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Posted Date: October 27th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-11194429/v1>

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Additional Declarations: No competing interests reported.

The effect of soft phase on magnetic properties and magnetization reversal process of Nd-Fe-B based multilayers

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Abstract: The soft phase significantly influenced the magnetic properties of Nd-Fe-B based nanocomposite multilayers, the effect structure and type of soft phase on magnetic properties and magnetic reversal process of multilayers were systematically investigated based on micromagnetic method. NdFeB is the hard phase and α -Fe is the soft phases to form soft/hard bilayer, hard/soft/hard and soft/hard/soft trilayers, the calculation results indicated that the B_r increases monotonically while the H_c decreases with increase of soft layer thickness L_s . However, the $(BH)_{\max}$ of three multilayers all show an initial increase followed by a decrease with increase of L_s , the soft/hard/soft trilayers has the highest $(BH)_{\max}$ value of 84.54 MGOe at $L_s = 4$ nm compared to other multilayers. The increasing trend of $(BH)_{\max}$ with soft phase structure mainly attribute to the increase in B_r . The magnetic moment distributions in three multilayers demonstrate the soft/hard layer interfaces differ leading to the distinct forms and magnitudes of magnetic moments. The $(BH)_{\max}$ of Nd-Fe-B based trilayers with Fe_{16}N_2 , $\text{Fe}_{65}\text{Co}_{35}$, and Fe_3B as soft phase reaching maximum values at $L_s = 4$ and 3 nm, respectively; with $(BH)_{\max} = 109.38, 92.01, \text{ and } 65.40$ MGOe, primarily attributable to the high M_s and B_r of soft phase. The magnetic moment distributions in the NdFeB/ Fe_{16}N_2 trilayers display a progress of three-step magnetic reversal, which the coercivity mechanism is pinning. The results demonstrate that the soft/hard/soft trilayers has high B_r and $(BH)_{\max}$, while the NdFeB/ Fe_{16}N_2 trilayers is candidate for high magnetic properties materials.

Keywords: Micromagnetic simulations; Soft/hard phase exchange coupling; Coercivity; Maximum energy product

Introduction

In 1984, Sagawa and Croat fabricated the $\text{Nd}_2\text{Fe}_{14}\text{B}$ permanent magnet with high saturation magnetization (1.61 T), high magnetic crystal anisotropy field (70 kOe), and high maximum energy product $(BH)_{\max} = 64$ MGOe, this permanent magnet has

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the highest magnetic performance available today and is renowned as the “Magnetic King”¹⁻⁵. To meet the demands of modern scientific and technological advancements, researchers have consistently pursued higher energy product. The experiment prepared Nd₂Fe₁₄B permanent magnet exhibits a $(BH)_{\max}$ of 59.5 MGOe which achieving 93% of the theoretical value, representing the highest energy product⁶. Once the energy product of Nd₂Fe₁₄B permanent magnet is raised to a certain level, it hits a bottleneck, leaving little room for further improvement. A major leap in energy product will requirt the emergence of a new material. In 1989, Coehoorn et al. first reported that the remanence enhancement effect in Nd₂Fe₁₄B-based double nanocrystalline composite materials which improves the coercivity and energy product of permanent magnets⁷. Skomski and Coey pointed out that the theoretical $(BH)_{\max}$ value for the Sm₂Fe₁₇N₃/Fe₆₅Co₃₅ multilayers can reach 1 MJ/m³ (125 MGOe)⁸. Therefore, soft/hard exchange-coupled composite permanent magnets are poised to become the fourth-generation rare-earth permanent magnet and thus have emerged as a focal point of research⁹⁻¹⁶. However, the experimental measured $(BH)_{\max}$ value is significantly lower than the theoretical value; this is primarily attributed to the inverse relationship between remanent and coercivity. The high saturation magnetization of soft phase enhances the remanent, while the low magnetic crystalline anisotropy field reduces the coercivity. The soft phase has a significant impact on the magnetic performance and energy product of soft/hard exchange-coupled composite magnets.

For soft/hard exchange-coupled nanocrystalline magnets, both theoretical and experimental studies have shown that when the grain size of soft phase is approximately twice the width of the Bloch domain wall in the hard phase¹⁷⁻¹⁹, the composite magnet exhibits excellent exchange coupling interaction; the remanence enhancement effect increases the energy product. When Nd₂Fe₁₄B is used as the hard phase ($\delta_B = \pi\sqrt{A/K} = 4.2$ nm), the optimal grain size for the soft phase ranges from 5 to 10 nm. However, it is experimentally challenging to prepare the soft phases with grain sizes below 10 nm and regular, uniform grain shapes, as well as hard phases that exhibit an ideal, uniformly oriented grain arrangement in nanocrystalline composite magnets. Thanks to advances in thin-film fabrication technologies, soft/hard exchange-coupled composite multilayers allow for precise control over the composition, microstructure, and film thickness of the soft and hard phases; by

adjusting experimental parameters to align the easy-axis orientations of the two phases in a consistent manner, this represents an effective approach to achieving the high energy product²⁰⁻²⁷. The $(BH)_{\max}$ value of the $\text{Nd}_2\text{Fe}_{14}\text{B}/\text{Fe}_{67}\text{Co}_{33}$ nanocomposite multilayers synthesized by Cui et al. reached 61 MGOe²⁸. Via the micromagnetic simulation, the $(BH)_{\max}$ value for $\text{Nd}_2\text{Fe}_{14}\text{B}/\alpha\text{-Fe}$ multilayers with soft layer thickness of 10 nm is 80 MGOe²⁹, for the $\text{Nd}_2\text{Fe}_{14}\text{B}/\text{Fe}_{65}\text{Co}_{35}$ bilayer film with soft layer thickness of 1 nm, $(BH)_{\max} = 561.61 \text{ kJ/m}^3$ (70.6 MGOe)³⁰; and for the SmCo/Fe bilayer film with soft layer thickness of 2 nm, $(BH)_{\max} = 40.2 \text{ MGOe}$ ³¹. No fixed soft layer thickness has been identified in soft/hard exchange-coupled multilayers that can achieve the optimal $(BH)_{\max}$ value, the $(BH)_{\max}$ is influenced by various factors including the hard and soft phase, film structure, and simulated parameters.

This study employs a micromagnetism simulation method, using NdFeB as the hard phase, and focuses on soft/hard exchange-coupled bilayer and trilayer films to investigate the influence of the soft phase structure on remanence, coercivity, energy product, and magnetization reversal, as well as to determine the conditions under which the optimal $(BH)_{\max}$ value can be achieved. By transforming the soft phases into $\alpha\text{-Fe}$, Fe_{16}N_2 , $\text{Fe}_{65}\text{Co}_{35}$, and Fe_3B , the influence of soft phase type on energy product of the multilayers was investigated. The results presented in this paper can provide theoretical support for the design and fabrication of thin films with high magnetic properties.

Calculation model and method

The Mumax3 micromagnetism simulation software was employed to calculate soft/hard exchange-coupled multilayers; the two- and three-dimensional model are shown in Fig. 1. The yellow color represents the soft layer; the blue color represents the hard layer, type a represents the typical soft/hard bilayer; type b (hard/soft/hard trilayers) is formed by adding a soft phase to type a; similarly, type c (soft/hard/soft trilayers) is formed by adding a soft layer to type a. Set the hard layer thickness to $L_h = 10\text{nm}$ and the soft layer thickness to $L_s = 1 \sim 10 \text{ nm}$; the subscripts h and s denote the hard and soft layer, respectively. NdFeB represents the hard phase, while $\alpha\text{-Fe}$, Fe_{16}N_2 , $\text{Fe}_{65}\text{Co}_{35}$ and Fe_3B represent the soft phases; the magnetic parameters used in the simulation are listed in Table 1, where M_s , K , A , and M_s^s / M_s^h are the saturation magnetization, magneto-crystal anisotropy constant, exchange coupling constant, and

saturation magnetization ratio of soft and hard phase.

