Numerical Study of Enhanced Coercivity of a Magnetically Hard Grain with Thin Surface Layers due to Antiferromagnetic Coupling

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The effects of a thin ferromagnetic layer covering a magnetically hard grain on the coercivity were calculated based on micromagnetic theory. Antiferromagnetic exchange coupling between this thin ferromagnetic surface layer (SF layer) and the hard grain was found to prevent nucleation of reverse domains at the surface of the hard grain and to increase the coercivity. This was effective for hard grains with a layer of reduced magnetic anisotropy on their surfaces whose thickness is comparable with the exchange length. It was also found that a SF layer thickness of several nanometers is suitable. The effects were simulated for a hard grain with SF layers coupled both ferromagnetically and antiferromagnetically. It was found that the coercivity can be enhanced by antiferromagnetically coupled SF layers lying parallel to the easy magnetization axis of the hard grain.

Index Terms—Antiferromagnetic coupling, coercive force, exchange coupling, nucleation, permanent magnet.

I. INTRODUCTION

The effect of grain boundary phases on the coercivity H_c of sintered Nd-Fe-B magnets has recently been studied. When nonmagnetic boundaries disrupt the exchange coupling between Nd₂Fe₁₄B grains, magnetization reversal propagates to the whole magnet through only the magnetostatic interaction [1]. This is expected to give a higher H_c than that for a magnet with magnetic boundaries [2, 3], when H_c is determined by the magnetic field required for the propagation of reverse domains. On the other hand, composition analysis by a three-dimensional atom probe suggests that sintered Nd-Fe-B magnets have ferromagnetic boundary phases [4]. In this case, the magnetization reversal will spread to the whole magnet though exchange and magnetostatic interactions. Thus, exchange coupling between the Nd₂Fe₁₄B grain and boundary phases is expected to greatly affect H_c .

One of important parameters determining the role of boundary phases is the exchange interaction J between boundary and magnetically hard phases. Depending on the on J , grain boundaries can function as nucleation sites, a propagating medium for magnetization reversal, and pinning sites for the domain wall. It is also known that grain boundaries coupling antiferromagnetically with a hard grain induce exchange anisotropy and increase H_c [5–7]. Some ferromagnetic materials couple antiferromagnetically through the RKKY interaction; Co–Ru–Co has been reported to have the strongest RKKY interaction (-5×10^{-3} J/m²) [8]. This phenomenon change magnetization process of the coupled substances [9] and is used as an exchange bias for memory materials [9,10]. In α -Fe(001)/Fe₃O₄(001), the two ferromagnetic phases directly exchange couple antiferromagnetically (antiparallely) and their magnetization process is affected by the exchange interaction [11].

The possibility of antiferromagnetic exchange coupling between α -Fe(110) and Nd₂Fe₁₄B(100) has recently been predicted by first-principles calculations [12]. The exchange anisotropy induced by the antiferromagnetic coupling is expected to affect H_c of Nd₂Fe₁₄B similarly to the cases in the

previous materials [5–10]. We may enhance H_c of magnets by using antiferromagnetic coupling between boundary and hard magnetic phases. However, the magnetic anisotropy of recent hard magnetic materials is much higher than that of the previous materials mentioned above, and it is not clear if we can enhance H_c of magnets with high magnetic anisotropy.

In the present study, we calculated the demagnetization process for a magnetically hard grain with a thin ferromagnetic surface layer (a SF layer) which is antiferromagnetically exchange coupled with the magnetization of the hard grain. It was found that H_c can be enhanced by a thin SF layer antiferromagnetically coupled with the hard grain when the exchange coupling is sufficiently strong.

II. PRINCIPLE OF ENHANCED COERCIVITY BY AN ANTIFERROMAGNETICALLY COUPLED LAYER

To evaluate the effect of an antiferromagnetically coupled layer on H_c , we consider the one-dimensional model magnet shown in Fig. 1, which extends infinitely in the y - and z -directions. The magnetically hard grain has uniaxial anisotropy, except at the imperfect layer (the IP layer) and the easy magnetization axis lies in the z -direction. The IP layer, which is the surface of the hard grain, is thought to lose its magnetic anisotropy due to the presence of crystal imperfections within it. Aharoni [13, 14] assumed imperfect crystal anisotropy to explain H_c of a magnet. Furthermore, the existence of imperfect crystal anisotropy has been predicted for a Nd-Fe-B magnet [15]. The hard grain is covered by a SF layer, which has a thickness D_s , and it directly exchange couples antiferromagnetically with the hard grain.

