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Self-pinning: Dominant coercivity mechanism in exchange-coupled permanent/composite magnets

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Our micromagnetic calculation demonstrates that the dominant coercivity mechanism is self-pinning in most exchange-coupled permanent and composite magnets. Such a pinning is attributed to the change of the intrinsic parameters associated with the phase change at the interface. From this self-pinning some more specific formulas on pinning field can be derived. In particular, for sufficiently large soft grains/defects, the pinning field can be expressed as $H_p = \alpha H_K$, where $H_K = 2k/M_S$ is the anisotropy field and α depends on the material parameters and micromagnetic structures. For an exchange-coupled $\text{Nd}_2\text{Fe}_{14}\text{B}-\alpha\text{Fe}$ system with abrupt change of parameters in the interface, $\alpha \approx 0.1$. Reducing the size of the soft grain will increase the coercivity, while the smooth change of the parameters in the interface will lead to a reduction in the coercivity. Comparison with experimental data justifies our calculation. © 2007 American Institute of Physics.

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I. INTRODUCTION

The coercivity mechanism in permanent magnets has been debated for decades.¹⁻⁴ Currently there are mainly two coercivity mechanisms, nucleation and pinning, based on the role of the defects in the process of the magnetic reversal.

It has been proposed recently that the coercivity mechanism changes with the thickness of the soft layer in a hard/soft multilayer system.^{3,4} By comparing the calculated nucleation and pinning fields directly, it was found that the coercivity mechanism for a multilayer with the thin soft layer is nucleation, while that with the thick one is pinning.^{3,4} In this paper, the results are extended to the nanocrystalline permanent/composite magnets with microstructures considered. As there is some distribution of the soft defects/grains and the coercivity is defined at the large soft grains where the coercivity is small, we put forward that self-pinning is the dominant coercivity mechanism in such materials.

II. NUCLEATION AND PINNING FIELD

We start from a simple triple-layer system with easy axis parallel to the applied field. As the soft layer can be regarded as a planar soft defect, the result here can also be applied in single-phased permanent magnets. The total energy density of such a system can be written as^{3,4}

$$\gamma = \int_0^{L^h} \left[A^h \left(\frac{d\theta}{dz} \right)^2 + K^h \sin^2 \theta - M_S^h H \cos \theta \right] dz + \int_{-L^s/2}^0 \left[A^s \left(\frac{d\theta}{dz} \right)^2 + K^s \sin^2 \theta - M_S^s H \cos \theta \right] dz, \quad (1)$$

where A and K are the exchange and anisotropy energy constants, respectively. θ is the angle between the magnetization and the applied field H and M_S is the spontaneous magnetization. L is the thickness of the layer and the superscripts h and s denote the hard and soft layers, respectively.

By applying the variational method to the total energy density with suitable boundary conditions,^{3,4} we obtain the following nucleation field:

$$\frac{\pi L^s}{2\Delta^s} = \frac{\tan^{-1}[\sqrt{A^h K^h(1+h_N^h)}/\sqrt{A^s K^s(-1-h_N^s)}]}{\sqrt{-h_N^s-1}}, \quad (2)$$

where $h_N = -H/H_K$ is the reduced nucleation field and $H_K = 2K/M_S$ is the anisotropy field. $\Delta = \pi\sqrt{A/K}$ is the Bloch wall width. The superscripts h and s stand for the hard and soft layers, respectively.

Nucleation is a necessary but not sufficient condition for complete magnetic reversal. It sets a lower limit to the coercivity. Figure 1 compares the calculated nucleation field with the coercivity for $\text{Nd}_2\text{Fe}_{14}\text{B}/\alpha\text{-Fe}/\text{Nd}_2\text{Fe}_{14}\text{B}$ trilayer system. The material parameters are listed in Table I. It can be seen that the coercivity is identical to the nucleation field only for L^s less than a certain critical size L_{crit} , where the

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FIG. 1. Comparison of nucleation field with numerical and experimental coercivities (given by Ref. 6) for a $\text{Nd}_2\text{Fe}_{14}\text{B}/\text{Fe}/\text{Nd}_2\text{Fe}_{14}\text{B}$ trilayer system.

coercivity mechanism is totally nucleation. For $\text{Nd}_2\text{Fe}_{14}\text{B}/\alpha\text{-Fe}/\text{Nd}_2\text{Fe}_{14}\text{B}$ trilayer system, $L_{\text{crit1}} = 4$ nm. For large L^s there is a gap between the two fields and the coercivity equals the pinning rather than the nucleation field, indicating that pinning is the dominant coercivity mechanism. For sufficiently large L^s , H_c saturates at 6.82 kOe, the smallest pinning field, which can be derived analytically.

$$H_p^{\min} = \frac{2(A^h K^h - A^s K^s)}{(\sqrt{A^s M^s} + \sqrt{A^h M^h})^2}. \quad (3)$$

Similar result has been obtained by Kronmüller and Goll,⁵ but by using a different method.