The three-dimensional scheme of soft/hard exchange-coupled multilayers is shown in Fig. 1(d), the vertex at the bottom surface is taken as the origin O , and an $o-xyz$ coordinate system is established. The film surface lies within the x - y plane, with both its length and width measuring 500 nm. The easy axes of both phases denoted by e and the applied field H are assumed to be in the $+y$ direction. Nd-Fe-B Bloch wall width $\delta_B = 4.2$ nm, the grid size is set to $4 \text{ nm} \times 4 \text{ nm} \times 1 \text{ nm}$ which is close the δ_B .

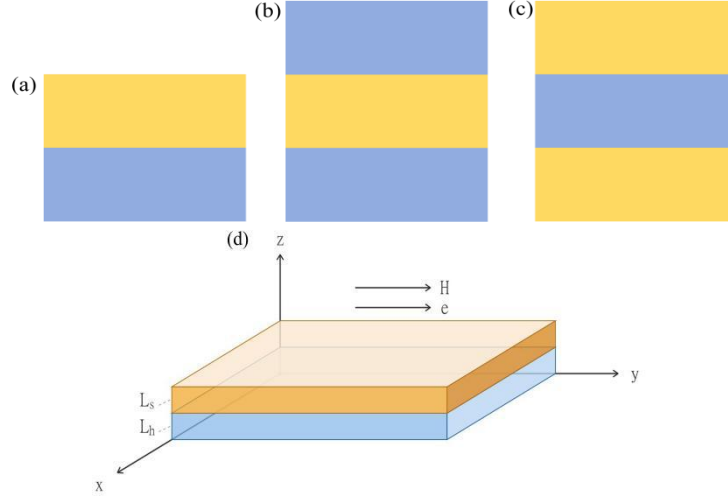


Fig. 1 Two- and three-dimensional scheme for three soft/hard exchange-coupled multilayers, (a) type a: soft/hard bilayer, (b) type b: hard/soft/hard trilayers, (c) type c: soft/hard/soft trilayers, and (d) basic scheme in this work.

Table 1 Simulated magnetic parameters of the hard and soft phase

Magnetic phase	$M_s / (10^6 \text{ A/m})$	$K / (10^6 \text{ J/m}^3)$	$A / (10^{-12} \text{ J/m})$	M_s^s / M_s^h
Nd ₂ Fe ₁₄ B	1.28	4.3	7.7	
α -Fe	1.71	0.046	25	1.34
Fe ₁₆ N ₂	2.235	0.25	32.5	1.75
Fe ₆₅ Co ₃₅	1.934	0.01	16.7	1.51
Fe ₃ B	1.30	0.20	8.11	1.02

Results and discussions

The influence of structure of soft phase

α -Fe is the soft phases, which forms NdFeB/ α -Fe nanocomposite bilayers and trilayers, the influence of the structure of soft phase was investigated. Fig. 2 compares the demagnetization curves of types a, b, and c, the shapes of demagnetization curves for the three multilayers are similar; as soft layer thickness L_s increases, the shape of

curve gradually change from rectangular to elliptical. The rectangular curve indicates that the soft layer is strongly coupled to the hard layer, and the multilayers exhibits a single hard phase externally. When L_s increases to a certain value, the demagnetization curve begins to exhibit a knee-shaped turn, indicating that the exchange coupling between the soft and hard phase starts to weaken, and the magnetic reversal processes of the two phases become separated. Curves of types a, b, and c all have a good remanence ratio, $m_r \approx 1$.

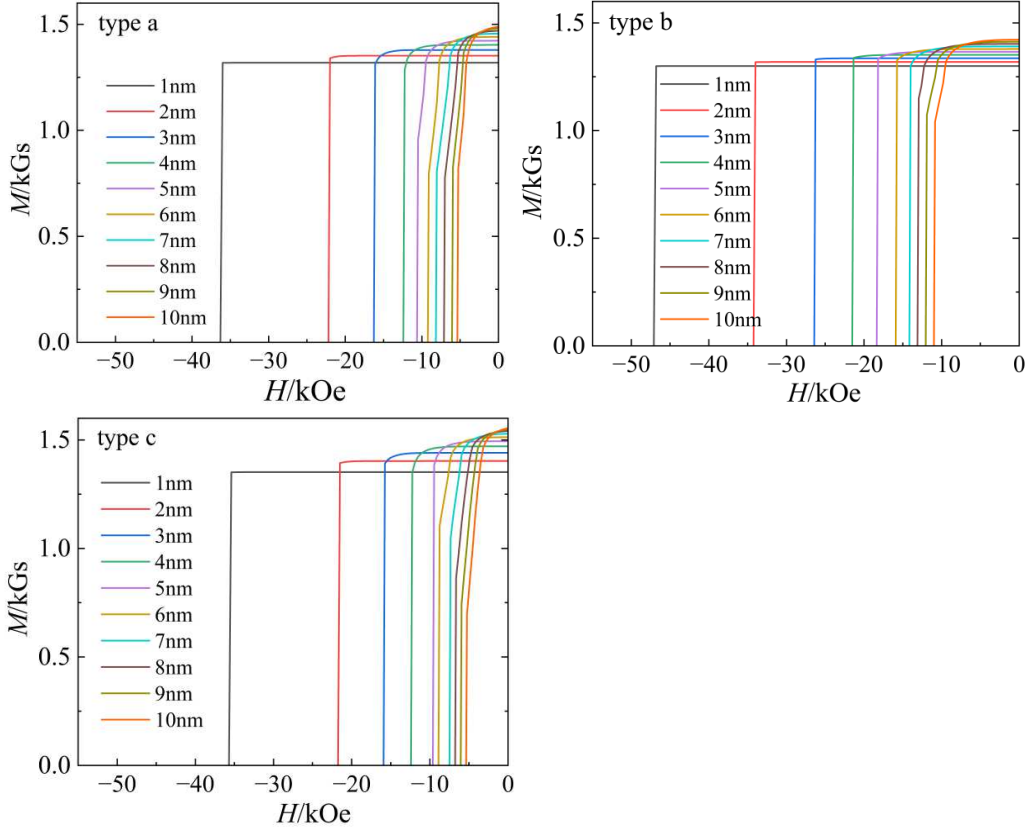


Fig. 2 Demagnetization curves for types a, b, and c of the NdFeB/ α -Fe nanocomposite multilayers

Based on the demagnetization curves, the variation trends of the soft phase volume fraction f_s , remanence B_r , coercivity H_c , and maximum energy product $(BH)_{\max}$ for types a, b, and c with L_s can be obtained, as shown in Fig. 3. The f_s and B_r of the three multilayers both monotonically increase with the increase of L_s , and the variation trends of f_s and B_r are consistent. Type b has two hard layers and one soft layer, resulting in the lowest f_s value; whereas type c has two soft layers and one hard layer, resulting in the highest f_s value. Since the M_s of soft phase α -Fe is higher than that of hard phase NdFeB, type b has the lowest B_r , whereas type c has the highest B_r . When L_s increases from 1 nm to 10 nm, the B_r of types a, b, and c increase from 1.65,

1.63, and 1.70 T to 1.87, 1.79, and 1.95 T, respectively; with increases of 13.3%, 9.8%, and 14.7%. The increase in B_r will be conducive to improving the energy product.

