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Research Article

Structural Dependent Ferromagnetic-Nonmagnetic Phase Change in FePtRu Films

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Herein, we studied correlations between crystal structures and magnetic properties of $FePt_{1-x}Ru_x$ films. At room temperature, the chemical disordered $A1$ films with $0 \leq x < 0.20$ and $0.20 \leq x \leq 1.00$ exhibited ferromagnetic properties and paramagnetic properties, respectively. Curie temperature of the disordered film with $x = 0.30$ was 200 K. In contrast, the ordered $L1_0$ films had ferromagnetic properties in a wider range of $0 \leq x < 0.80$ with the magnetic easy-axis perpendicular to the film plane. For $0 \leq x \leq 0.50$, with the ordered structure, the films had high magnetization and high uniaxial magnetic anisotropy of over 10^7 erg cm^{-3}. For $x = 0.60$–0.70 with the ordered structure, a temperature-dependent magnetic phase transition appeared, and magnetization reached its maximum value at around 200 K. Using this material system, we proposed a nanopatterning method involving a ferromagnetic-paramagnetic phase change due to the ordered-disordered structural transformation.

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 2 Tb 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_BT; 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 $(K_uV/k_BT) > 60$ (this metric was derived using the Sharrock equation [10]) which is typically regarded as a minimum requirement for magnetic data storage [1, 11].

$L1_0$ FePt (CuAu-I type, fct, $a = b > c$) films with ordered structures are possible candidates for BPM due to their high K_u of 7.0×10^7 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 $L1_0$ 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 $L1_0$ 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_0) 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 $L1_0$ $Fe_{1-x}Mn_xPt$ films with $x \leq 0.44$ exhibit FM properties corresponding to $K_u > 2.1 \times 10^7$ 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 $L1_0$ 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 $L1_0$ 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 $L1_0$ 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^{-5} 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 2×10^{-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) with a maximum field of 18 kOe and a superconducting quantum interference device (SQUID) magnetometer with a maximum field of 50 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, and 0.30, measured by the VSM with a maximum field of 18 kOe applied perpendicular ($\perp$, dashed line) and parallel (//, 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 FM 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 ~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 ~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 radiuses of Pt, Ru, and Fe (Pt > Ru > Fe), the

