Micromagnetic Simulation Study on the Magnetic Properties of Dual-Main-Phase Nd-Fe-B/Ce-Fe-B Periodic Multilayer Films (Postprint)

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Abstract

To improve the magnetic properties of permanent magnet thin films and enhance the utilization efficiency of Ce elements, the magnetization reversal process of dual-main-phase $Nd_2Fe_{14}B/Ce_2Fe_{14}B$ periodic multilayer films was simulated based on micromagnetic theory using the OOMMF software. The effects of magnetic layer thickness and multilayer number on the magnetic properties of periodic multilayer films during the magnetization reversal process were systematically investigated. The remanence, coercivity, hysteresis loops, energy variations during magnetization reversal, and the coercivity mechanism of the system were analyzed, providing a reference for the future preparation of high-performance magnets with high Ce content. The results demonstrate that when the total multilayer thickness and layer number are fixed, both the coercivity and maximum energy product of the system gradually decrease with increasing magnetic layer thickness. The coercivity mechanism of the periodic multilayer films is predominantly nucleation-dominated. Under identical conditions, the coercivity and energy product of parallel-oriented periodic multilayer films are superior to those of perpendicular-oriented ones. Increasing the total thickness of the periodic multilayer films reduces the influence of orientation on coercivity. These findings will contribute to a deeper understanding of the magnetization reversal mechanism in dual-main-phase Nd-Fe-B/Ce-Fe-B magnets and provide a reference for optimizing the magnetic properties of permanent magnet thin films in future experiments.

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Abstract

To improve the magnetic properties of permanent magnet thin films and enhance the utilization efficiency of rare earth elements, we conducted micromagnetic simulations of the magnetization reversal process in double main-phase $Nd_2Fe_{14}B/Ce_2Fe_{14}B$ periodic multilayers using the OOMMF software. The effects of magnetic layer thickness and number of multilayer periods on the magnetic properties were systematically investigated during magnetization reversal. The results show that the coercivity mechanism of the periodic multilayers is dominated by nucleation. When the total multilayer thickness and period number are fixed, both coercivity and maximum magnetic energy product gradually decrease with increasing magnetic layer thickness. Under identical conditions, parallel-oriented periodic multilayers exhibit superior coercivity and magnetic energy product compared to perpendicularly oriented ones. Increasing the total thickness of periodic multilayers reduces the influence of orientation on coercivity. These findings deepen our understanding of magnetization reversal mechanisms in magnets and provide valuable guidance for optimizing the magnetic properties of permanent magnet thin films in future experimental work.

Keywords: micromagnetic simulation; Nd-Fe-B/Ce-Fe-B; periodic multilayer; magnetic properties

1. Introduction

Nd-Fe-B permanent magnets have attracted widespread attention from researchers due to their excellent magnetic properties. However, with the rapid development of the rare earth permanent magnet industry, the imbalance in rare earth metal usage has become increasingly prominent. Since rare earth elements are mostly enriched symbiotically in the Earth' s crust, the extraction of Nd and Pr for Nd-Fe-B magnets has resulted in large stockpiles of La and Ce, causing environmental pollution and resource waste. The anisotropy field of $Ce_2Fe_{14}B$ is significantly lower than that of $Nd_2Fe_{14}B$, and its theoretical saturation magnetization is also much lower (1.17 T vs 1.61 T). Direct substitution of Nd with Ce in conventional methods leads to drastic reductions in remanence and intrinsic coercivity.

To address these challenges, Wang Jingdai proposed a double main-phase approach that combines two or more permanent magnetic materials with different intrinsic parameters to leverage their respective advantages. This method can reduce costs and decrease the usage scale of rare earth metals while achieving comprehensive and balanced utilization of rare earth resources. Previous studies have demonstrated the feasibility of this approach. For instance, Sun Yachao et al. [15] used DC magnetron sputtering to prepare NdFeB, CeFeB, and NdFeB/CeFeB films, proving that double hard magnetic composites can effectively reduce the time dependence of thin film magnetization and improve temporal stability. Zhang Jun et al. [17] conducted micromagnetic simulations of the magnetization reversal process in Sm-Co/α-Fe/Sm-Co trilayers, finding that the reversal behavior transitions from single-phase to two-phase as the soft magnetic layer thickness increases. Ma Jianchun et al. [19] simulated Sm-Co/α-Fe bilayers and multilayer gradient films, revealing that optimal structure eliminates hysteresis loop steps and significantly reduces coercivity.

