The formation of closely packed grains with $L_{10}$ ordering in gas-flow-sputtered FePt thin films

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The $L_{10}$ ordering and microstructures of FePt films fabricated by gas flow sputtering using two heating methods, i.e., heating during deposition and post-annealing in vacuum, were studied. A 60-nm-thick film with an order parameter above 0.6 was obtained at a heating temperature of 300°C using both methods. While the film that is heated during sputtering consists of closely packed fibrous grains, the post-annealing method produces a film containing voids. The differences in the microstructures of the films fabricated by the two heating methods are explained by means of a structure zone model.

Key words: FePt, Gas flow sputtering, Ordering, Microstructure, Coercivity, Structure zone model

1. INTRODUCTION
The high magnetocrystalline anisotropy—of the order of $10^7$ erg/cm$^3$—in FePt alloys with an $L_{10}$ structure [1] is an attractive property for magnetic recording applications. However, $L_{10}$ ordering is attained only by heat treatment during film deposition or by post-annealing. Heat treatments (including post-annealing) generally result in the growth of FePt grains, which is not conducive for the downsizing of the recording bits in magnetic recording applications.

One technique for fabricating fine FePt grains with $L_{10}$ ordering involves burying the FePt grains in matrix materials such as SiO$_2$ or C, and Al$_2$O$_3$ [2-5]. However, such granular thin films frequently suffer from problems that arise from the low density of granules in the matrix and wide particle-size distribution. In this paper, we present an alternative technique for obtaining fine and dense FePt grains with $L_{10}$ ordering using microstructures formed during the sputter deposition process. We discuss the formation of the microstructures using a structure zone model (SZM) [6-8].

We used gas-flow sputtering (GFS) for the deposition of FePt thin films. GFS is carried out at a high pressure of around 100 Pa using a hollow cathode discharge. The mean free paths of both Ar atoms and sputtered atoms are as short as 0.1 mm as a result of the high pressure. Energetic particles, including sputtered atoms and recoiled Ar atoms, lose their energies by collisions with ambient Ar atoms. Therefore, the mobility of adatoms on the growing film surface is very low, and it can be precisely controlled by heating the substrate.

2. Experiment
FePt films were deposited on SiO$_2$ glass substrates by GFS. The sputtering target was a tube (inner diameter of 3 cm and length of 6 cm) made of Fe, in which Pt plates were positioned. The target-to-substrate separation was 75 mm. The GFS chamber was evacuated to below $2.6 \times 10^{-5}$ Pa prior to sputtering, and film deposition was carried out at a pressure of 130 Pa and an Ar flow rate of 600 sccm. The sputtering power was approximately 360 W.

Two methods, A and B, were employed for heat treatment, and the samples were accordingly classified into two groups depending on the method employed. In method A, the substrate was heated during sputtering, whereas in method B, the film and the substrate were post-annealed in vacuum at a pressure below $7.0 \times 10^{-3}$ Pa for 30 min. Film deposition was carried out for 6 min and 2 min, producing a film with thicknesses of approximately 150 nm and 60 nm, respectively. The heating (annealing) temperature was 300°C and 250°C for each film thickness. Although the substrate is not intentionally heated in method B, heating due to plasma, which accompanies sputtering, occurs typically at temperatures below 80°C. Groups A and B were each further divided into two subgroups of thick- and thin-film thickness, and the eight samples that were fabricated were classified into these subgroups, as shown in Table 1.

