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Dependence of coercivity on phase distribution and grain size in nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets*

Feng Wei-Cun(冯维存)[a)b)], Gao Ru-Wei(高汝伟)[b)], Li Wei(李 卫)[a)†], Han Guang-Bing(韩广兵)[b)], and Sun Yan(孙 艳)[b)]

[a)] *Division of Functional Materials, Central Iron and Steel Research Institute, Beijing* 100081, *China*

[b)] *School of Physics & Microelectronics, Shandong University, Ji'nan* 250100, *China*

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The dependence of coercivity on the grain size in nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets with different distributions of magnetically soft and hard phases is investigated by means of statistical mean. The calculations show that when there exists no soft phase, the coercivity of magnets decreases monotonically with hard grain size reducing. For a given volume fraction of hard phase, the coercivity of nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets with a random distribution of soft and hard grains shows a peak value as a function of hard grain size. When the hard grain size is larger than an optimum value of soft grain size (15nm), the nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets with the multilayer structure of soft and hard grains can possess a higher coercivity than that with the random distribution of soft and hard grains.

Keywords: nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets, coercivity, grain size, phase distribution, exchange-coupling interaction

PACC: 7530E, 7530G, 7560G, 7560J

1. Introduction

In recent years, much attention has been paid to the grain size effect on magnetic properties of nanocomposite permanent materials.[1–9] Some papers reported that the coercivity of single-phase nanometre magnetic materials decreases[1,2,5] but the coercivity of nanocomposite magnets increases peculiarly with grain size reducing.[3,5] These phenomena are considered to originate from the exchange-coupling interactions between magnetic grains. Generally, intergrain exchange-coupling interactions tend to align the magnetic moments at the grain boundaries and are not favourable for the improvement of magnetic crystalline anisotropy and coercivity. Therefore, the increase in coercivity with grain size reducing in nanocomposite permanent magnets cannot be simply attributed to the exchange coupling interactions between magnetically soft and hard grains.

In fact, the phase distributions in nanocomposite permanent materials depend on their preparation procedures. For example, Liu *et al* reported that good magnetic properties were achieved in a nanocomposite (Nd, Dy)(Fe, Co, Nb, B)$_{5.5}/\alpha$–Fe magnet by sputtering and heat treatment,[10] and obviously exceeded those of nanocomposites with the same compositions, but prepared by use of mechanical alloying technique.[4] They attributed these phenomena to a more ideal microstructure with a uniform distribution of soft and hard phases obtained by sputtering and heat treatment. Similarly, the theoretical value of energy product, which is as high as 1MJ/m^3, is obtained based on the physical model with a multilayer structure composed of alternating hard and soft phases.[11] Therefore, the phase distribution should be one of the key factors controlling the magnetic properties of nanocomposite permanent magnets. In this paper, based on a simple effective anisotropy model, with the different phase distributions taken into account,[12] the dependence of coercivity on grain size in nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets is investigated. This

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†E-mail: fengweicun@126.com

work may provide a theoretical reference for the understanding of the grain size effects on coercivity in nanocomposite permanent magnetic materials.

2. Calculation method and models

There exist three kinds of grain boundaries, i.e. soft–soft, hard–hard and hard–soft grain boundaries in nanocomposite magnets consisting of magnetically soft and hard phases. The exchange-coupling interactions between the grain interfaces should be different, and the phase distributions make the exchange-coupling interactions fluctuated. Based on a simple effective anisotropy model of individual magnetic grains, Feng *et al*[12] put forward that the effective anisotropy of nanocomposites, $K_{\rm eff}$, was determined by the statistical mean of the anisotropies of all grains located in different coupling states. $K_{\rm eff}$ can be expressed by the following equation:

$$\begin{aligned} K_{\rm eff} =& f_{\rm ss}\langle K_1^{\rm ss}\rangle + f_{\rm sh}\langle K_1^{\rm sh}\rangle \\ &+ f_{\rm hs}\langle K_1^{\rm hs}\rangle + f_{\rm hh}\langle K_1^{\rm hh}\rangle \\ =& \sum_{i,j} f_{ij}\langle K_1^{ij}\rangle, \qquad (i,j={\rm s},\ {\rm h}), \end{aligned} \tag{1}$$

where $\langle K_1^{ij}\rangle$ denotes the effective anisotropy of individual magnetic grains located in different coupling states, i and j represent the soft and hard grain, respectively . The values of $\langle K_1^{ij}\rangle$ can be acquired from Ref.[12]. f_{ij} is the corresponding proportion of $\langle K_1^{ij}\rangle$, and determined by the phase distribution.