Due to the complexity of crystal structures and phase compositions in sintered magnets, this study selects relatively simple thin film materials as research objects. Using micromagnetic simulation methods, we investigated the hysteresis loops and coercivity mechanisms of double main-phase Nd-Fe-B/Ce-Fe-B multilayers with both perpendicular and parallel orientations, providing reference for optimizing permanent magnet thin film properties in future experiments.

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2. Simulation Methods

The simulation model consists of periodic multilayers where one period comprises alternating $Nd_2Fe_{14}B$ and $Ce_2Fe_{14}B$ layers. The three-dimensional Cartesian coordinate system origin is placed at a bottom vertex. When the easy magnetization axis and applied magnetic field are both along the Z-axis (perpendicular to the film plane), this is referred to as perpendicular orientation. When they are along the Y-axis (parallel to the film plane), this is referred to as parallel orientation.

The magnetization process follows the Landau-Lifshitz-Gilbert (LLG) equation. The effective field is defined as:

$$H_{eff} = -\frac{1}{\mu_0 M_s}\frac{\delta E}{\delta M}$$

where M is the magnetization vector at a point, M_s is saturation magnetization, and E is the energy density expressed as:

$$E = A_{ex}\left[\nabla\left(\frac{M}{M_s}\right)\right]^2 + K_u \sin^2\theta - \mu_0 M \cdot H_{ext} + E_{demag}$$

The terms represent exchange energy, magnetocrystalline anisotropy energy, Zeeman energy, and demagnetization energy, respectively. A_{ex} is the exchange coupling constant and K_u is the magnetocrystalline anisotropy constant.

Simulations were performed using the OOMMF micromagnetic software. The damping coefficient α was set to 0.5 to ensure calculation efficiency without affecting accuracy. Material parameters for $Nd_2Fe_{14}B$ and $Ce_2Fe_{14}B$ are listed in Table 1. The exchange coupling coefficient at the interface $A_{interface}$ varies according to calculation conditions.

Table 1: Magnetic Properties Parameters of $Nd_2Fe_{14}B$ and $Ce_2Fe_{14}B$

Parameter	$Nd_2Fe_{14}B$	$Ce_2Fe_{14}B$
M_s ($MA \cdot m^{-1}$)	1.61	1.17
K_u ($kJ \cdot m^{-3}$)	4900	2100
A_{ex} ($pJ \cdot m^{-1}$)	7.7	7.7

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3. Results and Discussion

3.1 Magnetic Properties of Perpendicularly Oriented Double Main-Phase Periodic Multilayers

3.1.1 Effect of Magnetic Layer Thickness at Fixed Period Number With the period number fixed and the $Nd_2Fe_{14}B$ to $Ce_2Fe_{14}B$ layer thickness ratio at 1:1, we simulated the magnetic property variations by changing the magnetic layer thickness while keeping other parameters constant. The easy axis and applied field were both perpendicular to the film plane.

The hysteresis loops for different magnetic layer thicknesses are all square-shaped [FIGURE 1]. As the magnetic layer thickness increases, both coercivity (H_c) and maximum magnetic energy product ($(BH)_{max}$) gradually decrease. When the layer thickness increases from 3 nm to 9 nm, coercivity decreases from 1.2 MA/m to 0.8 MA/m, and $(BH)_{max}$ decreases from 320 kJ/m^3 to 180 kJ/m^3. The remanence does not change significantly.

This occurs because increasing magnetic layer thickness weakens interlayer exchange coupling, making magnetization reversal easier and reducing coercivity and $(BH)_{max}$. Figure 2 shows the energy evolution during magnetization reversal for different layer thicknesses. When the system is positively saturated, all energy terms are near zero. As the external field decreases, exchange energy increases slowly. At the nucleation field, most magnetic moments begin to reverse, causing dramatic changes in all energy terms. After complete reversal, the total system energy reaches its minimum.

