Fabrication of two-dimensional assembly of L10FePt nanoparticles

doi: 10.1063/1.1558053

Fabrication of two-dimensional assembly of $L1_0$FePt nanoparticles

T. Miyazaki,[a] S. Okamoto, O. Kitakami, and Y. Shimada
Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan

(Presented on 14 November 2002)

We searched for an optimum thermal treatment of double or quadruple layers of (Fe 2.5 nm/Pt 2.5 nm) in ordered to obtain a monolayer assembly of ordered FePt particles. It was found that by controlling the thickness, the substrate temperature, and using MgO(100) substrates, monolayer assemblies of ordered FePt particles with the crystal axes [001] aligned vertically can be obtained at relatively low substrate temperatures around 400 °C. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1558053]

I. INTRODUCTION

In the field of longitudinal magnetic recording, thermal agitation of the media is becoming a serious obstacle and the fundamental solution to the problem is to develop a monolayer of high anisotropy nanoparticles with aligned crystal axes. The ordered $(Co,Fe)_{50}Pt_{50}$ alloy films have been investigated intensively because of their very high uniaxial anisotropy.[1,2] Although a number of papers have been published on granular structures, ultrathin films, and single crystal films of this alloy, the material is still far from practical use. The reason is that to attain the ordered phase of the alloy very high-temperature treatment (550–650 °C) is indispensable and no practical process has been developed to cope with the compatibility between the nanoparticle structure and the high anisotropy of the ordered $L1_0$ phase. On the other hand, a few studies aiming to reduce the ordering temperature have been reported[3,4] Luo and Sellmyer reported that the ordering process of Fe/Pt multilayers occurs in the appreciably reduced temperature range of 300–400 °C.[4]

In this article, the ordering process of double or quadruple layers of Fe/Pt is studied. We will search for an optimum thermal treatment by which a fine monolayer of ordered FePt particles is available. The island formation process of Volmer–Weber (V–W)-type is introduced in the thermal treatment experiment of Fe/Pt multilayers in order to obtain monolayer particles. For the crystal alignment, the effect of single crystal MgO(100) substrate is investigated.

II. EXPERIMENTS

Double $[(Fe/Pt)_1]$ and quadruple $[(Fe/Pt)_2]$ layers were prepared by a dc magnetron sputtering system onto thermal oxidized Si or single crystal MgO(100) substrates. The thickness of each Fe and Pt layer was fixed to be 2.5 nm. The back pressure was less than 5×10^{-9} Torr and Kr was used for sputtering at a pressure of 5 mTorr. The substrate temperature was adjusted in the range from room temperature to 500 °C. Annealing was made in a vacuum at 300–500 °C for 1 h. The surface topology of samples was observed using an atomic force microscope (AFM) as well as a scanning electron microscope (SEM). The crystal structure was analyzed by x-ray diffraction (XRD) and reflection high-energy electron diffraction (RHEED). The measurements of magnetic properties were carried out with a vibrating sample magnetometer ($H_{max}=2.2$ T).

III. RESULTS AND DISCUSSION

We will describe the ordering process of double and quadruple layers of (Fe/Pt) with regard to an optimum thermal treatment for a monolayer of ordered FePt particles. Figure 1 shows XRD patterns of $(Fe/Pt)_2$ deposited onto Si substrates held at ambient temperature and postannealed at various temperatures (T_a). In the as deposited film, the fundamental and satellite peaks due to the periodic structure were observed and it turned out from these peak positions that the multilayers have a preferred crystal orientation of ⟨110⟩ of body-centered-cubic-Fe and ⟨111⟩ of face-centered-cubic-Pt along the growth direction.[5] After annealing at temperatures above 300 °C, the fundamental and satellite peaks disappear and instead the FePt(111) peak appears which indicates that the multilayer structure rapidly transformed into a FePt alloy. As shown in Fig. 2, the coercivity (H_c) starts to increase at appreciably low T_a (300–350 °C) compared with single-