Calculations by Aharoni [13] predict that the IP layer of the hard grain reduces the nucleation field. The solid curve in Fig. 2 shows a typical variation of the nucleation field for a hard grain without a SF layer as a function of the

logarithm of the IP layer thickness, D_i . When the hard grain is covered by the SF layer, a field applied in the z -direction stabilizes the magnetization configuration shown in Fig. 1 and prevents reverse domain nucleation at the IP layer. Assuming that the magnetization directions in the SF and IP layers, and inside the hard grain are tilted from the z -direction by angles of $\pi - \theta_s$, θ_s and θ_p , respectively, and neglecting the anisotropy of the SF layer, the magnetic energy W_{bk} stored in the model is given by

$$W_{bk} = H_a \left\{ \mu_0 M_i D_i \left(1 - \frac{M_s D_s}{M_i D_i} \right) \cos \theta_i + M_p D_p \cos \theta_p \right\} + J_{pi} \{ 1 - \cos(\theta_i - \theta_p) \} + D_i K_{ui} \sin^2 \theta_i, \quad (1)$$

where K_u is the anisotropy constant, $\mu_0 M$ is the saturation polarization, and D is the layer thickness. The subscripts indicate the layer. Furthermore, H_a is the applied field and J_{pi} is the exchange constant between the IP layer and the inside of the hard grain (the perfect (PF) region). As we assume a strong negative exchange interaction between the SF and IP layers and an antiparallel configuration for M_s and M_i , the exchange energy, W_{AF} , stored in the interface between these layers is independent of the magnitude of the applied field. We thus omit W_{AF} in (1), which shows that the presence of the SF layer reduces the effective thickness of the IP layer from D_i to $D_i \{ 1 - M_s D_s / (M_i D_i) \}$. Therefore, the presence of the SF layer increases the nucleation field, as shown in Fig. 2.

III. THREE-DIMENSIONAL SIMULATION

1) Calculation Model and Method

We applied the above concept in three-dimensional calculations. The two models depicted in Fig. 3 were employed in these calculations. The hard grain has an IP layer

Fig. 1 Model magnet with a thin ferromagnetic layer antiferromagnetically coupled with a magnetically hard grain.

Fig. 2 Effect of an antiparallel coupled layer on the nucleation field.

so that it is not magnetically anisotropic and its surfaces are covered by SF layers. The magnetization of the SF layer antiferromagnetically exchange couples with that of the hard grain at all the interfaces in model A (Fig. 3(a)). Since the calculations in Ref. 11 predict that the exchange interaction between α -Fe and $\text{Nd}_2\text{Fe}_{14}\text{B}$ will be negative in the a -plane but positive in the c -plane, we also employed model B (Fig. 3(b)). In model B, the magnetization of the SF layer couples antiferromagnetically with that of the hard grain at interfaces lying parallel to the xz and yz planes, although it couples ferromagnetically at interfaces lying parallel to the xy plane. The assumed positive and negative values of J_{s-i} indicate ferromagnetic and antiferromagnetic couplings, respectively. Both models are composed of 32,768 cubic elements and periodic boundary conditions were applied in the x -, y -, and z -directions. A field was applied in the z -direction.

TABLE I lists the simulation parameters used in this investigation. We used $\text{Nd}_2\text{Fe}_{14}\text{B}$ as the hard grain. We set K_u of the IP layer to zero as a first-order approximation [13], although various distributions of K_u have been employed in previous investigations [13, 14, 16, 17]. Employing another distribution may change the optimal SF layer thickness but it will not affect the general tendency of H_c . K_u was also set to zero for the SF layer to eliminate the effect of the magnetic anisotropy.

