)_{\max}^{\mathrm{theor}}$. $(BH)_{\max}^{\mathrm{theor}}$ is an upper limit to the energy product that can be realized for any material.[11] This is based on the assumption of a rectangular hysteresis loop with $H_N=M_0/2$, where M_0 is the saturation magnetization. The results are presented in Fig. 3. We see that the thickness of the soft phase remains constant (∼13 nm) while the thickness of the hard phase decreases with increasing $(BH)_{\max}^{\mathrm{theor}}$. Around hard layer thickness of 2 nm, the soft layer thickness begins to decrease in thickness. One should keep in mind that the use of the continuum model is questionable for 2 nm and thinner layers. One needs to use discrete models to deal with such structures. We see from Fig. 3 that $(BH)_{\max}^{\mathrm{theor}}$ is 65 MGOe for hard and soft multilayers with thicknesses of 5 and 13 nm, respectively. Such a multilayer is expected to have a T_c of ∼1200 K, making it a good candidate for a high-temperature permanent-magnet material. Stacking of hcp Co and $SmCo_5$ in the (1100) direction also has a very good match at the interface and such a layered structure may form with easy axis perpendicular to the layering direction. In this case the relevant exchange stiffness constant of the hard phase is smaller than that in the c direction due to the anisotropy of the exchange coupling in $SmCo_5$. However, this will only marginally decrease the optimal thickness of the soft phase. We hope the present calculations will stimulate experimental studies of this very promising hard/soft composite. Finally, the exchange stiffness parameters calculated here can be used to perform micromagnetic simulations of any other experimentally realizable nanostructures of this composite.

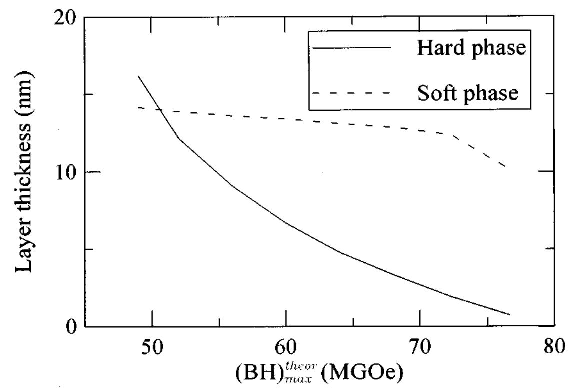

FIG. 3. The optimal thicknesses of hard and soft phases versus the theoretical energy product $(BH)_{\max}^{\mathrm{theor}}$ for $SmCo_5/Co_3Fe$ multilayers.

IV. CONCLUSIONS

We have carried out first-principles studies of the magnetic properties of $SmCo_5/Co_{1-x}Fe_x$ multilayers. An addition of a small amount of Fe to Co makes the soft phase fcc. The multilayer stacking along the c-axis of the hard phase and the (111) direction of the soft phase is well matched structurally. A micromagnetic analysis of the composite gives the thickness of the soft phase as 13 nm, which is independent of the thickness of hard phase. An optimal value of the theoretical energy product is ∼65 MGOe for hard/soft thicknesses of 5/13 nm. Curie temperature for this composite is expected to be ∼1200 K making $SmCo_5/Co_{1-x}Fe_x$ a very good candidate as a high-temperature hard magnet.

ACKNOWLEDGMENTS

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