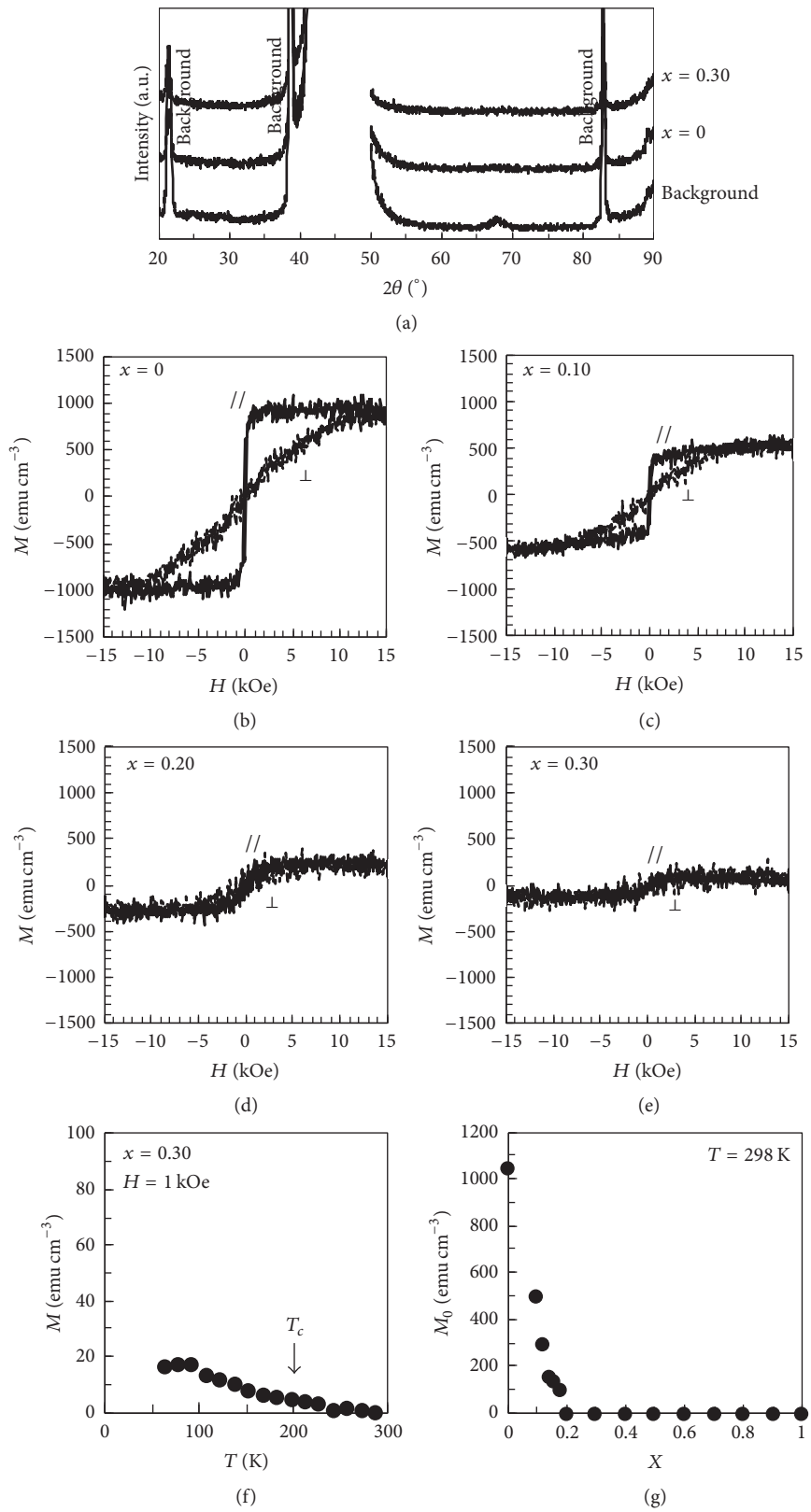

FIGURE 1: Crystal structure and magnetic properties of disordered $A1$ $FePt_{1-x}Ru_x$ films before annealing (as-deposited films). (a) Out-of-plane XRD patterns. Magnetization curves of the films with x = (b) 0, (c) 0.10, (d) 0.20, and (e) 0.30. Magnetic fields (H) were applied perpendicular ($\perp$, 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.

