Numerical simulation of random magnetic anisotropy with solid magnetization grains

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Abstract

Random anisotropy model (RAM) was investigated by means of numerical simulation. Magnetization of magnetically interacting grains with randomly oriented uniaxial anisotropy was calculated using the Landau-Lifshitz-Gilbert equation where the magnetization in a particular grain is assumed to align in the same direction (single spin model). Calculations were carried out for $10 \times 10 \times 10$ three dimensional cells changing cell sizes from 5 to 25 nm. The relation between coercive forces and grain sizes was obtained to be $H_C \sim D^{5.7}$ from the simulated magnetization curves. This result fits to the primitive theory $H_C \sim D^6$ and the experimental results.

Key words: random magnetic anisotropy, micromagnetic simulation, single domain, coercive force, exchange interaction

1. Introduction

Nanocrystalline materials have been intensively studied as one of the most typical examples of nanotechnology.[1] In these materials, nono-sized crystalline grains were densely gathered and have a bulk form. Especially for magnetic materials, these sorts of materials realize excellent soft magnetic properties and have great potentials as realistic materials.[2] Basically, random orientation of nano-sized ferromagnetic grains with dense packing is essentially important. Exchange interaction working between fine grains averaged out the magnetic anisotropy and lowered their amplitudes remarkably and it resulted in soft magnetic characteristics.

In the model proposed by Herzer[3, 4], the magnetic anisotropy energy is averaged out in a magnetically coherent area coupled with exchange interaction, and the relation between the grain size, D, and coercive force, H_C, is predicted to be $H_C \sim D^6$. The model is called random magnetic anisotropy model (RAM) and is the most popular one for interpreting soft magnetic characteristics. Many experimental reports support this simple theory not only for single phase materials but also for multi-phase substances and granular substances (see Fig 1.). Recently we have reported on coercive forces of pure Ni nanocrystals and confirmed D^6 dependence in a diameter range between 8 and 13 nm.[5] However, the different expressions form the 6th power law, D^6, were reported, in which the shape of the sample and the distribution of the stress might affect to the dependency of the coercive force.[6, 7] Therefore, to reconfirm the relation between D^6 and H_C within the framework of RAM, an analysis purely including the exchange interaction is necessary.

An analytic model of Herzer, where averaging effect of the magnetic anisotropy is dealt with the statistical technique, has
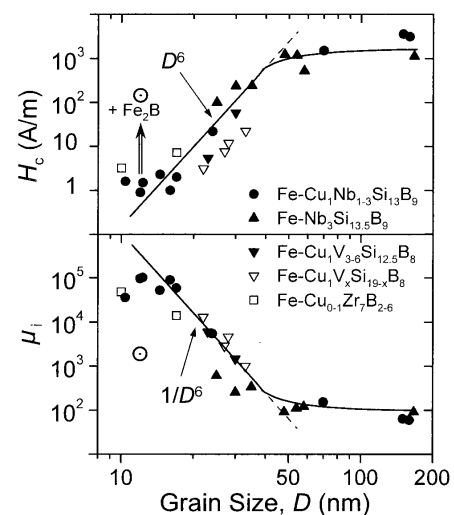

Figure 1: Example of the relation of coercive force and initial permeability to grain size in fine grain magnetic material.[3] D^6 dependence of coercive force is clearly demonstrated in a nanocrystalline region.

pointed out that coercive force becomes 0 keeping D^6 low even if grain size becomes very small.[8] In order to avoid this discrepancy between the model and experimental results, effects of defects and stresses[7, 9, 10] with the decrease in grain sizes have been introduced in analysis. However, there are still unclear parts in the effect of stress on the relation of D^6 and the range of grain size and minimum values of coercive forces resembled materials with different magnitudes of magnetostriction. These facts require us to propose a new model interpreting the change of coercive force only by the diameter of grains.

