Microstructure and Magnetic Properties for Highly Coercive FePt Sputtered Films

Toshiyuki Shima¹, Koki Takanashi¹, Guo Qing Li² and Shunji Ishio²

¹Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
²Faculty of Engineering and Resource Science, Akita University, Akita 010-8502, Japan

In this paper, we review our recent investigations on the magnetic properties and microstructure of highly coercive FePt films prepared on both MgO (001) and MgO (110) substrates with various film thicknesses (tN). Perpendicularly magnetized FePt (001) film were obtained on MgO (001) substrates, while FePt (101) films with easy magnetization axis canted from the perpendicular direction to the film plane were obtained on MgO (110) substrates. The film morphology changes from a particulate to a continuous state depending on tN. A drastic decrease in the coercivity has been observed with the percolation of particles for FePt (001) films, when tN exceeds a critical thickness, tN = 45 nm. On the other hand, the gradual decrease of Hc with tN has been observed in FePt (101) films and no remarkable change is seen at the critical thickness where the film morphology changes from a particulate to continuous state.

(Received April 18, 2003; Accepted June 11, 2003)

Keywords: FePt, thin film, sputtering, microstructure, magnetic domain structure

1. Introduction

Magnetization processes in nanometer-sized magnets are of great technological and scientific interest. They are important for understanding the high coercivity mechanism of micromagnets with potential applications in various future magnetic devices such as ultrahigh-density recording media and bias magnets in monolithic microwave integrated circuits. An assembly of nanomagnets with large uniaxial anisotropy is required for these applications. The magnetization processes of fine particles and their assemblies have been extensively investigated for a long time.¹⁻³ The magnetization process, and therefore the coercivity should depend strongly on the characteristic size and the morphology of materials. However, there have been only a few experimental investigations on the correlation between the measured magnetization behavior and the actual nanostructure of ultra thin films.

L1₀ ordered FePt alloy has attracted much attention in recent years because of the large uniaxial magnetocrystalline anisotropy.⁴ The epitaxial growth of FePt (001) films with the c axis normal to the film plane was investigated by conventional deposition techniques such as sputtering and molecular beam epitaxy (MBE). Recent interest of this field have been focused on the reduction of the ordering temperature,⁵⁻¹³ the mechanism for coercivity¹⁴⁻¹⁷ and the fabrication of a self-organized magnetic assembly.¹⁸ In a previous paper,¹⁹ we reported that a huge coercivity as high as 3.2 MA/m was obtained for perpendicularly magnetized FePt (001) films with small island structure, and we explained the variation of magnetic properties based on the film morphology change from a particulate to a continuous state that was observed by transmission electron microscopy (TEM). In addition, we also reported that the different magnetization behavior was observed between the FePt films grown on MgO (001) and MgO (110) substrates at the critical thickness where the film morphology changed.¹⁰ In this paper, we review our recent investigations on the relationship between microstructure and coercivity for both the FePt particulate thin films grown on MgO (001) and MgO (110) substrates.

2. Experimental Procedure

Samples were prepared in a high vacuum system (base pressure ~6.7 × 10⁻⁶ Pa) using multiple dc-sputtering with co-deposition of Fe and Pt directly onto polished single crystalline MgO (001) or MgO (110) substrates. The substrates were successively cleaned in a mixture of nitric acid and ethanol, then ethanol and acetone. The targets were commercial products with purities higher than 99.99 at% for Fe and 99.9 at% for Pt. High-purity argon of 1.06 Pa was flown during sputtering. The substrate temperature (Ts) during deposition was fixed at 973 K. The nominal thickness of the FePt film (tN) was varied in the range between 10 and 100 nm. The composition of the films was determined to be Fe₅₂Pt₄₈ (at%) by electron probe X-ray microanalysis (EPMA). The typical growth rate for FePt was 0.12 nm/s. X-ray diffraction with Cu-Kα radiation was performed for structural characterization. Magnetic properties were measured by a superconducting quantum interference device (SQUID) magnetometer in the field up to 4.4 MA/m at room temperature, and the uniaxial magnetic anisotropy energy K_u was determined from the area enclosed between the magnetization curves in applied fields parallel (H∥) and perpendicular (H⊥) to the film plane.