<table>
<thead>
<tr>
<th>Sample group</th>
<th>Sample name</th>
<th>Heating method</th>
<th>Heating temperature (°C)</th>
<th>Film thickness (nm)</th>
<th>$x$ in Fe$<em>{50}$Pt$</em>{50}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A-thick</td>
<td>ACH</td>
<td>A</td>
<td>300</td>
<td>130</td>
<td>0.51</td>
</tr>
<tr>
<td>A-thick</td>
<td>ACL</td>
<td>A</td>
<td>250</td>
<td>162</td>
<td>0.64</td>
</tr>
<tr>
<td>A-thin</td>
<td>ANH</td>
<td>A</td>
<td>300</td>
<td>66</td>
<td>0.43</td>
</tr>
<tr>
<td>A-thin</td>
<td>ANL</td>
<td>A</td>
<td>250</td>
<td>49</td>
<td>0.44</td>
</tr>
<tr>
<td>B-thick</td>
<td>BCH</td>
<td>B</td>
<td>300</td>
<td>144</td>
<td>0.55</td>
</tr>
<tr>
<td>B-thick</td>
<td>BCL</td>
<td>B</td>
<td>250</td>
<td>146</td>
<td>0.54</td>
</tr>
<tr>
<td>B-thin</td>
<td>BNH</td>
<td>B</td>
<td>300</td>
<td>52</td>
<td>0.61</td>
</tr>
<tr>
<td>B-thin</td>
<td>BNL</td>
<td>B</td>
<td>250</td>
<td>63</td>
<td>0.56</td>
</tr>
</tbody>
</table>

Table 1. Classification of samples.
X-ray diffraction (XRD) patterns (θ–2θ scan) produced by Cu-Kα radiation were used for estimating the L1₀ order parameter (Bragg–Williams order parameter) [9]. The integrated intensities of the 001 superstructure peak and the 002 fundamental peak were obtained by fitting Gaussian curves to the Kα₁ and Kα₂ lines. The integrated intensities were corrected using the Lorentz-polarization factor for randomly oriented crystallites [10], the thermal factor (where the overall isotropic atomic displacement parameter was assumed to be 0.5 Å²), and the film-thickness factor [11] (where the linear absorption coefficient was assumed to be the mean value of those of Fe and Pt). The chemical composition of the samples was determined using inductively coupled plasma-atomic emission spectroscopy. The microstructure and film thickness were evaluated using field-emission scanning electron microscopy (FE-SEM). The coercivity of the films was determined from the in-plane magnetization curves measured using a vibrating sample magnetometer with a maximum field of 20 kOe. The film thickness and composition of the eight samples are shown in Table 1.

3. Results

XRD patterns of the samples are shown in Fig. 1. The peak-to-peak intensity ratios in all samples are almost equal to that of a powdered sample, suggesting that the crystallites are randomly oriented in the films. The order parameter estimated from the XRD patterns is shown in Fig. 2 as a function of heating (including annealing) temperature. The films fabricated by both methods show similar behavior. The order parameter of the thinner films is zero at a heating temperature of 250°C, whereas that of the thicker films is approximately 0.6. The thickness-dependent order parameter is generally observed in FePt films [12]. At a heating temperature of 300°C, the order parameter of all the films lies between 0.6 and 0.8. It is noted that the order parameter of the films fabricated both by methods A and B is higher than that reported in our previous paper [13]; the order parameter was 0.46 for a 50-nm-thick FePt film post-annealed in 3% H₂/Ar flow at 363°C.

Fig. 3(a) shows the coercivity of the films as a function of heating temperature. The coercivity increases with heating temperature for all sample groups. This increase in coercivity is mainly due to an increase in magnetocrystalline anisotropy with increasing order parameter [14]. Fig. 3(b) shows the coercivity as a function of order parameter. The coercivity increases with order parameter in each group; however, the correlation is poor when all data are viewed together. The change in the magnetization reversal mechanism originating from a change in the microstructure may also contribute to the change in coercivity.