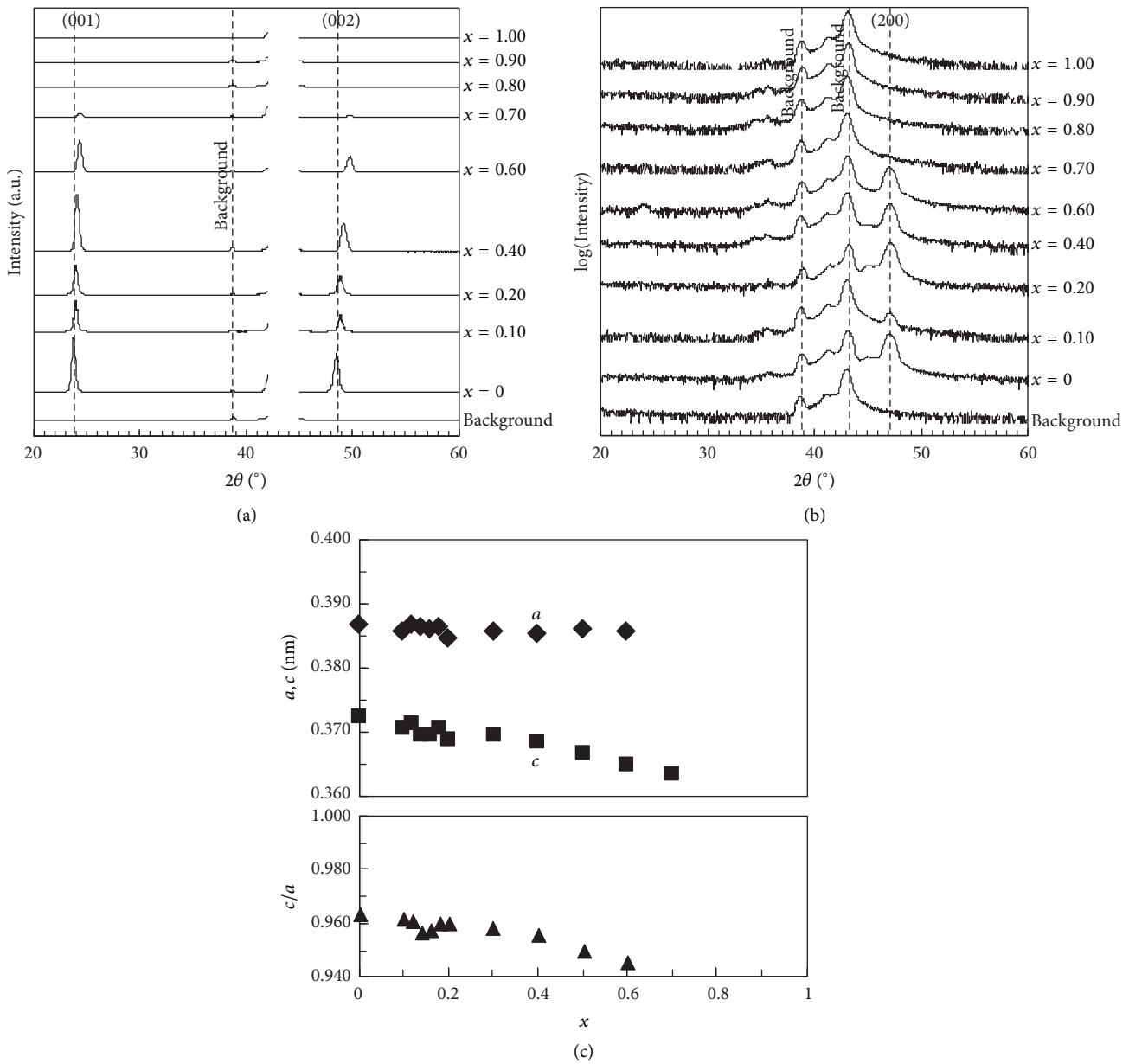

FIGURE 2: Crystal structure of $FePt_{1-x}Ru_x$ films after annealing at 1173 K for 4 h. (a) Out-of-plane XRD patterns and (b) in-plane XRD patterns. (c) Lattice constants (a, c, and c/a) as a function of x.

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 (//, 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_k) 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 $K_u = (M_s \times H_k/2) + K_{\text{shape}}$, where K_{shape} is the shape anisotropy calculated using the demagnetization factors (N) of the film shape ($N_\perp = 4\pi$, $N_{//} = 0$). The films with $0 \le x \le 0.50$ have high magnetization of $500 \le M_0 \le 800$ emu cm^{-3}, high coercivity of $15 \le H_c \le 43$ kOe, and high anisotropy of $2.6 \times 10^7 \le K_u \le 5.0 \times 10^7$ erg cm^{-3} in the perpendicular direction, and these results are broadly consistent with the previous report [23].