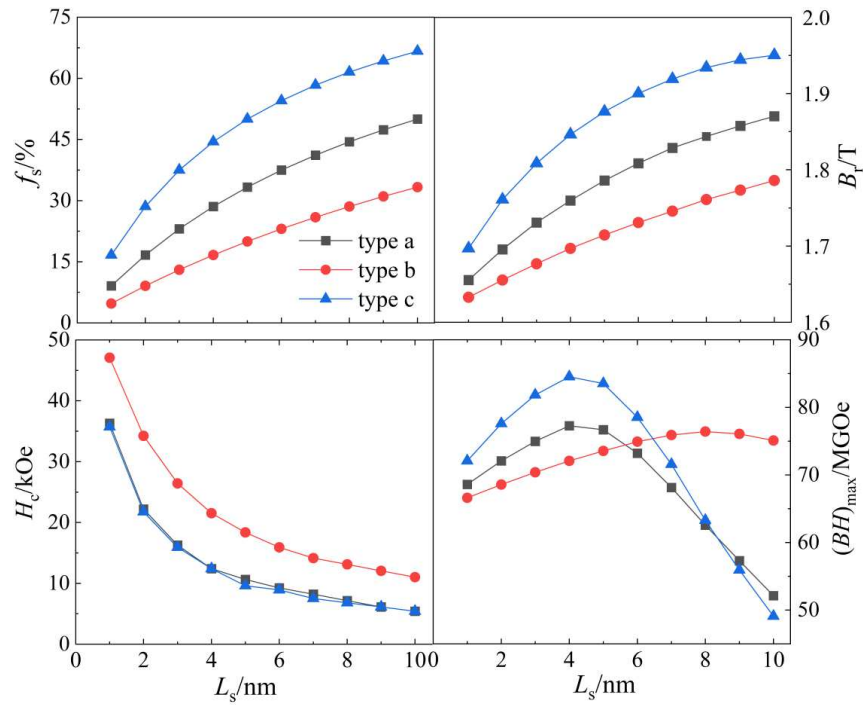


Fig. 3 The variation trends of f_s , B_r , H_c and $(BH)_{\max}$ for types a, b, and c of the NdFeB/ α -Fe multilayers as a function of L_s

Table 2 The $(BH)_{\max}$ values for types a, b, and c of the NdFeB/ α -Fe multilayers for various L_s

soft layer thickness L_s/nm	type a	type b	type c
	$(BH)_{\max}/\text{MGOe}$		
1	68.61	66.61	72.09
2	72.09	68.58	77.60
3	74.98	70.40	81.85
4	77.28	72.08	84.54
5	76.70	73.55	83.52
6	73.22	74.92	78.52
7	68.14	75.91	71.57
8	62.58	76.42	63.28
9	57.31	76.08	55.92
10	52.14	75.11	49.10

As shown in Fig. 3, the coercivity of types a, b, and c all decrease with the increase of L_s . At all soft layer thicknesses, type b has the highest coercivity, while

types a and c have similar coercivity. The H_c of types a, b, and c decreases from 36.30, 47.10, and 35.70 kOe at $L_s = 1$ nm to 5.42, 11.02, and 5.42 kOe at $L_s = 10$ nm, representing reductions of 85.2%, 76.6%, and 84.8%, respectively. The lower magneto-crystalline anisotropy field of soft phase dilutes the magneto-crystalline anisotropy field of hard phase, leading to the decreasing trend of H_c . Type b has one soft layer and the lowest f_s , thus having the minimum impact of the soft phase on the reduction of coercivity, resulting in the highest H_c value. Exchange coupling occurs at the interface between the soft and hard phases, this exchange coupling causes the magnetic moments on both sides of interface to deviate from their respective easy axes, allowing the magnetic moments of both phases to align with the applied field H , which weakens the ability of the multilayers magnetic moments to resist reversal by the H , leading to a significant reduction in coercivity. Compared to type a, type c has two soft layers, but its coercivity does not decrease significantly, which plays a positive role in improving the energy product.

As shown in Fig. 3, the $(BH)_{\max}$ of types a, b, and c all show an initial increase followed by a decrease with the increase of L_s , reaching maximum values of 77.28, 76.42, and 84.54 MGOe at $L_s = 4$ nm, 8 nm, and 4 nm, respectively; the specific values are listed in Table 2. The soft layer thickness at which types a and c exhibit the maximum $(BH)_{\max}$ value is approximately equal to the Bloch wall width of NdFeB ($\delta_B = 4.2$ nm), while the soft layer thickness at which type b exhibits the maximum $(BH)_{\max}$ value is approximately twice that. The increased remanence and decreased coercivity jointly affect the energy product. When remanence and coercivity reach an equilibrium at a certain soft layer thickness, the NdFeB/ α -Fe multilayers achieves the optimal $(BH)_{\max}$. This soft layer thickness is not a fixed value but is influenced by the soft phase structure. The increasing trend of $(BH)_{\max}$ with L_s mainly stems from the increase in remanence, while the decreasing trend of $(BH)_{\max}$ is primarily attributed to the significant reduction in coercivity. When $L_s \leq 6$ nm, the $(BH)_{\max}$ value of type c is higher than that of types a and b, which is mainly due to the higher remanence. Literature 29 reported the $(BH)_{\max}$ of 80 MGOe for Nd₂Fe₁₄B/ α -Fe magnetic multilayers at $L_s = 10$ nm, which is lower than the $(BH)_{\max}$ value of type c in this study (84.54 MGOe). The difference in simulated values may arise from differences in calculation methods, magnetic film thickness, and simulation parameters. Therefore, the type c trilayers can be used to prepare high-performance magnetic

films.

