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<td>Author(s)</td>
<td>Tou, Akinori; Morimura, Takao; Nakano, Masaki; Yanai, Takeshi; Fukunaga, Hirotoshi</td>
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Magnetic properties and microstructure of Sm-Co/\(\alpha\)-Fe nanocomposite thick film-magnets composed of multi-layers over 700 layers

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

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

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We synthesized Sm-Co/\(\alpha\)-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 \(\alpha\)-Fe layers with the well-controlled \(\alpha\)-Fe thickness of approximately 10–20 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 kJ/m\(^3\) was obtained at the averaged compositions of Sm/(Sm + Co) = 0.16 and Fe/(Sm + Co + Fe) = 0.47. The \(\alpha\)-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. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4866842]

I. INTRODUCTION

A Sm-Co/\(\alpha\)-Fe nanocomposite magnet is one of attractive candidates for high performance magnets used at high temperatures because of high Curie temperatures of Sm-Co alloys.\textsuperscript{2} Thus, Sm-Co/\(\alpha\)-Fe nanocomposite magnets have been intensively studied.\textsuperscript{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\(_5/\alpha\)-Fe films,

\(a\)Author to whom correspondence should be addressed. Electronic mail: bb52112226@cc.nagasaki-u.ac.jp.

\(\frac{1}{4}, \frac{1}{2}, \text{or }\frac{3}{4}\) and the Sm content \(x\) of Sm\(_x\)Co\(_5\) segment

II. EXPERIMENTAL PROCEDURE

Composite targets, which are composed of Sm\(_x\)Co\(_5\) (\(x = 1-5\)) and \(\alpha\)-Fe segments, were prepared and the area fractions of the \(\alpha\)-Fe segment were set to 0, 1/4, 1/2, and 3/4. Subsequently, Sm-Co/\(\alpha\)-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 \(\alpha\)-Fe layers) and did not exhibit hard magnetic properties. Consequently, they were crystallized by annealing. Detailed preparation condition of Sm-Co/\(\alpha\)-Fe films has been reported elsewhere.\textsuperscript{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.\textsuperscript{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,\textsuperscript{14} it was found that the increase in the area fraction of the \(\alpha\)-Fe segment from 1/4 to 1/2 increases \((BH)_{\text{max}}\). On the other hand, the micromagnetic simulations for SmCo\(_5/\alpha\)-Fe powder\textsuperscript{1} and multi-layered films\textsuperscript{15} predicted that \(\alpha\)-Fe can be included up to 80 vol. %. Therefore, we deposited films with varying area fraction of the \(\alpha\)-Fe segment (1/4, 1/2, or 3/4) and the Sm content \(x\) of Sm\(_x\)Co\(_5\) 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 \((BH)_{\text{max}}\) value versus Sm/(Sm + Co) curve showed a clear peak, and the peak shifted to the Sm-rich composition with increasing the area fraction of \(\alpha\)-Fe segment. In contradiction to the micromagnetic...
the increase in the area fraction of the $\alpha$-Fe segment over $1/2$ decreased $(BH)_{\text{max}}$. Resultantly, the highest $(BH)_{\text{max}}$ value of 100 kJ/m$^3$ was obtained for the averaged composition of $\text{Sm/(Sm} + \text{Co)} = 0.16$ and $\text{Fe/(Sm + Co + Fe)} = 0.57$. This value is much smaller than that predicted by the micromagnetic simulation ($\text{C24}/(BH)_{\text{max}} \approx 800$ kJ/m$^3$ for anisotropic magnets and $\text{C24}/(BH)_{\text{max}} \approx 300$ 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 ($H_c \approx 800$ 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 inset schematically shows the method of calculating the energy for magnetizing a film from the demagnetizing state to saturation.

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 $(BH)_{\text{max}}$ value can be obtained for the thickness of $\alpha$-Fe layers around 10–15 nm. 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, 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,\(^8,\) 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.

**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>
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<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>

**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.