高エネルギー加速器研究機構大型シミュレーション研究成果報告書（平成 25-26 年度）

(Brief report of the program)　　2014 年　12 月　　1 日

研究責任者名 Name	小野 寛太 Kanta Ono	所属機関 Affiliation	高エネルギー加速器研究機構 物質構造科学研究所
受理番号 Proposal No.	大型 13/14-08	研究課題名 Program title	磁気構造シミュレーションによる永久磁石の保磁力モデルの構築

研究を終了しましたので、下記の通り報告します。

成果の概要

Abstract

（和文）

磁性材料の磁化過程の理解は、磁気特性を知る上で重要である。次世代高性能磁石の開発などで大規模シミュレーションへの期待が高まっているが、従来のワークステーションや GPU を用いたシミュレーションでは、CPU 性能やメモリ量、通信帯域幅などの問題で、数十万セル程度の小規模な計算に留まっていた。われわれは、スーパーコンピュータを用いることにより、反磁場計算を含み、さらに 1 億セルを超える大規模シミュレーションが可能なマイクロ磁気シミュレータを開発した。KEK のスーパーコンピュータ Hitachi SR16000/M1 を使用し、自動並列コンパイラを用いた共有メモリ型並列化と MPI による分散メモリ型並列化を併用するハイブリッド並列化を行った。巨視的な反磁場の影響を避けるため周期境界条件を課し、反磁場の計算は 3 次元 FFT を使用した。本研究により、スーパーコンピュータを用いることで、1 億セルを超えるような大規模計算が 1 日程度という現実的な時間で可能であることが明らかとなった。大規模セル数での計算が可能になったことで、実際の磁石材料サイズのシミュレーションによる磁性材料開発への展開が期待できる。また、J-PARC などでの中性子実験との比較を行うための中性子小角散乱シミュレータを実装し、実験との比較を行った。

（英文）

We have successfully performed a large-scale micromagnetics simulation for more than 100 million cells with long-range dipolar interaction using the fast Fourier transform method. The recent demand for a coercivity mechanism in permanent magnets requires an extremely large simulation size, requiring a large-scale micromagnetics simulator. We have developed a large-scale micromagnetics simulator in which a magnetostatic energy calculation is implemented using fast Fourier transform. A hybrid parallel algorithm, which is a combination of shared-memory and distributed-memory parallel algorithms, is used to handle large data arrays. The simulation was carried out on a Hitachi SR16000/M1 supercomputer. Our simulator enables the calculation of realistic size of crystal grains and grain boundaries in a permanent magnet. We have also implemented a neutron scattering experiment simulator and compares the calculated results with experiments.

研究成果を公開しているホームページアドレス

研究成果の公表	口頭研究発表件数	査読つきの学術論文数	プロシーディング論文数	その他（投稿中を含む）
	2	2	0	0

成果の公表リスト（それぞれの枠に番号をつけて記入願います。）

口頭研究発表　Presentations at scientific meetings concerning the program

1. Hiroshi Tsukahara, Kaoru Iwano, Nobuhito Inami, Chiharu Mitsumata, Masao Yano, Tetsuro Ueno, Kotaro Saito, Tetsuya Shoji, Akira Manabe, Akira Kato, and Kanta Ono, “Micromagnetic simulation of magnetic small angle neutron scattering of Nd-Fe-B nanocrystalline magnet”, The 59th Annual Magnetism and Magnetic Materials (MMM) Conference

2. 塚原宙，岩野薫，井波暢人，三俣千春，矢野正雄，上野哲朗，斉藤耕太郎，庄司哲也，真鍋明，加藤晃，小野寛太，マイクロマグネティックシミュレーションによる Nd-Fe-B 磁石の磁化反転過程と SANS パターンの解析、第 38 回 日本磁気学会学術講演会

査読つきの学術論文(雑誌名等には 巻、頁、発表年を記載)（＊）不足する場合には追加願います。

Refereed Journal Articles (name of journal, volume, page, year)

