STRUCTURE AND MAGNETIC PROPERTIES OF EPITAXIAL CrPt₃ AND Mn_{0.2}Cr_{0.8}Pt₃ ALLOY FILMS

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Abstract - Epitaxial CrPt₃ and Mn_{0.2}Cr_{0.8}Pt₃ films were grown by MBE system and the structure and the magnetic and magneto-optical properties of these films have been investigated. RHEED observation suggests that the CrPt₃(111) film epitaxially grows on MgO(111) and forms Cu₃Au type ordered structure. The easy axis of magnetization of CrPt₃ on MgO(111) lies in the film plane although the CrPt₃ on fused quartz substrate exhibits large perpendicular anisotropy ($K_u = 1.5 \times 10^6$ erg/cm³). The perpendicular magnetic anisotropy of Mn_{0.2}Cr_{0.8}Pt₃ is induced when the 30 nm Pt buffer layer is inserted at the interface between the alloy layer and the MgO(111).

Key word: magnetic anisotropy, Kerr spectra, epitaxial film, CrPt₃, MnCrPt₃

1. Introduction

CrPt₃ and MnPt₃ ordered alloys with Cu₃Au structure exhibit large magneto-optical Kerr effects around 1.2 eV[1, 2]. Recently, we reported that the polycrystalline CrPt₃ alloy films prepared by sputter-deposition and post annealing at 800 °C exhibited a large perpendicular magnetic anisotropy[3] although sputter-deposited MnPt₃ films did not show perpendicular anisotropy, i.e., the easy axis of magnetization lay in the film plane[1]. Ordered CrPt₃ phase has highly symmetric structure of Cu₃Au type where Cr and Pt atoms are located at the corner and face center, respectively. This suggests that the uniaxial magnetocrystalline anisotropy of CrPt₃ is considered to be small.

In this paper, we have attempted to prepare highly oriented epitaxial CrPt₃ films by using a molecular beam epitaxy (MBE) system, and investigated the relationship between the magnetic anisotropy and the crystal structure. In the preliminary study, the preparation of epitaxial CrPt₃ films requires a heat treatment at 850 °C for ordering. This makes it difficult to insert a buffer layer into the interface between the CrPt₃ and the substrate without intermixing. In order to lower the ordering temperature, we prepared Mn_{0.2}Cr_{0.8}Pt₃ alloy films since the ordering temperature of bulk MnPt₃ is lower than that of CrPt₃[4]. In addition, highly ordered epitaxial MnPt₃ films were found to grow by MBE at a temperature of 300 °C[5] which is much lower than order-disorder transition temperature of ~1000 °C[4].

2. Experiment

CrPt₃ alloy films were prepared by using an MBE apparatus at a pressure less than 3×10^{-9} Torr in the following way. MgO(111) and fused quartz were used as the substrates for the growth of epitaxial and polycrystalline

samples, respectively. The substrates were kept at 600 °C during the deposition. The alloy films were deposited by the co-evaporation from independently controlled Cr (purity 99.9%) and Pt (purity 99.99%) e-beam sources at rates of 0.005 and 0.019 nm / sec, respectively. The total thickness of CrPt₃ films is 60 nm and the films were post-annealed at 850 °C for ordering in the MBE chamber. Mn_{0.2}Cr_{0.8}Pt₃ films were grown at a temperature of 500 °C onto the MgO(111) and fused quartz substrates without post annealing. In the growth of Mn_{0.2}Cr_{0.8}Pt₃, two kinds of samples were prepared: with and without 30 nm Pt buffer layer.

The structure of the films was characterized by *in-situ* Reflection High Energy Electron Diffraction (RHEED) observations and *ex-situ* X-ray Diffraction (XRD) measurements. The reciprocal space mapping was made by the measurement of rocking curves in an asymmetric geometry: the angle between the incident X-ray beam direction and the film plane was varied for fixed positions of the detector[6]. The film composition was determined by Electron Probe Microanalysis (EPMA). Hysteresis loops and the magnetic anisotropy of the samples were measured by Alternating Gradient Magnetometer (AGM) and Torque magnetometer, respectively. Kerr rotation θ_K was measured from the film side by a polarized angle modulation method.

3. Results and Discussion

Figure 1 shows the RHEED images during the deposition of (a) CrPt₃, (b) Mn_{0.2}Cr_{0.8}Pt₃, (c) Mn_{0.2}Cr_{0.8}Pt₃ / Pt buffer films for the incident electron beam along MgO<10 $\bar{1}$ > and <11 $\bar{2}$ > directions. The presence of elongated streaks indicates the sample surface to be very flat, and the films are confirmed to be epitaxially grown on the MgO(111). In Fig. 1 (a), superlattice lines were clearly observed as indicated by arrows in the figure, which suggests that the samples have the ordered Cu₃Au structure. The superlattice lines were also observed in the patterns during the growth of Mn_{0.2}Cr_{0.8}Pt₃ alloy films at a temperature of 500 °C as shown in Fig. 1 (b). This suggests that the ordered phase of Mn_{0.2}Cr_{0.8}Pt₃ is grown at lower temperatures than that of CrPt₃ with the post annealing. In the case of Mn_{0.2}Cr_{0.8}Pt₃ / Pt, the intensity of the superlattice lines are very weak (See Fig. 1 (c)), which implies the lack of long range ordering probably because of some intermixing at the Mn_{0.2}Cr_{0.8}Pt₃ / Pt interface.