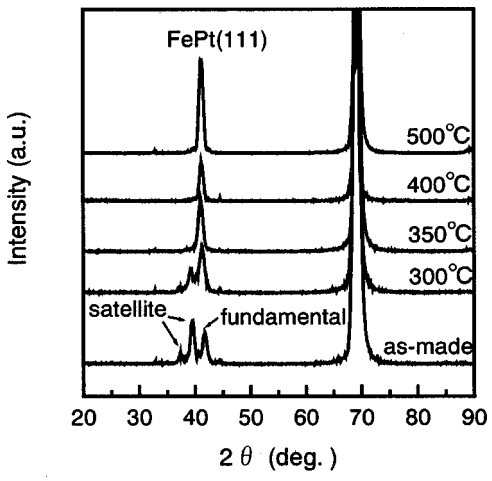

FIG. 1. XRD patterns for (Fe 2.5 nm/Pt 2.5 nm)$_2$ on oxidized Si substrates at various annealing temperatures.

[a]Author to whom correspondence should be addressed; electronic mail: miyataka@mail.tagen.tohoku.ac.jp

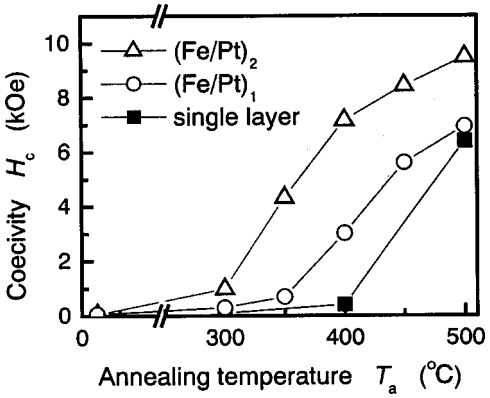

FIG. 2. Variation of coercivity by annealing for double and quadruple layers of Fe(2.5 nm)/Pt(2.5 nm) on oxidized Si substrates.

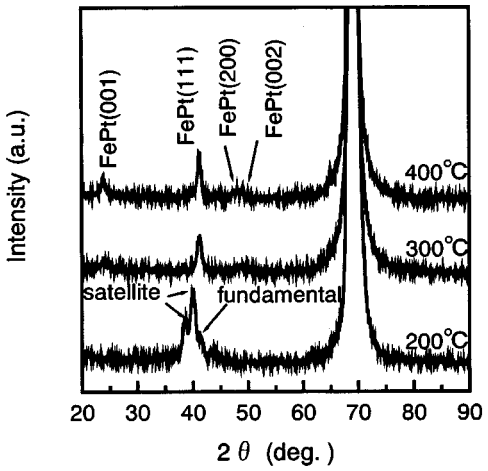

FIG. 4. XRD patterns for $(\text{Fe } 2.5 \text{ nm/Pt } 2.5 \text{ nm})_2$ on oxidized Si at various substrate temperatures (T_s).

layer films and continues to increase with T_a, which agrees with formation of the ordered $L1_0$ phase as seen in Fig. 1. Therefore, the ordering process of the multilayers starts near 300 °C and the reduction of ordering temperature for double and quadruple layers has been confirmed to occur analogously to the thicker multilayers.[6] The $B-H$ curves of these samples have no in-plane anisotropy. This is because the crystal plane of (111) tends to grow parallel to the film plane. However, in the electrical resistivity (ρ) measurements, all the multilayers turned out to be conductive, suggesting that in the Fe/Pt multilayers deposited without heating the substrates, the atomic mobility is not high enough to allow for island formation of V–W type.

Then, a search for the optimum substrate temperature to realize V–W-type island formation was made. Multilayers of $(\text{Fe/Pt})_1$ and $(\text{Fe/Pt})_2$ were deposited on Si substrates with a substrate temperature (T_s) of 200–500 °C. Figures 3(a) and 3(b) are images of AFM and SEM for samples deposited with T_s of 400 °C. The islands with the size of 25–35 nm are formed and they are well separated although some of them are still connected to each other. The resistivity shown in Fig. 3 is very high which confirms perfect separation of the particles. It should also be noted that the structure and the size of the particles are sensitively dependent on the total thickness of the multilayers. Subsequently, the degree of ordering, the crystal structure, and its orientation were studied by XRD. Figure 4 shows variation of XRD patterns for $(\text{Fe/Pt})_2$ with T_s. It suggests that T_s of 300 °C is high enough to allow for alloying and ordering of FePt since the fundamental and satellite peaks of the multilayer structure are replaced by (001) of superlattice diffraction from $L1_0$. This process of ordering is analogous to that in Fig. 1 and the study reported by Endo *et al.*[7] where the multilayers were deposited at ambient temperature and annealed afterward. Thus, it should be stressed that the monolayer formation process and the $L1_0$ ordering process are compatible with each other by adjusting the substrate temperature of multilayer deposition. Since the XRD is not intensive enough for an exact analysis of $(\text{Fe/Pt})_1$ double layer, the ordering process was monitored by an increase of H_c. And, it was confirmed that an almost identical behavior of H_c to those of the postannealed multilayers[7] was observed for these double layers. However, the magnetization loops and RHEED observation, together with the XRD, reveal that the particles have no preferred orientation of crystal axes.

