Effect of particle size on the magnetization process in lithographic arrays of Nd2Fe14B

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Effect of particle size on the magnetization process in lithographic arrays of $Nd_2Fe_{14}B$

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Controlled arrays of $Nd_2Fe_{14}B$ were fabricated via sputtering and lithography. The arrays were fabricated via optical and electron-beam lithography combined with dry etching technique. Designed particle size varies from 10 μm×10 μm to 0.5 μm×0.5 μm. We found an increase in the coercive field with decreasing particle size. Results of the micromagnetic calculations suggest that the coercive-field increase in smaller-sized particles is originated from the effective decrease in the exchange-coupling energy owing to the relative increase in the fraction of surface grain. © *2000 American Institute of Physics.* [S0021-8979(00)74608-1]

I. INTRODUCTION

There has been much interest in artificially engineering the magnetic properties of the thin magnetic films by lithographic structuring.[1–3] The ability to design and control magnetic properties is very useful not only from a technological point of view, but also opens up the possibility of experimentally studying fundamental magnetic properties of the material. The experimental study of the magnetic properties of microfabricated magnets are focused mainly on soft magnetic materials. Smyth *et al.*[1,2] have reported that, in permalloy particles, the coercive field H_c increase with decreasing particle size. They explained the results by taking account of the change in the switching mechanism.[2] As for the hard magnets, only one report[4] exists as far as we know, although the magnetization process is expected to change more significantly than in soft magnets. Since the characteristic length scale such as critical size of single-domain particle is larger than that in soft magnets and reaches the submicron range, we can observe the effect of the particle size on their magnetic properties more easily in hard magnets. In the present work, controlled arrays of $Nd_2Fe_{14}B$ were fabricated via sputtering and lithography. The effect of particle size on the magnetic properties has been examined by magnetization measurements and micromagnetic calculations.

II. EXPERIMENT

The films of Ti(50 nm)/$Nd_2Fe_{14}B$ (500 nm)/Ti(50 nm) were synthesized by rf magnetron sputtering on the thermally oxidized Si substrates at an Ar pressure of 0.8 Pa. They were then annealed at 923 K for 30 min under a pressure of 6×10^{-4} Pa. Designed particle size varies from 10 μm×10 μm to 0.5 μm×0.5 μm with the constant spacing of 10 μm. Optical lithography was used for over 1 μm particles, which was combined with the Ar ion milling technique. Electron-beam lithography was utilized for smaller particles, which was combined with the reactive ion etching (RIE) with CF_4+H_2 mixture and the Ar ion milling technique. We used the SiO_2 film as a hard mask, because the selectivity of the negative resist for electron-beam lithography to Ti and $Nd_2Fe_{14}B$ films is very low for the Ar ion milling. The RIE was used to remove the SiO_2 layer (1.2 μm) of the trilayer. Since the size of the array was 5 mm×5 mm, the total number of particles varied from 5×10^4 to 2×10^5 depending on the particle size. The phase structure was checked by x-ray diffraction (XRD) measurements with Cu$K\alpha$ radiation. The artificial periodic structures formed were observed by SEM. Magnetization curves were measured by a SQUID magnetometer at room temperature in fields of up to 50 kOe which was applied parallel to the array surface.

III. RESULTS AND DISCUSSION

By means of the XRD measurements, the films were confirmed to be randomly oriented polycrystals of $Nd_2Fe_{14}B$ and the averaged grain size to be about 15 nm in diameter, which are consistent with our previous report.[5] Figure 1

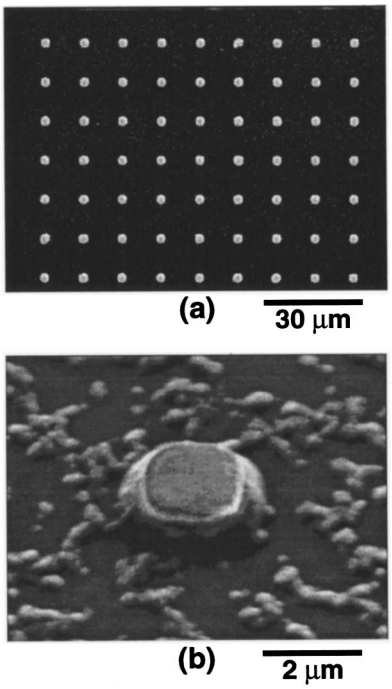

FIG. 1. SEM image of a particle array with the particle size of 2×2 μm^2.