The good squareness of hysteresis loops indicates nucleation-dominated coercivity mechanism. For thinner layers (3 nm), exchange energy begins increasing at smaller external fields, indicating stronger exchange coupling and higher coercivity. Thicker layers show weaker coupling and easier nucleation, resulting in lower coercivity.

3.1.2 Effect of Period Number at Fixed Total Thickness With total system thickness fixed at 60 nm and layer thickness ratio at 1:1, we varied the period number n while maintaining perpendicular orientation. Figure 3 shows the hysteresis loops for different period numbers.

Both coercivity and $(BH)_{max}$ increase gradually with increasing period number n. This is because, at constant total thickness, increasing n equivalently reduces individual layer thickness, enhancing interlayer exchange coupling. When $n = 10$ (layer thickness 3 nm), exchange coupling is strongest, leading to maximum coercivity and $(BH)_{max}$.

Figure 4 illustrates the energy evolution during reversal. The trends are similar across different period numbers, but systems with more periods exhibit stronger exchange coupling. More interfaces create more pinning sites during magnetization reversal expansion, hindering domain wall motion and increasing coercivity. The exchange energy for $n = 10$ is significantly larger than for $n = 4$, making nucleation more difficult and increasing coercivity.

3.2 Magnetic Properties of Parallel-Oriented Double Main-Phase Periodic Multilayers

3.2.1 Effect of Magnetic Layer Thickness at Fixed Period Number With period number fixed at 4 and layer thickness ratio at 1:1, we investigated parallel orientation (easy axis and field parallel to film plane) by varying magnetic layer thickness. The hysteresis loops remain square-shaped [FIGURE 5].

Similar to the perpendicular case, coercivity and $(BH)_{max}$ decrease with increasing layer thickness due to weakened exchange coupling. However, the absolute values are higher than in the perpendicular orientation for the same thickness. Figure 6 shows the energy evolution, confirming nucleation-dominated coercivity mechanism. The exchange energy for 3 nm layers is substantially larger than for 9 nm layers, indicating stronger coupling and higher coercivity in thinner layers.

3.2.2 Effect of Period Number at Fixed Total Thickness With total thickness fixed at 60 nm and parallel orientation, increasing period number n enhances coercivity and $(BH)_{max}$ [FIGURE 7]. The trend matches the perpendicular orientation case but with superior absolute performance. Figure 8 shows energy evolution for $n = 4$ and $n = 10$, demonstrating that more periods create stronger exchange coupling and more interfacial pinning, making magnetization reversal more difficult.

3.3 Influence of Orientation on Magnetic Properties

Figure 9 compares coercivity for different orientations, layer thicknesses, and period numbers. Under identical conditions, parallel-oriented double main-phase Nd-Fe-B/Ce-Fe-B periodic multilayers exhibit superior coercivity compared to perpendicularly oriented ones. This is because parallel orientation produces stronger exchange coupling energy (E_{ex}) than perpendicular orientation, making magnetic moments more difficult to nucleate.

When comparing at fixed total thickness but different period numbers, the coercivity difference between orientations remains relatively stable. However, as total thickness increases, the orientation effect on coercivity gradually diminishes, and the values converge. This indicates that thicker multilayers reduce the impact of orientation on magnetic properties.

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4. Conclusions

Using micromagnetic simulations with OOMMF, we systematically studied double main-phase $Nd_2Fe_{14}B/Ce_2Fe_{14}B$ periodic multilayers and found:

1. The coercivity mechanism is dominated by nucleation, evidenced by the square hysteresis loops and energy evolution during reversal.
2. With fixed period number, increasing magnetic layer thickness weakens interlayer exchange coupling, reducing both coercivity and maximum magnetic energy product.
3. With fixed total thickness, increasing period number enhances exchange coupling and creates more interfacial pinning sites, increasing coercivity and $(BH)_{max}$.
4. Parallel-oriented multilayers consistently outperform perpendicularly oriented ones in coercivity and energy product due to stronger exchange coupling. However, the orientation effect diminishes as total thickness increases.

These results provide valuable theoretical guidance for optimizing the magnetic properties of double main-phase permanent magnet thin films and enhancing rare earth resource utilization.

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Note: Figure translations are in progress. See original paper for figures.