Magnetic Anisotropy and Behaviors of Fe Nanoparticles

Takahiro Ibusuki, Seijyu Kojima, Osamu Kitakami, and Yutaka Shimada

***Abstract*—We have studied the magnetic properties of bcc-Fe fine particles with a diameter D of $3 \sim 40$ nm prepared by a vapor condensation process. The particles exhibit uniaxial anisotropy of $2.0 \sim 3.0 \times 10^6$ erg/cc which is much larger than that of bulk Fe (0.5×10^6 erg/cc). Numerical calculations taking into account dipolar interaction and thermal agitation reproduce the particle size dependence of coercivity very well. According to the present work, magnetization reversal of Fe particles proceeds via coherent rotation for $D < 15 \sim 20$ nm and via incoherent rotation for $D > 15 \sim 20$ nm. It is, however, still uncertain why the magnetic anisotropy of Fe fine particles is uniaxial and so large.**

***Index Terms*—Coercivity, dipolar interaction, Fe, fine particle, magnetic anisotropy.**

I. Introduction

FINE magnetic particles show peculiar behaviors quite different from the bulk ones. For instance, ferri- and antiferro-magnetic particles exhibit anomalous magnetic properties, such as large net moments and coercivity due to a finite size effect [1], [2]. As for metallic ferromagnetic particles, they show very large coercivity compared with the value predicted by the simple Stoner–Wohlfarth model under the assumption of bulk anisotropy [3]–[6], [9]. Moreover, it is recently found that bcc-Fe and fcc-Co nanoparticles exhibit strong uniaxial effective anisotropy in spite of their cubic crystal symmetry [7]–[9]. Surface anisotropy due to broken symmetry or spin disorders at the particle surfaces is believed to play a dominant role in such fine particle systems, although the details are still unknown. In the present work, we discuss about magnetization reversal process of bcc-Fe fine particles based on the measured magnetic anisotropy constants.

II. Experimental

Fe fine particles were prepared at ambient temperature by dc-magnetron sputtering, with high Ar gas pressure P_{Ar} of $0.1 \sim 0.5$ Torr [10]. No heat treatment was performed. The samples were successively covered by a 200 nm-thick Ag or SiO_2 overcoat in order to avoid surface oxidation. The particle diameter D was varied in the range of $3 \sim 40$ nm by controlling P_{Ar}. In addition, in order to study dipolar interaction, we have also prepared 10 period bilayer stackings consisting of a monodisperse Fe particles layer and a 200 nm thick-Ag or SiO_2 layer. All the samples showed bcc-Fe 110 diffraction without any distinct diffraction peaks from Fe oxides. Particle diameter was mainly evaluated from FWHM (full width at half maximum) of Fe 110, the values almost agreed with TEM observations. Fig. 1 shows a typical size distribution of Fe particles. The distribution almost follows the log-normal distribution function [11]. The magnetic properties were measured using a VSM in the temperature range of $10 \sim 300$ K. Since Fe particles are 3D randomly oriented, we adopted the law of approach to saturation (LAS) for determination of effective magnetic anisotropy [12]. For a 3D random assembly of uniaxial and cubic anisotropy particles, magnetic susceptibility can be easily derived as

$$\chi = \partial M/\partial H \cong \alpha K_{\mathrm{eff}}^2/M_s H^3, \tag{1}$$

where $\alpha = 0.533$ for uniaxial anisotropy and 0.152 for cubic anisotropy. Fig. 2(a) shows a typical $\chi - 1/H^3$ plot for Fe particles. We can evaluate K_{eff} from the slope using (1). K_{eff} was measured for various external fields H_m, as shown in Fig. 2(b). Note that accurate evaluation of K_{eff} is achieved by LAS with the error of several percent.

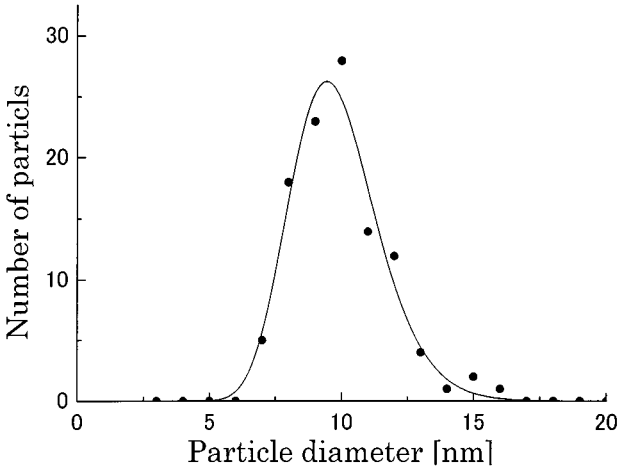

Fig. 1. Number of particles counted per size interval vs. diameter for Fe particles. The dots represent midinterval values obtained from the size histogram. The curve shows the frequency distribution calculated from the log-normal distribution function.

