あくもに 六方稠密 (hcp) 構造を持つ Co 合金の単結晶薄膜形成を酸化物単結晶基板を用いて行った。基板材料の種類と面方位を選択することにより、磁化容易軸 (hcp 構造の c 軸) が基板面と平行および垂直な単結晶薄膜が得られた。X 線解析法によって磁性薄膜と基板の結晶成長方位関係を決定し、透過型電子顕微鏡により薄膜の微細構造を調査された。
これらの単結晶磁性薄膜を用いることによって、Co 合金薄膜の磁気異方性定数などの磁気物性を正確に決定することができるようになった。

キーワード Co 合金、単結晶薄膜、酸化物単結晶基板、エピタキシャル成長、微細構造

---

Preparation and Characterization of Single-Crystal CO-alloy Magnetic Thin Films


Central Research Laboratory, Hitachi, Ltd., Kokubunji, Tokyo 185-8601, Japan
Phone:+81-42-323-1111, E-mail:futamoto@crl.hitachi.co.jp
*Tokyo University of Agriculture and Technology, Koganei, Tokyo 184-8588, Japan

Abstract: Single-crystal thin films of Co-alloy materials with hcp-crystal structure are prepared on oxide single crystal substrates. The orientation of easy magnetization axis (c-axis in hcp-crystals structure) can be controlled either parallel or perpendicular to the film plane by selecting the crystallographic orientation of the substrate. The epitaxial relationship between the magnetic layer, the underlayer, and the substrate are determined. The microstructures of magnetic thin films are investigated by using a high-resolution transmission electron microscope. Using these single crystal thin films, important basic magnetic properties like magneto-crystalline anisotropy constants (Ku) can be determined.

Key words: single-crystal Co-alloy thin films, single-crystal oxide substrates, epitaxial growth, hcp-crystal structure
1. Introduction

Co-based alloy thin films are widely used as magnetic recording media in hard disk drives (HDD). The areal density and the transfer rate of HDD will continuously increase up to 100 Gb/in\(^2\) and 500 – 1000 Mb/s within a few years. The thermal stability and the magnetic switching speed under such ultra-high density magnetic recording have been investigated through mainly computer simulation. In such investigations, it is necessary to use accurate magnetic properties of Co-based alloy materials such as uniaxial magneto-crystalline anisotropy constants and Gilbert’s damping constants. However it has been not easy to determine these values using polycrystalline magnetic thin films which consist of very small crystalline grains with complicated compositional microstructures.

In order to investigate the basic magnetic properties, we developed technologies to prepare single-crystal magnetic thin films employing single-crystal substrates [1-4]. Using the well-defined single-crystal magnetic thin films, it became possible to determine the basic magnetic properties that are crucial to design the future high density recording media. This paper briefly describes the preparation and the characterization of Co-alloy single-crystal magnetic thin films.

2. Preparation of single-crystal magnetic thin films

2.1 Single-crystal thin film with the c-axis parallel to the substrate

In order to investigate the magnetic properties of polycrystalline thin film media by using computer simulation, for example, the knowledge of basic magnetic properties of Co-alloy material in the form of thin film are required. It is known that the magnetic properties of thin film are sensitively influenced by the crystallographic stress and strain coming from the lattice mismatch with the underlayer. A detail of this topic investigated for a CoCrPt magnetic thin film is reported in the reference [5]. We found that Co-alloy single-crystal thin films can be grown on single-crystal substrate using an epitaxial technique employing a similar underlayer material which is used to prepare thin film recording media [1-4]. Co-alloy single-crystal thin films with the c-axis parallel to the substrate surface were prepared using MgO single crystal substrates. When a (Co or Co-alloy)/(Cr or Cr-alloy) bilayer film was deposited on a MgO (100) substrate, Co or Co-alloy film consisting of bicrystalline domains was formed [1]. The bicrystalline domains consist of two-types of crystals with the c-axes perpendicular to each other. A plan-view high-resolution TEM micrograph of bicrystalline CoCrPt thin film is shown in Fig. 1. The epitaxial relationship is determined to be (11-20)Co // (100)Cr // (100)MgO, [0001]Co // <011>Cr // [010]MgO.