The coercivity is determined by the anisotropy, saturation polarization, and microstructures.[8] Arcas *et al* described the coercivity of nanometre permanent magnets by Eq.(1)[13]

$$H_{\rm c} = p_{\rm c}\frac{K_{\rm eff}}{J_{\rm s}}, \tag{2}$$

where $p_{\rm c}$ is a dimensionless factor which depends on the particular magnetization process and microstructures of materials. The values of $p_{\rm c}$ are generally smaller than 1, and $p_{\rm c}$=1 represents the nanocomposite possessing the ideal magnetization process and microstructure.

Supposing the volume fractions of soft and hard phases are $v_{\rm s}$ and $v_{\rm h}$ ($v_{\rm s}+v_{\rm h}$=1), respectively, the saturation polarization $J_{\rm s}$ can be expressed by[14]

$$J_{\rm s} = v_{\rm s}J_{\rm s}^{\rm s} + v_{\rm h}J_{\rm s}^{\rm h}, \tag{3}$$

where $J_{\rm s}^{\rm s}$ and $J_{\rm s}^{\rm h}$ represent the saturation polarization of soft and hard phases, respectively. Equation (2) can be rewritten as

$$H_{\rm c} = p_{\rm c}\sum_{i,j} f_{ij}\langle K_1^{ij}\rangle \Big/ \sum_i v_i J_{\rm s}^i, \quad (i,j={\rm s,h}). \tag{4}$$

In the present calculation, we adopt the random distribution and multilayer distribution of soft and hard phases mentioned in Figs.4 and 5 of Ref.[11], where shadow and blank areas represent the soft and hard phases, respectively. For simplicity, both shadow and blank magnetic regions are considered to be composed of regular cubes with each edge being a length D, which are joined closely in space. For the soft and hard grains with different sizes, the influences of the impurities and interspace among grains on the calculations are ignored due to their smaller volume fractions. In the nanocomposites with the random distribution, the value of f_{ij} can be obtained from Ref.[9]. For the multilayer nanostructure, we assume that the thickness of single layer is equal to the corresponding hard grain size $D_{\rm h}$ or soft grain size $D_{\rm s}$. So, the value of f_{ij} in the multilayer distribution is comprised of two items $f_{\rm s}$ and $f_{\rm h}$, which can be described by the volume fractions of soft and hard phases, respectively.

3. Results and discussion

Based on the random distribution, we investigate the dependence of the coercivity of nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets, $H_{\rm c}$, on the grain size and volume fraction of hard phase with soft grain sizes being 10nm and 15nm respectively, as shown in Fig.1 ($p_{\rm c}$=1). Figure 1 shows that when there exists no soft phase, $H_{\rm c}$ decreases monotonically with hard grain size reducing, and changes quickly only when $D_{\rm h} < 20$nm. For a given proportion and grain size of soft phase, $H_{\rm c}$ shows a maximum value as a function of the hard grain size, and augments with the volume fraction of soft phase increasing, but $D_{\rm h}$ corresponding to the maximum of $H_{\rm c}$ decreases. These results are basically in accordance with those in Refs.[12] and [15]. Ref.[12] pointed out that the decrease of coercivity of nanocomposite magnets is mainly due to the reduction in effective anisotropy of material, which is proved further by our calculations in this work. The calculations and experimental results in Ref.[15] indicate that both anisotropy and exchange coupling at boundaries decrease and then the coercivity, remanence and $(BH)_{\rm max}$ diminish, which is consistent with our results.

By comparing Figs.1(a) and (b) with the values of $D_{\rm s}$ being 10nm and 15nm respectively, the variations

of characteristics of all curves with D_s are basically similar, but the corresponding value of H_c in the case of D_s being 15nm is larger than that in the case of D_s being 10nm, which indicates that large D_s is in favor of the improvement of H_c under the precondition that decoupling behaviour does not occur. The experimental result in Ref.[4] shows that when the soft grain size is smaller than an optimum size of 15nm, the coercivity decreases rapidly with the soft grain size reducing. This phenomenon supports our calculations.