3. Results and Discussion

3.1 FePt (001) films with perpendicular magnetization

X-ray diffraction (XRD) patterns for Fe–Pt films grown on MgO (001) substrates with various tN = (a) 10 nm, (b) 20 nm, (c) 45 nm, (d) 50 nm, (e) 60 nm and (f) 100 nm are shown in Fig. 1. In addition to the fundamental (002) and (004) peaks, (001) and (003) superlattice peaks of the L₁₀ phase are clearly observed for all the samples. The unlabelled sharp peaks are due to the MgO substrate. From the integrated intensities of the fundamental and superlattice peaks extracted from numerical fitting, the degree of long-range chemical order S was evaluated to be 0.95 ± 0.05 for all the films. The detailed procedure for the evaluation was described elsewhere.¹³
Figure 2 shows a TEM image for $t_N = 10$ nm. The films were grown with the island growth mode. Strongly faceted islands of FePt particles are observed with large size distribution. The major facet planes are (100) and (010), and the minor facet plane is (110), indicating that the surface energy of the (100) planes is the lowest. When particles are small, the surface and interfacial energy is large with respect to the volume free energy, and thus the small particles would show clear faceting. The stripe contrast observed in the particles are the Moire pattern originating from the lattice parameter difference between FePt particles and MgO substrates, and their distance is approximately 2.2 nm as expected from the lattice mismatch between FePt and MgO with $\alpha_{\text{FePt}} = 0.40$ nm and $\alpha_{\text{MgO}} = 0.42$ nm, respectively. The selected area diffraction pattern shows that the orientation relationship (OR) between the FePt particles and the MgO substrate is cube-cube OR, that is (001)$_{\text{FePt}}$ || (001)$_{\text{MgO}}$ and (100)$_{\text{FePt}}$ || (100)$_{\text{MgO}}$. The sizes of particles are widely distributed with the typical size of about 50 nm in side. In addition, much smaller particles are observed. This suggests that a number of small particles were formed in the initial stage of the film deposition, then they coalesce to form big particles.

A remarkable change in the morphology of the film is observed from the films with different thicknesses as shown in Fig. 3. With increasing the film thickness, the typical size of particles increases from ~50 nm for $t_N = 10$ nm to about 400 nm for $t_N = 20$ nm. With further increase of $t_N$, particles grow and coalesce forming an interconnected isotropic maze-like pattern. The maze-like particles do not percolate for $t_N \leq 45$ nm. However, the percolation occurs for $t_N = 50$ nm, and the film changes from the discontinuous to continuous morphology. The electron transport measurements also reveal that there is a drastic change in electrical resistance between $t_N = 45$ nm and 50 nm. The resistances are 800 MΩ and 810 Ω for $t_N = 45$ nm and 50 nm, respectively. With further increase of $t_N$, the percolated network expands at the expense of the voids.

Figure 4 shows magnetization curves for $t = 10$ nm (a), 20 nm (b), 45 nm (c), 50 nm (d), 60 nm (e) and 100 nm (f). The easy magnetization axis is perpendicular to the film plane for all the samples. A huge $H_c$ of about 3.2 MA/m, which was measured in the perpendicular direction to the film plane, has been obtained for $t_N = 10$ nm at room temperature.

Figure 2 shows a TEM image for $t_N = 10$ nm. The films were grown with the island growth mode. Strongly faceted islands of FePt particles are observed with large size distribution. The major facet planes are (100) and (010), and the minor facet plane is (110), indicating that the surface energy of the (100) planes is the lowest. When particles are small, the surface and interfacial energy is large with respect to the volume free energy, and thus the small particles would show clear faceting. The stripe contrast observed in the particles are the Moiré pattern originating from the lattice parameter difference between FePt particles and MgO substrates, and their distance is approximately 2.2 nm as expected from the lattice mismatch between FePt and MgO with $\alpha_{\text{FePt}} = 0.40$ nm and $\alpha_{\text{MgO}} = 0.42$ nm, respectively. The selected area diffraction pattern shows that the orientation relationship (OR) between the FePt particles and the MgO substrate is cube-cube OR, that is (001)$_{\text{FePt}}$ || (001)$_{\text{MgO}}$ and (100)$_{\text{FePt}}$ || (100)$_{\text{MgO}}$. The sizes of particles are widely distributed with the typical size of about 50 nm in side. In addition, much smaller particles are observed. This suggests that a number of small particles were formed in the initial stage of the film deposition, then they coalesce to form big particles.