Experimental results⁶ show similar trend in the change of coercivity mechanism. However, it is noted that the experimental coercivity is only about 40% of the theoretical pinning field. This discrepancy is attributed mainly to the abrupt change of the material parameters at the interface assumed in our calculation. Theoretically, a smoother change of magnetic parameters might be considered through adding an interface region between the soft interlayer and the hard layer. The magnetic parameters in this region, denoted by a superscript i , are expected to be between those of soft and hard phases. The propagation of the domain wall involves the movement from the soft zone to the interface region and then continues to the hard phase. The pinning fields of these two processes are given by Eq. (3), which can be rewritten as

TABLE I. Magnetic properties for various hard and soft magnetic materials at room temperature.

Material	M_S (kG)	K ($\times 10^{-7}$ ergs/cm ³)	A ($\times 10^{-7}$ erg/cm)	Δ (nm)	H_K (kOe)
$\text{Nd}_2\text{Fe}_{14}\text{B}$	1.28	4.3	7.7	4.2	67.2
$\text{Pr}_2\text{Fe}_{14}\text{B}$	1.25	5.6	7.7	3.7	89.6
$\text{Sm}_2\text{Fe}_{17}\text{N}_3$	1.23	12	10.7	3.0	195
SmCo_5	0.84	17.1	12.	2.6	407
$\text{Sm}_2\text{Co}_{17}$	1.19	2.91	9.5	5.7	48.9
$\alpha\text{-Fe}$	1.71	0.046	25.	73.2	0.54
Co	1.43	0.43	10.3	15.4	6.0

$H_{p1} = H_K^i(1 - \xi_1)/(1 + \sqrt{\eta_1})^2$ and $H_{p2} = H_K^h(1 - \xi_2)/(1 + \sqrt{\eta_2})^2$, where $\xi_1 = A^s K^s / (A^i K^i)$, $\xi_2 = A^i K^i / (A^h K^h)$, $\eta_1 = A^s M^s / (A^i K^i)$ and $\eta_2 = A^i M^i / (A^h K^h)$. The parameters in the interface region can be taken as a linear combination of the soft and hard ones, i.e., $K^i = K^h \zeta + K^s(1 - \zeta)$, $A^i = A^h \zeta + A^s(1 - \zeta)$, and $M_S^i = M_S^h \zeta + M_S^s(1 - \zeta)$. By setting $H_{p1} = H_{p2}$ we obtain $H_{p1} = H_{p2} = 0.4 H_K^h$ for a $\text{Nd}_2\text{Fe}_{14}\text{B}/\alpha\text{-Fe}/\text{Nd}_2\text{Fe}_{14}\text{B}$ trilayer system. This value agrees very well with the experimental data in Fig. 1.

III. SELF-PINNING: THE DOMINANT COERCIVITY MECHANISM

Now let us discuss the coercivity mechanism in exchange-coupled permanent and composite magnets with various microstructures, where many nucleation centers exist. According to the micromagnetics,⁷ the first nucleation starts where the nucleation field is the smallest, i.e., the defect sites for the permanent magnets. Only those reversed domains that can overcome the pinning fields and propagate through the whole material will lead to observable magnetic reversal and contribute to the coercivity. For every reversed domain nucleated, there are generally two kinds of pinning in the material. The first one is associated with the interface between the soft and hard phases, which surrounds the nucleation centers. The reversed domain walls have to overcome this pinning field to propagate into the hard phases. This pinning is inherently linked with the nucleation centers and is called as self-pinning in this paper. Once the domain walls entered the hard phases, it has to overcome the other pinning centers to spread to the whole material. The pinning field given by these traditional pinning centers is generally smaller than the self-pinning field for permanent and composite magnets. Thus the traditional pinning can be ignored in the following discussions.

On the one hand, as shown in Fig. 1 and discussed in the previous section, the nucleation field is generally smaller than the self-pinning field, in particular, for large defect size/thickness. For an exchange-coupled material consists of many nucleation centers, the coercivity is determined by the smallest self-pinning field, which corresponds to the largest defect size. In this defect size region, as shown in Fig. 1, the coercivity mechanism is pinning rather than nucleation. Therefore, the dominant coercivity mechanism in exchange-coupled permanent magnets is self-pinning and the coercivity is given by the smallest self-pinning field of the material, which, in many cases, is approximately given by Eq. (3).

If we ignore the K^s , which is about two orders of magnitude smaller than K^h , Eq. (3) can be rewritten as

$$H_p = \alpha H_K^h, \quad (4)$$

where $\alpha = 1/(1 + \sqrt{\eta})^2$ and $\eta = A^s M^s / A^h M^h$. Taking $\text{Nd}_2\text{Fe}_{14}\text{B}$ as an example, due to the existence of $\alpha\text{-Fe}$ as a soft defect, $\eta = 4.33$ (see Table I), the coercivity is roughly 10% of that of a perfect material. As discussed in the Sec. II, this coercivity could be less if the change of the parameters from the soft to hard phases are more gradual.