III. Results and Discussion

According to the Stoner–Wohlfarth (S–W) model, H_c and M_r/M_s for an assembly of noninteracting 3D random particles are given by

$$H_c = 0.958 K_1/M_s, \qquad M_r/M_s = 0.5 \quad \text{for uniaxial anisotropy} \tag{2}$$

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The authors are with the Research Institute for Scientific Measurements, Tohoku University, 980-8577 Sendai, Japan (e-mail: ibusuki@rism.tohoku.ac.jp).

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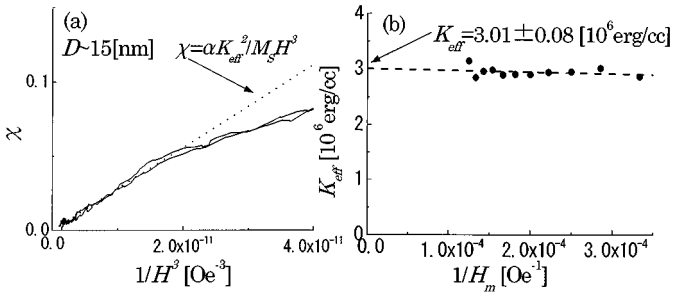

Fig. 2. (a) χ vs. $1/H^3$ and (b) $K_{\rm eff}$ vs. $1/H_m$ plots for Fe particles with the diameter of 15 nm. The dashed line in (b) is a guide to the eye.

and

$$H_c = 0.64K_1/M_s, \qquad M_r/M_s = 0.832, \quad \text{for cubic anisotropy } (K_1 > 0) \tag{3}$$

if thermal agitation is entirely ignored. A very simple calculation gives $H_c = 180$ Oe and $M_r/M_s = 0.83$ for no-interacting bcc-Fe single-domain particles with cubic anisotropy of $K_1 = 5 \times 10^5$ erg/cc. In general, however, this simple theory does not explain magnetic properties of real fine particle systems even when particle size is in the single-domain size region [3]–[6], [9].

Fig. 3(a)–(c), respectively, show the particle size dependence of $K_{\rm eff}$, M_r/M_s, and H_c at room temperature. Note that M_r/M_s is almost equal to that for uniaxial anisotropy [(2)], which is much smaller than that for cubic anisotropy [(3)]. This result suggests that magnetic anisotropy of Fe fine particles is uniaxial in its nature. Since M_r/M_s may be also affected by dipolar interaction and thermal agitation, we have investigated M_r/M_s of a nearly noninteracting system ($p \sim 0.05$) consisting of a [monodisperse bcc-Fe particle layer/SiO_2(200 nm)]$_{10}$ multilayer, and found that M_r/M_s became equal to 0.5 at $T = 10$ K. Therefore, we believe that Fe fine particles possess uniaxial anisotropy in spite of their cubic crystal symmetry, as has been discussed in previous works [5], [7]–[9]. Moreover, their effective anisotropy is much larger than that of bulk Fe, as can be noticed in Fig. 3(a). Several groups have also found very strong uniaxal anisotropy in magnetic fine particles and speculated that surface anisotropy enhances the effective anisotropy of fine particles, although the details are still unknown [7]–[9]. Using the values of $K_{\rm eff}$ in Fig. 3(a), we can roughly calculate H_c for a 3D random assembly of bcc-Fe particles. At first, we evaluate dipolar interaction effect on H_c by numerical computations. The system we use for the calculation is a simple cubic lattice with a lattice constant equal to particle size. Fe particles with effective uniaxial anisotropy $K_{\rm eff}$ occupy the lattice points in a random manner with the probability equal to the packing density p. $32 \times 32 \times 5$ lattice points was enough for good convergence. The calculations revealed that H_c $(p = 0.5)$ is about 40% smaller than that for a noninteracting system $(p = 0)$. Since the above calculation entirely ignores thermal agitation, we have to take into account thermal agitation at the next step. For a 3D random assembly of uniaxial single-domain

