SPREAD OF INTERACTION IN NANOCOMPOSITE HARD/SOFT NANOSTRUCTURED MAGNETS

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ABSTRACT

In this study, the magnetic properties of 3D modeled two-phase hard/soft nanocomposite nanostructured magnets were simulated by means of the Monte-Carlo method. The dependences of the energy product and coercivity on the grain size and magnetically soft phase content were investigated. The influence of the interaction spreading in the soft phase on the magnetic properties was also discussed. The obtained results revealed that the energy product reaches an optimal value when the soft phase content ranges around 50 vol.%, and a strong magnetic interaction spreading locally along the Kneller-Hawig exchange length seems to be more important than a weak but widely spreading interaction.

Keywords: 3D simulation, nanocomposite magnets, Monte-Carlo method, two-phase magnetically hard and soft, hardening interaction.

1. INTRODUCTION

Two-phase hard/soft nanocomposite nanostructured magnetic materials are combinations of a highly coercive and moderate spontaneous magnetization hard phase and a high spontaneous magnetization soft phase. According to the Kneller-Hawig theory [1], by the exchange interaction between two nanostructured magnetic phases, the soft phase is hardened in the region contiguous to the hard phase leading to the energy product *(BH)_{max}* improvement in comparison with that of the single hard phase. Several theoretical calculations showed that the energy product can reach 120 MGOe (1000 kJ/m^3) with the presence of very high soft phase content ~ 90 vol.% [2, 3]. Therefore, the nanocomposite nanostructured magnetic materials have been under intensive investigation during the past twenty years.

However, until now, the experimental value of *(BH)_{max}* of the nanocomposite nanostructured magnets is lower than 25 MGOe (200 kJ/m^3), far below the expected value [4 - 9]. Moreover, the optimal soft phase content (the content giving rise to the best value of *(BH)_{max}*) was only in the range of 20 ÷ 30 vol.% [10 - 15]. Because of this large discrepancy between the theoretical diagnose and experimental results, a more realistic simulation of two-phase hard/soft nanocomposite nanostructured magnetic materials should be implemented to orient experiments.

Our previous simulations [4, 16] based on the Kneller-Hawig criterion and the random grain size distribution generated by the Monte-Carlo procedure partly described the magnetic properties of two-phase nanocomposite materials. The obtained results revealed the important roles of the grain size distribution, the soft phase content as well as the grain dispersion of the system. However, in this simulation, the Kneller-Hawig hardening criterion manifesting an exchange interaction between hard and soft phases was kept fixed along the exchange length x_0. This can cause a discrepancy with that of the reality where the exchange interaction can decrease gradually from the boundary into the inner region of soft phase grains.

Currently, Saiden et al. have done the micro-magnetic simulation for NdFeB/α-Fe/Fe$_3$B systems [17] and observed the normal behavior where the remanence increase and coercivity decrease ocurred by increasing the soft phase content. However, the simulated demagnetization curves are kink-free even with 40 vol.% of the soft phase content. This result should be reconsidered since their simulation was performed with 22 nm averaged grain size which is larger than the value 10 nm obtained from Kneller-Hawig criterion for this system. Moreover, the real dispersion of grains can build clusters which reduce the ability of hardening effect. Thus, the results obtained by Saiden and co-authors can give raise to the effect that the exchange interaction should be spreaded over the length estimated by Kneller-Hawig criterion and this effect must be affirmed.

To study the spread of the exchange interaction in the soft phase and its effect on the magnetic properties of the nanocomposite magnetic materials, the 3D Monte-Carlo simulation was performed and presented in this paper. The result reveals a strong interaction located along the exchange length is necessary to reach high-performance magnetic properties.

2. SIMULATION ALGORITHM

In this study, the Nd$_2$Fe$_{14}$B/α-Fe nanocomposite nanostructured system was considered. The intrinsic parameters of phases were given in Table 1.

Table 1. Magnetic parameters of two magnetic phases [18].

	J_s (T)	$_iH_c$ (kOe)	K (kJ/m^3)	A (pJ/m^3)
Nd$_2$Fe$_{14}$B	1.61	10	4900	8
α-Fe	2.15	0	48	22

The microstructure of the two-phase nanocomposite nanostructured magnets is described by the 3D model of polyhedron grains as presented in Fig. 1. This microstructure was built based on the Voronoi structure, where Voronoi sites are distributed randomly in the magnet region sized $a \times b \times c = 50$ nm $\times$ 50 nm $\times$ 50 nm. Every polygon should be randomly assigned as hard or soft grains, this assigning process depends also on the soft phase content.