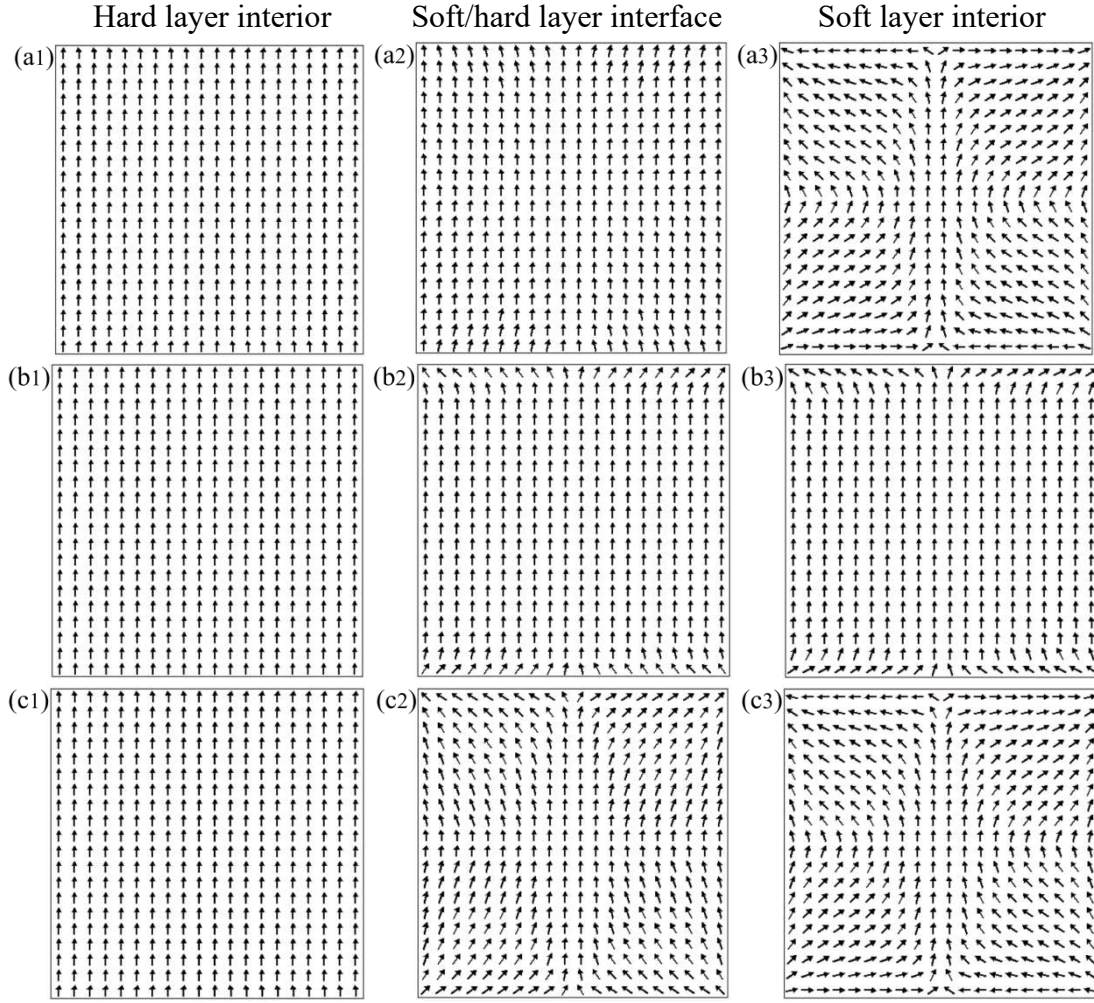


Fig. 4 Magnetic moment distributions before magnetization reversal for types a, b, and c of the NdFeB/ α -Fe multilayers at $L_s = 4$ nm, (a1–a3): type a, $H = -12.25$ kOe; (b1–b3): type b, $H = -21.00$ kOe; (c1–c3): type c, $H = -11.90$ kOe

The demagnetization curve characterizes the macroscopic magnetic properties of magnet, while the magnetic moment distributions clearly reflects the microscopic features during the magnetization reversal process. Fig. 4 shows the magnetic moment distributions of types a, b, and c at various applied field H before magnetization reversal at $L_s = 4$ nm. Due to the high magneto-crystalline anisotropy field of NdFeB, its magnetic moments are difficult to deflect, whereas α -Fe has three easy axes and its magnetic moments are easily deflected. Magnetization reversal nucleation occurs in the soft layer, and then drives the deflection of the magnetic moments in hard layer. As shown in the Fig. 4 (a1) – (a3), when $H = -12.25$ kOe, most of the magnetic moments inside the soft layer of type a have already deflected, exhibiting a left-right

symmetric "C"-shaped distribution; meanwhile, the magnetic moments inside the hard layer still maintain their initial magnetization state, i.e., along the $+y$ axis. Driven by the magnetic moments in the soft layer, the magnetic moments at the four corners and sides of the interface between the soft and hard layers begin to deflect.

In type b, the two hard layers are strongly coupled to the soft layer, and exchange coupling occurs at two interfaces. As shown in Fig. 4 (b1) – (b3), the high anisotropy of the hard phase fixes the magnetic moments of soft phase along the easy axis, until $H = -21.00$ kOe the magnetic moments at the four corners and sides of the film surface inside the soft layer deflect, driving the deflection of the magnetic moments at the interface, while the magnetic moments inside the hard layer maintain their initial magnetization state. As shown in Fig. 4 (c1) – (c3), in type c the two soft layers are coupled to one hard layer at the two interfaces, most of the magnetic moments inside the soft layer can deflect at a relatively small applied field of $H = -11.90$ kOe, driving most of the magnetic moments at the interface to deflect, with a higher degree of deflection than type a. Therefore, the soft phase structure significantly affects the magnetization reversal process of NdFeB/ α -Fe multilayers.

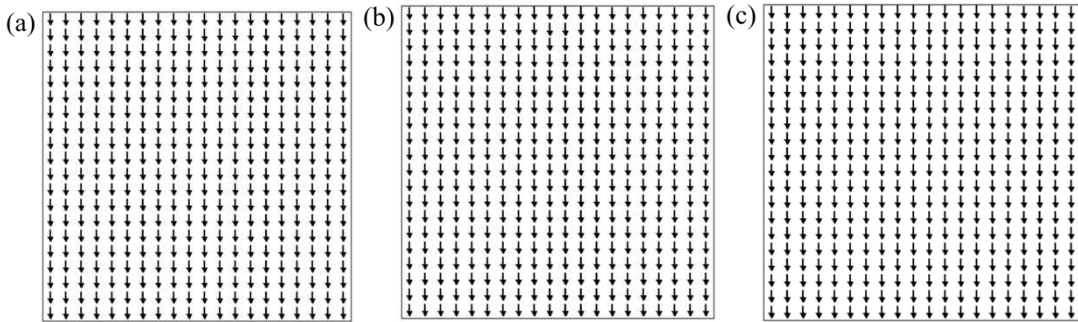


Fig. 5 Magnetic moment distributions after magnetization reversal for types a, b, and c of the NdFeB/ α -Fe multilayers at $L_s = 4$ nm, (a) type a: $H = -12.60$ kOe; (b) type b:

$H = -21.35$ kOe; (c) type c: $H = -12.25$ kOe

When the applied field before magnetization reversal of types a, b, and c is slightly reduced, the magnetic moments can complete their deflection. Fig. 5 shows the magnetic moment distributions of the three multilayers after magnetization reversal. When $H = -12.60$, -21.35 , and -12.25 kOe, all magnetic moments of types a, b, and c deflect 180° from their initial saturated magnetization state (along the $+y$ axis) to the $-y$ axis direction. The negative values of the average applied fields before and after magnetization reversal for types a, b, and c are $H_a = 12.43$ kOe, $H_b = 21.17$

kOe, and $H_c = 12.08$ kOe, respectively. These average values are close to the coercivity obtained from the demagnetization curves: $H_{ca} = 12.42$ kOe, $H_{cb} = 21.52$ kOe, and $H_{cc} = 12.42$ kOe. Therefore, the NdFeB/ α -Fe multilayers undergo magnetization reversal near the coercivity.

The influence of type of soft phase

To study the influence of type of soft phase on magnetic properties of Nd-Fe-B based multilayers, Fe_{16}N_2 , $\text{Fe}_{65}\text{Co}_{35}$, and Fe_3B were taken as the soft phase to form type c trilayers, and compared with type c where α -Fe was the soft phase. Fig. 6 shows the hysteresis loops of Nd-Fe-B based trilayers with Fe_{16}N_2 and Fe_3B as the soft phase. As shown in Fig. 6, with the increase of L_s , the hysteresis loop of the NdFeB/ Fe_{16}N_2 trilayers gradually changes from rectangular to elliptical, similar to the types of a, b, and c NdFeB/ α -Fe multilayers as shown in Fig. 2, and also similar to the NdFeB/ $\text{Fe}_{65}\text{Co}_{35}$ trilayers. When $L_s = 1 \sim 2$ nm, the hysteresis loop of NdFeB/ Fe_3B trilayers is rectangular. Starting from $L_s = 3$ nm, a knee -shaped turn appears in the hysteresis loop. When $L_s = 6 \sim 10$ nm, the hysteresis loop shows a significant waist, exhibiting typical soft magnetic behavior, with the soft and hard phases undergo decoupling. The high anisotropy of hard phase can no longer fix the magnetic moments of soft phase, leading to the deflection of the soft phase magnetic moments at low fields, resulting in the separation of the magnetization reversal of the two phases.