1	著者名 Author	N. Inami, Y. Takeichi, C. Mitsumata, K. Iwano, T. Ishikawa, S.-J. Lee, H. Yanagihara, E. Kita, and K. Ono,
	タイトル title	“Three-Dimensional Large-Scale Micromagnetics Simulation Using Fast Fourier Transformation”,
	雑誌名 name of journal	IEEE Trans. Magn. 50, 1400304 (2014).
	URL	http://ieeexplore.ieee.org/stamp/stamp.jsp?arnumber=6692942
2	著者名	S.-J. Lee, N. Inami, H. Yanagihara, E. Kita, C. Mitsumata, and K Ono,
	タイトル	“Inter-grain Interaction in Random Magnetic Anisotropy Simulation in Magnetic Nanocrystals”
	雑誌名等	J. Appl. Phys. (in press)
	URL	
3	著者名	
	タイトル	
	雑誌名等	
	URL	

プロシーディング論文(雑誌名等には 巻、頁、発表年を記載)（＊）不足する場合には追加願います。

International Conference Proceedings (name of journal, volume, page, year)

1.	著者名 Author	
	タイトル title	
	雑誌名等 name of journal	
	URL	
2.	著者名	
	タイトル	
	雑誌名等	
	URL	
3.	著者名	
	タイトル	
	雑誌名等	
	URL	

その他（学位論文、紀要、投稿中の論文を含む）（著者、タイトル、論文種別、URL を記載）

Others (thesis for a degree, bulletin, papers to be published, etc.)

1. Author, title, sort of article, URL:

特記（本研究に関係した、新聞記事・著作、受賞など）（過去に遡っても構いません。）

Special Notes (newspaper article, literary works, awards, etc.)

1.

2.

実施報告書

課題グループ名：scmag
高エネルギー加速器研究機構
小野寛太

①研究組織

小野寛太（おの　かんた）
　高エネルギー加速器研究機構 物質構造科学研究所 准教授
　研究責任者、シミュレーションと放射光・中性子実験結果の解析と保磁力メカニズム

岩野薫（いわの　かおる）
　高エネルギー加速器研究機構 物質構造科学研究所 研究機関講師
　非平衡統計物理シミュレーションによる保磁力モデルの構築

井波暢人（いなみ　のぶひと）
　高エネルギー加速器研究機構 物質構造科学研究所 特任助教
　大規模磁気構造シミュレーション

塚原宙（つかはら　ひろし）
　高エネルギー加速器研究機構 物質構造科学研究所 研究員
　大規模磁気構造シミュレーション

三俣千春（みつまた　ちはる）
　高エネルギー加速器研究機構 物質構造科学研究所 共同研究員
　計算モデルの構築、非平衡統計物理とマイクロ磁気学との融合

柳原英人（やなぎはら　ひでと）
　筑波大学 数理物質系 准教授
　計算モデルの設定、シミュレーションデータ解析

喜多英治（きた　えいじ）
　筑波大学 数理物質系　教授
　計算モデルの設定、シミュレーションデータ解析

時井真紀（ときい　まき）
　筑波大学 図書館情報メディア系　講師
　新規アルゴリズム開発、シミュレーション結果の３次元可視化

李昇珍（Sun-jin Lee）
　筑波大学 数理物質系　研究員
　計算モデルの設定、シミュレーションデータ解析

②当該期間の実施報告の詳細

Inter-grain Interaction in Random Magnetic Anisotropy Simulation in Magnetic Nanocrystals

S.-J. Lee,[1] Nobuhito Inami,[2] Hideto Yanagihara,[1] Eiji Kita,[1] Chiharu Mitsumata,[3] and Kanta Ono[2]
[1)] *Institute of Applied Physics, University of Tsukuba, Ibaraki 305-8573, JAPAN*
[2)] *High Energy Accelerator Research Organization (KEK), Tsukuba, Ibaraki 305-0801, JAPAN*
[3)] *National Institute for Materials Science, Tsukuba, Ibaraki 305-0047, JAPAN*