The x-dependence of M_0 and K_u is plotted in Figure 3(f). M_0 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, $L1_0$

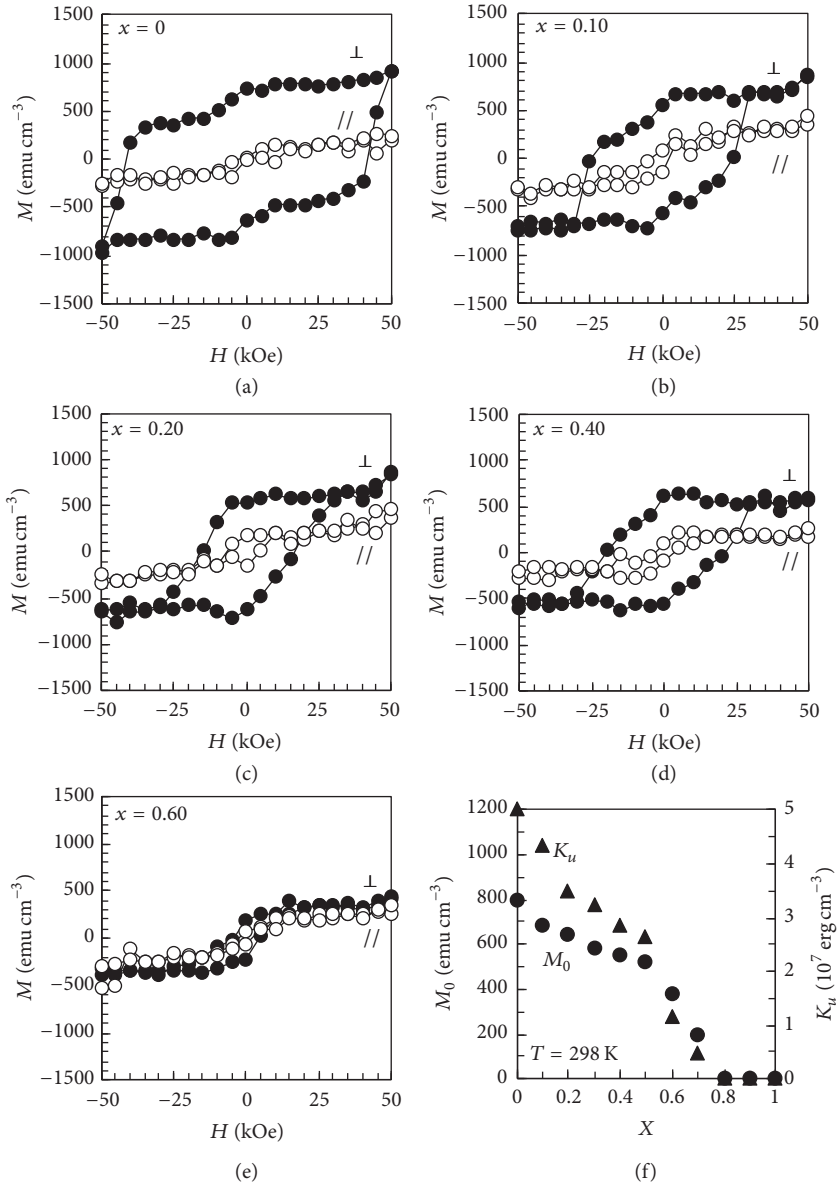

FIGURE 3: Magnetic properties of the $FePt_{1-x}Ru_x$ films after annealing at 1173 K for 4 h. Magnetization curves of the ordered $L1_0$ $FePt_{1-x}Ru_x$ films with x = (a) 0, (b) 0.10, (c) 0.20, (d) 0.40, and (e) 0.60. H was applied perpendicular ($\perp$, filled symbols) and parallel (//, open symbols) to the film plane at 298 K. (f) x-dependence of M_0 and K_u at 298 K.

film with $x = 0.20$ has FM properties with a $K_u = 3.5 \times 10^7$ erg cm^{-3}, whereas, the disordered $A1$ film with $x = 0.20$ has PM properties at room temperature. The film can change from a FM to PM state as the $L1_0$ structure transforms into the $A1$ structure due to ion irradiation. By using the thermal stability factor $(K_uV/k_BT) > 60$ and $K_u = 3.5 \times 10^7$ erg cm^{-3}, the smallest diameter of the spherical shape was estimated to be ~5.1 nm, which indicates that this material system has sufficient thermal stability for use in high-density magnetic data storage media of ~10 Tb in^{-2}.