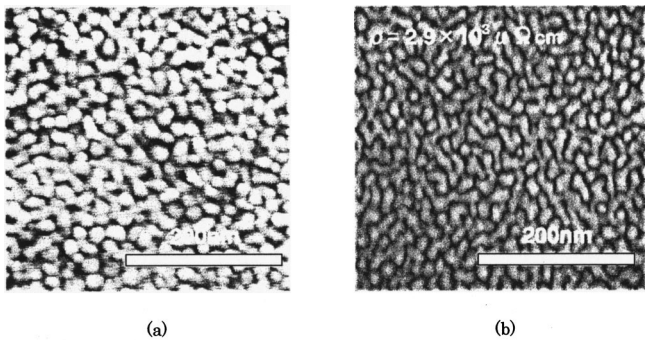

FIG. 3. (a) AFM and (b) SEM images of $(\text{Fe } 2.5 \text{ nm/Pt } 2.5 \text{ nm})_1$ layers deposited at 400 °C. The inset shows the resistivity (ρ).

In the next stage of this study our interest is focused on whether it is possible to align the crystal axes of the particles in the process described herein. The same experiments were repeated just by replacing Si substrates with MgO and the possibility of epitaxial growth of $L1_0$ particles was examined. In the past, Fe/Pt double layers deposited on NaCl were investigated.[8] Epitaxial growth of FePt ordered particles was confirmed although the annealing temperature is relatively high and {001} variants [namely, coexistence of (001) and (100) oriented crystals in a single particle] were frequently observed. So, our interest is whether the process is reproduced or improved on MgO (100) surface. Figure 5 shows SEM images of $(\text{Fe/Pt})_1$ deposited on MgO at 400 and 500 °C. Together with the very high resistivity as in Fig. 5, the images confirm that isolated FePt particles are formed analogously and the size of the particles is also the same with the ones on Si substrates. XRD is shown in Fig. 6. It is noted that for T_s above 400 °C, the superlattice diffraction (001) is clearly observed but the peaks (002) and (200) are not separated explicitly. This suggests the possibility of variants that have c axis of ordered FePt lying in the film plane. Figure 7 shows the magnetization loops for $(\text{Fe/Pt})_1$ deposited on T_s of 400 and 500 °C. The H_c along the vertical direction is

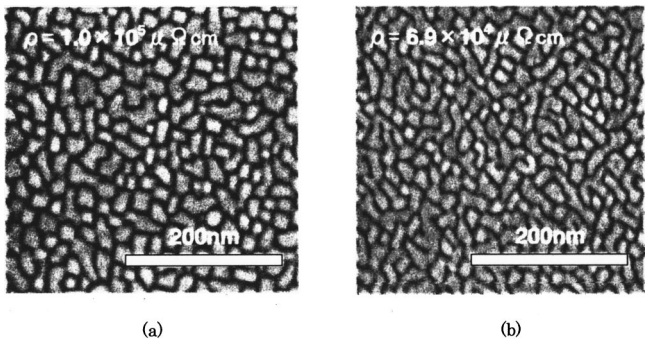

FIG. 5. SEM images of $(\text{Fe } 2.5\text{ nm/Pt } 2.5\text{ nm})_1$ deposited on MgO(100) substrate at (a) 400 °C and (b) 500 °C. The inset shows the resistivity (ρ).

higher than 3 kOe and that in the plane is a few hundred Oe. So it is very likely that only a little amount of (100) variants are formed in our process. It should also be noted that H_c for the sample deposited at 500 °C is reduced compared to that for the one deposited at 400 °C. This should be attributed to the occurrence of coalescence of the particles due to excessively heated substrates. The δM measurements were performed for the samples deposited at 400 °C in order to clarify the degree of magnetic interaction. The curves show no peaks except for a slight deviation to the negative. This may be due to a weak magnetostatic interaction between the particles.