(Dated: 7 November 2014)

Effect of inter-grain exchange interaction on the coercive forces was analyzed with a numerical simulation in magnetic materials with random magnetic anisotropy. The magnetization of an assembly of magnetically-interacting grains with randomly-oriented uniaxial anisotropy was calculated using the Landau-Lifshitz-Gilbert equation. We supposed a single spin model where the magnetizations in a grain were aligned in the same direction, for simplicity. Calculations were carried out for an $N \times N \times N$ system, where the number of grains on a side, N ranged from 16 to 128. The relation between the coercive forces $H_{\rm C}$ and the grain size D is represented by $H_{\rm C} \propto D^k$. With the increase of N, k decreased gradually and tended to reach a saturated value around $k = 4.5 \sim 5$, which dose not correspond to the primitive theory of the RAM where $k = 6$. The deviation was discussed in terms of the inter-grain interaction, essentially proportional to the inverse of D.

I. INTRODUCTION

Randomness in magnetic materials has been payed attention from the begining of the development of amorphous materials.[1,2] Magnetic nanocrystals, in which randomly-oriented, nano-sized and ferromagnetic crystalline grains are densely gathered, exibit excellent soft magnetic properties and have potential for use in practical application.[3,4] In order to elusidate this soft magnetism, random magnetic anisotropy has been studied from both fundamental and practical points of view.[5]

Herzer[6] has proposed a theory where the magnetic anisotropy energy is averaged out in a magnetically-coherent area coupled with exchange interactions. As a result, the relation between the grain size D and the coercive force $H_{\rm C}$ is predicted to be $H_{\rm C} \sim D^6$. This is called the random anisotropy model (RAM) and it is the most popular model for interpreting soft magnetic characteristics. Many experimental reports support this simple theory not only for single-phase materials but also for multi-phase and granular substances. We have reported on the coercive force of pure Ni nanocrystals and confirmed the D^6 dependence in the particle diameter range of 8-13 nm.[7]

In spite of a convincing argument in the RAM theory, some studies of nanocrystalline materials for practical applications showed different dependence of coercive forces from the D^6. The shape of the sample and the distribution of the stress were considered to affect the dependency of the coercive force[8,9]. Additionally, a few magnetic systems do not show the D^6 relationship, but rather a D^3 dependence, because of the increasing effects of defects and stresses with decreasing grain sizes[9–11]. Therefore, the relationship between D^6 and $H_{\rm C}$ within the framework of the RAM is worth reconsidering with an analysis that includes the exchange interaction.

We have performed the numerical analysis of RAM with a formalization of the exchange magnetic field that operates between particles by using the Landau - Lifshitz - Gilbert (LLG)equation.[12,13] The coercive forces were obtained from a simulation with calculation cells of $N = 10$, namely $10 \times 10 \times 10$, and the D^6 dependence was confirmed. For making the simulation accurate, it is natural to increase the number of calculation size, N. With an increase of N, k, the index in the D^k, determined by the averaged slope in the $\log H_{\rm C} - \log D$ curve decreased and deviated from 6 for $N = 25$, while the peak value of derivatives of the curve show less deviation from 6. It was suggested that the simple model proposed by Herzer still works in the limited range of D.[13]

In this study, we extend the simulation to examine the inter-grain interaction in a random magnetic anisotropy system. The larger scale simulation than previous works was carried out. To obtain the D^6 dependence as one from the fundamental theory, correction in the inter-grain interaction is numerically examined by adjusting the inter-grain interaction.

