Coercivity exceeding 100 kOe in epitaxially grown FePt sputtered films

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Microstructure and magnetization processes of highly ordered FePt (001) films with large perpendicular magnetic anisotropy have been studied. The film morphology was controlled from assemblies of single-domain nanoparticles to those of multidomain islands by varying the nominal thickness (t_N) of the FePt films sputter-deposited on a heated MgO (001) substrate. The change in the magnetization process from magnetization rotation to domain wall displacement is clearly demonstrated by the initial magnetization curves. Huge coercivities as high as 70 and 105 kOe have been achieved in the film with single-domain particles at room temperature and 4.5 K, respectively. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1794863]

The magnetization process of assemblies of ferromagnetic nanoparticles with a large uniaxial magnetocrystalline anisotropy is of great scientific and technological interest, since they are expected to be applied in forthcoming magnetic devices such as high density magnetic recording media and high performance biasing nano-magnets. It is well known that the magnetization process and the coercivity depend strongly on the characteristic size and the morphology of the assemblies of ferromagnetic particles. Recently, a lot of investigations have been made to process FePt films and particles by conventional thin film preparation techniques[1–6] and a chemical synthesis technique,[7] since the $L1_0$ ordered FePt phase possesses a large uniaxial magnetocrystalline anisotropy. However, only a few investigations have realized high coercivity[8–10] together with highly aligned crystal orientation. In our previous work,[9] we reported that high coercivity (H_c) exceeding 40 kOe was achieved in highly ordered FePt (001) films with the island structure that were epitaxially grown on MgO (001) substrates. However, H_c was still only one-third of the value expected from the coherent rotation model[2] ($2K_u/M_s \sim 120$ kOe; K_u: uniaxial magnetic anisotropy, M_s: saturation magnetization). The present study focuses on a further improvement in coercivity by approaching the ideal morphology with the coherent rotation of single-domain particles. High magnetic field measurements were performed to evaluate the huge coercivity obtained from isolated single domain particles, and the magnetization processes are discussed based on the initial magnetization curves.

FePt films were prepared by co-sputtering Fe and Pt targets directly onto single crystalline MgO (001) substrates using a high vacuum (base pressure $\sim 5 \times 10^{-10}$ Torr) multiple dc-sputtering system. High-purity argon of 1.4 m Torr was flown during sputtering. The substrates were heated to 780 °C during the deposition. The nominal thickness of FePt film (t_N) was varied in the range between 1 and 40 nm. The compositions of the films were determined to be $Fe_{52}Pt_{48}$ (at. %) by electron probe x-ray microanalysis (EPMA). The typical growth rate for FePt was 0.12 nm/s. The structural analysis was performed by transmission electron microscopy (TEM) and x-ray diffraction with Cu-Kα radiation. Magnetization curves were measured by a vibrating sample magnetometer (VSM) equipped with a superconducting magnet (maximum magnetic field of ±140 kOe) at temperatures of 4.5 and 295 K.

In-plane TEM bright field images of the FePt films with different t_N deposited on a MgO (001) substrate are shown in Fig. 1. t_N is 1 nm (a), 3 nm (b), 5 nm (c), and 20 nm (d). Note that the scale of each micrograph is different. The films are grown with the island growth mode at $T_s = 780$ °C as reported in our previous work for $T_s = 700$ °C.[9] The particle size of the film with $t_N = 1$ nm is smaller than 5 nm. With increasing t_N, the typical size of particles increases from 10 to 20 nm for $t_N = 3$ nm to 20 to 30 nm for $t_N = 5$ nm. Strongly faceted islands of FePt particles with large size distribution are observed for $t_N = 3$ and 5 nm. With further increase of t_N, a number of small particles coalesced to form big particles. The distribution of the particles with $t_N = 20$ nm is bimodal, consisting of larger ones with a typical size of about 200 nm