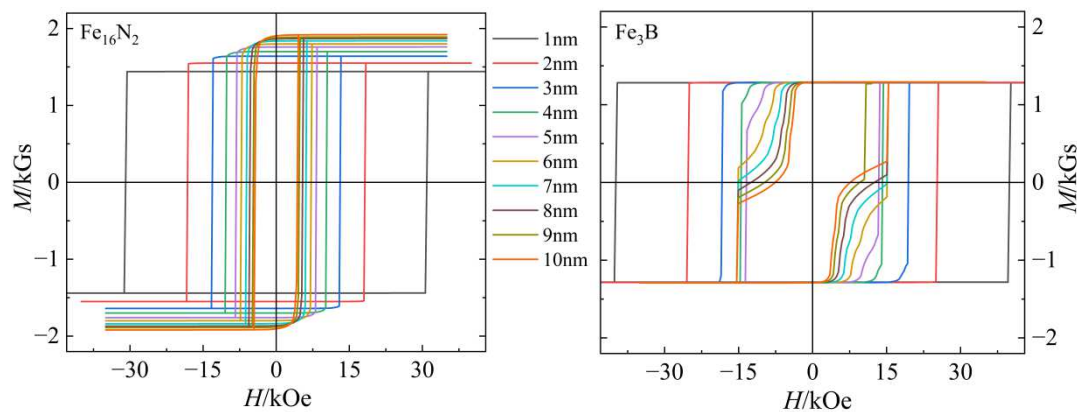


Fig. 6 Hysteresis loops of Nd-Fe-B based trilayers with Fe_{16}N_2 and Fe_3B as soft phases

The trends of B_r , H_c , and $(BH)_{\max}$ of Nd-Fe-B based trilayers with soft phases of Fe_{16}N_2 , $\text{Fe}_{65}\text{Co}_{35}$, α -Fe, and Fe_3B as a function of L_s are obtained from the hysteresis

loops, as shown in Fig. 7. The M_s value of NdFeB/Fe₃B trilayers is close to that of NdFeB, $M_s^s/M_s^h = 1.02$, the increase in B_r of Fe₃B with L_s is relatively small, increasing from 1.61 T at 1 nm to 1.62 T at 10 nm, an increase of only 0.62%. The B_r values of the trilayers with Fe₁₆N₂, Fe₆₅Co₃₅, and α -Fe as soft phases all increase monotonically with L_s , and the increasing trends are consistent with the values of M_s^s/M_s^h . The M_s and $M_s^s/M_s^h (= 1.75)$, as shown in Table 1) values of Fe₁₆N₂ are the highest, so the NdFeB/Fe₁₆N₂ trilayers has the highest B_r , increasing from 1.81 T at 1 nm to 2.39 T at 10 nm, an increase of 32.04%.

The H_c of the trilayers with Fe₁₆N₂, Fe₆₅Co₃₅, and α -Fe as soft phases all decrease monotonically with L_s . At all soft layer thicknesses, the H_c value of the NdFeB/ α -Fe trilayers is slightly higher than those of the NdFeB/Fe₁₆N₂ and NdFeB/Fe₆₅Co₃₅ trilayers, with the H_c values of the latter two being very close. The NdFeB/Fe₃B trilayers has the highest H_c , which first decreases and then increases with L_s , reaching a peak at $L_s = 6$ nm ($H_c = 15.11$ kOe) before decreasing again. The intermediate increase in H_c is mainly due to the decoupling effect between the soft and hard phases at high soft layer thickness.

As shown in Fig. 7, the $(BH)_{\max}$ of Nd-Fe-B based trilayers with Fe₁₆N₂, Fe₆₅Co₃₅, and Fe₃B as soft phases all show an initial increase followed by a decrease with L_s , reaching maximum values at $L_s = 4$ nm and 3 nm, respectively; with $(BH)_{\max} = 109.38, 92.01, \text{ and } 65.40$ MGOe. The specific values are listed in Table 3. The NdFeB/Fe₁₆N₂ trilayers has the highest $(BH)_{\max}$ value, which is mainly attributed to its largest B_r ; the increase from $L_s = 1$ nm to 4 nm is 33.91%. The $(BH)_{\max}$ value of NdFeB/Fe₆₅Co₃₅ trilayers increases by 21.02% from $L_s = 1$ nm to 4 nm. The $(BH)_{\max}$ value of NdFeB/Fe₃B trilayers shows the smallest increase, only 0.83% from $L_s = 1$ nm to 3 nm, which is mainly due to the small increase in B_r . The $(BH)_{\max}$ of the soft/hard/soft c-type NdFeB/Fe₁₆N₂ trilayers for $L_s < 8$ nm is superior to the $(BH)_{\max}$ of the Nd₂Fe₁₄B/ α "-Fe₁₆N₂ bilayer in literature 32 (81.7 MGOe). The $(BH)_{\max}$ of the NdFeB/Fe₆₅Co₃₅ trilayers for $L_s < 6$ nm is superior to the $(BH)_{\max}$ of the Nd₂Fe₁₄B/Fe₆₅Co₃₅ bilayer in literature 30 (70.6 MGOe), and also superior to the $(BH)_{\max} = 630 \text{ kJ/m}^3$ (79.14 MGOe) of the Nd₂Fe₁₄B/Fe₆₅Co₃₅/Nd₂Fe₁₄B trilayers reported in literature 33.

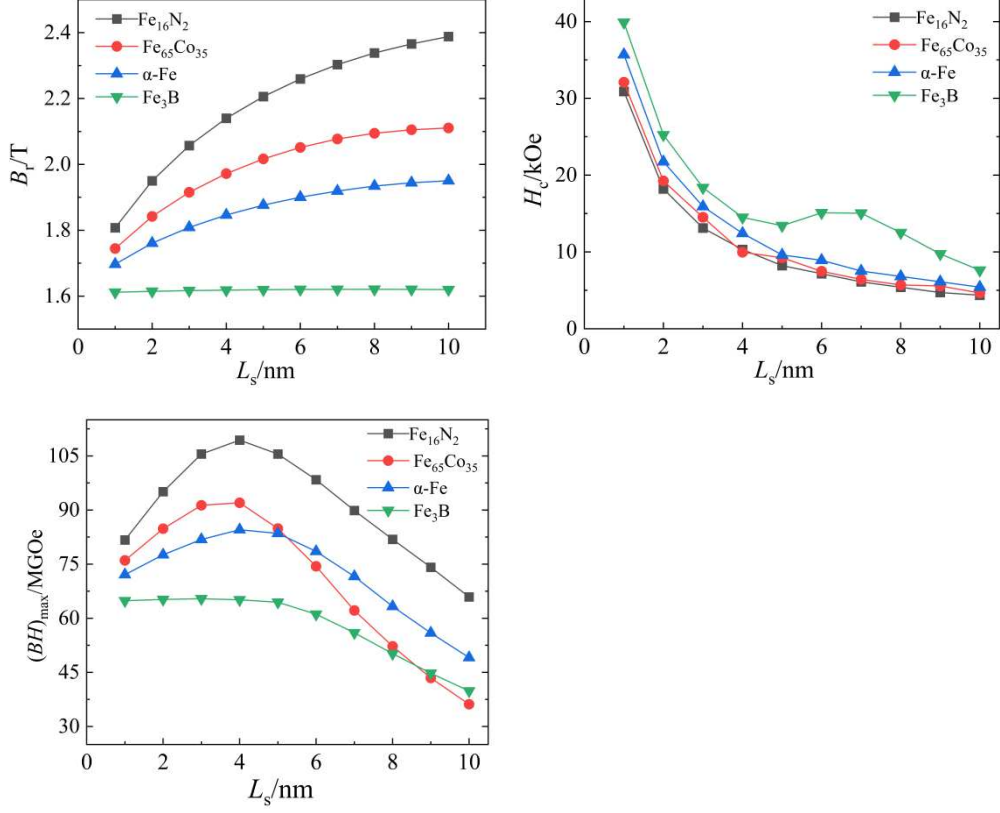


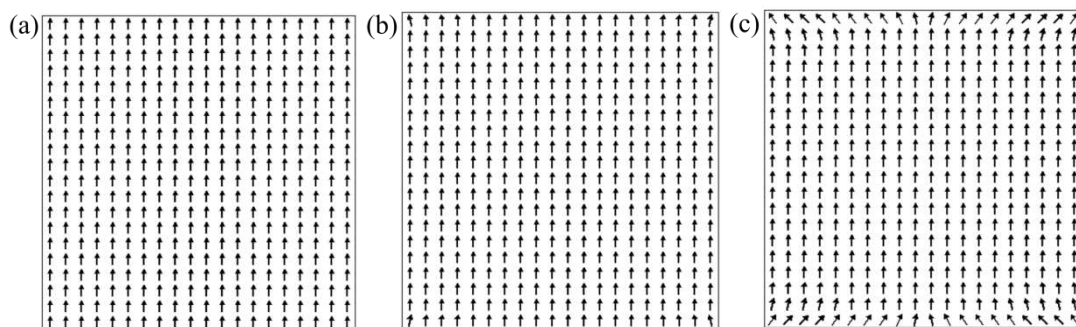
Fig. 7 The variation trend of magnetic properties of Nd-Fe-B based trilayers with Fe_{16}N_2 , $\text{Fe}_{65}\text{Co}_{35}$, $\alpha\text{-Fe}$, and Fe_3B as soft phases as a function of L_s