II. SIMULATION MODEL

The model of magnetic nano-grains with the exchange interaction at their interface, where the grains align on a simple cubic lattice, is shown in Fig. 1. One particular i-th grain has a cubic shape surrounding by six adjacent cubic grains. The cube is assumed to have $N_{\rm S}$ atoms on a side and a lattice constant of a, therefore total number of the atoms in the grain $N_{\rm T}$ and the grain size D is expressed as $N_{\rm T} = N_{\rm S}^3$ and $D = N_{\rm S} a$. The exchange energy of the i-th grain, $E_{\rm ex}^{\rm I}$, is expressed as follows:

$$E_{\rm ex}^{\rm I} = -\sum_{\langle \rm spin \rangle} J(\mathbf{S_i} \cdot \mathbf{S_{k \in \{i, j_1, \cdots, j_n\}}}) = E_{\rm gr} + E_{\rm int}. \quad (1)$$

Here, j, $E_{\rm gr}$ and $E_{\rm int}$ denote the grain adjacent to the i-th grain, the energy in the i-th grain and the interac-

tion energy between adjacent grains, respectively. The summation in Eq. 1 is carried out for n = 6 from j_1 to j_6 .

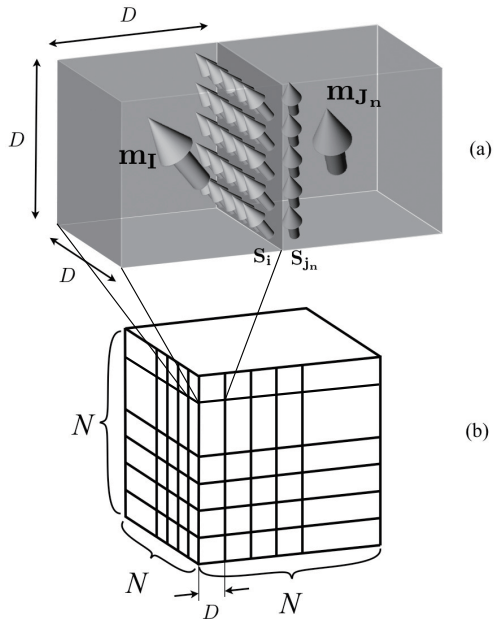

FIG. 1. Model of the random anisotropy nanomagnet system. Spins at the interface between two magnetic grains i and $j_{\rm n}$ with exchange interaction and the calculation size N are schematically illustrated.

In a case the magnetic spins in the cubic grains are aligned in the same direction and have the same amplitude (single spin model), then the magnetization of i-th grain, $\mathbf{m_i}$, is expressed by $\mathbf{m_i} = \sum \mathbf{S} = N_{\rm T}\mathbf{S}$. The interaction energy in Eq. 1 is expressed simply as

$$E_{\rm int(i,j_n)} = -\sum_{\mathbf{k}\in\{\mathbf{j_n}\}} J(\mathbf{S_i}\cdot\mathbf{S_k}) = -JN_s^2(\mathbf{S_i}\cdot\mathbf{S_{j_n}}). \quad (2)$$

Rewriting the terms in Eq. 1, the following equations are obtained:

$$E_{\rm int} = \sum_{n=1}^{6} E_{\rm int(i,j_n)} = -\sum_{n=1}^{6} JN_s^2(\mathbf{S_i}\cdot\mathbf{S_{j_n}}). \quad (3)$$

The effective field at the i-th grain, $\mathbf{H}^{\rm I}_{\rm eff}$, that results from the interaction between grains can be represented by the relation $\mathbf{H}^{\rm I}_{\rm eff} = -\partial E^{\rm I}_{\rm ex}/\partial\mathbf{m_i}$. Because the derivative of the first term in Eq. 1 should be 0, the effective field is derived as follows:

$$\mathbf{H}^{\rm I}_{\rm eff} = -\frac{\partial}{g\mu_B\partial\sum_i \mathbf{S_i}} \sum_{n=1}^{6} -JN_{\rm s}^2(\mathbf{S_i}\cdot\mathbf{S_{j_n}}) \quad (4)$$

$$= \frac{J}{N_s g\mu_B}\sum_{n=1}^{6}\mathbf{S_{j_n}}. \quad (5)$$

Finally the effective field is proportional to the inverse of the grain size.[12]

$$\mathbf{H}^{\rm I}_{\rm eff} = \frac{Ja}{Dg\mu_B}\sum_{n=1}^{6}\mathbf{S_{j_n}}. \quad (6)$$

