Magnetic properties and microstructure of Sm-Co/\(x\)-Fe nanocomposite thick film-magnets composed of multi-layers over 700 layers

A. Tou, a) T. Morimura, M. Nakano, T. Yamai, and H. Fukunaga

Graduate School of Engineering, Nagasaki University, 1-14 Bunkyo-machi, Nagasaki 852-8521, Japan

(Submitted 6 November 2013; received 23 September 2013; accepted 22 November 2013; published online 12 March 2014; corrected 17 March 2014)

We synthesized Sm-Co/\(x\)-Fe nanocomposite film-magnets, approximately 10 \(\mu\)m in thickness, composed of 780 layers by the pulse laser deposition method. Transmission electron microscopic observations revealed that the synthesized film is composed of Sm-Co and \(x\)-Fe layers with the well-controlled \(x\)-Fe thickness of approximately 10–20\(\text{nm}\), which is suitable one predicted by the micromagnetic simulation. In spite of the enhanced interlayer diffusion of Fe and Co by annealing for crystallization, the \((BH)_{\text{max}}\) value of 100\(\text{kJ/m}^3\) was obtained at the averaged compositions of Sm/(Sm + Co) = 0.16 and Fe/(Sm + Co + Fe) = 0.47. The \(x\)-Fe fraction for obtaining the highest \((BH)_{\text{max}}\) value was smaller than that expected from the micromagnetic simulation. Although the annealing for crystallization lay the easy direction of magnetization in the plane, the film is not expected to have strong crystallographic texture.

I. INTRODUCTION

A Sm-Co/\(x\)-Fe nanocomposite magnet is one of attractive candidates for high performance magnets used at high temperatures\(^1\) because of high Curie temperatures of Sm-Co alloys.\(^2\) Thus, Sm-Co/\(x\)-Fe nanocomposite magnets have been intensively studied.\(^3\)–\(^12\) In particular, \((BH)_{\text{max}}\) values exceeding the theoretical limit of SmCo\(_5\) have been reported in multi-layered and tri-layered SmCo\(_5/\text{-Fe}\) films,\(^11,12\) which suggests that a layered structure has an advantage in controlling a microstructure.

On the other hand, we need to increase the film thickness for applying them to magnetic devices such as MEMS. We have already reported that we could synthesize multi-layered thick film-magnets composed of several hundred layers by the pulse laser deposition (PLD) method.\(^13\) Therefore, an application of this method to the synthesis of Sm-Co/\(x\)-Fe nanocomposite magnets is expected to enable us to obtain multi-layered Sm-Co/\(x\)-Fe nanocomposite thick film-magnets applicable to magnetic devices such as MEMS.

From the above point of view, we synthesized Sm-Co/\(x\)-Fe nanocomposite thick film-magnets and reported magnetic properties for the films including approximately 25 and 50 vol.\% \(x\)-Fe as well as the microstructure of an as-deposited film.\(^14\) It was also confirmed that the crystallized film-magnets had \((BH)_{\text{max}}\) values higher than those of Nd-Fe-B/\(x\)-Fe nanocomposite thick film magnets and (Nd, Dy)-Fe-B flakes at 423 K.\(^14\)

In this contribution, we report magnetic properties of multi-layered Sm-Co/\(x\)-Fe nanocomposite thick film-magnets including approximately 25–75 vol.\% \(x\)-Fe, their magnetic anisotropy, and the microstructure after crystallization. The obtained magnetic properties are discussed in terms of the results of our micromagnetic simulation.

II. EXPERIMENTAL PROCEDURE

Composite targets, which are composed of Sm\(_x\)Co\(_5\) (\(x = 1\)–5) and \(x\)-Fe segments, were prepared and the area fractions of the \(x\)-Fe segment were set to 0, 1/4, 1/2, and 3/4. Subsequently, Sm-Co/\(x\)-Fe films were deposited on a Ta substrate. The target was rotated during the ablation. The as-deposited films, approximately 10 \(\mu\)m in thickness, were composed of 780 layers (amorphous Sm-Co and crystalline \(x\)-Fe layers) and did not exhibit hard magnetic properties. Consequently, they were crystallized by annealing. Detailed preparation condition of Sm-Co/\(x\)-Fe films has been reported elsewhere.\(^13,14\)

Magnetic properties and thickness of the films were measured with a vibrating sample magnetometer (VSM) and a digital point micrometer, respectively. Details have been also reported elsewhere.\(^13\) Morphology and microstructure were analyzed with an X-ray diffractometer, a scanning electron microscope (SEM), and a transmission electron microscope (TEM). The composition of the films was analyzed with an energy dispersive X-ray spectrometer (EDS).