Table 3 The $(BH)_{\text{max}}$ values of Nd-Fe-B based trilayers with Fe_{16}N_2 , $\text{Fe}_{65}\text{Co}_{35}$, and Fe_3B as soft phase

soft layer thickness L_s/nm	Fe_{16}N_2	$\text{Fe}_{65}\text{Co}_{35}$	Fe_3B
	$(BH)_{\text{max}}/\text{MGOe}$		
1	81.68	76.03	64.86
2	95.09	84.83	65.22
3	105.54	91.31	65.40
4	109.38	92.01	65.12
5	105.54	84.88	64.45
6	98.38	74.40	61.11
7	89.87	62.15	55.97
8	81.88	52.23	50.19
9	74.11	43.43	44.75
10	65.90	36.14	39.85

To analyze the coercivity mechanism of NdFeB/ Fe_{16}N_2 trilayers, Fig. 8 shows the magnetic moment distributions at the interface between soft and hard layer at $L_s =$

4 nm. When the applied field ($H = 30.00$ kOe) is in the positive saturation state, the magnetic moment direction is consistent with the applied field direction, i.e., along the $+y$ axis. When the applied field decreases to -2.45 kOe as shown in Fig. 8 (b), the magnetic moments at the four corners of film surface begin to deflect by a small angle, while the magnetic moments over a large area still maintain their initial magnetization state. At this point, the opposite of the applied field is called nucleation field, $H_N = 2.45$ kOe. As the applied field continues to decrease, the magnetic moments at the interface deflect and diffuse towards the edges. When $H = -8.05$ kOe as shown in Fig. 8 (c), the magnetic moments at the edges of the film surface deflect in-plane by about 45° . When applied field decreases to near the coercivity ($H = -9.80$ kOe, as shown in Fig. 8 (d)), the magnetic moments at the edges of the film surface have completed a 90° deflection and drag the surrounding magnetic moments to deflect, putting the trilayers in a critical state. As shown in Fig. 8 (e), a slight decrease in the applied field to -10.15 kOe leads to the irreversible movement of the domain wall, and the magnetic moments complete their reversal. At this point, the opposite of applied field is called pinning field, $H_P = 10.15$ kOe. When the applied field decreases to the reverse saturation field ($H = -30.00$ kOe), all magnetic moments complete the 180° reversal, i.e., along the $-y$ axis, and reach saturation in the reverse direction. The H_c ($= 10.32$ kOe) of the trilayers is greater than the H_N while approximately equal to the H_P , indicating that the coercivity mechanism is pinning. The magnetization reversal process of NdFeB/Fe₁₆N₂ trilayers undergoes demagnetization nucleation, reverse domain expansion, and irreversible reversal of the magnetization vector, which is consistent with the pinning mechanism proposed by Zhao et al³⁴.



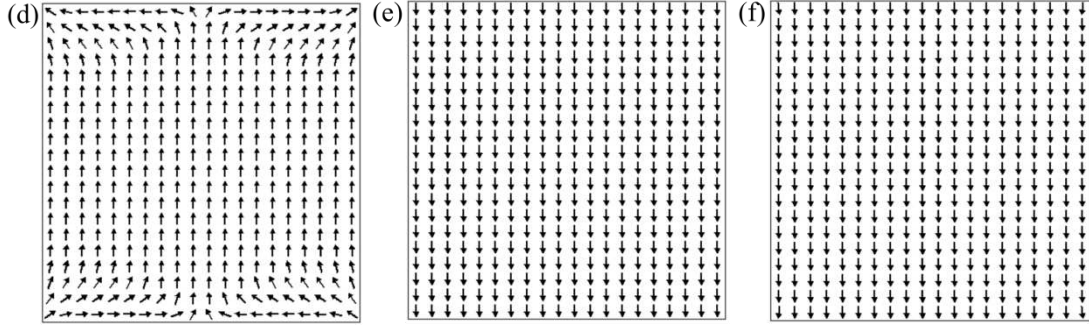


Fig. 8 Magnetic moment distributions of NdFeB/Fe₁₆N₂ trilayers under various applied fields at $L_s = 4$ nm, (a) $H = 30.00$ kOe; (b) $H = -2.45$ kOe; (c) $H = -8.05$ kOe; (d) $H = -9.80$ kOe; (e) $H = -10.15$ kOe and (f) $H = -30.00$ kOe

To study the magnetic reversal process and magnetic moment distributions of multilayers using the micromagnetic method, it is necessary to calculate the total Gibbs free energy of the trilayers and then solve for the minimum free energy. Generally, neglecting the elastic energy of the trilayers, the Gibbs free energy formula is defined as follows:³⁵

$$E_{tot} = E_{ex} + E_{an} + E_{dem} + E_{zeem} \quad (1)$$

where E_{tot} , E_{ex} , E_{an} , E_{dem} , and E_{zeem} are the total, exchange, magneto-crystalline anisotropy, demagnetization, and Zeeman energies. The most of Nd-Fe-B based trilayers with different soft phases tend to achieve the optimal $(BH)_{max}$ value at $L_s = 4$ nm, Fig. 9 consequently shows the variation trends of the energies terms for the four soft phase trilayers at this thickness. As shown in the Fig. 9, the magnetic moment is aligned with the applied field direction when $H = 30.00$ kOe, the angle $\theta = 0^\circ$, and the magnetic moment reaches an equilibrium state under the action of E_{ex} , E_{an} , E_{dem} , and E_{zeem} , which point the energy is at the minimum. When applied field decreases to near the negative value of coercivity, the energy terms exhibit positive or negative peaks. The applied fields corresponding to the soft phases of α -Fe, Fe₁₆N₂, Fe₆₅Co₃₅, and Fe₃B are $H = -12.25$, -10.15 , -10.85 , and -14.35 kOe, respectively; indicating that the energy terms undergo abrupt changes when the magnetic moment reverses. Under this applied fields, E_{ex} , E_{an} , and E_{zeem} reach their maximum values, while E_{dem} reaches its minimum value, and the total energy E_{tot} reaches its maximum value. When the total energy of trilayers reaches its maximum, the magnetic moment distributions becomes unstable, an irreversible reversal occurs, and the magnetic moment reaches another equilibrium state, $\theta = 180^\circ$. As the applied field continues to

decrease, E_{ex} , E_{an} , and E_{zeem} decrease, while E_{dem} increases; finally, at $H = -30.00$ kOe, the E_{tot} reaches a new minimum state.

