Research Article

Structural Dependent Ferromagnetic-Nonmagnetic Phase Change in FePtRu Films

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1. Introduction

Increased power consumption in data centers having millions of hard disk drives (HDDs) has emerged as a serious issue. Decreasing the number of HDDs by increasing their magnetic storage density can be a simple and efficient means to mitigate increased power consumption. Ferromagnetic (FM) thin films prepared using nanofabrication techniques can be used for high-density magnetic data storage [1–3] and spintronic devices [4].

Bit-patterned media (BPM) is a candidate for the creation of ultra-high-density magnetic data storage devices [1, 2]. BPM consist of multiple phases, which include the FM phase (dots) characterized by a high uniaxial magnetic anisotropy along with, in principle, diamagnetic, antiferromagnetic (AF), and paramagnetic (PM) phases. Ion milling is conventionally used for nanofabrication of two-dimensional FM films [5, 6], which produces physically separated FM regions. After ion milling, backfill and polishing stages are required to obtain BPM with smooth surfaces, which is a prerequisite for read/write heads to be able to fly at a few nanometers above the medium surface [1].

Another BPM fabrication method, using ion irradiation, has also been proposed [7, 8]. This method can replace the three processes of ion milling, backfill, and polishing steps, thereby streamlining the production of BPM. Ion irradiation results in structural disordering. Co/Pt multilayers [7, 8] and $L1_0$ CrPt$_3$ films [9] with an uniaxial magnetocrystalline anisotropy constant ($K_u$) of ~$10^6$ erg cm$^{-3}$, which is not sufficient for the realization of high magnetic data storage density of over 2Tb in$^{-2}$, are transformed from FM to PM phase after ion irradiation.

A problem faced by high-density magnetic data storage is the thermal fluctuation effect of magnetic grains or dots. The magnetic anisotropy energy ($K_uV$; $V$: volume of an isolated magnetic grain or dot) becomes lower relative to the thermal fluctuation energy ($k_B T$; $k_B$: Boltzmann constant, $T$: temperature) when the grain and/or dot size is reduced for increasing storage density. The thermal fluctuation can be sufficiently reduced using materials with high $K_u$, satisfying the thermal stability factor requirement of ($K_uV/k_B T$) $> 60$ (this metric was derived using the Sharrock equation [10]) which is typically regarded as a minimum requirement for magnetic data storage [1, 11].
$L_1_0$ FePt (CuAu-I type, fct, $a = b > c$) films with ordered structures are possible candidates for BPM due to their high $K_u$ of $70 \times 10^6$ erg cm$^{-3}$, high saturation magnetization ($M_s$) of $1100$ emu cm$^{-3}$, high corrosion resistance, and low resistivity [12–14], which leads to excellent thermal stability of magnetization in nanometer-size structures. After ion irradiation, the ordered $L_1_0$ FePt films undergo transformation from the hard-FM phase (high $K_u$) to the soft-FM phase (low $K_u$) with the disordered $A1$ structure (fcc, $a = b = c$) [15–20]. Although the $L_1_0$ FePt is suitable for BPM, modifying its properties using ion irradiation is difficult due to the insensitivity of magnetization with ion irradiation. The high spontaneous magnetization ($M_s$) of the soft-FM region (interdot spacing), wherein the ions are irradiated, leads to spike noise in the storage media. Therefore, developing high-$K_u$ and high-$M_s$ materials, whose $K_u$ and $M_s$ values decrease upon ion irradiation with high sensitivity, is required.

In our previous study, nonmagnetic phases were observed by replacing Fe with Mn [21] and Pt with Rh [22] in FePt films, and the former improved the sensitivity of $M_s$ to ion irradiation. The $L_1_0$ Fe$_{1-x}$Mn$_x$Pt films with $x \leq 0.44$ exhibit FM properties corresponding to $K_u > 2.1 \times 10^6$ erg cm$^{-3}$, whereas, disordered $A1$ films with $x \geq 0.44$ possess PM properties at room temperature. These films change from a FM to PM state as the $L_1_0$ structure transforms into the $A1$ structure due to ion irradiation. However, the high Mn contents could decay corrosion resistance and $K_u$. By replacing Pt with Ru in the $L_1_0$ FePt films, which could be uninfluential to its corrosion resistance, a reduction of Curie temperature ($T_c$) has been reported [23].