Kronmüller *et al.*[11] made the precise calculation of magnetic coercive force in a magnetic particle assembly using a finite element method. They reproduced the increase of the coercive force with the increase of the grain size for small grain

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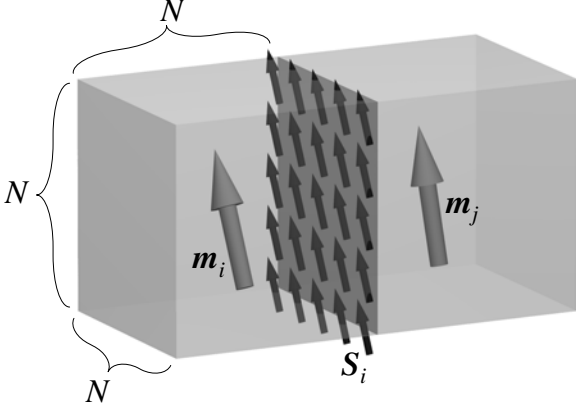Email address:* kita@bk.tsukuba.ac.jp (E. Kita)

Figure 2: Model of inter-particle exchange interaction based on single spin approximation.

size region and a decrease in grain diameters becoming larger. However, this model was insufficient because the aim of the research was focused on the analysis of magnetic coercive force of the permanent magnet and the resulting relation between the coercive force and the grain size was far from the form of D^6.

In this study, an analytic model only taking into account the effects of the grain diameter is built for the analysis of the random anisotropy model. The numerical analysis with formalization of the exchange magnetic field which works in between particles was carried out to obtain the relation between the coercive force and the grain diameter.

2. Expression of exchange coupling field under single spin approximation

When we calculate the distribution of local magnetic moment with numerical modeling, it is necessary to use discrete analysis techniques such as a difference equation method and a finite element method. As a simple discrete model, the following relation:

$$\frac{d\mathbf{M}(x)}{dx} \approx \frac{\mathbf{M}(x + \delta x) - \mathbf{M}(x)}{(x + \delta x) - x} \tag{1}$$

can be considered for the efficient approach to numerical calculation by taking the maximum amplitude of δx. This means that eq.(1) is generally kept when the locally defined magnetization $\mathbf{M}(x)$ is constant in a range of δx. Then local magnetization, $\mathbf{M}(x_i)$, at the position x_i is represented by the averaged amplitude over the range, δx. This approach where the averaged value represents the local magnetization is called single spin approximation and is generally used in the numerical analysis such as micromagnetic simulation.[12] Based on this assumption, the single spin model can be adopted in the magnetic particle assembly composed of magnetic single domain particles.

The model of nanocrystalline magnetic materials proposed here is shown in Fig. 2. Each crystalline grain has a cube form in which the magnetic moment distributes uniformly. The size of the particle is sufficiently small to behave as single domain

ferromagnets. Magnetization at the i-th particle is written as $\mathbf{m}_i$ and keeps a relation with individual spin $\mathbf{S}_i$, $\mathbf{m}_i = g\mu_B \sum \mathbf{S}_i$. Namely, magnetic ions with $\mathbf{S}_i$ spin align on a simple cubic allay having $N \times N \times N$ lattice points. The distance between magnetic ions is supposed to be a. Magnetic moments on two adjacent grains are coupled with magnetic exchange interaction through the interface atoms. It is supposed that Heisenberg type direct exchange interaction takes place between the interface atoms of the mother phase. An effective field working on the precise particle is evaluated by solving an energy equation.

Exchange interaction energy of the classical Heisenberg model upon magnetic atoms is expressed as $E = -J(\mathbf{S}_i \cdot \mathbf{S}_j)$ where J is an exchange constant among two spins. From the definition of the interface between two grains, $N \times N$ spins are faced in both i-th and j-th grains and form spin pairs. In each grain, all spins direct the same direction, therefore, exchange energy at the interface is

$$E_{\text{int}} = \sum -J(\mathbf{S}_i \cdot \mathbf{S}_j) = -JN^2(\mathbf{S}_i \cdot \mathbf{S}_j). \tag{2}$$

Here, summation is carried out on all possible spin pairs at the interface. Grain size D is equal to the sizes of the cube and $D = aN$, thus, it is clear that the energy is proportional to the area of the interface from eq.(2). There is also exchange energy among spins within a grain,

$$E_{\text{gr}} = \sum -J(\mathbf{S}_i \cdot \mathbf{S}_{i'}) = -\frac{JzN^3}{2}(\mathbf{S}_i \cdot \mathbf{S}_{i'}). \tag{3}$$

Summation is carried out over the all the possible spin pairs. Here, z and $1/2$ are the coordination numbers in the crystal and the correction factor during summation. Finally, the total energy related to exchange interaction on i-th grain is given by the sum of eq.(2) and eq.(3), $E_{\text{ex}}^i = \sum E_{\text{int}} + E_{\text{gr}}$. The term $\sum E_{\text{int}}$ is summed up over the adjacent grains.