Kneller-Hawig criterion is used for hardening a region inside the soft grain with an infinitesimal volume $d\Omega$, which depends on the distance from the region center to the boundary between the soft and adjacent hard grains. This criterion describes the exchange interaction spread by different shapes presented in Fig. 2. In the soft phase, the exchange interaction intensity is spread according to the following function of Fermi-Dirac:

$$\frac{I(x)}{I(x=0)} = \frac{1}{exp\left(\frac{x-x_0}{\beta}\right)+1},\tag{1}$$

where x is the distance counted from the boundary, x_0 is the exchange length $x_0 = \pi\sqrt{A_m/2K_c}$ with A_m being the exchange energy constant of the soft phase, K_c being the magnetocrystalline anisotropy constant of the hard phase, and β being the spreading constant. For $Nd_2Fe_{14}B/\alpha$-Fe, x_0 ≈ 5 nm was calculated with the parameters of Table 1. The exchange length x_0 was fixed while the parameter β was varied by the values 0.1, 0.5 and 1.5 to get different shapes of interaction spread.
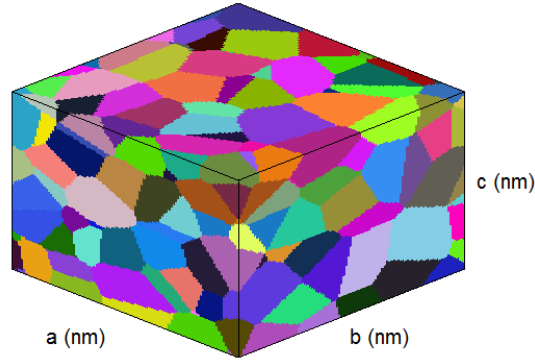

Figure 1. The 3D model simulating the microstructure of a two-phase nanocomposite nanostructured magnetic material. Different grains are differently colored.
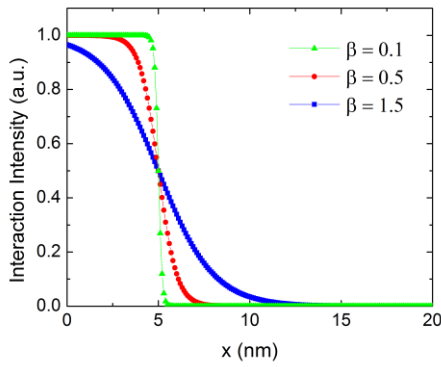

Figure 2. Different shapes of exchange interaction spreading inside the soft phase. The average exchange length 5 nm corresponds to the value of x_0 of $Nd_2Fe_{14}B/\alpha$-Fe.
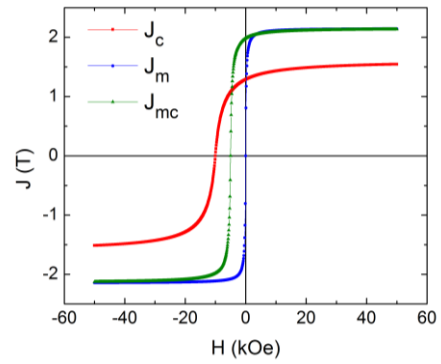
Figure 3. The demagnetization curves of magnetically hard (red) and soft (blue) phases and one example for the hardened soft phase (green).

So, under the effect of exchange interaction, the magnet considered as the assembly of infinitesimal regions of volume $d\Omega$ and magnetization J_Ω and these regions could be magnetically pure hard, pure soft or hardened soft phases. Their typical demagnetization curves are shown in Figure 3 and notated by J_c, J_m and J_{mc}, respectively. The total magnetization J of

the magnet is the averaged magnetization contributions of all these regions over the total magnet volume V:

$$J = \frac{1}{V} \int_V J_\Omega d\Omega. \tag{2}$$

The soft phase content of the magnet is signed as f_m, it is the sum of volumes of all the pure soft and hardened soft regions inside the magnet.