Figures 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.

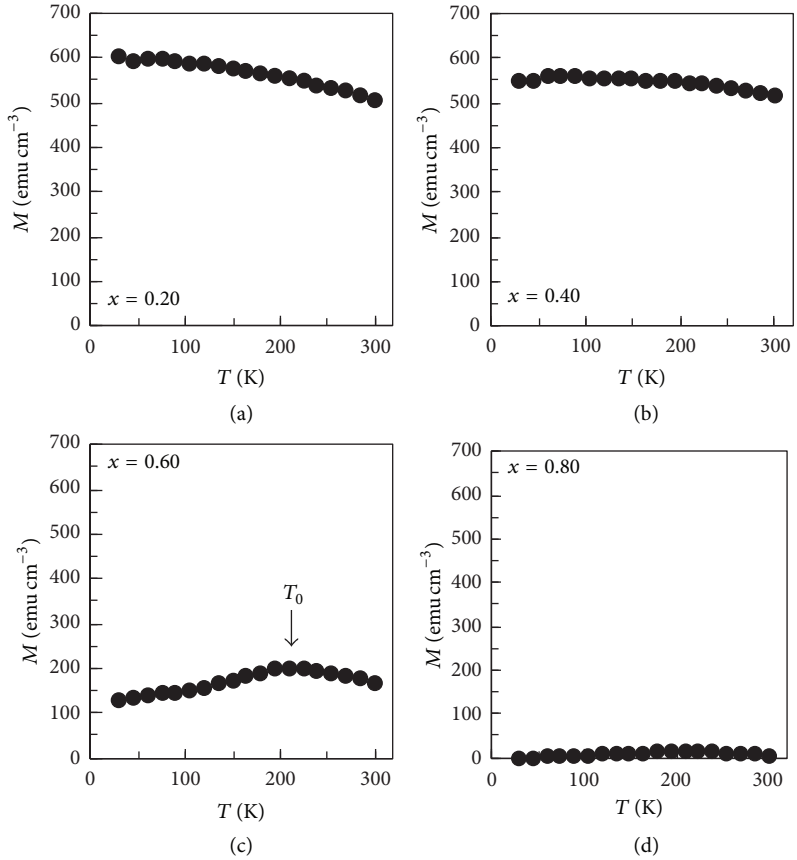

FIGURE 4: Temperature dependence of magnetization (M-T curves) for the $FePt_{1-x}Ru_x$ films after annealing at 1173 K for 4 h with x = (a) 0.20, (b) 0.40, (c) 0.60, and (e) 0.80, measured from 300 K to 30 K with a field of 1 kOe applied perpendicular to the film plane after saturating.

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.

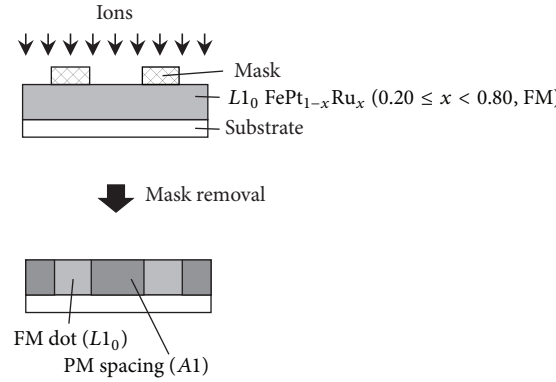

FIGURE 5: Schematics of the nanopatterning method using FM-PM phase change.

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 ($100 \leq M_0 \leq 1050$ 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 ($M_0 \approx 0$ 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 ($500 \leq M_0 \leq 800$ emu cm^{-3}, $15 \leq H_c \leq 43$ kOe, $2.6 \times 10^7 \leq K_u \leq 5.0 \times 10^7$ 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 $L1_0$ 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.

Acknowledgments

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