In model A, we did not specify the material of the SF layer and we assumed a saturation polarization of 1 T and an exchange stiffness constant of 2.5×10^{-11} J/m, both of which are typical values for a ferromagnetic material. We took the SF layer to be α -Fe in model B. As mentioned above, negative exchange interactions constants have been determined for α -Fe/ Fe_3O_4 interface and predicted for α -Fe/ $\text{Nd}_2\text{Fe}_{14}\text{B}$ one; the determined value is -2.2×10^{-3} J/m² for the α -Fe/ Fe_3O_4 interface and the maximum predicted value for the α -Fe/ $\text{Nd}_2\text{Fe}_{14}\text{B}$ interface is -1.3×10^{-2} J/m². We thus varied the negative exchange constant J_{s-i} between 10^{-2} and 10^{-4} J/m².

The stable direction of the magnetization vector in each element was determined for a given applied field by minimizing the total magnetic energy stored in the model magnet, which includes the anisotropy, exchange, Zeeman, and magnetostatic energies, as given by micromagnetic theory. Demagnetization curves were obtained by varying the magnitude of the applied field. Details of the calculation

Fig. 3 Model grains used in three-dimensional simulations. In model A, all the interfaces between the IP and SF layers have negative exchange interactions, whereas those lying parallel to the xy plane have a positive exchange interaction in model B.

TABLE I SIMULATION PARAMETERS.

Parameters	Values
Model size : L [nm]	48
SF layer	
Thickness : D_s [nm]	0 – 6
Exchange stiffness constant : A [J/m]	2.5×10^{-11}
Saturation polarization : $\mu_0 M$ [T]	
Model A	1.0
Model B	2.16
Anisotropy constant : K_u [MJ/m ³]	0
Hard grain	
Exchange stiffness constant : A [J/m]	8.7×10^{-12}
Saturation polarization : $\mu_0 M$ [T]	1.61
Anisotropy constant : K_u [MJ/m ³]	
Perfect region	4.5
Imperfect layer	0
Thickness of : imperfect layer : D_i [nm]	0 – 6
Exchange constant between SF layer and hard grain : J_{s-i} [J/m²]	
Model A	-1.66×10^{-1} – -1.66×10^{-4}
Model B	
Parallel plane	-1.3×10^{-1} – -1.3×10^{-4}
Perpendicular plane	6.5×10^{-3}

procedure have been reported elsewhere [18].

2) Results and Discussion for Model-A

Figure 4 shows the calculated demagnetization curves for model A. In the calculation, the SF and IP layer thicknesses were respectively set to 1.5 and 3.0 nm and the exchange constant between the SF and IP layers, J_{s-i} , was varied between -1.66×10^{-1} and -1.66×10^{-4} J/m². When the exchange interaction was weak ($J_{s-i} = -1.66 \times 10^{-4}$), the presence of a SF layer gave a lower H_c than that (0.226) when no SF layer was present. H_c gradually increased with increasing $|J_{s-i}|$ and was enhanced for $|J_{s-i}| > 1 \times 10^{-3}$ J/m². This value is equivalent to the exchange constant of Fe(100)/Fe₃O₄(100) [10], but is smaller than that predicted for α -Fe(110)/Nd₂Fe₁₄B(100) [11]. The enhancement of H_c can be attributed to stabilization of the magnetization at the IP layer due to antiferromagnetic coupling between the magnetizations of the SF and IP layers.

Figure 5 shows the effects of D_i and D_s on the coercivity h_c , which is the value of H_c normalized by the anisotropy field of the PF region of the hard grain. In these calculations, J_{s-i} was set to -1.66×10^{-2} J/m². As shown in Fig. 5, h_c decreases with increasing D_i and increases with increasing D_s . The observed enhancement of h_c saturated at around $D_s = 4.5$ nm, which suggests that a SF layer that is several nanometers thick is sufficient to suppress reverse domain nucleation from the IP layer. For the grain with $D_i = 3.0$ nm, the saturated h_c value is approximately two times higher than that for $D_s = 0$. On the other hand, the effect of the SF layer was not significant for a thick IP layer. This is thought to

be because magnetization reversal initiates inside the IP layer, which is far from the interface between the SF and IP layers. Thus, the presence of a SF layer is effective when the thickness of the IP layer is comparable to the exchange length of the hard grain (1.4 nm).