SEM images of the surfaces and cross sections of the films are shown in Fig. 4. In addition to the images of the eight samples, those of the films fabricated without heating during deposition or post-annealing are shown for comparison. The thicker (198 nm) and thinner (76 nm) films are named as NC and NN, respectively. We
term the smallest unit recognizable in the SEM images of the surfaces as a “grain,” which is likely to be a crystallite (see circles in Fig. 4). There is a clear difference between the films fabricated by method A and those fabricated by method B; the grains in the films fabricated by method A are closely packed, whereas there are voids in those fabricated by method B. A columnar-like structure is observed in the cross-sectional images of the films fabricated by both methods, but it is more clearly visible in the films fabricated by method B because of the existence of voids. The grain size was measured for 100 grains in each film. Fig. 5 shows the grain size as a function of heating temperature. In BCH (Fig. 4(e)) and BCL (Fig. 4(f)) samples, the coalescence of grains is promoted, and grain size cannot be measured. The average grain size of NC and NN samples was 7.4 nm and 6.8 nm, respectively. In all groups, the average grain size slightly increases with heating temperature; however, the increase is within 5 nm. For thinner films, the difference in grain size of the samples obtained by employing methods A and B is small; the average and standard deviation of the grain size are below 15 nm and 2.1 nm, respectively.

From the above results, method A is found to be suitable for the fabrication of a film with closely packed grains. An FePt film with an order parameter of 0.76 and a grain size of 15 nm is obtained by heating the substrate at 300°C during film deposition.

4. Discussion

The formation of a film with closely packed grains using method A is discussed using an SZM. An SZM is

Fig. 3. Coercivity as a function of (a) heating temperature and (b) order parameter.
Formation of closely packed grains with $L1_0$ ordering in gas-flow-sputtered FePt thin films

Fig. 5. Grain size as a function of heating temperature. The plots and error bars represent average and standard deviation of particle size, respectively.

A general model describing the evolution of a microstructure in a film deposited by several methods such as evaporation [6], sputtering [7,8], and the sol-gel method [15]. The primary determinant factor of the microstructure is the mobility of adatoms on the growing film surface, which can be controlled by temperature, pressure in the sputtering chamber, and bias voltage applied to the growing film. Since the variable parameter in this study is temperature, we limit our discussion to the effects of temperature. The reduced temperature $T/T_m$, where $T$ denotes temperature and $T_m$ denotes the melting point of the film material, is essential in the SZM. For the lowest value of $T/T_m$ (typically $T/T_m < 0.3$ for conventional sputtering [7]), the mobility of adatoms is very low and the self-shadowing effect manifests itself, resulting in columnar structures with a high density of voids (zone 1). When the temperature is slightly elevated, the voids disappear, and closely packed columnar structures appear (zone T). When the temperature is further increased (typically $0.3 < T/T_m < 0.5$ [7]), the widths of the columnar structures increase (zone 2). Surface and grain-boundary diffusions are major mechanisms for the transport of adatoms in zones T and zone 2. The melting point of the equiatomic FePt alloy is 1580°C [16], and the heating temperatures of 250°C and 300°C correspond to $T/T_m$ ratios of 0.16 and 0.19, respectively. The microstructures of the films fabricated without heating (samples NC and NN) correspond to typical zone 1 structures, in which high densities of voids are observed. For films fabricated by method A, the microstructures in which the voids disappear correspond to zone T structures.

The microstructures of the films fabricated by method B cannot be discussed using the SZM since method B includes an annealing process after deposition. In method B, film deposition is carried out under the same conditions as those for samples NC and NN, and the films are post-annealed. Therefore, the voids formed during deposition are retained after annealing.

Thus, moderate heating during deposition in method A results in the appearance of zone T structures, in which fibrous grains are closely packed by surface and grain-boundary diffusion. It should be noted that the grain-boundary diffusion also plays an important role in $L1_0$ ordering, as discussed in the previous report [13]. An $L1_0$-ordered phase nucleates at the grain boundaries and grows into the interior of the disordered grains [17]. This ordering mechanism is termed “discontinuous ordering” [12].

5. Conclusion

A 60-nm-thick FePt film with an order parameter above 0.6 was fabricated by heating the substrate during GFS-deposition or by post-annealing at 300°C. While heating during deposition produced a film with closely packed fibrous grains, 15 nm in size, the post-annealing method produced a film containing voids. The closely grained microstructure obtained by heating during deposition corresponded to zone T in the SZM.

Acknowledgement

This study was supported by KAKENHI (21560348).

References


(Received June 13, 2011; Accepted November 22, 2011)