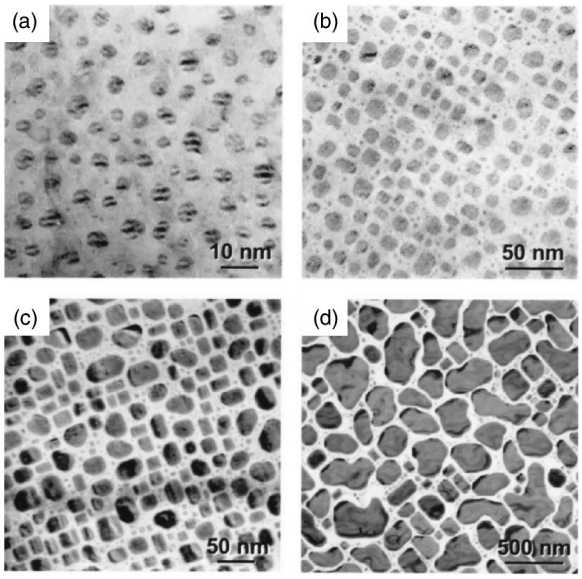

FIG. 1. In-plane TEM bright field images for the FePt films with different thickness: $t_N = 1$ nm (a), 3 nm (b), 5 nm (c), and 20 nm (d).

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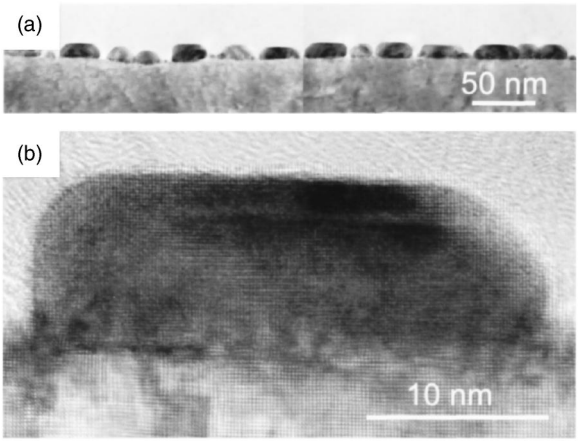

FIG. 2. Cross-sectional TEM images of the FePt film with t_N=5 nm.

in side and smaller ones with a {001} faceted morphology whose size ranges from 10 to 50 nm. In comparison with the microstructure of the films deposited at T_s=700°C,[9] there is little strain contrast in the particles grown at 780°C.

Cross-sectional TEM images of the film with t_N=5 nm are shown in Figs. 2(a) and 2(b). Although the sizes of particles are scattered as shown in Fig. 1, the heights of large particles are almost the same (~15 nm). The enlarged cross-sectional image [Fig. 2(b)] clearly shows the contrast corresponding to the alternating atomic stacking of Fe and Pt layers in the [001] direction of the $L1_0$ structure. Furthermore, no structural defects, such as twins, are observed and the surfaces of the particles are atomically flat. From the integrated intensities of the fundamental and superlattice peaks extracted from the numerical fitting of x-ray diffraction patterns (the figure not shown), the degree of long-range chemical order S was estimated to be 0.95±0.05 for all the samples.

Figure 3 shows magnetization curves for the films with t_N=1 nm (a), 3 nm (b), 5 nm (c), 20 nm (d), and 40 nm (e) measured at 295 K. The solid and broken lines are the magnetization curves with magnetic fields applied in the perpendicular and parallel directions to the film plane, respectively. The magnetic easy axes are perpendicular to the film plane for all the samples. Among the curves, the film with t_N =1 nm [Fig. 3(a)] shows a different nature: low saturation magnetization of less than 500 emu/cc and low H_c of 18 kOe. TEM observation revealed that the sizes of the particles for t_N=1 nm were mostly smaller than 5 nm in diameter. The different behavior of the magnetization curve is thought to arise from the insufficient $L1_0$ ordering, in other words the existence of small particles with poor chemical order. It was recently reported that FePt particles smaller than ~4 nm were not ordered to the $L1_0$ structure.[11] A huge H_c of 64 kOe was observed for the film with t_N=3 nm [Fig. 3(b)] in the perpendicular direction. The particle size of this film was smaller than 20 nm. The step of the perpendicular magnetization curve around zero magnetic field is thought to arise from the mixture of fully $L1_0$ ordered particles and small particles with poor chemical ordering as described before. However, with increasing t_N, the step of the magnetization curve around zero magnetic field disappears and the maximum H_c of 70 kOe is achieved for the film with t_N =5 nm [Fig. 3(c)]. With further increasing t_N, the H_c decreases slowly, but still keeps a quite large value of about