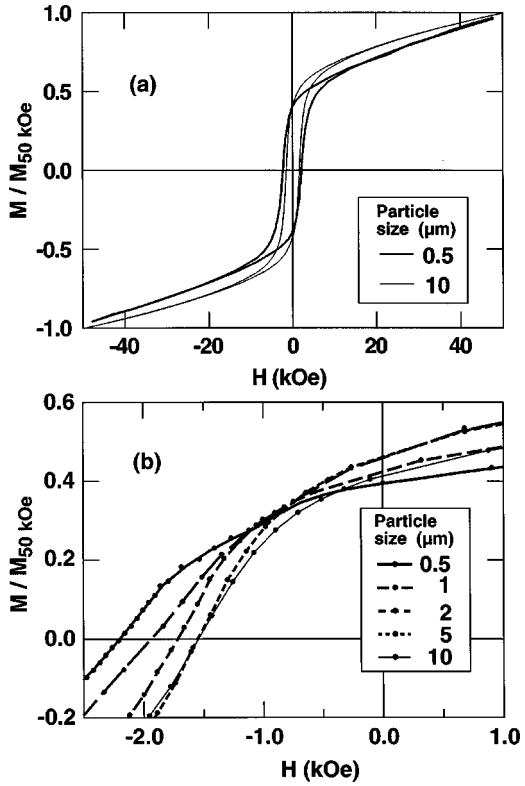

FIG. 2. Magnetization curves of the particle arrays with different particle size. (a) Full loops and (b) demagnetization curves.

shows the SEM image of a particle array with particle size of 2×2 μm^2. The $Nd_2Fe_{14}B$ particles show a clear periodic structure as shown in Fig. 1(a). Expanded image in Fig. 1(b) shows that observed shape of each particle is rounded in the corner, although the designed shape has been square. This rounding was found to be more significant in smaller-sized particles. The surface around the particle is very rough, which is often observed after ion milling process. Since the milling time was enough to remove the bottom Ti layer, the surface of the substrate is partly etched around the particles.

Figure 2(a) shows the magnetization curves of the particle arrays. Magnetization values are normalized by those at 50 kOe. The overall shape of the curves were found to be almost unchanged. The magnetization values do not saturate even at 50 kOe, although those for increasing and decreasing fields coincide for fields higher than about 20 kOe. The magnitude of H_c for the 10 μm particle was confirmed to be almost the same as that of the film before microfabrication. With decreasing particle size, H_c increases systematically from 1.5 kOe (10 $\mu m\times10$ μm) to 2.2 kOe (0.5 $\mu m\times0.5$ μm) as shown in Fig. 2(b).

In order to interpret the size dependence of H_c, we have made a micromagnetic calculation. The calculation is based on the method of LaBonte.[6] A cubic particle with a side dimension L is considered, which consists of cubic cells. Each cell was assumed to represent a $Nd_2Fe_{14}B$ grain with a size of 15 nm. The number of the cell thus depends on L, which, for example, is $100\times100\times100$ for $L=1.5$ μm. The total energy E of the system is assumed to be expressed by

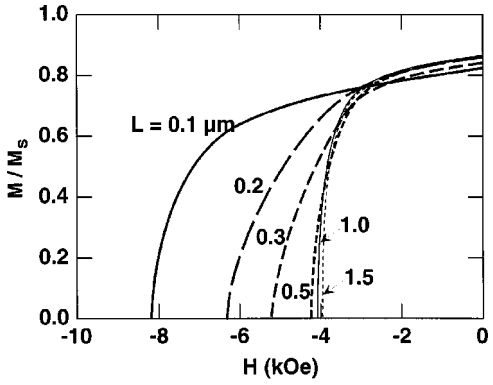

FIG. 3. Calculated magnetization curves for different values of L.

$$E=\sum_{\text{pair}} A[(\nabla m_x)^2+(\nabla m_y)^2+(\nabla m_z)^2] + K_1\sin^2\theta-\mathbf{M}\cdot\mathbf{H}, \quad (1)$$

where the first, second, and third terms denote the exchange energy between neighboring grains, anisotropy energy of each grain, and Zeeman energy, respectively. The magnetization vector $\mathbf{M}=M_s(m_x,m_y,m_z)$ was assumed to be uniform with each cell. The magnitudes of anisotropy constant K_1, saturation magnetization M_s, and exchange stiffness constant A are taken[7,8] to be $K_1=4.3\times10^7$ erg/cm^3, $M_s=1.3\times10^3$ emu/cm^3, and $J_e=16$ erg/cm^2, in which a relation $A=J_e\cdot\delta h$, where δh is the intergrain distance, was used. For computational economy, dipolar interactions are ignored, since they are considered to give unimportant contribution owing to the large anisotropy of the system.[8] We have actually checked the effect of dipolar interactions on the coercivity for several choice of L values. The reduction of H_c by the inclusion of dipolar interactions was found to be less than 5%, which did not depend on H_c.