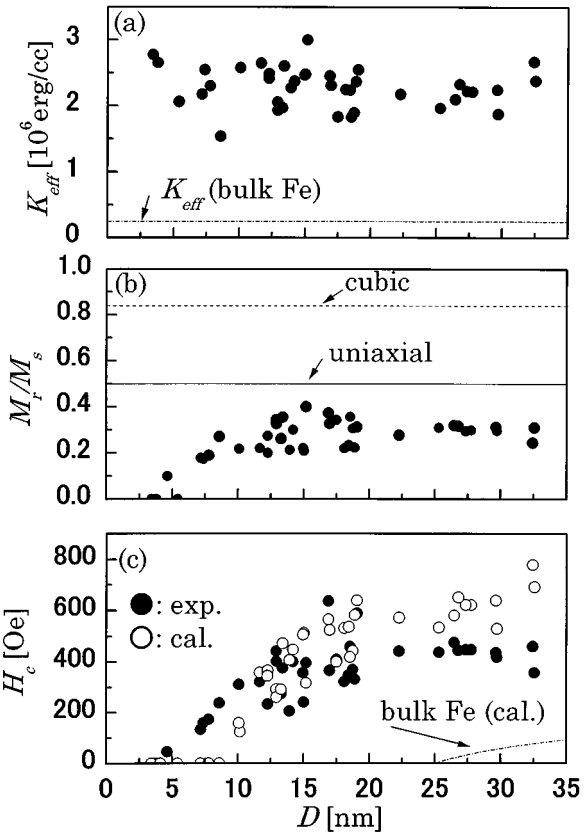

Fig. 3. Room temperature magnetic properties of 3D random bcc-Fe particles as functions of particle size D. (packing density $p \sim 0.5$): (a) effective anisotropy $K_{\rm eff}$, (b) remanence ratio M_r/M_s, and (c) coercivity H_c. Solid and empty circles in (c) indicate measured and calculated H_c. Dotted and solid lines in (b) show remanence ratios for 3D random noninteracting cubic and uniaxial particles, respectively. The dotted line in (c) is the coercivity calculated using the anisotropy constant of bulk-Fe (5×10^5 erg/cc).

particles, Pfeiffer derived the following approximate expression for coercivity based on the S–W model [13];

$$H_c(V, p, T) = H_c(V, p, 0)\left\{1 - \left(\frac{V_s}{V}\right)^{0.77}\right\}, \tag{4}$$

where V is the particle volume and V_s is the critical volume for a superparamagnetic limit. Setting an attempt frequency $f_0 \sim 10^9$ Hz and a relaxation time $\tau_0 \sim 100$ sec., $V_s = 25k_BT/K_{\rm eff}$, here k_B is the Boltzmann constant. Using (4), we can roughly calculate H_c at a finite temperature T. Strictly speaking, V should be effective volume enhanced by dipolar interaction, but we used real particle volume for the present calculation because of difficulty in evaluation of the effective volume. Therefore, the effect of thermal agitation may be somewhat overestimated. The calculated results are indicated by open circles in Fig. 3(c). Note that the calculated coercivity reproduces the experiments (closed circles) very well for $D < 15 \sim 20$ nm, indicating that magnetization reversals take place via simple coherent rotation. For larger particle size of $D > 15 \sim 20$ nm, H_c is somewhat smaller than calculated values. This difference may be due to the change in magnetization reversal process. From micromagnetics calculations, we found that the reversal mode for an Fe particle changes from coherent to incoherent around at $D \sim 20$ nm.

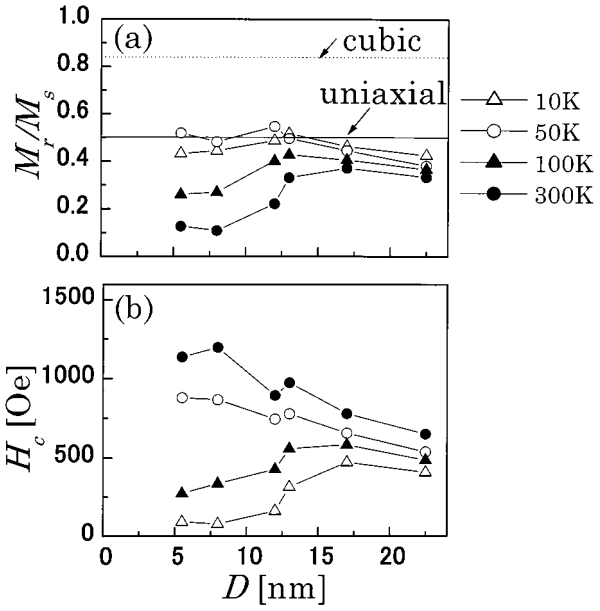

Fig. 4. M_r/M_s and H_c vs. D for 3D random bcc-Fe particles (packing density $p \sim 0.5$) as a parameter of temperatures T. Dotted and solid straight lines in (a) show remanence ratios for 3D random noninteracting cubic and uniaxial anisotropy particles, respectively.