Figure 2 represents $\theta - 2\theta$ X-ray profiles of (a) CrPt₃ after annealing at 850 °C, (b) Mn_{0.2}Cr_{0.8}Pt₃, and (c) Mn_{0.2}Cr_{0.8}Pt₃ / Pt films grown onto MgO(111) and fused quartz substrates. In the profiles of MgO(111) substrate,

Fig. 1 RHEED images of (a) CrPt₃, (b) Mn_{0.2}Cr_{0.8}Pt₃, and (c) Mn_{0.2}Cr_{0.8}Pt₃/Pt on MgO(111) for the incident e-beam along MgO<101> and <112> directions. Arrows in the figures indicate the superlattice lines due to the formation of the Cu₃Au ordered phase.

intense CrPt₃ {111} and {222} peaks are observed, while the peaks due to the grains with other crystal orientations are not seen. In the pattern of Mn_{0.2}Cr_{0.8}Pt₃ / Pt, peaks from the alloy and the buffer layer are distinguishable, which implies that the intermixing at the Mn_{0.2}Cr_{0.8}Pt₃ / Pt interface is not significant. In the case of the sample grown on fused quartz substrates, fundamental peaks such as {111} and {200} suggest that the samples have less {111} preferred orientation as reported for the sputter-deposited film[3]. In addition, the {100} and {110} superlattice lines due to the formation of the ordered phase in the {200} and {220} oriented grains, respectively, are seen in the patterns of (a) and (b). In the films with Pt buffer layer, the peaks from the alloy and the buffer layers are not so distinguishable for quartz substrate as those for MgO(111) substrates. This may be attributed that intermixing at the interface is more enhanced in the polycrystalline film.

The lattice parameters estimated from the X-ray profiles in the same manner as Ref. 7 are listed in Table I together with the value of M_s . The CrPt₃ on MgO(111) substrate has almost the same lattice constant as that of the bulk alloy[8]. On the other hand, CrPt₃ on fused quartz substrate has a smaller lattice parameter in the film normal direction than the bulk alloy. This might reflect the difference in the linear thermal expansion coefficients between the MgO (1×10^{-5} which is the same order as that of Pt) and the fused quartz (0.4×10^{-6}).

Figure 3 shows the hysteresis loops of CrPt₃ and Mn_{0.2}Cr_{0.8}Pt₃ films applying a magnetic field in the film

Fig. 2 θ -2 θ X-ray profiles of (a) CrPt₃, (b) Mn_{0.2}Cr_{0.8}Pt₃, and (c) Mn_{0.2}Cr_{0.8}Pt₃/Pt. Black and gray lines represent the patterns for the samples on MgO(111) and fused quartz, respectively.

Table I Lattice constant a and saturation magnetization M_s of CrPt₃, Mn_{0.2}Cr_{0.8}Pt₃, and Mn_{0.2}Cr_{0.8}Pt₃/Pt.

Substrate	film construction	a [nm]	M_s [emu/cm ³]
fused quartz	CrPt ₃	0.384	232
	Mn _{0.2} Cr _{0.8} Pt ₃	0.386	264
	Mn _{0.2} Cr _{0.8} Pt ₃ / Pt	0.387	76
MgO(111)	CrPt ₃	0.387	264
	Mn _{0.2} Cr _{0.8} Pt ₃	0.388	132
	Mn _{0.2} Cr _{0.8} Pt ₃ / Pt	0.387	81
bulk CrPt ₃		0.387	237.3

plane (gray line) and the film normal (black line) directions. The CrPt₃ / fused quartz exhibits large perpendicular anisotropy ($K_u = 1.5 \times 10^6$ erg/cm³ from the torque measurement) as shown in Fig. 3(a). (The sputter-deposited film in Ref. 3 have slightly larger K_u of 2.4×10^6 erg/cm³ than the present sample.) However, the easy axis of CrPt₃ / MgO(111) lies in the film plane (See Fig. 3(a')). The CrPt₃ on both substrates exhibit the saturation magnetization, M_s , of about 240 emu/cm³, which is roughly same

Fig. 3 M - H curves of (a) CrPt_3 , (b) $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$, and (c) $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{Pt}$ on fused quartz, (a') CrPt_3 , (b') $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$, and (c') $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{Pt}$ on $\text{MgO}(111)$ measured by applying a field in the film plane (gray line) and the film normal (black line) directions.

as the bulk value (237.3 emu/cm^3).