Indirect exchange coupling between the IP and SF layers via the RKKY interaction [8] seems to have a similar effect as direct interaction. Regardless of direct or indirect exchange coupling, a large $|J_{s-i}|$ value exceeding -1×10^{-3} J/m² is required to enhance H_c .

3) Results and Discussion for Model-B

As the magnetization configuration of model A has not been realized for real magnets, we also calculated H_c for hard grains with both antiferromagnetically and ferromagnetically coupled SF layers, based on the prediction for α -Fe/Nd₂Fe₁₄B

Fig. 4 Effect of magnitude of exchange interaction constant at the interface between SF and IP layers on the demagnetization curves of model A. The SF and IP layer thicknesses were set to 1.5 and 3.0 nm, respectively. The magnetization and applied field are normalized by the saturation magnetization and the anisotropy field of the PF region of the hard grain, respectively.

Fig. 5 Effect of thicknesses of (a) IP layer D_i and (b) SF layer D_s on the normalized coercivity of model A. The upper axis represents the thickness normalized by the exchange length ($= 1.4$ nm).

Fig. 6 Effect of magnitude of negative exchange constant between IP and SF layers on the normalized coercivity. The IP and SF layer thicknesses were set to 3 nm.

[12]. Figure 6 shows a plot of h_c as a function of the strength of negative exchange interaction. In these calculations, D_s and D_i were fixed at 3.0 nm. h_c increased with increasing $|J_{s-i}|$ and it exceeded 0.226 (the h_c value for the model without an SF layer) at $|J_{s-i}| = 1.3 \times 10^{-3} \text{ J/m}^2$. This result strongly suggests that the SF layer can enhance H_c , even if some of the SF layers have ferromagnetic coupling. This behavior can be explained as follows. As our model has IP layers on the hard grain, reverse domain nucleation occurs at some IP layers. IP layers lying parallel to the easy axis of the PF region have a smaller nucleation field H_N than IP layers lying perpendicular to the easy axis because nucleation at the perpendicular layers generates a high demagnetization field. In model B, the SF layers lying parallel to the easy axis antiferromagnetically exchange couple with the IP layers with a small H_N value, preventing nucleation within them and enhancing H_c .

Figure 7 shows h_c as a function of D_s , where D_i was set to 3.0 nm. As seen in the figure, h_c exhibits a maximum at about 2.0 nm. This behavior of h_c differs from that shown in Fig. 4 and suggests that strict control of D_s is required to enhance H_c . The presence of a maximum can be attributed to the effect of ferromagnetic coupling, which tends to reduce H_c [19]. For $J_{s-i} = -1.3 \times 10^{-3} \text{ J/m}^2$, h_c was approximately 1.8 times higher due to the presence of a 2.0-nm-thick SF layer.

At the present, only α -Fe is reported to couple antiferromagnetically with $\text{Nd}_2\text{Fe}_{14}\text{B}$, and its role may be clarified experimentally by using thin film process or grain boundary diffusion process.

IV. CONCLUSIONS

The effects of a thin ferromagnetic layer covering a magnetically hard grain on the coercivity H_c were calculated based on micromagnetic theory. A thin surface (SF) layer exchange coupled antiferromagnetically with the hard grain was found to prevent reverse domain nucleation at the surface of the hard grain, and this can enhance H_c when the negative exchange interaction is sufficiently strong. The enhancement of H_c was remarkable for hard grains with a thin layer that destroys the magnetic anisotropy on their surfaces when its thickness is comparable with the exchange length. A suitable SF layer thickness was found to be several nanometers. We also calculated the effect of SF layers for both ferromagnetic and antiferromagnetic coupling with a hard grain and found that SF layers effectively enhance H_c when they are parallel to the easy axis of magnetization of the hard grain.

Fig. 7 Effect of SF layer thickness D_s on the normalized coercivity h_c of model B. D_i was set to 3.0 nm.

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