Formation of SmFe$_5$(0001) Ordered Alloy Thin Films on Cu(111) Single-Crystal Underlayers

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Abstract. SmFe$_5$(0001) single-crystal thin films are prepared by molecular beam epitaxy employing Cu(111) single-crystal underlayers on MgO(111) substrates. The Cu atoms diffuse into the Sm–Fe layer and substitute the Fe sites in SmFe$_5$ structure forming an alloy compound of Sm(Fe,Cu)$_5$. The Sm(Fe,Cu)$_5$ film is more Cu enriched with increasing the substrate temperature. The Cu underlayer plays an important role in assisting the formation of the ordered phase.

1. Introduction

High $K_u$ magnetic thin films of rare earth–transition metal alloys like SmCo$_5$ have attracted much attention to applications for magnetic recording media, thin film magnets, etc. For such applications, high-quality epitaxial thin films are the strong candidates since the film uniformity and the magnetic anisotropy are well controlled, and can be fabricated into nano-patterns. For magnetic recording media applications, high $K_u$ magnetic thin films with perpendicular magnetic anisotropy have been investigated. SmCo$_5$ polycrystalline [1–4] and SmCo$_5$ epitaxial [5–7] thin films with (0001) texture have been prepared by employing Cu(111) underlayers. Cu atom dissolution is known to stabilize the ordered phase formation through forming an alloy compound of Sm(Co,Cu)$_5$ [4,5,8]. Recently, SmFe$_5$ ordered phase, which is metastable and does not appear in the Sm–Fe phase diagram, has been successfully prepared in the forms of film and powder samples [9–11]. The material showed high magnetic anisotropy field of 40 kOe [11]. However, there are no reports on the formation of epitaxial thin film with (0001) texture. In the present study, Sm–Fe films were prepared by employing Cu(111) single-crystal underlayers. The effect of substrate temperature on the ordered phase formation was investigated.

2. Experimental procedure

Thin films were prepared on polished MgO(111) substrates using a molecular beam epitaxy system under base pressures lower than 7×10$^{-9}$ Pa. Pure Fe (99.99%) was evaporated by electron beam heating, while pure Sm (99.99%) and Cu (99.99%) were evaporated by using Knudsen cells. The film layer structure was Sm–Fe(20 nm)/Cu(20 nm)/MgO(111). A 20-nm-thick Cu(111) single-crystal underlayer was prepared by hetero-epitaxial growth on the MgO substrate at 500 °C. The epitaxial
A 20-nm-thick Sm–Fe layer was formed on the Cu underlayer at a substrate temperature between 300 and 500 °C by co-evaporation of Sm and Fe sources. The layer composition was adjusted to be within Fe – 16.7±0.5 at.% Sm (SmFe₅ stoichiometry) and it was confirmed by energy dispersive X-ray spectroscopy (EDX).

The surface structure during film deposition was studied by in-situ reflection high energy electron diffraction (RHEED). The film structure was investigated by out-of-plane (2θ/ω-scan), in-plane (2θχ/φ-scan), and φ-scan X-ray diffraction (XRD) with Cu–Kα radiation (λ=0.15418 nm).

3. Results and discussion

The effects of substrate temperature on RHEED pattern observed during Sm–Fe layer deposition are shown in figures 1(a)–1(e). The RHEED results show that SmFe₅(0001) epitaxial thin films with CaCu₅ structure are successfully prepared on Cu(111) underlayers at a substrate temperature between 300 and 500 °C by co-evaporation of Sm and Fe sources. The layer composition was adjusted to be within Fe – 16.7±0.5 at.% Sm (SmFe₅ stoichiometry) and it was confirmed by energy dispersive X-ray spectroscopy (EDX).