Calculated magnetization curves for different values of L are shown in Fig. 3. It is seen that remanence value do not depend on L very much, while H_c significantly increases with decreasing L. We have then plotted H_c as a function of L, together with the observed H_c, as shown in Fig. 4. It

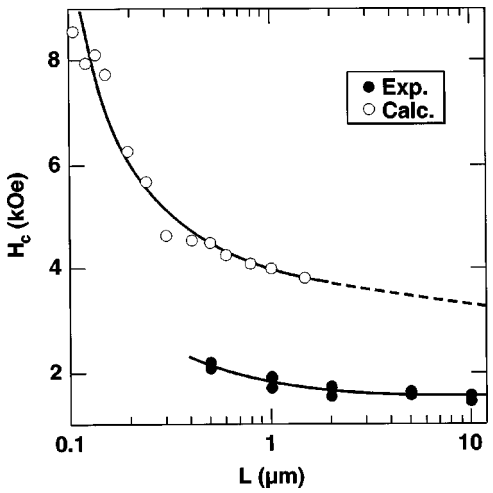

FIG. 4. Experimental and calculated coercive fields H_c as a functions of L.

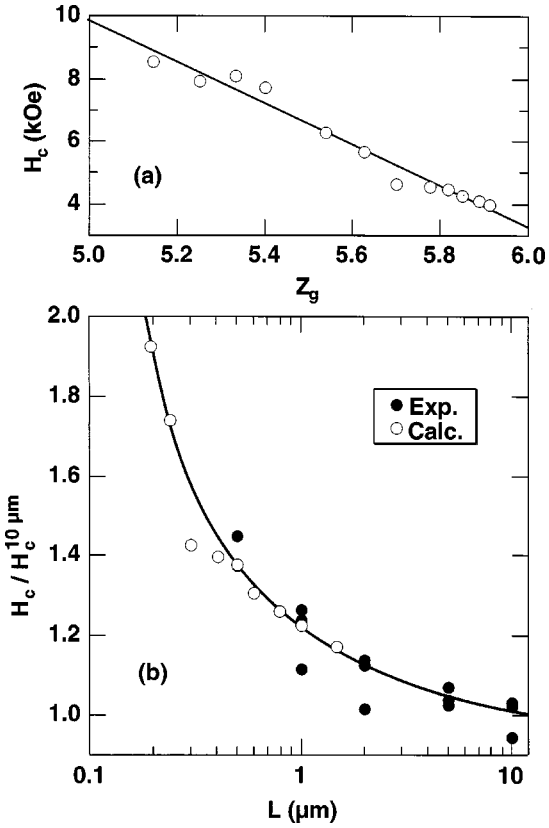

FIG. 5. (a) Calculated H_c as a function of the average number of neighboring grains Z_g. (b) Normalized coercive fields as a function of L.

should be noted that the calculated H_c is systematically larger than the observation. This difference is considered to come from the simplified modeling of present calculation, in which any possibility of incoherent magnetization changes within the grains and domain wall motion is neglected.[9] Although overlapped range of L for calculated and observed H_c is narrow, it appears that L dependence of H_c is similar with each other. In smaller-sized particles, a ratio of surface to volume becomes larger, which would modify the exchange coupling between grains. We thus examined the effect of neighboring grain number on the H_c value. For example, a number of neighboring grains is six for grains inside a particle, while for surface grains it becomes less than six. In general, the average number of neighboring grains Z_g is equal to $6x^2(x-1)/x^3=6-(6/x)$ for each particle with $x \times x \times x$ cubic grains. In Fig. 5(a), calculated H_c values given in Fig. 4 are replotted against Z_g, which are found to give linear dependence with Z_g. By using this plot, the extrapolated value of the calculated H_c for the 10 μm particle was derived as 3.3 kOe. Figure 5(b) shows the L dependence of observed and calculated H_c, both of which are normalized by the 10 μm values. Calculated values appear to connect smoothly with the observation. This result thus suggests that the effective decrease in the exchange coupling enhances the independence of randomly oriented grains, which results in the increase of H_c in smaller-sized particles.

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