3. RESULTS AND DISCUSSION

Several demagnetization curves calculated for various contents of the soft phase are presented in Fig. 4, assumed that the interaction spreads with the parameter $\beta = 1.5$. Notably, these simulated results describe well the experimental demagnetization curves obtained in [12, 14, 15]. Specifically, for $f_m < 70$ vol.%, the demagnetization curves seem to be similar to that of the pure single hard phase, which happens in the case of low contents of the soft phase and very strong interaction between two phases (hard and soft), thus there is no or only small fraction of the non-hardened soft phase in the microstructure. By increasing the soft phase content over 70 vol.%, the fraction of the non-hardened or weakly hardened soft phase regions increases leading to the kink-behavior demagnetization curves and significant decrease of $(BH)_{max}$. The features of the coercivity $_iH_c$ and the remanence M_r are well described: while the coercivity decreases monotonically, the remanence increases then decreases by increasing the content of the soft phase.
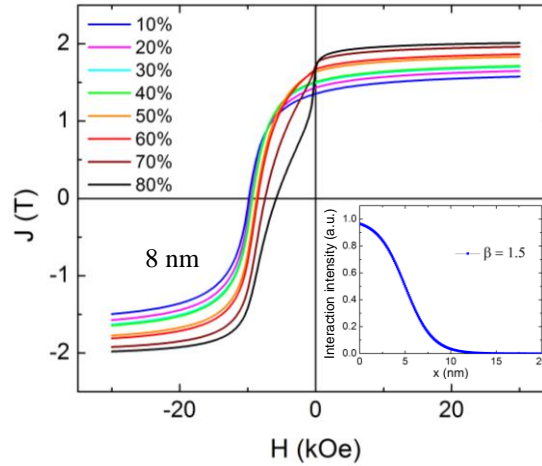

Figure 4. Demagnetization curves of the magnets with different soft phase contents ranged from 10 to 80 vol.%, the grain size is 8 nm and the interaction spread shape with the parameter $\beta = 1.5$ were used and shown in the inset.

Figure 5 presents the changes of the $(BH)_{max}$ versus soft phase content for three cases of interaction spread. In the case of small-size soft grains (< 9.5 nm), all the soft grains could be completely hardened resulting in the enhancement of $(BH)_{max}$ in comparison with that of the case of single hard phase and the above mentioned optimal content of soft phase can reach the value 50 vol.%. In the case of larger size of grains, the tendency of reducing this optimal content is observed, and the value of $(BH)_{max}$ is also decreased quickly. The reduction of energy product by

increasing soft phase content or grain size is caused by decreasing the fraction of the non-hardened soft phase.
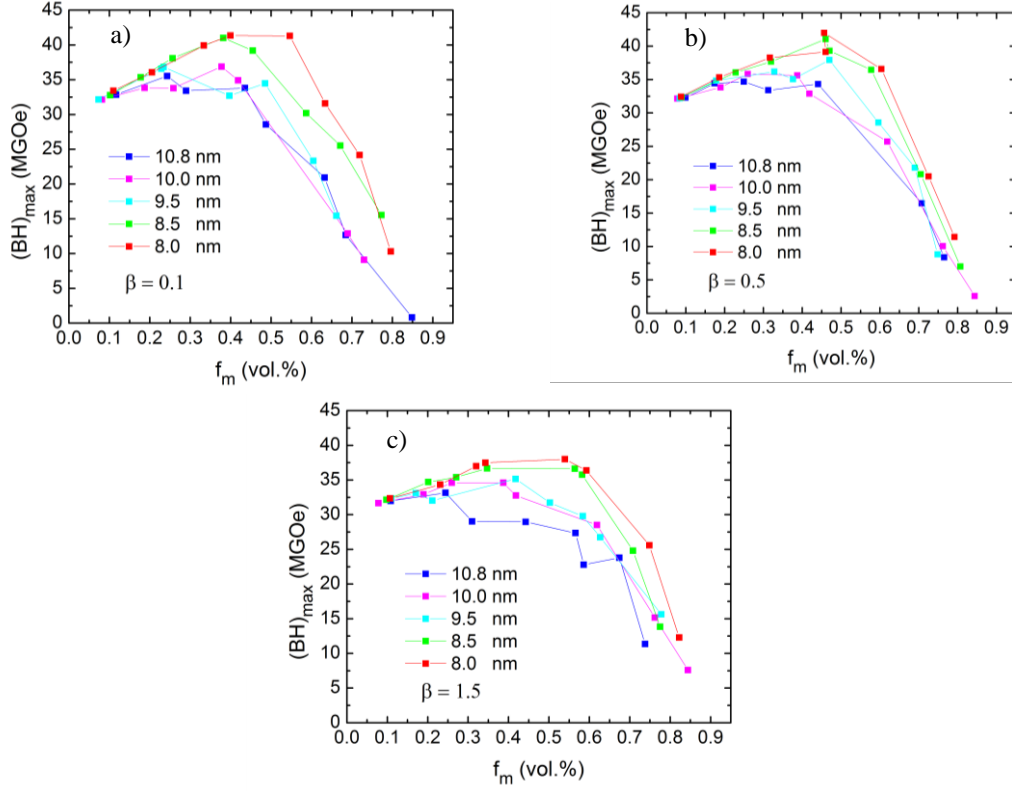