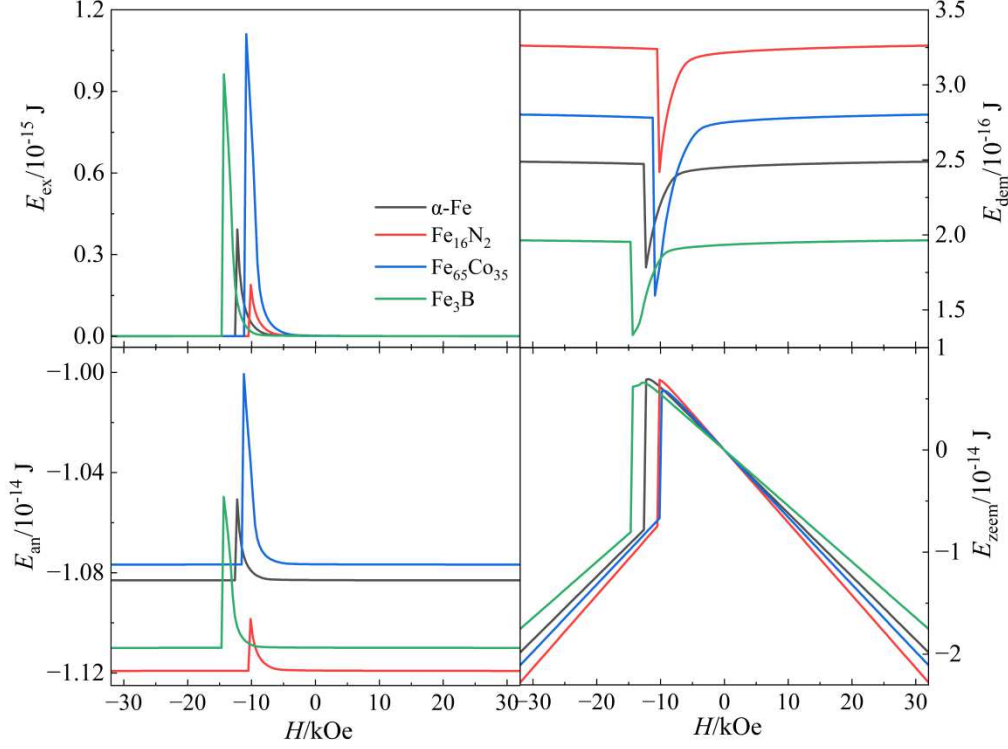


Fig. 9 The variation trend of energies for Nd-Fe-B based nanocomposite trilayers with α -Fe, Fe_{16}N_2 , $\text{Fe}_{65}\text{Co}_{35}$, and Fe_3B as soft phase at $L_s = 4$ nm

As shown in Fig. 9, the E_{zeem} of four trilayers remain around 0.655×10^{-14} J, so the changes in E_{ex} , E_{an} , and E_{dem} are the main reasons for the change in E_{tot} . Near the coercivity, the exchange energy and magneto-crystalline anisotropy energy of the NdFeB/ Fe_{16}N_2 trilayers are the lowest values, with $E_{\text{ex}} = 0.189 \times 10^{-15}$ J and $E_{\text{an}} = -1.098 \times 10^{-14}$ J; while the NdFeB/ $\text{Fe}_{65}\text{Co}_{35}$ trilayers has the highest values for these two energies, with $E_{\text{ex}} = 1.11 \times 10^{-15}$ J and $E_{\text{an}} = -1.001 \times 10^{-14}$ J; while the NdFeB/ Fe_3B trilayers has lower values for these two energies than the NdFeB/ $\text{Fe}_{65}\text{Co}_{35}$ trilayers, with $E_{\text{ex}} = 0.963 \times 10^{-15}$ J and $E_{\text{an}} = -1.049 \times 10^{-14}$ J. The exchange energy tends to make the magnetic moment of adjacent atoms align parallel, while the magneto-crystalline anisotropy energy causes the magnetization inside the magnet to align along the easy axis. The minimum E_{ex} of NdFeB/ Fe_{16}N_2 trilayers indicates a strong exchange coupling, making it difficult for the magnetic moment of the soft phase to nucleate reverse magnetization; the low E_{an} value indicates a small coercivity. The NdFeB/ Fe_3B trilayers has higher E_{ex} and E_{an} , showing a opposite trend to that the NdFeB/ Fe_{16}N_2 trilayers. The demagnetization energy is the interaction

energy between the demagnetizing field and magnetization of the trilayers, and the demagnetizing field favors the reverse magnetization. The NdFeB/Fe₁₆N₂ trilayers has the highest demagnetization energy, $E_{\text{dem}} = 2.42 \times 10^{-16}$ J; while the NdFeB/Fe₃B trilayers has the lowest demagnetization energy, $E_{\text{dem}} = 1.33 \times 10^{-16}$ J. The increase in demagnetization energy leads to a decrease in coercivity, so the NdFeB/Fe₃B trilayers has the largest coercivity.

Conclusions

In this paper, the demagnetization curves, magnetic moment distributions, and energy change trends of Nd-Fe-B-based multilayers were calculated using the Mumax3 micromagnetic simulation software. The soft phase structure and type significantly affect the magnetic properties and magnetization reversal of the multilayers. The calculation results show that:

(1) For the multilayers of types a, b, and c, as soft layer thickness L_s increases, the remanence B_r of the three multilayers increases, while the coercivity H_c decreases; meanwhile, the maximum energy product $(BH)_{\text{max}}$ shows a trend of first increasing and then decreasing. When $L_s < 7$ nm, the $(BH)_{\text{max}}$ value of the type c trilayers is higher than those of types a and b, which is mainly attributed to the higher B_r and H_c ; the optimal $(BH)_{\text{max}} = 84.54$ MGOe is obtained at $L_s = 4$ nm.

(2) E_{ex} , E_{an} , E_{dem} , and E_{zeem} undergo abrupt changes near the coercivity, and the magnetic moment distributions of all trilayers changes from one equilibrium state to another. The E_{ex} and E_{an} of NdFeB/Fe₁₆N₂ trilayers are the lowest, while the NdFeB/Fe₃B trilayers has higher E_{ex} and E_{an} , indicating that these two trilayers have opposite magnetic properties.

(3) Among the type c trilayers with α -Fe, Fe₁₆N₂, Fe₆₅Co₃₅, and Fe₃B as soft phase, the $(BH)_{\text{max}}$ of the NdFeB/Fe₁₆N₂ trilayers is higher than that other trilayers in the range of $L_s = 1 \sim 10$ nm, which can provide a reference for the experimental design and preparation of high-performance magnetic thin films.

Data availability

All data supporting the findings of this study are available within this article.

Funding

This research was supported by the Open Fund of the National Experimental

References

1. Sagawa, M., Fujimura, S., Togawa, N., Tamamoto, H. & Matsuura, Y. New material for permanent magnets on a base of Nd and Fe(invited). *J. Appl. Phys.* **55**(6), 2083-2087(1984).
2. Croat, J.J., Herbst, J.F. & Lee R., W.R. Pr-Fe and Nd-Fe-based materials: A new class of high-performance permanent magnets(invited). *J. Appl. Phys.* **55**(6), 2078-2082(1984).
3. Ram, S. Synihesis, magnetic properties and formalism of magnetic properties of high-quality refined Nd₂Fe₁₄B powders for permanent magnet devices. *J. Mater. Sci.* **32**(15), 4133-4148(1997).
4. Wang, X.F., Chen, X.Y. Jiang, Z.L. Chen, Y. & Chen, H.M. Study on the nanostructure and magnetic properties of NdFeNbB permanent magnets. *Mater. Sci. Forum.* **884**(638-642), 1749-1754(2010).
5. Uysal, I.E., Nishino, M. & Miyashita, S. Magnetic field threshold for uncleation and depinning of domain walls in the neodymium permanent magnet Nd₂Fe₁₄B. *Phys. Rev. B* **101**(9), 094421(2020).
6. Matsuura, Y. Recent development of Nd-Fe-B sintered magnets and their applications. *J. Magn. Magn. Mate.* **303**(2), 344-347(2006).
7. Coehoorn, R., De Mooij, D.B. & Waard, C. Meltspun permanent magnet materials containing Fe₃B as the main phase. *J. Magn. Magn. Mate.* **80**(1), 101-104(1989).
8. Skomski, R. & Coey, J.M.D. Giant energy product in nanostructured two-phase magnets[J]. *Phys. Rev. B* **48**(21), 15812-15816(1993).
9. Ping, D.H., Hono, K., Kanekiyo, H. & Hirosawa, S. Mechanism of grain size refinement of Fe₃B/Nd₂Fe₁₄B nanocomposite permanent magnet by Cu addition. *J. Appl. Phys.* **85**(4), 2448(1999).
10. Li, Y.Q., Yue, M., Zhao, G.P. & Zhang, H.G. Effect of magnetic soft phase on the magnetic properties of bulk anisotropic Nd₂Fe₁₄B/ α -Fe nanocomposite permanent magnets. *Mater. Res. Express.* **5**(1), 016108(2018).
11. Cha, H.G., Kim, Y.H., Kim, C.W., Kwon, H.W. & Kang, Y.S. Preparation for exchange-coupled permanent magnetic composite between α -Fe (soft) and Nd₂Fe₁₄B (hard). *Curr. Appl. Phys.* **7**(4), 400-403(2007).
12. Mahato, B.K., Piramanayagam, S.N., Rawat, R.S. & Laha, P. Magnetization