We have estimated magnetostriction constant, λ_s , of polycrystalline CrPt_3 film in the same manner as Ref. 9. The λ_s value was estimated to be 9.9×10^{-5} . From the λ_s value and the deviation of the lattice constant from the bulk value, the magnetoelastic contribution to the perpendicular anisotropy can be estimated to be $2 \times 10^5 \text{ erg/cm}^3$, which is only about 10 % of measured value of K_u ($1.5 \times 10^6 \text{ erg/cm}^3$).

The $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{MgO}(111)$ film has the easy magnetization direction in the film plane just as the case of the epitaxial CrPt_3 film. The M_S value of $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{MgO}$ decreases to 132 emu/cm^3 , which is considered to be due to insufficient substrate temperature of 500°C for the growth of highly ordered alloy. In contrast to the films on MgO , the perpendicular magnetic anisotropy was induced in $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ films with Pt buffer layer as shown in Fig. 3 (c') although K_u of this film is rather small ($K_u = 0.6 \times 10^6 \text{ erg/cm}^3$). The M_S value which decreased to 81 emu/cm^3 , might be due to alloying with the buffer layer.

Figure 4 shows the magneto-optical Kerr spectra for $\text{CrPt}_3/\text{fused quartz}$, $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{MgO}(111)$, and $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{Pt}/\text{MgO}(111)$ films. The spectrum of bulk CrPt_3 is also shown in the figure as a reference[2]. The spectrum of CrPt_3 film exhibits negative and positive peaks at 1.4 eV and 3.1 eV , respectively. The spectral shape of the epitaxial CrPt_3 film agrees well with those of the bulk

Fig. 4 Kerr spectra of $\text{CrPt}_3/\text{fused quartz}$, $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{MgO}(111)$, and $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{Pt}/\text{MgO}(111)$ measured from the film side.

alloy and the sputter-deposited films[2, 3] although there is a minor difference in magnitude of θ_K . Significant difference in the Kerr spectra between CrPt_3 and $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ films was not observed. This might be related to the fact that the Kerr spectrum of CrPt_3 has a similar shape to that of MnPt_3 [2]. The M_S value of $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{MgO}$ is much smaller than that of CrPt_3 , but the θ_K value of $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ (60 nm) has almost the same magnitude as that of CrPt_3 . The might be due to the fact that the Kerr signal is mainly originated from the sample surface of $\sim 10 \text{ nm}$ thick. The magnetization in the surface region of the film might be larger than in the bottom region because of the intermixing between the alloy and the buffer layers.

In order to investigate the origin of the perpendicular magnetic anisotropy in the $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ film induced by inserting Pt buffer layer on MgO substrate, we carried out the two-dimensional scan in the reciprocal space of epitaxial $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ films with and without the Pt buffer layer. Figure 5 shows the reciprocal space mapping measured for (a) $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$, and (b) $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{Pt}$, where Q_z and Q_x are the perpendicular component ($[111]$ direction) of the momentum transfer vector and the in-plane component parallel to $[11\bar{2}]$ directions of the $\text{MgO}(111)$, respectively. By scanning the $Q_z - Q_x$ plane, superlattice reflections such as $\{221\}$, $\{332\}$ are clearly observed as in Fig. 5 (a) and (b), which indicates the formation of a Cu_3Au ordered phase. In the patterns of $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ (Fig. 5 (a)), $\{331\}$ and $\{221\}$ peaks comes from the twined domain whose $[111]$ direction is antiparallel to that of MgO . On the other hand, these peaks do not appear in the figure for $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{Pt}$ (Fig. 5 (b)), which is thought to be related to the growth mode of the buffer layer of Pt onto $\text{MgO}(111)$. In Fig. 5(b), it is noted Pt layer on $\text{MgO}(111)$ also does not form the twined structure. However, it is difficult to find the relationship between the uniaxial anisotropy observed for $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$

Fig. 5 Reciprocal space mappings of (a) $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ and (b) $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3 / \text{Pt}$ grown on $\text{MgO}(111)$.

/ Pt and the twinned domain.

4. Summary

We prepared epitaxial CrPt_3 and $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ films, and investigated the structure and the magnetic and magneto-optical properties. From the RHEED observation and the X-ray analysis, it was found that CrPt_3 and $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ films have epitaxially grown on the $\text{MgO}(111)$ substrates and formed Cu_3Au type ordered structure. The Kerr spectrum of epitaxial CrPt_3 film exhibited similar shape to those of the bulk alloy and the sputtered films. The significant difference in the Kerr spectra between CrPt_3 and $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ films was not observed. The easy axis of magnetization of CrPt_3 on $\text{MgO}(111)$ lay in the film plane, although CrPt_3 on fused quartz exhibited large perpendicular anisotropy ($K_u = 1.5 \times 10^6 \text{ erg/cm}^3$). The perpendicular magnetic anisotropy of $K_u = 0.6 \times 10^6 \text{ erg/cm}^3$ was induced in $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{MgO}$ films by inserting a 30 nm Pt buffer layer. However, no obvious structural difference which can explain the induced perpendicular magnetic anisotropy was observed between $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3$ and $\text{Mn}_{0.2}\text{Cr}_{0.8}\text{Pt}_3/\text{Pt}$ in the reciprocal space mappings.

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