Synthesis and Structure of Isolated $L1_0$FePt Particles

Bo Bian, David E. Laughlin, Kazuhisa Sato, and Yoshihiko Hirotsu

***Abstract*—Isolated FePt particles were fabricated by taking advantage of overgrowth of Fe on Pt particles and subsequent annealing at 873K. The annealing lead to the formation of ordered tetragonal ($L1_0$) FePt particles with three orthogonal variants. The average size of FePt particles could be adjusted to less than 10 nm and no significant grain coarsening occurred upon annealing due to the existence of an amorphous Al_2O_3 layer. Nano-beam electron diffraction revealed that the lattice constant ratio c/a was approximately 0.97 for the annealed FePt particles with Fe composition close to 50 at.%.**

***Index Terms*—FePt, isolated particles, $L1_0$phase, ordering, TEM.**

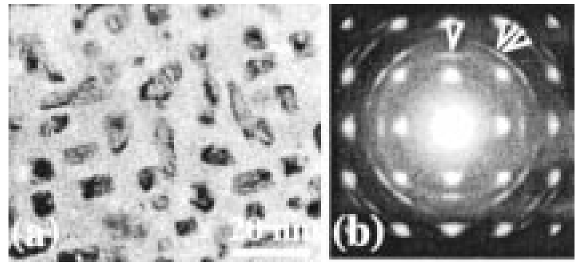

Fig. 1. TEM image and SAED pattern of a-Al_2O_3/Fe/Pt film. The 002 and 112 reflections of α-Fe are marked by single and double arrows respectively in (b).

I. Introduction

THE ORDERED equiatomic CoPt and FePt phases with tetragonal $L1_0$ structure are attractive materials for future magnetic recording media due to their high magnetocrystalline anisotropies. Future high-density magnetic recording media require thermally stable grains with size much less than 10 nm. In this grain size range, magnetic grains of conventional Co-based media with the hexagonal structure become unstable at room temperature. On the other hand, the high anisotropy of ordered tetragonal ($L1_0$) CoPt and FePt phases allows the grains with size much less than 10 nm to remain thermally stable [1]. Former work on $L1_0$CoPt and FePt systems reported high coercivity in CoPt [2] and FePt [3] films, CoPt/Ag [4] and FePt/SiO_2 [5] granular films, and CoPt particles grown on quartz substrate [6]. For application of the $L1_0$CoPt and FePt systems in future high-density magnetic recording media, it is important to fabricate magnetically isolated $L1_0$FePt particles with smaller size. In this study, we have prepared separated $L1_0$ FePt particles with size close to 10 nm and investigated their microstructures.

II. Experimental

Sample preparation was performed in an electron-beam deposition system with base pressure less than 3×10^{-7} Pa. First, Pt was deposited on (100) NaCl and MgO substrates kept at 673K. Next, Fe was deposited onto the substrates with Pt. A cover layer of amorphous $(a-)$ Al_2O_3 with a thickness larger than 4 nm was then deposited without breaking vacuum. The deposition rate for Pt and Fe was in the range of 0.1–0.4 nm/min. The total thickness of FePt particle layer was about 3 nm. The relative composition of Fe in the FePt layers was analyzed by an energy dispersive x-ray (EDX) analysis system. The relative Fe concentration in the FePt layers was approximately 45 at.% unless specified. To examine the structure of the pure Pt particle, a part of the substrate was shielded from Fe flux after Pt deposition. Annealing of a-Al_2O_3/Fe/Pt films was done at 873K for different time intervals in a vacuum better than 2×10^{-5} Pa. A part of the NaCl (100) substrate with as-deposited a-Al_2O_3/Fe/Pt films was immersed in distilled water and the films were mounted onto copper microgrids for later transmission electron microscope (TEM) observation. In-plane magnetization hysteresis loops of the as-deposited and annealed films were measured at room temperature with a superconducting quantum interference device magnetometer (SQUID).

III. Results and Discussion

TEM observation of the a-Al_2O_3/Pt film showed a morphology of isolated Pt particles with facets. The Pt particles are highly $\langle 100 \rangle$ oriented due to their epitaxial growth on the NaCl (100) substrate. Fig. 1 shows the TEM image and corresponding selected area electron diffraction (SAED) pattern of an as-deposited a-Al_2O_3/Fe/Pt film. The FePt particles appear to be island-like. Many regions with dark contrast are visible in the images. By comparing TEM images of as-deposited a-Al_2O_3/Pt and a-Al_2O_3/Fe/Pt films, it is determined that the dark contrast regions correspond to Pt particles in as-deposited a-Al_2O_3/Pt film. Obviously, the Pt particles acted as nucleation sites for the growth of Fe crystallites. From Fig. 1(b), it is known that Fe particles grown on $\langle 100 \rangle$ oriented Pt crystals have a bcc structure. The Fe particles are textured at $\langle 100 \rangle$ and $\langle 110 \rangle$ two different directions.