The motion of the i-th grain's magnetization under an effective magnetic field $\mathbf{H}_{\rm eff} = -\partial\mathcal{H}_{\rm i}/\partial\mathbf{m}_{\rm i}$ can be represented using the Landau–Lifshitz–Gilbert equation

$$\frac{d\mathbf{m}_{\rm i}}{dt} = -\gamma[\mathbf{m}_{\rm i}\times\mathbf{H}_{\rm eff}] - \frac{\alpha}{m}\big[\mathbf{m}_{\rm i}\times[\mathbf{m}_{\rm i}\times\mathbf{H}_{\rm eff}]\big]. \quad (7)$$

Here, $\mathcal{H}_{\rm i}$ is a sum of the exchange energy, anisotropy energy, and Zeeman energy.

The parameters were assumed as those used in previous works[12,13]: an atomic magnetic moment of $g\mu_B S = 5.56\times10^{-21}$ emu, a distance between atoms of $a = 3.4\times10^{-8}$ cm, an exchange energy between interface atoms of $J = 0.1\times10^{-14}$ erg ($J = 1.6\times10^{-14}$ erg for bulk Ni), and a magnetic anisotropy constant of $K = 4\times10^4$ erg/cm^3.[12] These conditions were set to correspond to the case of Ni nanocrystals, and a uniaxial magnetic anisotropy is assumed. A periodic boundary condition was applied to eliminate the edge effect.

The first step of simulation is to generate an orientation distribution of the magnetic anisotropy in $N^3(N\times N\times N)$ cells (see Fig.1). To establish the random orientation, we created pairs of random numbers by using a conventional large-scale random-number generator algorithm[14]. The direction of the principal axis was specified with polar coordinates θ and ϕ. The probability on the coordinate system was weighted by $\cos^{-1}\theta$ to obtain an equal distribution on the polar coordinate system. The simulation cell size N was varied from 16 to 128. The motion equation based on Eq. (7) was numerically solved[12] by using a Hitachi SR16000/M1 supercomputer installed in KEK.[15] By changing the amplitude of the external magnetic field, we reproduced the hysteresis loops, and we determined the coercive force of the system.

III. RESULTS AND DISCUSSION

Grain size dependence of the coercive force were examined for N = 16 - 128. Each plot in Fig.2 corresponds to its respective abscissa. The results of the linear fits are displayed by the solid lines, and the corresponding number k is given near each plot.

For one particular calculation size of N, k was deduced according to the range of interest; for example, k = 4.94, 4.65 and 4.28 for the fitting range of $H_{\rm C}$ = 3-30 Oe, 2-30 Oe and 1-30 Oe, respectively. These results are summarized in Fig.3.

To conduct a detailed analysis we have used the differential amplitude against the grain size for the estimation of k.[13] The same way of smoothing the data and

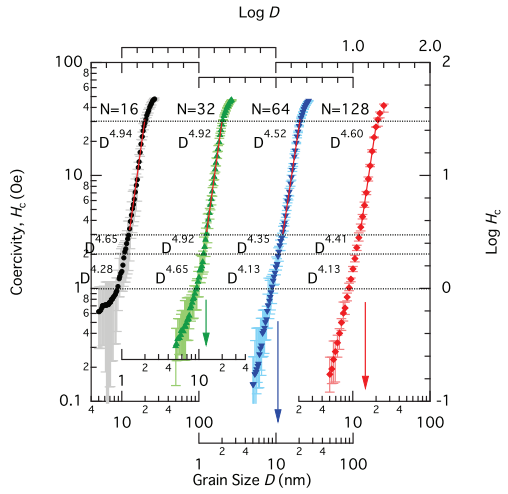

FIG. 2. Grain size dependence of the coercive force for N = 16 - 128. Each plot corresponds to its respective abscissa. The results of the linear fits are displayed by the solid lines, and the corresponding number k is written beside each plot.