In this study, by replacing Pt with Ru in $A1$ and $L_1_0$ FePt films, correlations between crystal structures and magnetic properties were investigated.

2. Materials and Methods

Fe$_{50}$(Pt$_{1-x}$Ru$_x$)$_{50}$ films with the thickness of 6.0 nm were deposited by magnetron cosputtering at a base pressure of $10^{-7}$ Pa using Ar gas at 0.5 Pa on a single-crystalline MgO (100) substrate at 298 K. The Ru composition ($x$) was controlled by varying the sputtering rates of the Pt and Ru targets and was detected using an electron probe X-ray microanalyzer. The films were annealed by rapid thermal annealing (RTA) with a heating rate of 300 K s$^{-1}$ under a vacuum of $2 \times 10^{-4}$ Pa. The crystalline structure was studied using out-of-plane X-ray diffraction (XRD) measurement and in-plane XRD measurement with CuKα radiation. A vibrating sample magnetometer (VSM) and a superconducting quantum interference device (SQUID) magnetometer with a maximum field of 18 kOe were used to assess the magnetic properties of the films.

3. Results and Discussion

Figure 1(a) shows out-of-plane XRD patterns for the as-deposited FePt$_{1-x}$Ru$_x$ films of thickness 6.0 nm. The Ru composition $x$ of the FePt$_{1-x}$Ru$_x$ films was changed from 0 to 1.00 and is shown from 0 to 0.30 with the background pattern (sample holder and substrate) in the figure. Only the background peaks were observed for all the films, since the fundamental (111) reflection peaks for the disordered $A1$ structure appear at the same angle as the MgO (100) substrate.

Figures 1(b)–1(e) show magnetization curves of the films with $x = 0, 0.10, 0.20, 0.30$, measured by the VSM with a maximum field of 18 kOe applied perpendicular (\(\perp\)) as dashed line and parallel (\(/\)) as solid line to the film plane at 298 K. The magnetic easy-axes of all the films are parallel to the film plane. Figure 1(f) shows the temperature ($T$) dependence of magnetization ($M$) ($M$-$T$ curve) for the film with $x = 0.30$ at temperatures at or below room temperature (300 K down to 30 K), measured using a SQUID magnetometer with a field of 1 kOe applied parallel to the film plane after saturating the magnetization by a field of 50 kOe at 298 K. $M$ decreases with an increase in $T$ and approaches 0 at around $T_c$ of 200 K. $T_c$ was estimated from the $M^2$-$T$ curve (not shown). This indicates that the film with $x = 0.30$ is in the PM phase at room temperature.

The $x$-dependence of $M_0$, determined using the Arrott plot [24] at 298 K, is shown in Figure 1(g). $M_0$ decreases with increasing $x$, and an abrupt decrease in $M_0$ is found at $x = 0.20$, caused by the decrease in $T_c$. This implies that the films with $0 \leq x < 0.20$ are in the PM phase, and the films with $x \geq 0.20$ are in the PM phase at room temperature.

Figures 2(a) and 2(b) show out-of-plane XRD patterns and in-plane XRD patterns, respectively, for the FePt$_{1-x}$Ru$_x$ films after annealing at 1173 K for 4 h with a heating rate of 300 K s$^{-1}$ by RTA. In Figure 2(a), only superlattice (001) and fundamental (002) reflection peaks for the ordered $L1_0$ structure are observed for the films with $0 \leq x \leq 0.70$, whereas only background peaks were observed for the films with $0.80 \leq x \leq 1.00$ due to overlapping of the fundamental (111) reflection peak for the disordered $A1$ structure and the reflection peaks for the MgO (100) substrate. In Figure 2(b), only the fundamental (200) reflection peak is observed for the films with $0 \leq x \leq 0.60$, whereas only background peaks are observed for the films with $0.70 \leq x \leq 1.00$. These reflection patterns indicate that the (001) crystalline texture is normal to the film plane in the films with $0 \leq x \leq 0.70$, whereas the disordered $A1$ structure is obtained in the films with $0.80 \leq x \leq 1.00$. This result is broadly consistent with the previous report [23]. The degrees of long-range chemical order parameter were estimated to be $\sim 0.90$ in the films with $0 \leq x \leq 0.60$.