A remarkable change in the morphology of the film is observed from the films with different thicknesses as shown in Fig. 3. With increasing the film thickness, the typical size of particles increases from ~50 nm for $t_N = 10$ nm to about 400 nm for $t_N = 20$ nm. With further increase of $t_N$, particles grow and coalesce forming an interconnected isotropic maze-like pattern. The maze-like particles do not percolate for $t_N \leq 45$ nm. However, the percolation occurs for $t_N = 50$ nm, and the film changes from the discontinuous to continuous morphology. The electron transport measurements also reveal that there is a drastic change in electrical resistance between $t_N = 45$ nm and 50 nm. The resistances are 800 MΩ and 810 Ω for $t_N = 45$ nm and 50 nm, respectively. With further increase of $t_N$, the percolated network expands at the expense of the voids.

Figure 4 shows magnetization curves for $t = 10$ nm (a), 20 nm (b), 45 nm (c), 50 nm (d), 60 nm (e) and 100 nm (f). The easy magnetization axis is perpendicular to the film plane for all the samples. A huge $H_c$ of about 3.2 MA/m, which was measured in the perpendicular direction to the film plane, has been obtained for $t_N = 10$ nm at room temperature.
Since the maximum magnetic field of 4.4 MA/m in SQUID is not sufficient to saturate the magnetization, the real values of $H_c$ and saturation magnetization might be even higher. With increasing $t$, the coercivity decreases slowly, but still keeps quite a large value of about 2.0 MA/m for $t_N = 45$ nm. However, a drastic change by one order of magnitude is observed between $t_N = 45$ and 50 nm. This critical region corresponds to the change in the film morphology from the discontinuous to continuous state. With further increase of $t$, the magnetization becomes easier to be saturated for the perpendicular direction. $K_u$ shows no remarkable change with $t_N$, and it is $6.0 \pm 0.5 \times 10^6$ J/m$^3$ which is very close to that of the fully ordered FePt alloy ($7 \times 10^6$ J/m$^3$). Therefore, the change in $H_c$ with $t_N$ is not attributed to that in $K_u$, but closely related to the morphology change.

The initial magnetization curve with the saturation hysteresis loop for $t_N = 20$ nm is shown in Fig. 5. The initial slope of magnetization increase is steep and the saturation is reached at the field much lower than the coercive force. This suggests that domain walls exist in each particle in the initial demagnetized state and the domain walls easily propagate over the particle without strong pinning. The magnetization process is classified as a nucleation-type, and not a pinning-type.

The magnetic domain structures were observed by MFM, and atomic force microscopy (AFM) observation was also performed in order to correlate the magnetic domain structure with the morphology.\textsuperscript{20,21} Figure 6 shows TEM (a), AFM (b) and MFM (c) images separately, and the overlapped images of AFM and MFM (d) for $t_N = 20$ nm. The AFM image is consistent with the TEM image, although the particulate sizes observed by AFM are somewhat larger than those observed by TEM. This difference is thought to be due to the finite size effect of AFM probe. The MFM observation was performed from the identical area with that observed by AFM. From the overlapped image, magnetic domains within a particle are clearly observed. Figure 7 shows the overlapped images of AFM and MFM for $t_N = (a) 10$ nm, (b) 20 nm, (c) 45 nm, (d) 50 nm, (e) 60 nm and (f) 100 nm. For $t_N = 10$ nm, most of particles have single domain structure. However, for $t_N = 20$ nm, multiple domain particles increase. The critical size between single and multiple domain particles is estimated to be approximately 200 nm. It is noted that no remarkable difference between the domain structures for $t_N = 45$ and 50 nm, where the coercivity changes drastically, is observed. These results indicate that the drastic change of coercivity does not occur when most of the particles change from single domain to multiple domain structure, but occurs when the percolation starts. It is considered that there are very few pinning sites for the domain wall movement for epitaxial FePt(001) films which are monocrystalline with the $c$-axis aligned perpendicular to the film plane. Therefore, the domain walls easily propagate all over the film through the percolated network, resulting in the remarkable drop of coercivity.

