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Inter-grain Exchange Interactions for Nanocrystalline $Nd_{2.33}Fe_{14}B_{1.06}Si_{0.21}$ Magnets

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The strengths of the inter-grain exchange interaction were evaluated for nanocrystalline $Nd_{2.33}Fe_{14}B_{1.06}Si_{0.21}$ magnets of different grain size by comparing the ${}_iH_c$ calculated by micromagnetics with the experiments. With increase of the grain boundary thickness to that of the magnet of grain diameter 12.4, 24.8, 37.2 and 49.6 nm, the strengh of the interaction in reference to that without the grain boundary phase decreases to 83%, 69%, 54% and 42%.

Key words : rare-earth permanent magnet, micromagnetics, coercivity, exchange interaction, nanocrystal

1. Introduction

It has been found by this paper's authors that the coercivity ${}_iH_c$ for nanocrystalline $Nd_2Fe_{14}B$ magnets calculated by the micromagnetics depends on the field direction. The average of the ${}_iH_c$ calculated along different field directions for a given grain number N decreases with increase of N and approaches a limit ${}_iH_c(\infty)$. ${}_iH_c(\infty)$ for small grain diameter D coincide with the experimental values of the nanocrystalline $Nd_{2.33}Fe_{14}B_{1.06}Si_{0.21}$ [1] very well [2, 3]. With increase of D, ${}_iH_c(\infty)$ increases in accordance with the experiments, but the increase is smaller and the discrepancy becomes larger. The difference should be attributed to the neglect of the grain boundary phase in the calculations. In the $Nd_{2.33}Fe_{14}B_{1.06}Si_{0.21}$ magnets, there exists a Nd-rich paramagnetic grain boundary phase, and the thickness of the boundary increases in proportion to D, thus causing decrease of the inter-grain exchange interaction and hence additional increase in ${}_iH_c$. This work presents a more accurate relation between the calculated $H_c(\infty)$ and D, and evaluate the variation of the strength of the inter-grain exchange interaction with the grain boundary thickness.

2. Models and Methods of Calculations

The cubic magnet consists of $n \times n \times n$ ($=N$) cubic $Nd_2Fe_{14}B$ grains of edge $L(=(\pi/6)^{1/3}D)$. The c-axes of the grains are randomly distributed. Each grain is divided into $m \times m \times m$ cubic regions of edge L/m. For model G, a region is a single domain element exchange coupled with the six adjacent elements. For model S, the region is subdivided into 24 tetrahedral elements of same size, and the magnetic polarization vector $\vec{J_s}$ varies linearly within each element. The dimensions of the elements for both models are between 1/3~1/2 of the domain wall thickness 4.2 nm of $Nd_2Fe_{14}B$. The periodical boundary conditions of magnetic properties hold at the magnet surfaces. The energy of the magnet consists of the magnetocrystalline anisotropy energy, Zeeman energy and the exchange interaction energy with $J_s = 1.63$ T, $A = 7.7 \times 10^{-12}$ J/m, $K_1 = 4.3 \times 10^6$ J/m^3 and $K_2 = 0.65 \times 10^6$ J/m^3 [4]. The stray field energy, which affects the value of ${}_iH_c$ little [5] while increasing the computational time to more than twice, is neglected. The exchange interaction per unit surface between the adjacent (i+1, j, k) and (i, j, k) elements within a grain for model G is approximated by

$$E_{ex}=A\int_i^{i+1}\sum_\gamma^{x,y,z}(\nabla\alpha_\gamma)^2dx$$

$$\approx A\sum_\gamma^{x,y,z}\left[\frac{\alpha_\gamma(i+1,j,k)-\alpha_\gamma(i,j,k)}{L/m}\right]^2\frac{L}{m}$$

$$=-\frac{2Am}{L}\frac{\vec{J_s}(i+1,j,k)\cdot\vec{J_s}(i,j,k)}{J_s^2}+\text{const.},\qquad(1)$$

where α_γ ($\gamma = x, y, z$) are the direction cosines of $\vec{J_s}$, and

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the integration extends between the centers of the elements. The exchange interaction between the adjacent elements across the grain boundary is also formulated by equation 1 but replacing A by $A\beta$ ($0 \le \beta \le 1$). The field is decreased from 5 T to −5 T by step, and the magnetization at each field is obtained from minimization of the energy by use of the conjugate gradient method. See Ref. 3 for the details.

3. Results and Discussions

Figure 1 demonstrates $\mu_{0i}H_c$ calculated for different $1/n$ and field directions for L=10 nm for model S and model G with $\beta = 1$. The different small circles for a given $1/n$ are calculated along different field directions. The average of $\mu_{0i}H_c$ along different field directions decreases with decrease of $1/n$, and approaches the limit $\mu_{0i}H_c(\infty)$. $\mu_{0i}H_c(\infty)$ is 0.86 T for model S and 1.2 T for model G.