When a-Al_2O_3/Fe/Pt films were annealed in TEM at 773–823K, a gradual disappearance of the reflections from bcc Fe was observed, and the appearance of the diffraction pattern of $\langle 100 \rangle$ oriented fcc FePt structure began to appear. As the annealing was continued, 001 and 110 superlattice diffraction spots appeared and their intensities gradually increased. The annealed a-Al_2O_3/Fe/Pt films exhibited an island-like structure.

Manuscript received February 14, 2000.

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Publisher Item Identifier S 0018-9464(00)08475-2.

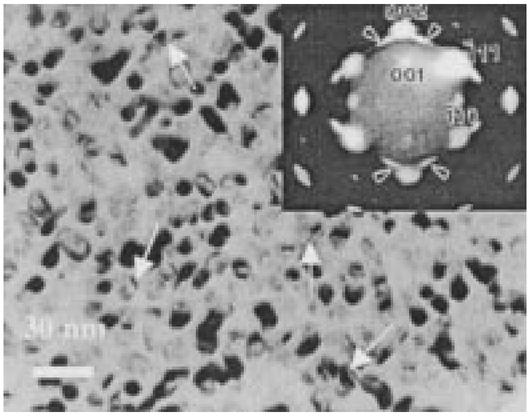

Fig. 2. Dark-field TEM image and SAED pattern of a 873K, 6 h annealed a-Al_2O_3/Fe/Pt film. Some $\{111\}$ twins are visible.

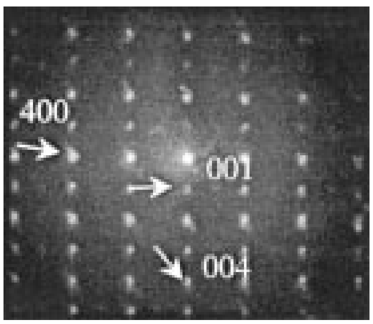

Fig. 3. NBD pattern of local region of a FePt particle. The c-axis of the FePt region is in the film plane and the c/a ratio was approximately 0.97.

No significant grain coarsening was observed in the annealed a-Al_2O_3/Fe/Pt films. Fig. 2 shows dark-field TEM image and corresponding SAED pattern of a 873K, 6 h annealed a-Al_2O_3/Fe/Pt film. The sample was tilted so the electron beam was parallel to $\langle 110 \rangle$ direction. Uniformly dispersed FePt particles are visible in the image. The average size of the FePt particles was estimated to be 12 nm. Some twins (as marked by white arrows) can be seen in the TEM image. From the direction of the streaks (as marked by dark arrows), we know they are $\{111\}$ twins. In the SAED pattern, besides the fundamental reflections, (001) and (110) type superlattice reflections due to $L1_0$ are visible. The co-existence of three-variant ordered domains in $L1_0$ FePt particles was confirmed by high-resolution electron microscopy observation [7].

Nano-beam electron diffraction (NBD) examination was also performed by focusing an electron beam probe of approximately 1–2 nm on local regions of various FePt particles. Image plates instead of negative films were used to record the NBD patterns and to measure the distance between diffraction spots. The application of image plates provided high accuracy in the distance measurements. The common existence of the three variants in the FePt particles was confirmed by the NBD examinations. Fig. 3 shows a NBD pattern taken from a local region in a 873K, 6 h annealed a-Al_2O_3/Fe/Pt film. This local region of FePt particle has its c-axis in the film plane. From measurement of the distances between (400) and (004) diffraction spots to transmission spot, it was determined that the lattice constant c/a ratio of the local region of FePt particle was approximately 0.97. The c/a ratios estimated from NBD patterns of 873K, 6 h annealed a-Al_2O_3/Fe/Pt films were in the range of 0.97–0.98, indicating the ordering reaction differed slightly from particle to particle.

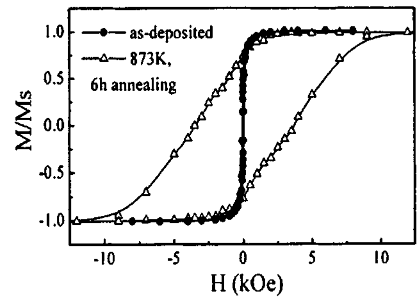

Fig. 4. In-plane hysteresis loops for a-Al_2O_3/Fe/Pt films on (100) NaCl substrate (unannealed and annealed at 873K for 6 h).

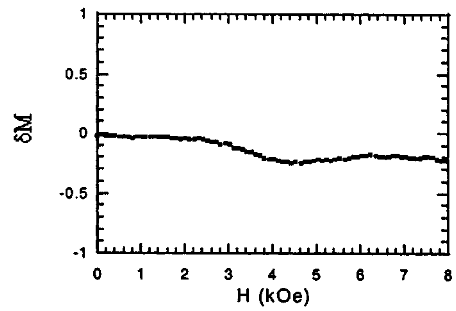

Fig. 5. δM plot for a 873K annealed a-Al_2O_3/Fe/Pt film.