![Fig. 4 Magnetization curves for FePt (001) thin films with different film thicknesses: (a) 10 nm, (b) 20 nm, (c) 45 nm, (d) 50 nm, (e) 60 nm and (f) 100 nm. The magnetic field was applied in the perpendicular direction to the film (solid line) and in the in-plane direction (broken line).](image1)

![Fig. 5 Initial magnetization curve with saturation hysteresis loop for FePt (001) thin film with $t_N = 20$ nm.](image2)

![Fig. 6 TEM, AFM, MFM and the overlapped images of AFM and MFM for FePt (001) film with $t_N = 20$ nm.](image3)
3.2 FePt (101) films with canted magnetic easy axis

XRD patterns for the FePt films deposited on MgO (110) substrate with 
$t_N = (a) 15 \text{ nm}, (b) 45 \text{ nm}, (c) 50 \text{ nm}, (d) 100 \text{ nm}$ are shown in Fig. 8. In addition to the fundamental
(220) peak, (110) and (330) superlattice peaks of the L1$_0$ phase are clearly observed for all the samples and the
intensities of these peaks increased with increasing $t_N$. However, the most intense peak is associated with FePt
(202), in other words, the $c$-axis of L1$_0$ structure is canted
from the perpendicular direction for the films grown on MgO (110) substrates. The films grown on MgO (110) substrate are
denoted as FePt (101) films, hereafter. It is considered that
the (202) texture is dominant in order to relax the strain
energy stored between the MgO (110) substrate and the FePt
layer because of the large lattice mismatch. It is noted that the
direction of the $c$-axis for the films grown on MgO (110)
substrates is different from previous results on the films with
Pt buffer layer.\textsuperscript{22,23} When the FePt films were deposited on buffer layer, orientation relationship between FePt (110) and
MgO (110) can be achieved.

TEM images for $t = 15$ and 45 nm are shown in Fig. 9. The
morphology change with $t_N$ for FePt (101) films is similar to
that for FePt (001) films. However, some regularity is seen in
the alignment of particles for $t_N = 15 \text{ nm}$ (Fig. 9(a)). One
possible origin for the regular alignment is the effect of facets
formed on the unstable (110) surface of MgO substrate.
Magnetization curves for $t_N = (a) 15 \text{ nm}, (b) 30 \text{ nm}, (c) 45 \text{ nm}, (d) 50 \text{ nm}, (e) 60 \text{ nm}$ and (f) $100 \text{ nm}$ are shown in Fig. 10. The easy magnetization axis exists between the perpendicular direction to the film plane and in-plane MgO [1C2210] direction for FePt (101) films, i.e., the easy magnetization axis is canted from the normal direction to the film plane.

$H_c$ measured in both in-plane MgO [1C2210] direction and perpendicular to the film plane direction are about 1.5 MA/m. TEM observation and electrical resistance measurements have revealed that the film with $t_N = 15 \text{ nm}$ has discontinuous morphology, while the film with $t_N = 50 \text{ nm}$ has continuous one with interconnected islands.

In contrast to the behavior in FePt (001) films, a gradual change of $H_c$ has been observed, i.e., a large $H_c$ of 1.0 MA/m is obtained even for a continuous state ($t = 60 \text{ nm}$), and no remarkable change is seen at the critical thickness. This indicates the presence of pinning sites for the domain wall movement. The pinning is thought to be due to planar defects like twins, which have been induced in a course of the growing process since the c-axis is canted from the perpendicular direction and they play an effective role as pinning sites for the domain wall movement.

4. Summary

FePt (001) films with perpendicular magnetization and FePt (101) films with a canted easy magnetization axis (c-axis) were prepared on MgO (001) and MgO (110) substrates, respectively. The film morphologies change from a particulate to a continuous state depending on $t_N$. At the critical thickness where the film morphology changes from a particulate to a continuous state, a drastic decrease in the coercivity $H_c$ has been observed for the FePt (001) film, while no remarkable change has been observed for the FePt (101) films. The difference is thought to arise from the presence of pinning sites for the domain wall movement.

Acknowledgements

This work is partly supported by the Special Coordination Funds for Promoting Science and Technology on “Nanohetero Metallic Materials” from the Ministry of Education, Culture, Sports, Science and Technology. The structural characterization was performed at Laboratory for Advanced Materials, IMR, Tohoku University. The authors would like to acknowledge Dr. K. Hono and Dr. Y. K. Takahashi for TEM observation and critical discussion, Dr. S. Mitani, Dr. S. Sugimoto for useful discussion and Mr. Y. Murakami for technical assistance.
REFERENCES