III. RESULTS AND DISCUSSION

A. Magnetic properties

In the previous report,\(^14\) it was found that the increase in the area fraction of the \(x\)-Fe segment from 1/4 to 1/2 increases \((BH)_{\text{max}}\). On the other hand, the micromagnetic simulations for SmCo\(_5/\text{-Fe}\) powder\(^1\) and multi-layered films\(^15\) predicted that \(x\)-Fe can be included up to 80 vol.\%. Therefore, we deposited films with varying area fraction of the \(x\)-Fe segment (1/4, 1/2, or 3/4) and the Sm content \(x\) of SmCo\(_5\) segment (\(x = 1\)–5), and crystallized them. The \((BH)_{\text{max}}\) values obtained in the in-plane direction is shown in Fig. 1 as a function of the Sm content, Sm/(Sm + Co). The \((BH)_{\text{max}}\) vs Sm/(Sm + Co) curve showed a clear peak, and the peak shifted to the Sm-rich composition with increasing the area fraction of \(x\)-Fe segment. In contradiction to the micromagnetic
prediction, the increase in the area fraction of the $\alpha$-Fe segment over $1/2$ decreased $(BH)_{max}$. Resultantly, the highest $(BH)_{max}$ value of 100 kJ/m$^3$ was obtained for the averaged composition of Sm/(Sm + Co) $= 0.16$ and Fe/(Sm + Co + Fe) $= 0.47$. This value is much smaller than that predicted by the micromagnetic simulation ($BH)_{max} / C^2 = 800$ kJ/m$^3$ for anisotropic magnets and $\sim 300$ kJ/m$^3$ for isotropic ones). These differences between the experimental and micromagnetic results can be attributed to the fact that $H_c$ obtained experimentally is much smaller than that of the simulation ($H_c / C^2 = 800$ kA/m for anisotropic magnets and 450 kA/m for isotropic ones).

In order to evaluate magnetic anisotropy of the films, we measured in-plane hysteresis loops in the two directions perpendicular to each other and an out-of-plane hysteresis loop. It was found that the prepared film-magnet is isotropic in the plane. The out-of-plane and in-plane hysteresis loops are shown in Fig. 2. The out-of-plane hysteresis loop is corrected by using the demagnetizing factor of 1. As shown in the table of Fig. 2, the remanence $J_r$ and $H_c$ of the in-plane hysteresis loop are larger than those of the out-of-plane hysteresis loop, which suggests that the easy direction of magnetization lies in the plane. This result is consistent with the previous report on a SmCo$_5$ film.16,17

We calculated the energy $E_{mag}$ needed to magnetize the film from the demagnetized state ($M = 0, H = H_c$) to saturation, as shown schematically in the inset. $E_{mag}$ values were determined to be approximately 760 and 1080 kJ/m$^3$ for the in-plane and out-of-plane loops, respectively. Therefore, the magnetic anisotropy energy between the in- and out-of-plane directions was approximately 320 kJ/m$^3$. This value is much smaller than the anisotropy constants of SmCo$_5$ (12 MJ/m$^3$), Sm$_2$Co$_7$ (12 MJ/m$^3$), and SmCo$_5$ (17 MJ/m$^3$), suggesting that the film does not have strong crystallographic texture. Although the definition of $E_{mag}$ indicated in the inset of Fig. 2 may overestimate the magnetizing energy to be calculated from the initial magnetization curve, the overestimated energies are much smaller than magnetic anisotropy energies of the Sm-Co alloys mentioned above. Thus, the overestimation does not affect the above conclusion on crystallographic texture.

**B. Microstructure**

Figures 3(a) and 3(b) show TEM micrographs before and after crystallization. The averaged Sm/(Sm + Co) and Fe/(Sm + Co + Fe) value determined by SEM-EDS are 0.16 and 0.57, respectively. The layered structure was kept after crystallization as shown in Fig. 3(b). The results of EDS line scan across layers shown in Fig. 4 also indicate the layered structure after crystallization. The thickness of $\alpha$-Fe layers is approximately 10–20 nm. We have carried out the micromagnetic simulation for multi-layered isotropic SmCo$_5$/$\alpha$-Fe magnets with varying thicknesses of $\alpha$-Fe thickness and predicted that a large $(BH)_{max}$ value can be obtained for the thickness of $\alpha$-Fe layers around 10–15 nm.18 Therefore, our magnets have a suitable $\alpha$-Fe layer thickness.