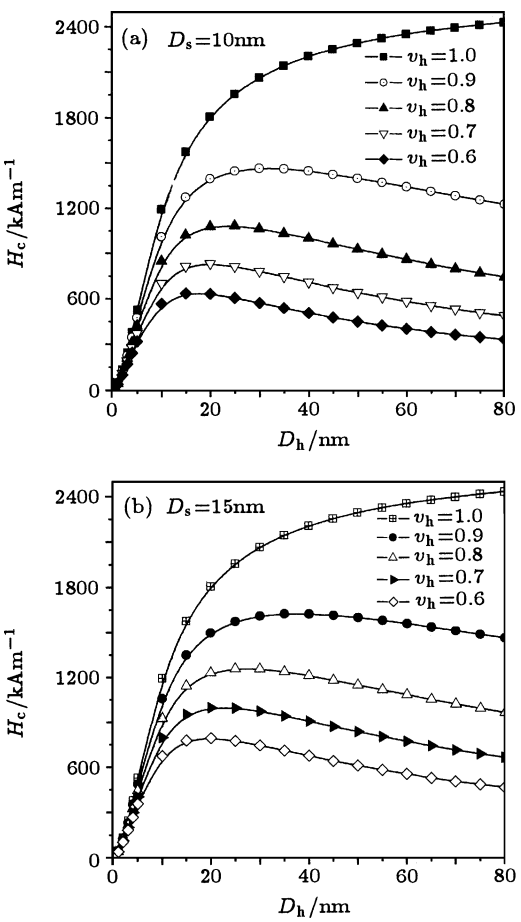


Fig.1. Dependence of the coercivity of a nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnet H_c on grain size D_h and volume fraction of hard phase v_h with different soft grain sizes being (a) D_s=10nm and (b)D_s=15nm.

Figure 2 shows a comparison of calculated coercivity H_c with the experimental results in Refs.[3, 5]. In the calculation, the relative values of the parameters, v_h and D_s, are identical with those in Refs.[3, 5]. When the fitting microstructure parameter p_c is in the range of 0.38–0.42 and the value of v_h equaling 0.70, or 0.85, or 1, the calculated dependence of H_c on the hard grain size,D_h, is basically in accordance with that given by Ref.[5]. When p_c is 0.32 and the value of v_h is 0.82, the calculated dependence of H_c on D_h is basically in accordance with that given by Ref.[3]. The values of p_c, which are determined by the magnetization processes and microstructure, should be identical under the same experimental condition, but different under different experimental conditions. In the present calculation, the soft and hard grains are idealized as the regular cubes. Actually, the grain shape is irregular and the grain size varies from several to a few tens of nanometres. In addition, the intergranular layers with a width of several nanometres should be considered in the calculation. So, the insertion of p_c is to characterize the effect of the factors mentioned above. In this calculation, p_c is obtained to be around 0.4 by fitting experimental results in Ref.[5], and 0.32 by fitting experimental results in Ref.[3]. From these comparisons, we find that the coercivity of nanocomposites is successfully described by Eq.(4) based on the simple effective anisotropy model.

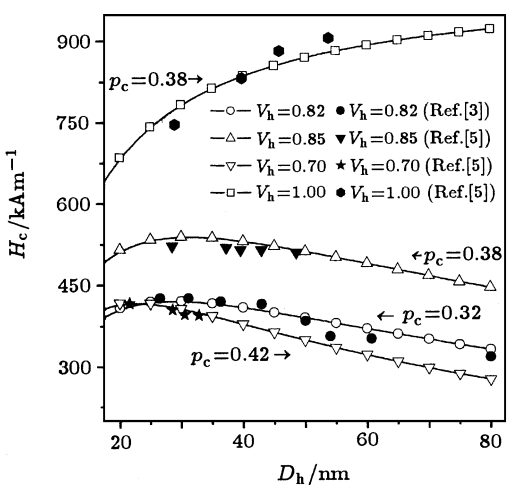


Fig.2. Comparison of the calculated coercivity H_c in this work with the experimental results; open symbols refer to experimental values of coercivity presented in Refs.[3, 5].

In order to better understand the effect of phase distribution on the coercivity of nanocomposites, we investigate the dependences of the coercivity of nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets H_c on grain size and volume fraction of hard phase with different phase distributions and soft grain sizes respectively being 10nm and 15nm. The results in Fig.3 show that the coercivities of a nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnet H_c increase monotonically with the increase of grain size and volume fraction of hard phase in