![Plan-view high-resolution TEM micrograph of bicrystalline CoCrPt film grown on MgO(100) substrate.](image)

When a (Co or Co-alloy)/(Cr or Cr-alloy) bilayer film was deposited on a MgO (110) substrate, single crystal magnetic thin film was obtained [1]. Thin film was prepared using a dc magnetron sputtering system with an Ar pressure of 3 mTorr and a substrate temperature of 573 K. Figure 2 shows the X-ray diffraction profile of a Co (30 nm)/Cr (30 nm)/MgO(110) sample. Figure 3 shows a plan-view TEM micrograph of single-crystal Co layer. The epitaxial relationship was determined to be (1100)Co//(211)Cr//(110)MgO, [0001]Co//[011]Cr//[001]MgO. A schematic model
of heteroepitaxial relationship in the Co/Cr/MgO(110) system is shown in Fig. 4.

Compositional microstructures of bicrystalline and single-crystalline Co–Cr–Ta thin films were investigated by using an EELS-TEM. In the case of bicrystalline CoCr\textsubscript{19}Ta\textsubscript{8}, magnetic thin film, the Cr concentration at the bicrystalline grain boundary was 18 at%, which is 3 at% greater than the average Cr composition [6]. It is clear that the distributions of Co, Cr, and Ta are fluctuating in a lateral direction in the bicrystalline specimen, though the amount of elemental segregation is far smaller compared with those observed in a polycrystalline Co–Cr–Ta thin film (20 – 30 at%) [6,7]. The presence of bicrystalline grain boundary is thought to have enhanced elemental segregation through giving stress or strain to the film during film growth process. In contrast, the elemental distributions in a single-crystal CoCr\textsubscript{19}Ta\textsubscript{8} magnetic thin film were confirmed by an EELS-TEM study to be almost homogeneous across the sample [8]. The Cr concentration was fluctuating within a small variation of ± 3 at% with respect to the average composition. The Co concentration showed a fluctuation opposite to that of Cr. The Ta distribution seemed to be uniform across the specimen. Similar homogeneous elemental distributions were confirmed for other Co-alloy compositions. The single-crystal Co-alloy magnetic thin films are confirmed to be compositionally almost homogeneous with a well-defined hcp crystallographic structure.

### 2.2 Single-crystal thin film with the c-axis perpendicular to the substrate

Preparation of Co-alloy single-crystal magnetic thin film with the c-axis perpendicular to the substrate was investigated by employing single crystals of Al\textsubscript{2}O\textsubscript{3}(0001), LaAlO\textsubscript{3}(0001), SrTiO\textsubscript{3}(111), and MgO(111) [9]. The oxide single crystals were chosen taking account of their lattice matching with the Co-alloy. A thin film consisting of CoCr\textsubscript{19}Pt\textsubscript{10}/non-magnetic CoCr\textsubscript{25}Ru\textsubscript{25} layers was deposited by dc magnetron sputtering at an Ar pressure of 3 mTorr by keeping the substrate at 533 K. Crystallographic properties were studied by X-ray diffraction and TEM. Figure 5 shows the X-ray diffraction spectrum observed for the sample deposited on Al\textsubscript{2}O\textsubscript{3}(0001) substrate. Figure 6 shows the X-ray pole figures measured for CoCrPt magnetic films deposited on various oxides. Table 1 summaries the experimental results. Single crystal magnetic films were obtained on the Al\textsubscript{2}O\textsubscript{3}(0001), the LaAlO\textsubscript{3}(0001), and the SrTiO\textsubscript{3}(111) substrates, while poly-crystalline thin films grew on the MgO(111) substrate. The crystatalllographic relationship between the c-axis oriented
CoCrPt single-crystal thin film and the substrate are respectively determined as:
Al₂O₃(0001)/CoCrPt(0001), Al₂O₃[10.0]/CoCrPt[21.0],
LaAlO₃(0001)/CoCrPt(0001),
LaAlO₃[10.0]/CoCrPt[10.0],
SrTiO₃(111)/CoCrPt(0001), SrTiO₃[1-10]/CoCrPt[10.0].
The best single-crystal thin film with small lattice distortion was obtained on the Al₂O₃(0001) substrate.
Cross-sectional TEM micrographs of a CoCrPt/non-magnetic-CoCrRu/Al₂O₃(0001) sample are shown in Fig. 7.
High resolution TEM micrographs of Fig. 7 (b) and (c) clearly show that atomically sharp interfaces are realized at the boundaries of the CoCrPt-layer/CoCrRu-layer and of the CoCrRu-layer/Al₂O₃(0001) substrate.
Figure 8 is a high-resolution TEM micrograph of single-crystal CoCrPt magnetic thin film with the c-axis perpendicular to the film plane.