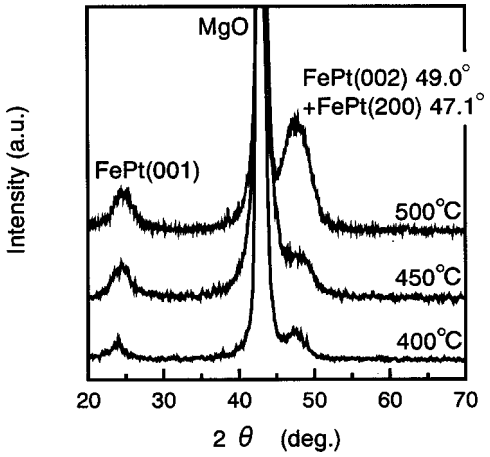

FIG. 6. XRD patterns for $(\text{Fe } 2.5\text{ nm/Pt } 2.5\text{ nm})_1$ on MgO(100) substrate at various substrate temperatures.

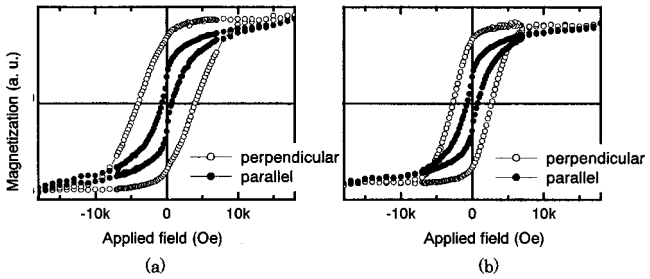

FIG. 7. Magnetization loops for $(\text{Fe } 2.5\text{ nm/Pt } 2.5\text{ nm})_1$ deposited on MgO(100) at substrate temperatures of (a) 400 and (b) 500 °C.

So, we conclude that it is possible to develop a process in which a monolayer assembly of ordered and well separated FePt particles can be formed at low temperatures around 400 °C. For further development of the process intensive search for a seed layer that realizes the reduction of the particle size together with a higher surface density and perfect crystal alignment is needed. The appreciable reduction of the substrate temperature makes the search easier because a variety of metallic underlayers may be considered as the candidates for the suitable seed layer.

IV. CONCLUSION

This study was aimed at developing a fabrication process of a monolayer assembly of FePt ordered particles without high-temperature treatments. It was confirmed that FePt ordered particles are available by applying the V–W-type mechanism to double and quadruple layers of (Fe 2.5 nm/Pt 2.5 nm) at relatively low temperatures around 400 °C. Using MgO(100) as substrates, single crystal particles with the crystal axis [001] aligned vertically are formed with appreciably reduced amount of (100) variants.

[1] O. A. Ivanov, L. Solina, V. A. Demshima, and L. M. Magat, Phys. Met. Metallogr. **35**, 81 (1973).
[2] R. F. C. Farrow, D. Weller, R. F. Marks, M. F. Toney, A. Cebolada, and G. R. Harp, J. Appl. Phys. **79**, 5967 (1996).
[3] C. Chen, O. Kitakami, S. Okamoto, Y. Shimada, K. Shibata, and M. Tanaka, IEEE Trans. Magn. **35**, 3466 (1999).
[4] C. P. Luo and D. J. Sellmyer, IEEE Trans. Magn. **31**, 2764 (1995).
[5] E. E. Fullerton, I. K. Schuller, H. Vanderstraeten, and Y. Bruynseraede, Phys. Rev. B **45**, 9292 (1992).
[6] R. A. Ristau, K. Barmak, L. H. Lewis, K. R. Coffey, and J. K. Howard, J. Appl. Phys. **86**, 4257 (1999).
[7] Y. Endo, N. Kikuchi, O. Kitakami, and Y. Shimada, J. Appl. Phys. **89**, 7065 (2001).
[8] B. Bian, Y. Hirotsu, K. Sato, T. Ohkubo, and A. Makino, J. Electron Microsc. **48**, 753 (1999).