Effective magnetic field originated from the exchange energies is deduced by the differentiation of the exchange energy,

$$
\begin{aligned}
\mathbf{H}_{\text{ex}} &= -\frac{\partial E_{\text{ex}}}{\partial \mathbf{m}_i} \\
&= -\frac{\partial \sum E_{\text{int}}}{\partial \mathbf{m}_i} - \frac{\partial E_{\text{gr}}}{\partial \mathbf{m}_i}
\end{aligned}
\tag{4}
$$

The second term should be zero because eq. 3 becomes constant due to the relation, $(\mathbf{S}_i \cdot \mathbf{S}_{i'}) = 1$ because of the parallel configuration of spins in the grains. The fact that the number of spins in a grain is $N \times N \times N$ and $\mathbf{m}_i = g\mu_B \sum \mathbf{S}_i$ leads to the next relation

$$
\begin{aligned}
\mathbf{H}_{\text{ex}} &= \frac{\partial \sum \sum J(\mathbf{S}_i \cdot \mathbf{S}_j)}{g\mu_B \partial \sum \mathbf{S}_i} \\
&= \frac{JN^2 \partial \sum (\mathbf{S}_i \cdot \mathbf{S}_j)}{g\mu_B N^3 \partial \mathbf{S}_i} \\
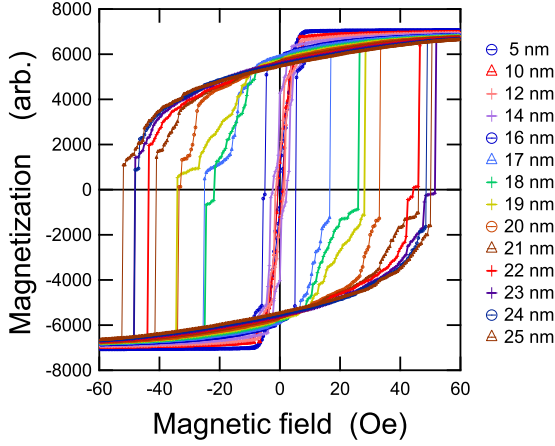&= \frac{J}{Ng\mu_B} \sum \mathbf{S}_j.
\end{aligned}
\tag{5}
$$

Figure 3: Numerically simulated hysteresis curves for ferromagnetic randomly oriented nano-grains. The grain size, D, was changed in the range from 5 to 25 nm. Inter-particle exchange interaction was taken into account.

The number of atoms at the side of the interface region N is $N = D/a$ then the following relation results in

$$\mathbf{H}_{\mathrm{ex}} = \frac{Ja}{Dg\mu_B} \sum \mathbf{S}_j. \tag{6}$$

This relation shows that exchange interaction proportional to its area results in the exchange field inversely proportional to the grain size. Since the definition of the exchange stiffness is identical to eq.(6), exchange stiffness decreases with the increase in size of grain, D.

3. Analysis of coercive force in nanocrystals based on the single spin approximation

Magnetization process of magnetic nanocrystal assembly was investigated using the single spin approximation mentioned above. The Hamiltonian for the i-th grain is written by the sum of the exchange energy, magnetic anisotropy energy, E_{an}, and Zeeman energy,

$$\mathcal{H}_i = \sum E_{\mathrm{int}} + E_{\mathrm{an}} + E_{\mathrm{Zeeman}}. \tag{7}$$

The anisotropy energy is given as

$$E_{\mathrm{an}} = -\frac{D^3 K}{m^2} (\mathbf{m}_i \cdot \mathbf{n}_i)^2, \tag{8}$$

where K and $\mathbf{n}_i$ are the magnetic anisotropy constant and unit direction vector for magnetic easy axis of the grain, respectively.

Motion of magnetization of the i-th grain under the effective magnetic field is supposed to obey Landau–Lifshitz–Gilbert equation,

$$(1 + \alpha^2)\frac{d\mathbf{m}_i}{dt} = -\gamma \mathbf{m}_i \times \mathbf{H}_{\mathrm{eff}} - \frac{\alpha}{m} \mathbf{m}_i \times (\mathbf{m}_i \times \mathbf{H}_{\mathrm{eff}}). \tag{9}$$