Figure 2 shows $\mu_{0i}H_c(\infty)$ as a function of D compared with the experimental values for the $Nd_{2.33}Fe_{14}B_{1.06}Si_{0.21}$ magnets [1]. The calculations by Fischer *et al.* [4] are also presented for reference. The calculations by model S for small D coincide with the experiments better than model G. The result would be related to that model S with continuous variation of $\vec{J}_s$ in the magnet is more

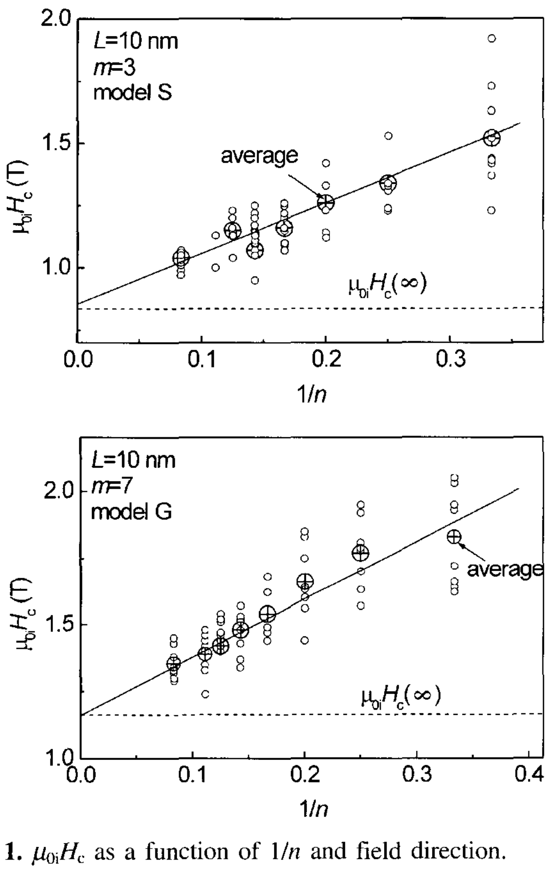

Fig. 1. $\mu_{0i}H_c$ as a function of $1/n$ and field direction.

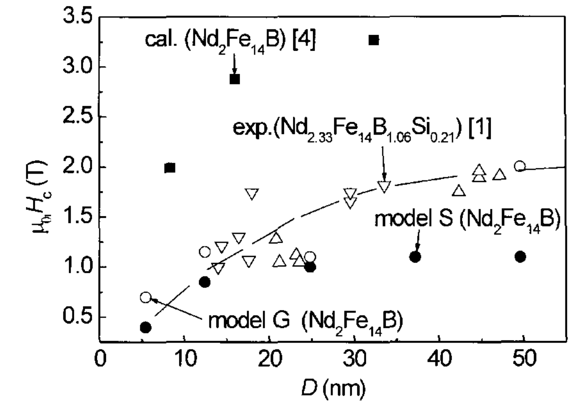

Fig. 2. $\mu_{0i}H_c$ (∞) and $\mu_{0i}H_c$ (exp.) as a function of D.

reasonable than model G with discrete variation. The $_iH_c$ calculated by model S is also less sensitive to m than model G [3], and $_iH_c(\infty)$ increases more smoothly with increase of D.

Model G, however, is useful for evaluation of the inter-grain exchange interaction when the effect of the grain boundary is taken into account. The inter-grain exchange interaction in this model is represented by the exchange interaction between the elements across the grain boundary. The parameter $2A\beta m/L$ represents the strength of the inter-grain exchange interaction. $2A$ is a constant, and the strengh is represented by $\beta m/L$ below. The coercivity for a given L should solely depends on the strengh of the inter-grain exchange interaction $\beta m/L$. As the grain boundary thickness approaches zero, β approaches 1, so m for β is near 1 should have a definite value which should be found. For smaller β, $_iH_c$ should be a function of βm but not m nor β. Figure 3 demonstrates $\mu_{0i}H_c$ as a function of $\beta m/L$ for L=20 nm, n=10 and m=14 and 16. The $\mu_{0i}H_c$ depends linearly on $\beta m/L$ in the range of $0 \le \beta \le 0.8$ for m=16. The results are the same for m=14, but the range of β extends to the whole range of $0 \le \beta \le 1$. It is believed

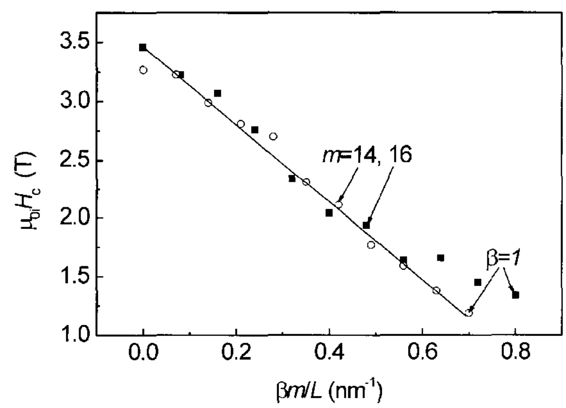

Fig. 3. $\mu_{0i}H_c$ as a functions of $\beta m/L$ calculated by model G. L=20 nm, n=10 and m=14 and 16.