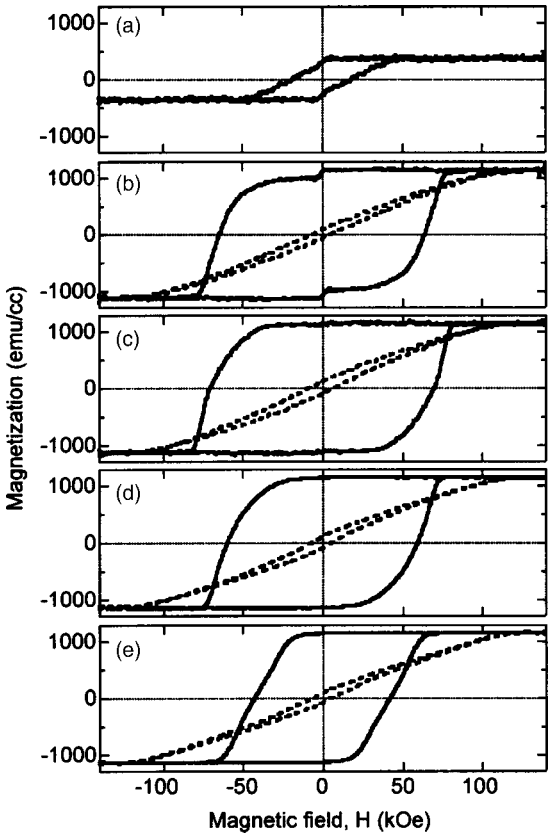

FIG. 3. Magnetization curves at 295 K for the FePt films with t_N=1 nm (a), 3 nm (b), 5 nm (c), 20 nm (d), and 40 nm (e).

43 kOe for t_N=40 nm [Fig. 3(e)]. The large H_c can be achieved until the percolation of the film morphology occurs between t_N=50 and 70 nm. The saturation magnetization for the films with $t_N \geqslant 3$ nm is about 1150 emu/cc and the maximum energy product $(BH)_{max}$ reaches about 50 MGOe, which is almost equal to the ideal value $(2\pi M_s)$.[2] The saturation magnetic fields H_s measured in the in-plane direction (magnetic hard axis) for the films with $t_N \geqslant 3$ nm are about 120 kOe. The uniaxial magnetic anisotropy K_u determined from the area enclosed between the magnetization curves in applied fields parallel and perpendicular to the film plane does not change with increasing t_N. The films for $t_N \geqslant 3$ nm showed a large value of $6.2\pm0.2\times10^7$ erg/cc, which is very close to the value of fully ordered FePt alloys (7.0×10^7 erg/cc).

From the magnetic force microscopy (MFM) observation, it has been revealed that the multidomain particles are formed when the particle size becomes larger than approximately 200 nm.[12] The difference between the magnetization processes in single-domain particles and multidomain particles is demonstrated clearly by the initial magnetization curves shown in Fig. 4. All the magnetization curves were measured at 295 K in the perpendicular direction to the film plane. The vertical axis indicates the magnetization normalized by the magnetization value at the magnetic field of 140 kOe. t_N is 3 nm (a), 5 nm (b), 8 nm (c), 10 nm (d), 12 nm (e), 15 nm (f), 18 nm (g), 20 nm (h), 25 nm (i), 30 nm (j), and 40 nm (k). The films with t_N=3 and 5 nm are very difficult to be magnetized [Fig. 4(a) and 4(b)], because they contain only single domain particles with the sizes of ten to a few tens of nanometers. Hence, the magnetization progresses only by the magnetization rotation in the par-

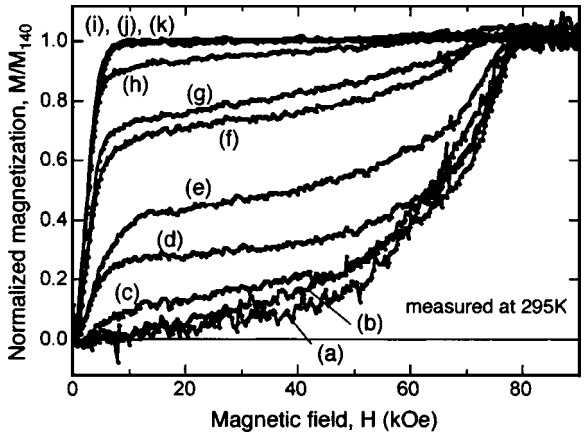

FIG. 4. Initial magnetization curves for the FePt films with t_N=3 nm (a), 5 nm (b), 8 nm (c), 10 nm (d), 12 nm (e), 15 nm (f), 18 nm (g), 20 nm (h), 25 nm (i), 30 nm (j), and 40 nm (k).