Figure 5. Dependences of the energy product on the soft phase contents calculated for different values of $\beta = 0.1$, 0.5 and 1.5.

The simulated results also proved that the energy product can be improved by several tens of percent. For example, for the case of the grain size of 8 nm and the optimal content of 50 vol.%, Figure 5a shows the energy product is enhanced up to the value of 42.5 MGOe, that means 41 % enhancement by comparing with the value of 30 MGOe of the case of single hard phase. Thus, a high content of soft phase around of 50 vol.% still can be used, of course the magnetically clean interface are critically required to hold an intensive exchange interaction spreading locally over the exchange length.

The above presented results agree with our previous ones (see [4, 16]) and the results obtained by Saiden et al., about the ability to increase the energy product of nanocomposite magnets with 40 vol.% of soft phase, although there is a discrepancy in grain size between Saiden's study and ours. If the experimental results support the simulation results of Saiden et al., this fact would imply that the averaged interaction length should be larger than Kneller-Hawig length $\pi\sqrt{A_m/2K_c}$.

To compare the results simulated by using different shapes of the interaction spread, notice that for the case of the small grain size the optimal content is independent of the parameter β, while the enhancement of energy product is declined by changing β from 0.1 to 1.5, which corresponds to the change from a strong but located in the small area of x_0 to a weaker but far

spreading interaction. This conclusion can be observed in Figs. 6a and 6b. Therefore, with the same interaction length x_0, the far spread but low intensive interaction does not significantly contributeto improve the magnetic performance.

Figures 6c and 6d show the dependences of the coercivity on the soft phase contents simulated for different shapes of the interaction spread. Governed by the strong and located interaction the coercivity is gradually reduced with the increase of the soft phase content. Conversely, by the weak but far spreading interaction the coercivity decreases faster. Based on this feature, the shape of interaction spread can be estimated by considering the manner of reduction of coercivity.
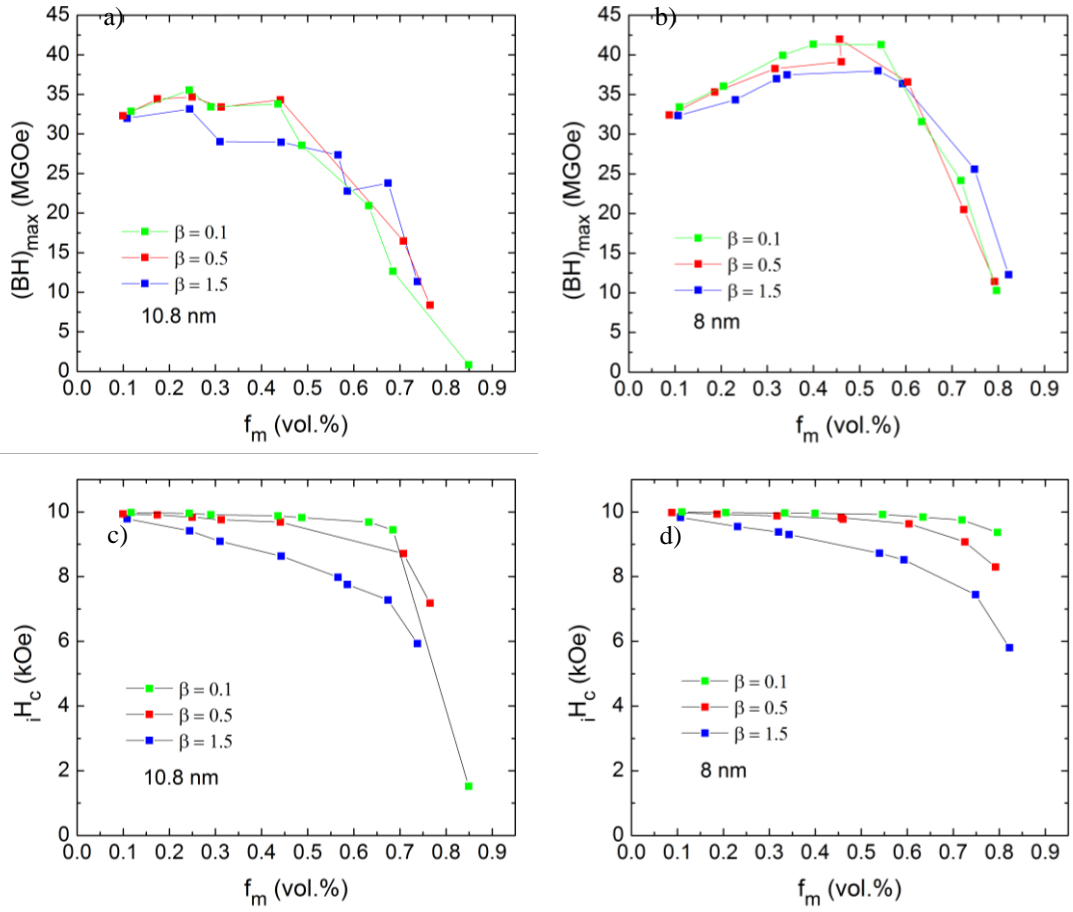