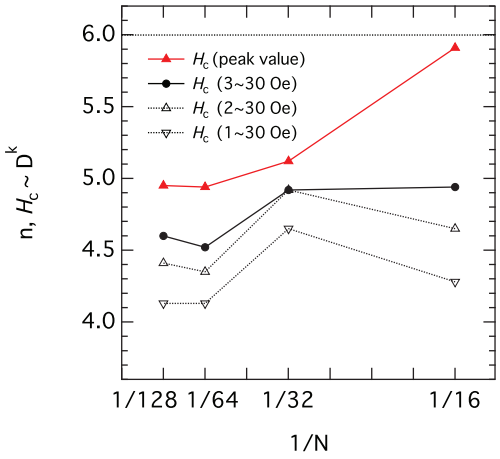

FIG. 3. Dependence of k on the inverse of the simulation size N. The indices k in D^k obtained from the different range of interest in coercive forces are plotted together with the peak values of gradient seen in Fig. 4(a).

calculation of $d\ \log H_C/d\ \log D$ as used in the previous report[13] was adopted. The result of k is depicted in Fig. 4(a) after smoothing the raw data plotted in Fig. 4(b). For each calculation size N, the derivative curve shows a peak at around D = 16 nm(Fig. 4(a)). The k, namely $d\ \log H_C/d\ \log D$, curves for $N = 64$ and 128 have almost identical feature over the whole range of D. The peak values are also plotted against $1/N$ in Fig. 3 and the k values derived from the linear fit for 3-30 Oe and from peak values of the curves tends to saturate to be 4.5 and 5, respectively, as N become larger. These facts indicate that the k saturates for the calculation size larger than 64.

It is noted that scattering of data at the small grain size region is getting thinner as the N increases as seen

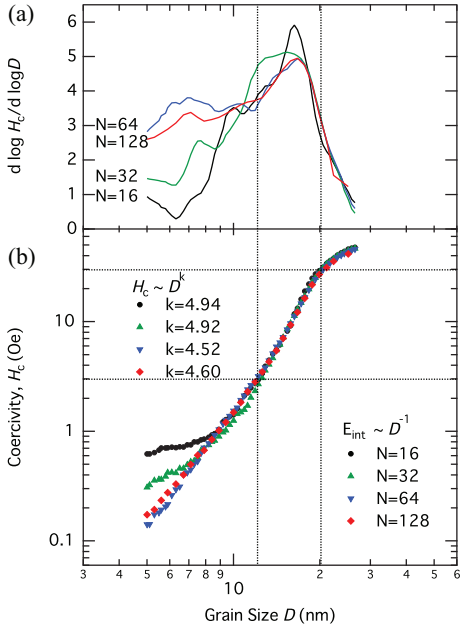

FIG. 4. (a) Index k in D^k evaluated from the derivatives of the data shown in (b) and plotted against grain size. (b) Coercive force before smoothing plotted against grain sizes for $N = 16 - 128$.

in the Fig. 2. It is considered to be due to the range of exchange covers wider region at smaller grain size.

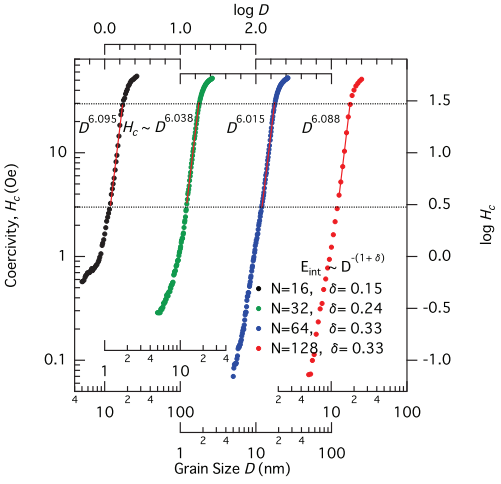

FIG. 5. Grain size dependence of the coercive force for N = 16 - 128 with correction factor δ to fit to the D^6 dependence. In the case of N = 128, the result fit to k = 6.1 is plotted. (See text.)