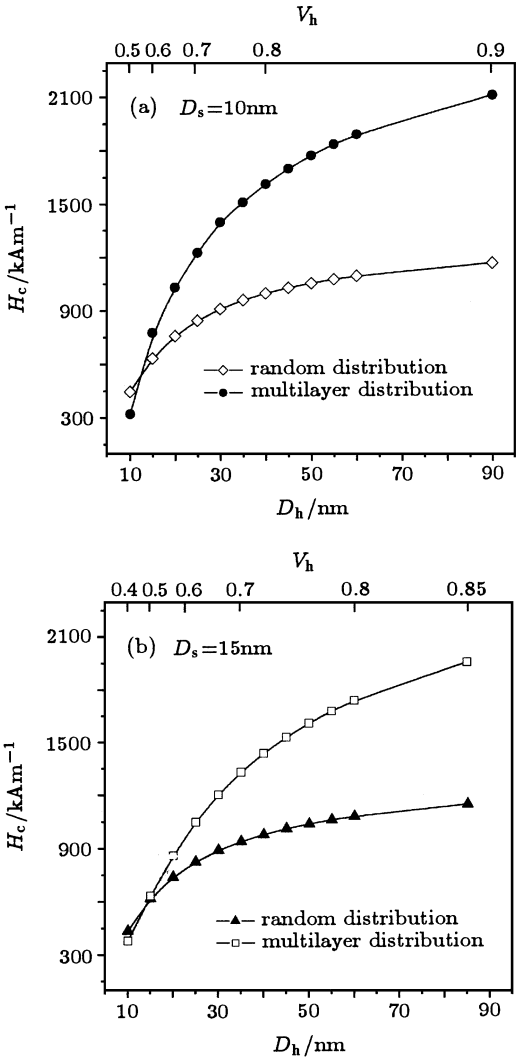


Fig.3. Dependences of the coercivity of a nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnet H_c on grain size D_h and volume fraction of hard phase v_h with different phase distributions and soft grain sizes being (a) D_s=10nm and (b)D_s=15nm.

both random and multilayer distributions. But the variation of H_c for a multilayer distribution tends to be steeper than that for a random distribution. The coercivity for the multilayer distribution is obviously larger than that for the random distribution when $D_h > D_s$ for the same grain size and volume fraction, and the coercivity for the multilayer distribution is about twice that for the random distribution while D_h >60nm. Therefore, the high coercivity is probably obtained by designing an ideal distribution of magnetically soft and hard phases.

4. Conclusion

There exist different exchange-coupling interactions between the different grain interfaces in nanocomposite magnets. Phase distributions make the exchange-coupling interactions fluctuate. In the absence of the soft phase, the coercivity of single phase $Nd_2Fe_{14}B$ magnets decreases monotonically with hard grain size reducing and changes quickly when $D_h <$ 20 nm. For a given volume fraction of the hard phase, the coercivity of nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets shows a peak value as a function of hard grain size for the random distribution of soft and hard grains. The fitting results with relative experiments indicate that the microstructure parameter p_c is around 0.3–0.4 and the statistical mean is useful to investigate the coercivity in nanocomposite permanent magnets. A comparison between the numerical results for the random and multilayer distributions indicate that the nanocomposite $Nd_2Fe_{14}B/\alpha$–Fe magnets with the multilayer structure of soft and hard grains can possess a higher coercivity than that with the random distribution when D_h is larger than D_s.

References

[1] Herzer G 1990 *IEEE Trans. on Mag.* **26** 1397
[2] Manaf A *et al* 1991 *J. Magn. Magn. Mater.* **101** 360
[3] Billona O V *et al* 1998 *J. Magn. Magn. Mater.* **187** 371
[4] Sun X K, Zhang J and Chu Y L 1999 *Appl. Phys. Lett.* **74** 1740
[5] Betancourt J I and Davies R H A 2002 *J. Magn. Magn. Mater.* **246** 6
[6] Zhang H W, Rong C B, Zhang J, Zhang S Y and Shen B G 2003 *Acta Phys. Sin.* **52** 718 (in Chinese)
[7] Jin H M, Wang X F, Zhao S F and Yan Y 2001 *Chin. Phys.* **10** 862
[8] Kronmüller H, Fischer R, Seeger M and Zern A 1996 *J. Phys. D: Appl. Phys.* **29** 2274
[9] Gao R W *et al* 2003 *J. Appl. Phys.* **94** 664
[10] Liu W, Zhang Z D, Liu J P, Dai Z R, Wang Z L, Sun X K and Sellmyer D J 2003 *J. Phys. D: Appl. Phys.* **36** 63
[11] Skomski R and Coey J M D 1993 *IEEE Trans. on Mag.* **29** 2860
[12] Feng W C, Gao R W, Han G B, Zhu M G and Li W 2004 *Acta Phys. Sin.* **53** 3171 (in Chinese)
[13] Arcas J, Hernando A and Barandiaran J M 1998 *Phys. Rev.* B **58** 5193
[14] Fischer R *et al* 1996 *J. Magn. Magn. Mater.* **153** 35
[15] Zhang H W *et al* 2004 *J. Magn. Magn. Mater.* **278** 127