The lattice constants ($a$, $c$, and $c/a$) are plotted as functions of $x$ in Figure 2(c). $c$ and $a$ values were obtained from the out-of-plane and in-plane XRD measurements, respectively. $a$ and $c$ values of the film with $x = 0$ are about 0.3867 nm and 0.3726 nm, respectively, and are close to those of the FePt bulk alloy ($a = 0.3852$ nm, $c = 0.3713$ nm; PDF number 43-1359). The values of $a$ are $\sim 0.39$ nm for each $x$ which could be kept by the epitaxial growth, whereas $c$ and the axial ratio ($c/a$) values decrease with increasing $x$. By considering the layer-by-layer atomic configuration in $L1_0$ FePt and the differences of atomic radii of Pt, Ru, and Fe ($Pt > Ru > Fe$), the
Figure 1: Crystal structure and magnetic properties of disordered $A1\text{FePt}_{1-x}\text{Ru}_x$ films before annealing (as-deposited films). (a) Out-of-plane XRD patterns. Magnetization curves of the films with $x =$ (b) 0, (c) 0.10, (d) 0.20, and (e) 0.30. Magnetic fields ($H$) were applied perpendicular ($\perp$, dashed line) and parallel ($//$, solid line) to the film plane at 298 K. (f) Temperature dependence of magnetization ($M$-$T$ curve) for the film with $x = 0.30$, measured from 300 K to 30 K with a field of 1 kOe applied parallel to the film plane after saturation. (g) $x$-dependence of $M_0$ at 298 K.
Figure 2: Crystal structure of FePt$_{1-x}$Ru$_x$ films after annealing at 1173 K for 4 h. (a) Out-of-plane XRD patterns and (b) in-plane XRD patterns. (c) Lattice constants ($a$, $c$, and $c/a$) as a function of $x$.

decrease in the lattice constant $c$ indicates the substitution of Ru for Pt.

Figures 3(a)–3(e) show magnetization curves of the films with $x = 0, 0.10, 0.20, 0.40$, and $0.60$, measured by the SQUID magnetometer with a maximum field of 50 kOe applied perpendicular ($\perp$, filled symbols) and parallel ($\parallel$, open symbols) to the film plane at 298 K. The magnetic easy-axes of all the films were perpendicular to the film plane. $K_u$ values were evaluated using the magnetization curves [25]. The magnetic anisotropy field ($H_A$) was determined from the intersection point of the extrapolated magnetization curves of the magnetic fields applied parallel and perpendicular to the film plane. Only the linear part of the in-plane magnetization curve was extrapolated. Consequently, $K_u$ was obtained using the relation $K_u = (M_s \times H_A/2) + K_{shape}$, where $K_{shape}$ is the shape anisotropy calculated using the demagnetization factors ($N$) of the film shape ($N_\perp = 4\pi, N_\parallel = 0$). The films with $0 \leq x \leq 0.50$ have high magnetization of $500 \leq M_s \leq 800$ emu cm$^{-3}$, high coercivity of $15 \leq H_c \leq 43$ kOe, and high anisotropy of $2.6 \times 10^7 \leq K_u \leq 5.0 \times 10^7$ erg cm$^{-3}$ in the perpendicular direction, and these results are broadly consistent with the previous report [23].