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variants, Nishiyama–Wasserman (NW) and Kurdjumov–Sachs (KS) relationships [12], are co-existing with the SmFe₅(0001) phase. The RHEED intensity from Fe(110) gradually decreases with increasing the Sm–Fe layer thickness. The increase of amorphous phase volume and the decrease of Fe(110) phase volume with increasing the layer thickness may be explained by considering the behavior of Cu atom diffusion from the underlayer into the Sm–Fe layer. Since the films are grown at elevated temperatures, the Sm–Fe films are expected to include Cu atoms diffused from the underlayer, and thus Sm(Fe,Cu)₅ ordered alloy layers are presumably formed, similar to the cases of Sm(Co,Cu)₅ ordered phase formation on Cu underlayers [3–6]. In the initial stage of Sm–Fe film growth on Cu underlayer, an Sm(Fe,Cu)₅ ordered phase is presumably formed through Cu atom diffusion and partial substitution of Fe sites in the SmFe₅ structure. Some Fe atoms which are replaced by Cu atoms in the ordered structure may be pushed out to form Fe(110) epitaxial crystals on the Cu underlayer. The number of Cu atoms that diffuse up to the Sm–Fe surface layer will decrease with increasing the Sm–Fe layer thickness, and thus the probability of both the Fe site replacement with Cu atom and the Fe(110) crystal formation will decrease. Cu atom diffusion will also decrease at lower substrate temperatures. Therefore, in these cases, the probability of amorphous phase formation will increase at lower substrate temperatures and when the layer thickness is increased. On the other hand, in the Sm–Fe film prepared at 500 °C, an amorphous phase is appearing with increasing the Sm–Fe layer thickness, though Cu atom diffusion into the Sm–Fe layer is considered to be enough to promote the Sm(Fe,Cu)₅ phase formation. This result suggests that too much Cu atoms tend to destabilize the Sm(Fe,Cu)₅ phase formation. To investigate an effect of Cu rich composition on the ordered phase formation, Sm–Cu layer was formed on the Cu(111) single-crystal underlayer at 500 °C. Here, the film was prepared by carefully adjusting the composition to be within 16.7±0.5 at.% Sm (SmCu₅ stoichiometry). Figure 1(f) shows the RHEED patterns observed during Sm–Cu layer formation. With increasing the Sm–Cu layer thickness, diffuse RHEED pattern typical for an amorphous phase starts to overlap with the RHEED pattern from the ordered phase. Considering this behaviour, too much Cu atom diffusion into the Sm–Fe layer could be a possible reason for the increase of amorphous phase in the Sm–Fe film prepared at 500 °C. From these results, in order to form the Sm(Fe,Cu)₅ phase, it is important to control the amount of Cu atoms diffused from the underlayer into the Sm–Fe layer.

The epitaxial orientation relationship between the Sm(Fe,Cu)₅ layer and the Cu underlayer is determined by RHEED observation and ex-situ φ-scan XRD analysis. Figure 2 (c) shows the enlarged view of RHEED pattern observed for a 20-nm-thick Sm–Fe layer grown on a Cu(111) underlayer at 450 °C. The superlattice spots of 0003, 1103, etc. are clearly observed. The epitaxial orientation
relationship of Sm(Fe,Cu)₅(0001)[1120] // Cu(111)[110] is determined by comparing the RHEED patterns observed for the Sm(Fe,Cu)₅ layer and the Cu underlayer. The φ scan XRD data shown in figure 3 also indicate that the Sm(Fe,Cu)₅(0001) single-crystal film is formed on the Cu(111) underlayer hetero-epitaxially grown on an MgO(111) substrate.

The lattice constants of the ordered phase are determined by out-of-plane and in-plane XRD analyses. For the Sm–Fe films prepared in a temperature range between 350 and 500 °C, the out-of-plane XRD spectra (figures 4(a)–4(d)) show that the SmFe₅(0001) superlattice peak is observed in addition to the SmFe₅(0002) fundamental peak, whereas the in-plane XRD spectra measured along the MgO[110] direction (figures 4(e)–4(h)) show SmFe₅(1120) peak. The SmFe₅(0001) and SmFe₅(1120) XRD peaks observed for the Sm–Fe film prepared at 450 °C are strongest as expected from the RHEED observation. Furthermore, the intensity of Fe(110) peak observed in the out-of-plane XRD increases with increasing the substrates temperature. The result suggests that the ratio of Fe atom substituted by Cu atom increases with increasing the substrate temperature. Figure 5 summarizes the lattice parameters of the Sm–Fe ordered phase formed on Cu underlayers. The lattice parameter values are between those of bulk SmFe₅ and bulk SmCu₅ alloys [11,13]. With increasing the substrate temperature, the lattice parameter, c, decreases (figure 5(a)), whereas the lattice parameter, a, increases (figure 5(b)). The lattice parameters of Sm–Fe film prepared at 500 °C are very close to those of bulk SmCu₅ alloy. These results apparently indicate that large amounts of Cu atom are diffusing into the Sm–Fe epitaxial film forming an alloy compound of Sm(Fe,Cu)₅ and that the Sm(Fe,Cu)₅ phase is more Cu enriched for samples prepared at higher substrate temperatures.

4. Conclusion
The epitaxial growth of SmFe₅ ordered crystal was investigated on Cu(111) underlayer. SmFe₅(0001) single-crystal films were successfully obtained. A high-quality SmFe₅ film is obtained at a substrate temperature of 450 °C. During film growth at elevated temperatures, Cu atoms diffuse from the underlayer into Sm–Fe layer and substitute parts of Fe sites in the SmFe₅ ordered structure forming an alloy compound of Sm(Fe,Cu)₅. The Sm(Fe,Cu)₅ layer is more Cu enriched with increasing the substrate temperature. The Cu atom diffusion into the Sm–Fe layer plays an important role in assisting the formation of Sm(Fe,Cu)₅ ordered phase.

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