It is thought that the shortage of a calculation size comparing with an exchange length of a system affects the artificial dependence of k on N. Clear dependence of k on the N obtained from the linear fits and differentiation was seen in Fig. 3. The k tends to saturate at a large value of N, therefore the smaller peak value around 5 than 6 is an intrinsic amplitude if we adopt the single

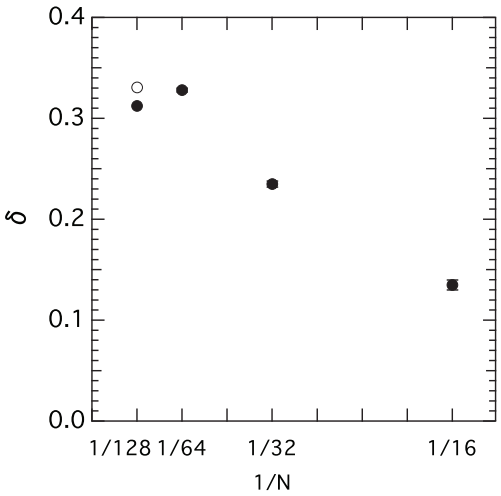

FIG. 6. Correction factor δ plotted against the inverse of the simulation size, $1/N$. The open and solid circles for $N = 128$ depict a value resulted from a simulation fit for $k = 6.1$ and an extrapolated value for $k = 6$, respectively. (See text.)

spin model to the random anisotropy system.

In the present RAM simulation, the power low $H_C \sim D^6$ is affected by the inter-grain interaction and k may change according to the condition of the interaction. We assumed that the total interaction between two adjacent grains is proportional to the number of spin pairs facing at the interface. This assumption leads to us that the interaction energy is proportional to D^{-1} seen in eq.(6). The deviation of k from 6 might be originated from the properties of interaction. To understand it phenomenologically, we determined the correction factor δ defined the interaction field by $\mathbf{H}^{\mathrm{I}}_{\mathrm{eff}} \sim D^{-(1+\delta)}$. Calculations were carried out to obtain D^6 dependence by adjusting δ and the results of $H_C - D^6$ relation are shown in Fig. 5. The obtained δ are plotted in Fig.6. Due to the limited calculation time for $N = 128$, the number of simulation was not enough to determine δ precisely. Therefore, the result of simulation fit to $k = 6.1$, closest to 6, was plotted in Fig. 5 and $\delta = 0.33$ was plotted by a open circle in Fig. 6. The extrapolated value for $k = 6$ is obtained to be $\delta = 0.31$ and is plotted by a solid circle in Fig. 6. Corresponding to the deviation of k from 6, δ become large and looks saturate when N increases.

The power low $H_C \sim D^6$ expressed the ideal averaging of magnetic anisotropy on all over the nano-sized grains, because RAM corresponded to the large limit of exchange coupling between grains. Therefore, correction factor δ simply realizes the obtained power low, $H_C \sim D^{4.5}$ on this single spin model to be $H_C \sim D^6$, and this δ does not guarantee the reproduction of realistic measurement.[7] In the realistic case, the exchange constant at the interface of nano-sized grains should be finite value, thus the magnetization in each grain possibly deviates from the averaged direction even under the influence of exchange coupling. The deviation of magnetization forms the twisted structure of the spin arrangement near the interface of grains.

Within a framework of the single spin model, the spin simply represents the direction of magnetization in a grain. Thus, it cannot express the twisted structure of the spin arrangement near the interface. The exchange energy near the interface is renormalized into the effective exchange field at the interface. These facts potentially cause the under estimation of an exchange energy. In other words, some sort of correction factor δ is required to reproduce the power low of realistic measurement.

ACKNOWLEDGMENTS

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