The effective field, $\mathbf{H}_{\mathrm{eff}}$ is obtained to be the differentiation of the model Hamiltonian, $\mathbf{H}_{\mathrm{eff}} = -\partial \mathcal{H}_i / \partial \mathbf{m}_i$.
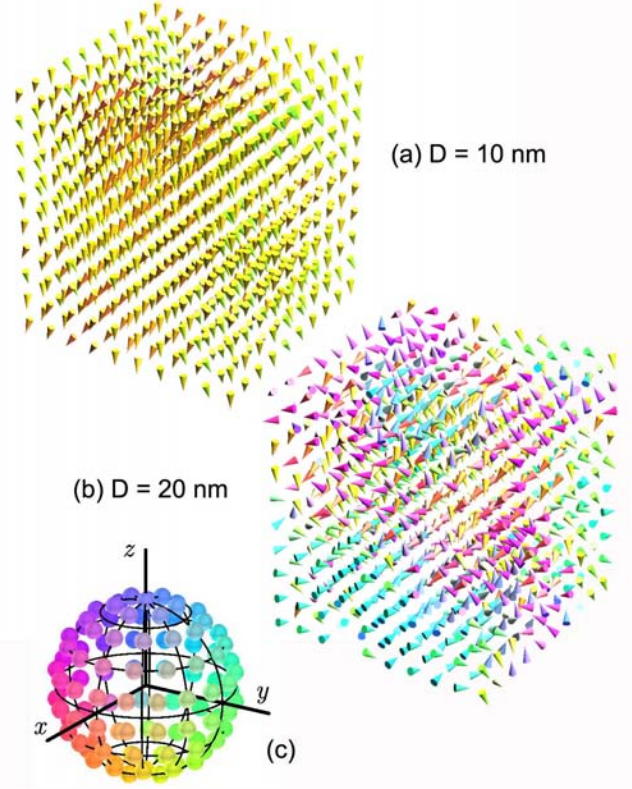

Figure 4: Illustration of magnetic moment distribution resulted from the simulation at $H_0 = H_c$. Distributions for grain sizes of $D = 10$ and $D = 20$ nm with inter–grain interaction of $J = 0.1 \times 10^{-14}$ erg are displayed in Figs. 4 (a) and (b), respectively. Directions of grain magnetization are expressed by the direction mapping sphere shown in (c).

We assumed atomic magnetic moment of $g\mu_B S = 5.56 \times 10^{-21}$ emu, distance of atoms of $a = 3.4 \times 10^{-8}$ cm, exchange energy between *interface* atoms of $J = 0.1 \times 10^{-14}$ ($J = 1.6 \times 10^{-14}$ erg for bulk Ni), and magnetic anisotropy constant of $K = 4 \times 10^4$ erg/cm^3. These conditions were set to fit to a Ni nanocrystals case, and uniaxial magnetic anisotropy is assumed for the sake of simplicity. Periodic boundary condition was applied to eliminate the edge effect.

Simulation was started from building up random orientation of magnetic anisotropy on $n^3(10 \times 10 \times 10)$ cells. The simulation cell size, n, was mainly 10 and was extended up to 15. The motion equation was numerically solved according to eq. (9). First, the energy in the eq. (7) on the each grain under the sufficiently high magnetic field was calculated. The effective field can determine the torque for each spin and distribution of the spins was obtained by the self-consistent calculation for all m_i. By changing the amplitude of the external magnetic field, hysteresis loops were reproduced and the coercive force of the system will be determined. Fig. 3 shows simulated magnetic hysteresis curves with different grain sizes. Jumps in MH curves at around $H \simeq H_C$ was observed in larger grain size $D \sim 20$ nm, while smooth change was shown in a smaller size nanocrystal. In Fig. 4, magnetization distribution at H_C is displayed where the direction of magnetization in each grain is expressed by an
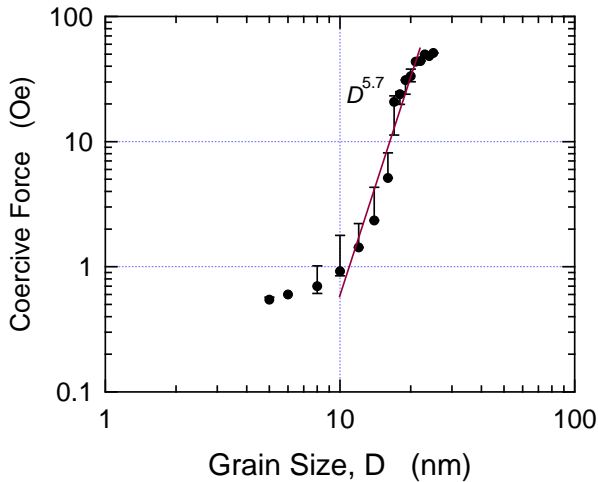
Figure 5: Dependence of coercivity on grain size in the magnetic nanoparticle system. Solid circles indicate coercive forces simulated with the simulation cell size of $n = 10$ and the solid curve is obtained by fitting data in a nanocrystalline range $D = 10 \sim 22$ nm. Bars indicate the maximum and minimum values of coercive forces simulated with cell sizes up to $n = 15$.