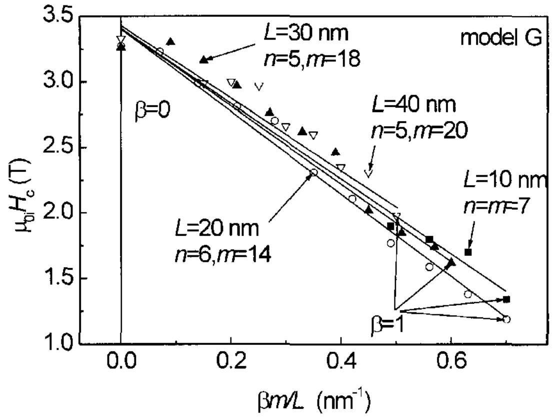

Fig. 4. $\mu_{0i}H_c$ (model G) as a function of *βm/L*.

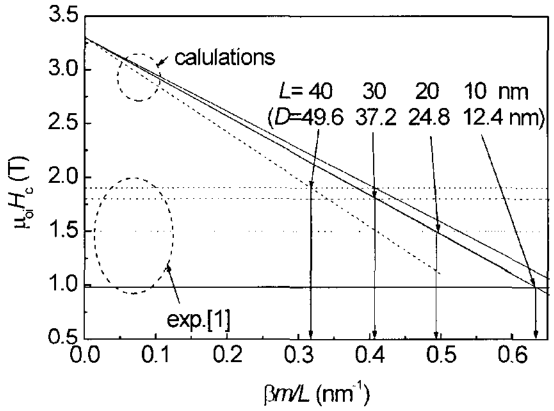

Fig. 5. $\mu_{0i}H_c$ (∞) (model S) as a function of *βm/L*, and $\mu_{0i}H_c$ of the experiments [1].

from the results that the straight line in the whole *β* range represents the relation between the coercivity and the strength of the inter-grain exchange interaction, and *βm*/*L* with $\beta = 1$ and m=14 represents the strengh of the inter-grain exchange interaction when the grain boundary thickness approaches 0.

Figure 4 demonstrates the similar linear relation between $\mu_{0i}H_c$ and *βm*/*L* for L=10 nm and n=m=7, L=20 nm, n=6 and m=14, L=30 nm, n=5 and m=18, and L=40 nm, n=5 and m=20. The straight line connects the $\mu_{0i}H_c(\beta=0)$ and $\mu_{0i}H_c(\beta=1)$ points. $\mu_{0i}H_c(\beta=0)$ is essentially of the Stoner-Wohlfarth model, and is little affected by the models, the values of n, m and L. On the other hands, $\mu_{0i}H_c(\beta=1)$ depends not only on L but also on n and field direction, and are always larger than corresponding $\mu_{0i}H_c(\beta=1, N=\infty)$ as is mentioned above. A more accurate relation could be obtained by replacing $\mu_{0i}H_c(\beta=1)$ by $\mu_{0i}H_c(\beta=1, N=\infty)$ for model S. Figure 5 shows the linear relations obtained in this way. In the Figure, the horizontal parallel lines represent the experimental values. From the intersect of the lines of the calculation and experiment for each grain size, *βm*/*L* as a function of the grain boundary thickness t (=cL, c: unknown constant) was obtained. Figure 6 shows *βm*/*L* as a function of t. With increase of of t from 0 to 10c, 20c, 30c and 40c nm, the inter-grain exchange interaction in reference to that with $t = 0$ decreases to 83%, 69%, 52% and 42% (if the volume fraction of the grain boundary is assumed to be 10% ($c \approx 0.033$), the inter-grain exchange interaction decreases to the above values for $t = 0.33$, 0.67, 1.0 and 1.3 nm).

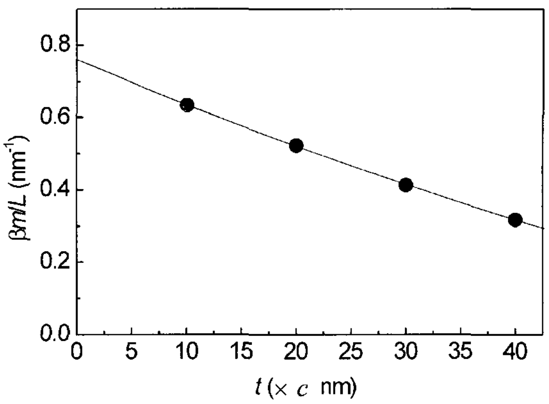

Fig. 6. *βm/L* as a function of *t*.

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References

[1] A. Manaf, R. A. Buckley, H. A. Davies, and M. Leonowicz, J. Magn. Magn. Mater. **101**, 360 (1991).

[2] H. M. Jin, S. F. Zhao, and X. F. Wang, J. Mater. Sci. Technol. **16**, 107 (2000).

[3] H. M. Jin, X. F. Wang, S. F. Zhao, and Y. Yan, Chin. Phys. **10**, 862 (2001).

[4] R. Fischer, T. Schrefl, H. Kronmuller, and J. Fidler, J. Magn. Magn. Mater. **153**, 35 (1996).

[5] H. M. Jin, Y. B. Kim, and X. F. Wang , J. Phys.: Condens. Matter **10**, 7243 (1998).