Such a self-pinning has both attributes of the traditional nucleation and pinning. On the one hand, similar equations

TABLE II. Calculated α and critical thickness of the soft phase for various groups of hard/soft materials.

Interface	$\frac{A^s M^s}{A^h M^h}$	α
Nd ₂ Fe ₁₄ B/ α -Fe	4.33	0.105
Pr ₂ Fe ₁₄ B/ α -Fe	4.44	0.104
SmCo ₅ /Co	1.46	0.205
Sm ₂ Fe ₁₇ N ₃ /Fe	3.25	0.127
Sm ₂ Co ₁₇ /Co	1.30	0.218

to that of Eq. (3) have been obtained by other groups, which is called as the pinning field. In addition, as illustrated in Ref. 3, from Eq. (3), some other formulas for pinning fields could be derived. On the other hand, such a self-pinning is due to the different parameters of the soft and hard phases and could be regarded as a part of nucleation. Kronmüller and co-workers from Max-Planck Institute, for example, obtain the same formula as Eq. (4) and took it as the nucleation field.^{1,8,9} In fact, for many experts in the field of micromagnetics, the nucleation field refers to the reverse field at which continuous, nonlocalized reversal processes such as coherent rotation, curling, or buckling initiate.⁸ From this point of view, the self-pinning field can be regarded as a kind of nucleation field.

Such a definition of the nucleation field, however, as pointed out by Livingston,¹⁰ is actually misleading since nucleation implies a localized (nucleus) rather than a nonlocalized behavior.

IV. COMPARISON WITH OTHER THEORIES AND EXPERIMENTAL DATA

Kronmüller and Goll,^{1,2} Bauer *et al.*,⁹ and Livingston have done a lot of valuable work in revealing the coercivity mechanisms in permanent magnets, both theoretically and experimentally. By careful fitting of temperature dependent of coercivity they found that the experimental coercivity is a linear function of H_k , i.e., $H = \alpha H_k + N_{\text{eff}} M_s$, for various permanent and composite materials. Apart from a correction term due to the shape effect, this equation is the same as Eq. (4). For the exchanged-coupled permanent/composite magnetic systems discussed here, the correction term is very small and could be ignored. The fitted α for those materials are listed in Table III.

It is interesting that they concluded that the dominant mechanism in NdFeB and PrFeB materials is nucleation, while that in SmCo is pinning based on these results.^{1,2,8,9} In fact, these data could be well explained by our proposed self-pinning mechanism. From Table III, it can be seen that for Nd₂Fe₁₄B/ α -Fe and Pr₂Fe₁₄B/ α -Fe systems all fitted values for α are around 0.1, which is consistent with our calculation (see Table II). As the volume occupation of the soft-phase increases, the largest soft grain size in the material also increases which leads to the drop of the coercivity.

As for the SmCo system, the fitted values of α are 0.07, 0.43, and 0.37, much different from that of the Sm₂Co₁₇/Co system shown in Table II. This discrepancy might be explained by the effect of the microstructures at different an-

TABLE III. Fitted α for various materials by Kronmüller and co-workers.

NdFeB		PrFeB		SmCo	
Composition	Fitted α	Composition	Fitted α	Annealing temperature (°C)	Fitted α
Stoichiometric	0.15	Stoichiometric	0.16	700	0.07
7.5% α -Fe	0.13	6.1% α -Fe	0.14	800	0.43
14.2% α -Fe	0.125	14% α -Fe	0.12	900	0.37
21.9% α -Fe	0.11	22.1% α -Fe	0.105		
30.0% α -Fe	0.095	30.4% α -Fe	0.09		
34.1% α -Fe	0.08	38.6% α -Fe	0.065		

nealing temperatures, especially the grain sizes. In fact, the value of α given by Table II Eq. (3) is just a guide value, which should be adjusted by the microstructures. It can be seen from Fig. 1 that the pinning field increases as the soft layer thickness (here it is the grain size of the soft phase) decreases. On the other hand, the gradual change of the parameters at the interface between the soft and hard phases will lead to smaller pinning field. (Actually, the interface quality could be partly attributed to the grain size effect. As the coercivity is given by the smallest pinning field, the larger the grain size, the larger the interface area and the more chance that some parts of the interface have poor quality which leads to the small coercivity. For a multilayer system, the interface area is infinite and the chance of poor interface quality is very high.)

It can be seen from Table III that the optimum annealing temperature is 800 °C corresponding to the largest α , which is about twice that given by Table II, and suggests that the materials are probably nearly stoichiometric with very small inclusion of Co. In contrast, the annealing temperature of 700 °C leads to very small α , which is about 34% of that given by Table II. The discrepancy here is close to that observed in Fig. 1, demonstrating that there are very large soft grains here. These grains function similarly to the role of a soft film in a multilayer system and decrease the coercivity significantly

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