arrow and its color. In the case of $D = 10$ nm, the magnetizations of all grains refer to parallel configuration. On the other hand, in the case of $D = 20$ nm, the magnetization randomly points to the arbitrary direction.

Coercive forces were determined in a grain size ranging from 5 to 25 nm for the simulation cell size of $n = 10$ and they are plotted as a function of the grain size, D, in Fig. 5. In a grain size range from 10 to 22 nm, the coercive force changed steeply. Those data points can be fitted by a curve of the the the $D^{5.7}$ relation. The number of power, 5.7, well agrees with the theoretical prediction by Herzer. It is concluded that the D^6 law is held in a bulk nanocrystal materials when the effect of demagnetization can be disregarded.

It is noted that the coercive forces deviated slightly when the simulation cell size increased up to $n = 15$ (displayed by the bars in Fig. 5). The initial distribution of magnetic anisotropy also influenced the magnitude of coercive forces with the same order as the case of cell size, however, both changes look not systematic but random.

4. Discussion

In this section, applicability of the model will be discussed on the grain size of magnetic nanocrystals. In the present model, the magnetically single domain particles are under consideration, therefore, the magnetization in a certain grain can be regarded as a single spin. That is, the coercive force of a single grain is simply determined by the Stoner-Wohlfarth model. The coercive force of the nanoparticle assembly is given by an average of the coercive force on such single domain particles, when there is no interaction between particles.[13] However, in the present model, the simulated coercive force is reduced from a simple average, and such reduction originates from the

inter-grain exchange interaction. Namely, the maximum coercive force should be realized in non-coupled assemblies as the single spin model. For example, the relation of $H_C = 0.479 \, H_k$ is produced by randomly oriented anisotropies in that system. Here, H_k depicts an anisotropy field.

The fitted curve of coercive forces on the log–log plot shown in Fig. 5 is expressed as

$$H_C = 10^{k \log D + h}. \tag{10}$$

As a fitting result, $k = 5.7 \pm 0.5$ and $h = -5.9 \pm 0.7$ are obtained. This approximate equation gives $H_C = 55.6$ Oe when D = 22 nm. This value coincides well with H_C for non-interacting assembly, corresponding to maximum coercive force of the simulated system. Thus, the particle diameter, $D = 22$ nm, determines the upper limit of applicability as the single spin approximation for RAM. Above that diameter, H_C changes more moderately and obey D^6 law no longer. The magnetic anisotropy energy dominates the behavior of coercive forces. In such a case, the distribution of magnetization (probably multi-domain structure) in a single grain should be taken into account. Beyond this single spin approximation, the model proposed by Kronmüller[11] will be suitable.

On the other hand, the minimum coercive force also exists in the system as mentioned, and a simple expression of RAM is not able to be adopted. The reduction of coercive force begins with the inter-grain exchange interaction. The minimum of exchange energy is obtained when the magnetization of all grains align in parallel. At the same time, the magnetic anisotropy energy reaches the maximum (the minimum state of anisotropy is given by the non-interacting assembly as mentioned above). Thus, it is thought that the minimum state of total energy exist in between the minimum states of exchange and anisotropy energies. Corresponding to that consideration, the magnetization in Fig. 4(a) shows the relaxation from the parallel state. Therefore, the calculated data of coercive forces displaces from the curve of D^6 when particles become too small. From the viewpoint of energy minimization, the present model is suitable to calculate the small coercive forces. The definitions of the minimum coercive force in this system need more details, and it will be discussed in a separate paper.

5. Conclusion

Numerical simulation for Random Anisotropy Model applied on the ferromagnetic nanoparticle assembly was performed. By taking into account inter-grain exchange interaction proportional to the interface area, single spin approximation was revealed to reproduce the relation between coercive force and grain sizes, D^6. As the grain size decreased down to the nanocrystal limit, the coercive force began to leave from the relation D^6 and approached to a constant value.

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