The $x$-dependence of $M_s$ and $K_u$ is plotted in Figure 3(f). $M_s$ and $K_u$ decrease with increasing $x$ and reach almost 0 at $x = 0.80$, which is the critical composition for the transition from the $L1_0$ structure to the $A1$ structure. For instance, $L1_0$
Figure 3: Magnetic properties of the FePt\(_{1-x}\)Ru\(_x\) films after annealing at 1173 K for 4 h. Magnetization curves of the ordered \(L1_0\) FePt\(_{1-x}\)Ru\(_x\) films with \(x = (a) 0, (b) 0.10, (c) 0.20, (d) 0.40,\) and (e) 0.60. \(H\) was applied perpendicular (\(\perp\), filled symbols) and parallel (\(/\), open symbols) to the film plane at 298 K. (f) \(x\)-dependence of \(M_0\) and \(K_u\) at 298 K.

Figure 4(a)–4(d) show the \(M-T\) curves of the films with \(x = 0.20, 0.40, 0.60,\) and 0.80 at temperatures at or below room temperature (300 K down to 30 K), measured using the SQUID magnetometer with a field of 1 kOe applied perpendicular to the film plane after saturating the magnetization by a field of 50 kOe at 298 K. In the films with \(x \leq 0.40\), \(M\) increases with a decrease in \(T\) due to the typical FM properties. However, at \(x = 0.60\), \(M\) reaches its maximum value at 210 K \((T_0)\) and decreases with a decrease in temperature below \(T_0\). This indicates that the film with \(x = 0.60\) can contain an antiferromagnetically ordered phase (spin-glass or canted-AF etc.) at temperatures below \(T_0\). In the films with \(x \geq 0.80\), \(M\) is almost 0 at each \(T\). These results imply that the films with \(0 \leq x \leq 0.40\) should have the FM–PM transition above 300 K, the films with \(0.40 < x < 0.80\) have the antiferromagnetically ordered phase FM–PM transition, and the films with \(0.80 \leq x \leq 1.00\) have the FM–PM transition below 300 K due to the \(A1\) structure.
Finally, we propose a nanopatterning method for this material system. According to our previous results, the ordered $L1_0$ structures of FePtX were easily transformed to the disordered $A1$ structure under ion irradiation [15–21]. Figure 5 shows the schematics of a nanopatterning method using an FM–PM phase change due to the $L1_0$–$A1$ structural transformation caused by ion irradiation. After ion irradiation, FM dots having $L1_0$ structure should be surrounded by PM spacing having $A1$ structure with a smooth disc surface.

4. Conclusions

Correlations between crystal structures and magnetic properties of FePt$_{1-x}$Ru$_x$ films (6.0 nm thick) were investigated. (1) Magnetic properties of $A1$ disordered structure (as-deposited films): $M_0$ decreased with increasing $x$, and an abrupt decrease in $M_0$ was found at $x = 0.20$ at room temperature, due to the decrease in $T_c$. In the range of $0 \leq x < 0.20$, the films had FM properties (100 ≤ $M_0$ ≤ 1050 emu cm$^{-3}$) with their magnetic easy-axes parallel to the film plane. In the range of $x \geq 0.20$, the films had PM properties ($M_0 = 0$ emu cm$^{-3}$) at room temperature.

(2) Magnetic properties of $L1_0$ ordered structure (annealed films): $M_0$ and $K_u$ decreased with increasing $x$. In the range of $0 \leq x \leq 0.50$, the films had FM properties (500 ≤ $M_0$ ≤ 800 emu cm$^{-3}$, 15 ≤ $H_c$ ≤ 43 kOe, 2.6 × 10$^7$ ≤ $K_u$ ≤ 5.0 × 10$^7$ erg cm$^{-3}$), with their magnetic easy-axis perpendicular to the film plane. In the range of 0.40 < $x$ < 0.80, the films could contain an antiferromagnetically ordered phase at temperatures below room temperature.

For instance, the $L1_0$ film with $x = 0.20$ had FM properties with a $K_u = 3.5 \times 10^7$ erg cm$^{-3}$, whereas the disordered $A1$ film with $x = 0.20$ had PM properties at
room temperature. The film could change from a FM to PM state as the L1₀ structure transforms into the A1 structure due to ion irradiation. These results suggest the possibility of applying the material system for nanopatterning method for high-density magnetic storage media.

**Conflicts of Interest**

The authors declare that there are no conflicts of interest regarding the publication of this article.

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