Fig. 4(a) and (b) shows M_r/M_s and H_c vs D at various temperatures. For the particles smaller than $D = 15 \sim 20$ nm, M_r/M_s and H_c rapidly increase with decreasing the temperature due to suppression of thermal agitation. Here we note M_r/M_s is 0.5 at $T = 10$ K, indicating that Fe particles possess uniaxial magnetic anisotropy. With the increase of particle size above $15 \sim 20$ nm, thermal agitation is entirely suppressed, resulting in no appreciable changes in M_r/M_s and H_c. Note that both M_r/M_s and H_c decrease with the particle size, probably due to the change in magnetization reversal mode from coherent to incoherent rotation.

IV. Conclusion

We have studied the magnetic properties of bcc-Fe particles with a diameter D of $3 \sim 40$ nm prepared by a vapor condensation method. Magnetic measurements showed that Fe particles exhibit very strong effective uniaxial anisotropy of $2 \sim 3 \times 10^6$ erg/cc in spite of its cubic crystal symmetry. Taking into account both dipolar interaction and thermal agitation, we have calculated particle size dependence of the coercivity using the measured anisotropy constants within the framework of the coherent rotation model. The calculation reproduced the experiments very well for $D < 15 \sim 20$ nm, indicating that magnetization reversals take place via coherent rotation for this small size region. For larger particle size of $D > 15 \sim 20$ nm, an incoherent rotation mode lower H_c than that expected from the coherent rotation mode.

References

[1] J. M. D. Coey, "Noncollinear spin arrangement in ultrafine ferrimagnetic crystallites," *Phys. Rev. Lett.*, vol. 27, p. 1140, 1971.
[2] See for example, R. H. Kodama, "Magnetic nanoparticles," *J. Magn. Magn. Mater.*, vol. 200, p. 359, 1999.
[3] E. F. Kneller and F. E. Luborsky, "Particle size dependence of coercivity and remanence of single-domain particles," *J. Appl. Phys.*, vol. 34, p. 656, 1963.
[4] W. H. Meiklejohn, "Experimental study of the coercive force of fine particles," *Rev. Mod. Phys.*, vol. 25, p. 302, 1953.
[5] S. Gangopadhyay, G. C. Hadjipanayis, B. Dale, C. M. Sonensen, K. Jkabunde, V. Papaefthymious, and A. Kastikas, "Magnetic properties of ultrafine iron particles," *Phys. Rev. B*, vol. 45, p. 9778, 1992.
[6] G. Xiao and C. L. Chen, "Giant magnetic coercivity and percolation effects in granular Fe–SiO_2 solids," *Appl. Phys. Lett.*, vol. 51, p. 1280, 1987.
[7] F. Bødker, S. Mørup, and S. Linderoth, "Surface effects in metallic iron nanoparticles," *Phys. Rev. Lett.*, vol. 72, p. 282, 1994.
[8] M. Respaud, J. M. Broto, H. Rakato, A. R. Fert, L. Thomas, B. Barbara, M. Verelst, E. Snoeck, P. Lecante, A. Mosset, J. Osuna, T. Ould Ely, C. Amiens, and B. Chaudret, "Surface effects on the magnetic properties of ultrafine cobalt particles," *Phys. Rev. B*, vol. 57, p. 2925, 1998.
[9] C. Chen, O. Kitakami, and Y. Shimada, "Particle size effects and surface anisotropy in Fe-based granular films," *J. Appl. Phys.*, vol. 84, p. 2174, 1998.
[10] O. Kitakami, H. Sato, Y. Shimada, F. Sato, and M. Tanaka, "Size effect on the crystal phase of cobalt fine particles," *Phys. Rev. B*, vol. 56, p. 13 849, 1997.
[11] C. G. Granqvist and R. A. Buhrman, "Ultrafine metal particles," *J. Appl. Phys.*, vol. 47, p. 2200, 1976.
[12] S. Chikazumi, *Physics of Ferromagnetism*. Oxford: Clarendon Press, 1997.
[13] H. Pfeiffer, "Determination of anisotropy field distribution in particle assemblies taking into account thermal fluctuations," *Phys. Stat. Sol. (a)*, vol. 118, p. 295, 1990.