- dynamics and cure temperature study in SmCo₅/Co core-shell nanostructures. *Appl. Phys. A* **131**(1), 10(2024).
13. Zhang, X.W., Li, J., Liu, C., Wang, R.Q., Liu, H.F., Bao, X.T. & Liu, Y. High-coercivity Nd₂Fe₁₄B/ α -Fe nanocomposites with ultrafine nanocrystalline structure via Zr-induced synchronous precipitation. *Rare Metals*. **45**(02), 135-137(2026).
 14. Kim, C.W., Kim, Y.H, Cha, H.G. & Kang, Y.S. Exchange-coupling effect of Nd₂Fe₁₄B/FeCo nanocomposite by colloidal method. *Mol. Cryst. Liq. Crys.* **464**(1), 1-7(2007).
 15. Fan, J.P., Zhang, X.Y., Jiang, Y.N., Liang, R.Y., Sun, J., Bai, Y.H. & Xu, X.H. Optimization of energy product and reversal process for Nd₂Fe₁₄B/ α "-(FeCo)₁₆N₂/Nd₂Fe₁₄B exchange-spring trilayer films. *J. Magn. Magn. Mate.* **441**, 43-48(2017).
 16. Fu, S., Alam, M.K., Raza, S., Han, G.B. & Kang, S.S. Effect of Nb content on texture evolution in Pr₂Fe₁₄B/ α -Fe melt-spun ribbons. *Physica B* **733**, 418595(2026).
 17. Kneller, E.F. & Hawig, R. The exchange-spring magnet: a new material principle for permanent magnets[J]. *IEEE Trans. Magn.* **27**(4), 3588-3560(1991).
 18. Yang, B., Zhang, L., Shen, B.G., Zhao, T.Y. & Yu, R.H. High-performance α -Fe/Pr₂Fe₁₄B-type nanocomposite magnets fabricated by direct melt spinning. *J. Rare Earths*. **31**(1), 49-53(2013).
 19. Li, W., Zhao, L.Z. & Liu, Z.W. Micromagnetic simulation on magnetic properties of Nd₂Fe₁₄B/ α -Fe nanocomposites with Fe nanowires as the soft phase. *Front. Mater. Sci.* **12**(4), 348-353(2018).
 20. Uehara, M., Gennai, N., Fujiware, M. & Tanaka, T. Improved perpendicular anisotropy and permanent magnet properties in Co-doped Nd-Fe-B films multilayered with Ta. *IEEE Trans. Magn.* **41**(10), 3838-3843(2005).
 21. Morala, J.S., Arnay, I., Gkouzia, G., Pedraz, P., Perna, P., Alff, L., Navio, C. & Bollero, A. Spin reorientation transition in epitaxial nanometric Nd-Fe-B films with large perpendicular magnetic anisotropy. *J. Alloys Compd.* **1006**, 176215(2024).
 22. Takayuki, T., Jun, F., Shin, S., Yamato, H. & Hirotsugu, T. C-axis texture of high-rate-sputtered Nd₂Fe₁₄B thin films: independence of sheet texture of Mo

- underlayer. *Jpn. J. Appl. Phys.* **61**(6), 065506(2022).
23. Zhang, M.G., Yang, L.L., Zhao, Y.C., Zhang, J. & Sun, G. Research on magnetic properties of Nd₂Fe₁₄B/ α -Fe/Nd₂Fe₁₄B exchange-coupled trilayer films. *Appl. Mech. Mater.* **1388**(84-85), 567-571(2011).
 24. Tran, L.B.T. & An-Cheng, S.A. Interface change of Pr-Fe-B thin films with different ferromagnetic compositions. *Inorg. Chem. Commun.* **147**, 110254(2023).
 25. Be, L.T.T. & Aidan, S.A. Manifestation of initial crystallization and c-axis texture in Pr-Fe-B magnetic thin films grown on glass substrates under volume effect. *Mater. Chem. Phys.* **275**, 125249(2022).
 26. Yang, C.J. & Kim, S.W. Exchange coupling in Nd₂Fe₁₄B/Fe/Nd₂Fe₁₄B sandwich films and their magnetic properties. *J. Appl. Phys.* **87**(9), 6134(2000).
 27. Suzuki, A., Hotta, Y., Yamada, M., Ohtake, M., Futamoto, M., Kirino, F. & Inaba, N. Structure and magnetic properties of SmCo/X(X=Fe, Fe-Co, Co) magnetic bilayer films on MgO(110) substrate. *J. Magn. Magn. Mater.* **440**, 74-78(2017).
 28. Cui, W.B., Takahashi, Y.K. & Hono, K. Nd₂Fe₁₄B/FeCo anisotropic nanocomposite films with a large maximum energy product. *Adv. Mater.* **24**(48), 6530-6535(2012).
 29. Xia, J., Zhao, G.P., Zhang, M. & Zuo, J.F. The effect of the easy axis orientation on the magnetic properties of hard/soft multilayers. *Sci Sin. Phys. Mech. Astr.* **43**(1), 39-47(2013).
 30. Peng, Y., Zhao, G.P., Wu, S.Q., Si, W.J. & Wang, X.L. Micromagnetic simulation and analysis of Nd₂Fe₁₄B/Fe₆₅Co₃₅ magnetic bilayered thin films with different orientations of the easy axis. *Acta Phys. Sin-CH ED.* **63**(16), 167505-8(2014).
 31. He, X.X. & Zhao, Q. Micromagnetic studies on influence of interface atomic diffusion on magnetic properties of SmCo/Fe exchange-spring bilayers. *Acta Phys. Sin-CH ED.* **70**(19), 197502-10(2021).
 32. Jiang, Y.N., Liang, R.Y., Sun, J., Yang, Y. & Fan, J.P. Investigation of magnetic properties and magnetic reversal process of Nd₂Fe₁₄B/ α "-Fe₁₆N₂ bilayers. *J. Atom. Mole. Phys.* **34**(4), 734-738(2017).
 33. Li, Y.Q., Yue, M., Wu, Q., Wang, T., Cheng, C.X. & Che, H.X. Investigation on magnetic properties of parallel and perpendicular oriented Nd₂Fe₁₄B/Fe₆₅Co₃₅/Nd₂Fe₁₄B films by the micro-magnetism finite element method. *J. Magn. Magn. Mater.* **394**(15), 117-120(2015).
 34. Zhao, G.P., Zhang, X.F. & Morvan, F. Theory for the coercivity and its

mechanisms in nanostructured permanent magnetic materials. *Rev. Nanosci. Nanotechnol.* **4**, 1-25(2015).

35. Landau, L.D. & Lifshitz, E. On the theory of the dispersion of magnetic permeability in ferromagnetic bodies. *Physikalische Zeitschrift der Sowjetunion*, **8**(153), 101-114(1935).