Figure 6. The influence of the shape of interaction spread on the magnetic properties versus the soft phase contents. The simulation results were done for two average grain size of 10.8 nm and 8 nm.

4. CONCLUSION

Monte-Carlo 3D simulation has been performed to study the effect of spreading of the exchange interaction in two-phase hard/soft nanocomposite nanostructured magnetic materials. The obtained demagnetization curves closely described the experimental ones. The energy product can be enhanced by 40 % more in the case of 50 vol.% of the soft phase content and the average grain size smaller than twice of the Kneller-Hawig exchange length, which corresponds

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to the case of the completely hardened soft grains. To attain this case, the magnetically clean grain boundaries are required to guarantee the strong interaction between two magnetic phases. This requirement should be taken into account for further improvement of the performance of nanocomposite magnets. Even in the case of high soft phase content up to 50 vol.% the strong local interaction can enhance the energy product easier than the weak spreading one. A shape of the interaction spread can be estimated through the rate of reduction of the coercivity.

REFERENCES

1. Kneller E. F. and Hawig R. - The Exchange-Spring Magnet: A new material principle for permanent magnets, IEEE Trans. Magn. **27** (1991) 3588-3600.

2. Skomski R. and Coey J. M. D. - Giant energy product in nanostructured two-phase magnets, Phys. Rev. B **48** (1993) 15812-15816.

3. Fukunaga H., Horikawa R., Nakano M., Yanai T., Fukuzaki T., and Abe K. - Computer simulations of the magnetic properties of Sm-Co/α-Fe nanocomposite magnets with a core-shell structure, IEEE Trans. Magn. **49** (2013) 3240-3243.

4. Truong N. X., Hieu N. T., Ky V. H., and Vuong N. V. - 2D simulation of $Nd_2Fe_{14}B$/α-Fe nanocomposite magnets with random grain distributions generated by a Monte Carlo procedure, J. Nanomater., Article ID 759750 (2012) 7 pages.

5. Liu Z. W. and Davies H. A. - The practical limits for enhancing magnetic property combinations for bulk nanocrystalline NdFeB alloys through Pr, Co and Dy substitutions, J. Magn. Magn. Mater. **313** (2007) 337-341.

6. Hou F., Cheng Y., Lou L., Li H., Guo D., Li W., Li X., and Zhang X. - Bulk α-Fe/$Nd_2Fe_{14}B$ nanocomposite magnets prepared by hot deformation of amorphous $Nd_{10.5}Fe_{82}Cu_{1.5}B_6$, Materials Letters, **108** (2013) 186-188.