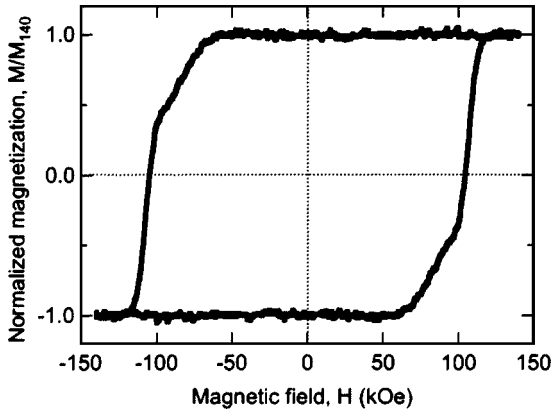

FIG. 5. Magnetization curves for the FePt film with t_N=5 nm measured at 4.5 K.

ticles. However, with increasing t_N [(c) 8 nm and (d) 10 nm], i.e., with increasing particle size, a steep increase of the magnetization at low magnetic field is observed although the complete saturation is still hard. The fractional magnetization at low magnetic field corresponds to the magnetic domain wall displacement; in other words, particles larger than 200 nm with multidomain structure are magnetized at low magnetic field. With further increasing t_N [(f) 15 nm and (h) 20 nm], the volume fraction of multidomain particles increases and consequently, the fraction of the magnetization that is magnetized at low magnetic field increases. Figure 4(h) indicates that the small particles less than 200 nm with single-domain structure still exist for the film with t_N =20 nm because there is a small fraction of the magnetization that is hard to saturate. This is consistent with the TEM observation as shown in Fig. 1(d). However, they disappear for the films with $t_N \geqslant 25$ nm (i,j,k). In this case, the magnetization can be easily saturated at lower than 10 kOe with the displacement of domain walls. These initial magnetization curves are usually classified as "nucleation-type." The observation of initial magnetization process gives significant information about the co-existence of both single-domain and multidomain structures, which cannot be distinguished from the MFM observation because of the present insufficient resolution. Furthermore, it is noted that the value of K_u keeps almost constant although the magnetic domain structure changes from single- to multidomain state, indicating that the magnetization process depends exclusively on the film morphology.

Although the largest H_c of 70 kOe was observed for t_N =5 nm at room temperature [Fig. 3(c)], it was further enhanced by measuring at a cryogenic temperature. Figure 5 shows the magnetization curve for the film with t_N=5 nm measured at 4.5 K. It was measured in the perpendicular direction to the film plane. With decreasing temperature, H_c increases remarkably and huge H_c of 105 kOe was achieved. This enhancement is thought to arise from the reduction of the thermal fluctuation, and also the increase in uniaxial magnetic anisotropy energy K_u with decreasing temperature. However, at this moment, it is hard to discuss the origin for the temperature dependence of H_c, because of the difficulty of precise determination of anisotropy field H_A at low temperature. Since the signal of the magnetization in the in-plane direction was too small and the contribution of the magnetic impurities in the MgO substrate was enormous at low temperature, we were not able to measure H_A at low temperature. The value of H_c and H_A for t_N=5 nm at room temperature is 70 and 120 kOe, respectively. The ratio of H_c/H_A is 0.58, which is considerably larger compared to the value obtained in commercial hard magnetic materials. This is because commercial magnets contain a lot of defects that can induce magnetic reversal. On the other hand, it is possible to produce nearly ideal ferromagnetic particle assemblies using the $L1_0$ FePt phase.

This work has revealed that a huge coercivity exceeding 100 kOe can be obtained in defect-free perfectly aligned single domain particles. From the initial magnetization behavior, the change in the magnetization process from rotation of magnetization to domain wall displacement has been clearly demonstrated.

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