Fig. 4 shows in-plane hysteresis loops for a-Al_2O_3/Fe/Pt films on (100) NaCl substrate (unannealed and annealed at 873K for 6 h). The as-deposited a-Al_2O_3/Fe/Pt film is magnetically soft with coercivity less than 100 Oe. The coercivities of the films increase dramatically upon annealing at 873K. The coercivity of the 873K, 6 h annealed a-Al_2O_3/Fe/Pt film reached 3.5 kOe, being consistent with the presence of chemically ordered $L1_0$ FePt in the annealed film. As annealing time increased, the coercivity increased but the magnetic squareness decreased. Our magnetic measurements showed that the magnetic properties of all the a-Al_2O_3/Fe/Pt films on both NaCl and MgO (100) substrates were similar.

Since the thickness of the FePt layer was about 3 nm and the relative composition of Fe was approximately 45 at.%, the areal moment density of the annealed a-Al_2O_3/Fe/Pt film was estimated to be in the range 0.3–0.4 memu/cm^2. Another important issue for noise reduction in the films is the magnetic isolation of the FePt particles. Fig. 5 shows a δM plot for a 873K annealed a-Al_2O_3/Fe/Pt film. The absence of data points in the positive range of δM indicates that no exchange-coupling exists between the FePt particles. The negative values of δM in the range of $-0.2 - 0$ imply that only a small amount of dipolar interaction is present.

Fig. 6 shows the composition dependence of magnetic coercivity of 873K annealed a-Al_2O_3/Fe/Pt films. The Fe composition in the films ranged from 31–62 at.%, as measured by an EDX analysis system. The coercivity had its maximum at

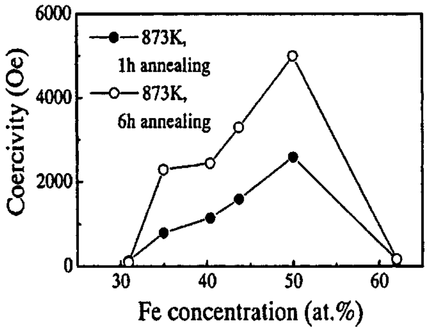

Fig. 6. Composition dependence of magnetic coercivity of 873K annealed a-Al_2O_3/Fe/Pt films.

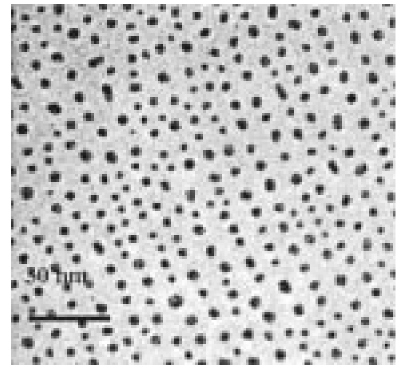

Fig. 7. TEM image of isolated Pt clusters.

around 50 at.% Fe and dropped significantly as the Fe concentration was close to 31 or 62 at.%. From our TEM observations, we knew that there still existed some bcc Fe phase in the annealed a-Al_2O_3/Fe/Pt films with 62 at.% of Fe. As the c/a ratio of 873K annealed a-Al_2O_3/Fe/Pt films with relative Fe composition close to 50 at.% was found to be in the range of 0.97–0.98, so the ordered $L1_0$ FePt phase was the reason for the high magnetic coercivity. The ratio $c/a = 0.97$ was reported in CoPt granular films with large coercivity and it was claimed that highly ordered tetragonal structure is a prerequisite for the large coercivity [8]. Gong *et al.* reported that the coercivity of FePt films was greatly dependent on composition and the highest coercivity was obtained around 53 at.% of Fe [9]. They also observed that the density of $\{111\}$ twins changes drastically with composition. Although similar $\{111\}$ twins were observed in the present FePt particles, their effect on magnetic coercivity needs further investigation.

Since the Pt particles acted as nucleation sites for the growth of Fe particles, we need to increase the particle density of Pt if we want to decrease the grain size and increase packing density of FePt particles. It was found in our experiment that Pt particles with high density could be prepared by increasing the deposition rate and reducing the substrate temperature. Fig. 7 shows a TEM image of Pt clusters deposited on NaCl (100) substrate at room temperature with a deposition rate of 1 nm/min. The distribution of the cluster size is very narrow. As the inter-cluster distance is less than or close to 10 nm, $L1_0$FePt particles with higher packing density and smaller grain size (<10 nm) can be prepared. The grain size and packing density of FePt particles can be further adjusted by applying multilayer structures [5].

IV. Summary

Isolated FePt particles with grain size 10 nm in size have been prepared by taking advantage of overgrowth of Fe on Pt particles and later annealing. The coercivity of the FePt particles is strongly dependent on composition and degree of ordering. The lattice constant ratio c/a of FePt particles with high coercivity is close to 0.97. The grain size and packing density can be controlled by experimental conditions.

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