Table 1. Summary of Epitaxial Thin Film Growth.

<table>
<thead>
<tr>
<th>Substrate material</th>
<th>Al₂O₃ (0001)</th>
<th>LaAlO₃ (0001)</th>
<th>SrTiO₃ (111)</th>
<th>MgO (111)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Orientation</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lattice parameter, (nm)</td>
<td>a=0.475</td>
<td>a=0.537</td>
<td>sqr2a=0.552</td>
<td>sqr2a=0.595</td>
</tr>
<tr>
<td>Mismatch (%) †</td>
<td>7.8</td>
<td>-4.7</td>
<td>-7.2</td>
<td>-13.9</td>
</tr>
<tr>
<td>XRD of CoCrPt layer</td>
<td>(0002) intensity (cps)</td>
<td>99200</td>
<td>13200</td>
<td>8300</td>
</tr>
<tr>
<td>∆θ 6° (deg)</td>
<td>0.6</td>
<td>2.6</td>
<td>5.1</td>
<td>6.3</td>
</tr>
<tr>
<td>Film quality</td>
<td>Single crystal</td>
<td>Single crystal</td>
<td>Single crystal</td>
<td>Poly-crystal</td>
</tr>
</tbody>
</table>

Sample structure: CoCrPt(25nm)/non-magnetic CoCrRu(50nm)/Oxide single-crystal.
† Mismatch is estimated for the lattice parameter of CoCrPt. (2a = 0.512 nm)
3. Determination of basic magnetic properties

Using well defined single-crystal thin film samples, it became possible to determine the basic magnetic properties of Co-alloy thin films. Figure 9 shows M-H loops of single-crystal Co film with the c-axis in plane. The easy magnetization axis is parallel to the substrate surface. Figure 10 shows M-H loops of 6-nm-thick single-crystal CoCrPt film with the c-axis perpendicular to the substrate surface. The M-H loop indicates that the easy magnetization axis is perpendicular to the substrate. Temperature dependence of magneto-crystalline anisotropy constants of $K_u$ and $K_{u3}$, and saturation magnetization values ($M_s$) are determined by using single-crystal magnetic thin films of Co-Cr-Ta and Co-Cr-Pt systems [3]. Other important magnetic properties such as Curie temperature, Gilbert’s damping constants, and exchange stiffness constants are also determined for Co-alloy thin films as a function of composition [3,10]. These magnetic properties are applied to investigate the possibility of future thin film recording media [11].
4. Summary

To investigate the magnetic properties using well-defined samples, Co-alloy single-crystal magnetic thin films are prepared using various oxide single-crystal substrates. On (110)MgO substrates, single crystal Co-alloy thin films with the c-axis parallel to the film plane could be obtained. The single-crystal thin films had good crystallographic quality and homogeneous distributions of alloying elements. On (0001)Al₂O₃ substrates, single crystal Co-alloy magnetic thin films with the c-axis perpendicular to the film plane were prepared. High resolution TEM study revealed that good heteroepitaxy is realized both between the (0001)Al₂O₃ substrate and the CoCrRu underlayer and between the CoCrRu underlayer and the CoCrPt magnetic layer. It became possible to accurately determine the basic magnetic properties such as magnetocrystalline anisotropy constants (Ku), Curie temperature (Tc), Gilbert’s damping constants (α), and so on. These basic magnetic properties are very useful to investigate the possibility of future high-density recording media.

Acknowledgment

A part of this work was carried out under the ASET program supported by NEDO.

References