As shown in Fig. 5, we can observe diffraction peaks from $\alpha$-Fe and several Sm-Co compounds in the X-ray diffraction patterns after crystallization. The peak around $2\theta = 42^\circ$ is very broad, which suggests that amorphous part and/or very small crystalline remain after crystallization.

The composition of each layer was analyzed by electron-beam, 10 nm in diameter (Table I). Before crystallization, the composition of $\alpha$-Fe layers is Sm/(Sm + Co) $= 0.16$ and Fe/(Sm + Co + Fe) $= 0.53$. The out-of-plane hysteresis loop was corrected by using the demagnetizing factor of 1.
crystallization, 6.0 at. % of Co and 15.0 at. % of Fe were detected in a \( \alpha \)-Fe and Sm-Co layers, respectively. The Co and Fe contents increased to 13.7% and 21.0% after crystallization. The above results strongly suggest that the interlayer diffusion of Co and Fe between \( \alpha \)-Fe and Sm-Co layers was enhanced by the annealing for crystallization. The results of EDS line scan shown in Fig. 4 also indicate the enhancement of the interlayer diffusion due to the annealing for crystallization. Similar behaviors have been reported for a layered structure,\(^6\) a bulk magnet,\(^8,19\) and a mechanically alloyed powder.\(^20\) In the Sm-Co layer, Sm/(Sm + Co) and Sm/(Sm + Co + Fe) were 0.228 and 0.181. These values are much higher than that of the stoichiometric composition of SmCo\(_5\), which may be one of reasons why Sm-rich Sm-Co phases such as SmCo\(_3\) and Sm\(_2\)Co\(_7\) were observed after crystallization.

**IV. CONCLUSION**

We synthesized Sm-Co/\( \alpha \)-Fe nanocomposite thick film-magnets by the PLD method. The film-magnets are approximately 10 \( \mu \)m in thickness and composed of 780 layers. Main results are summarized as follows:

1. The highest \((BH)_{\text{max}}\) value was obtained at the averaged compositions of Sm/(Sm + Co) = 0.16 and Fe/(Sm + Co + Fe) = 0.47. The obtained \( J_r \), \( H_c \), and \((BH)_{\text{max}}\) values were 1.17 T, 315 kA/m, and 100 kJ/m\(^3\), respectively. The obtained \((BH)_{\text{max}}\) value was smaller than that predicted by the micromagnetic simulation.

2. The obtained films were magnetically anisotropic, and their easy direction of magnetization lies in the plane. However, the films did not have strong crystallographic texture.

3. TEM observation revealed that the above film has the well-controlled \( \alpha \)-Fe thickness of 10–20 nm, which is a suitable thickness predicted by the micromagnetic simulation for an isotropic layered structure.

4. The annealing enhanced the interlayer diffusion of Fe and Co.

**TABLE I.** Composition of \( \alpha \)-Fe and Sm-Co layers before and after crystallization. The analysis was carried out with an electron beam with 10 nm diameter.

<table>
<thead>
<tr>
<th></th>
<th>Sm (at. %)</th>
<th>Co (at. %)</th>
<th>Fe (at. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-deposited</td>
<td>18.1</td>
<td>66.9</td>
<td>15.0</td>
</tr>
<tr>
<td>Sm-Co</td>
<td>As-annealed</td>
<td>18.1</td>
<td>61.0</td>
</tr>
<tr>
<td>As-deposited</td>
<td>0.6</td>
<td>6.0</td>
<td>93.4</td>
</tr>
<tr>
<td>( \alpha )-Fe</td>
<td>As-annealed</td>
<td>0.6</td>
<td>13.7</td>
</tr>
</tbody>
</table>

\(^3\)C. Rong et al., J. Appl. Phys. 109, 07A735 (2011).
\(^7\)C. Rong et al., J. Mater. Sci. 46, 6065 (2011).
\(^12\)Y. Neu et al., IEEE Trans. Magn. 48, 3599 (2012).
\(^17\)R. Horikawa et al., in the 58th MMM Conference, BQ-17, 2013.