7. Rong C. B., Wang D., Nguyen V. V., Daniil M., Willard M. A., Zhang Y., Kramer M. J., and Liu J. P. - Effect of selective Co addition on magnetic properties of $Nd_2(FeCo)_{14}B$/α-Fe nanocomposite magnets, J. Phys. D: Appl. Phys. **46** (2013) 045001-045005.

8. Bai Y., Lei Z., Baogen S., Tongyun Z., and Ronghai Y. - High-performance α-Fe/$Pr_2Fe_{14}B$-type nanocomposite magnets fabricated by direct melt spinning, J. Rare Earths **31** (2013) 49-53.

9. Poudyal N., Rong C., Nguyen V. V., and Liu J. P. - Hard-phase engineering in hard/soft nanocomposite magnets, Mater. Res. Express **1** (2014) 016103-016113.

10. Kato H., Kubota H., Koyama K., and Miyazaki T. - Fabrication of $SmFe_{12}$/α-Fe thin films as anisotropic nanocomposite magnet, J. Alloys Compd. **408–412** (2006) 1368-1372.

11. Zhang J., Zhang S., Zhang H., and Shen B. - Structure, magnetic properties, and coercivity mechanism of nanocomposite $SmCo_5$/α-Fe magnets prepared by mechanical milling, J. Appl. Phys. **89** (2001) 5601-5605.

12. Pop V., Dorolti E., Vaju C., Gautron E., Isnard O., le Breton J. M., and Chicinas I. - Structural and magnetic behaviour of $SmCo_5$/α-Fe nanocomposites obtained by mechanical milling and subsequent annealing, Rom. Journ. Phys. **55** (2010) 127-136.

13. Rong C., Zhang Y., Poudyal N., Xiong X., Kramer M. J., and Liu J. P. - Fabrication of bulk nanocomposite magnets via severe plastic deformation and warm compaction, App. Phys. Lett. **96** (2010) 102513-102515.

14. Rong C., Zhang Y., Poudyal N., Szlufarska I., Hebert R. J., Kramer M. J., and Liu J. P. - Self-nanoscaling of the soft magnetic phase in bulk SmCo/Fe nanocomposite magnets, J. Mater. Sci. **46** (2011) 6065-6074.

15. Rong C., Poudyal N., Liu X. B., Zhang Y., Kramer M. J., and Liu J. P. - High temperature magnetic properties of $SmCo_5/\alpha$-Fe(Co) bulk nanocomposite magnets, App. Phys. Lett. **101** (2012) 152401-152403.

16. Nguyen Trung Hieu, Master thesis, Modeling and simulation for nanostructure hard-magnetic materials, 2005.

17. Saiden N. M., Schrefl T., Davies H. A., Hrkac G. - Micromagnetic finite element simulation of nanocrystalline α-Fe/$Nd_2Fe_{14}B$/Fe_3B magnets, J. Magn. Magn. Mater. **365** (2014) 45-50.

18. Coey J. M. D., Magnetism and magnetic materials, Cambridge University Press, 2009.

TÓM TẮT

LAN TRUYỀN TƯƠNG TÁC TRONG NAM CHÂM TỔ HỢP HAI PHA TỪ CỨNG TỪ MỀM CẤU TRÚC NANO

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Trong nghiên cứu này, tính chất từ của nam châm tổ hợp hai pha từ cứng từ mềm cấu trúc nano ở mô hình 3 chiều đã được mô phỏng dựa trên phương pháp Monte-Carlo. Sự phụ thuộc của tích năng lượng cực đại, lực kháng từ vào kích thước hạt và tỉ phần pha từ mềm đã được khảo sát. Ảnh hưởng của dạng lan truyền tương tác trong pha từ mềm lên các tính chất từ cũng đã được nghiên cứu. Kết quả cho thấy rằng tích năng lượng cực đại đạt giá trị tối ưu trong vùng tỉ phần pha từ mềm ~50 vol.%, và tương tác từ mạnh trong khoảng chiều dài tương tác trao đổi Kneller-Hawig là quan trọng hơn một tương tác yếu nhưng có độ lan truyền cao.

Từ khóa: mô phỏng 3D, nam châm tổ hợp, phương pháp Monte-Carlo, hai pha từ cứng từ mềm, tương tác cứng hóa.

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