Interface Magnetic Anisotropy of Pd/Co$_2$Fe$_x$Mn$_{1-x}$Si/MgO Layered Structures

Takahide Kubota$^{1,*}$, Tomonari Kamada$^1$, Jinhyek Kim$^1$, Arata Tsukamoto$^2$, Shigeki Takahashi$^3$, Yoshiaki Sonobe$^3$ and Koki Takahashi$^1$

$^1$Institute for Materials Research, Tohoku University, Sendai 980–8577, Japan
$^2$Department of Electronic Engineering, Nihon University, Funabashi 274–8501, Japan
$^3$Samsung R&D Institute Japan, Yokohama 230–0027, Japan

Interface magnetic anisotropy of Co$_2$Fe$_x$Mn$_{1-x}$Si Heusler alloy thin films were studied quantitatively. Films of Co$_2$MnSi ($x = 1$, CMS), Co$_2$Fe$_x$Mn$_{0.5}$Si ($x = 0.5$, CFMS), and Co$_2$FeSi ($x = 1$, CFS) were fabricated onto MgO (001) substrates with an epitaxially grown Pd (001) under layer, and were capped by an MgO layer. The maximum thickness for the perpendicular magnetization was 0.8 nm for CMS and CFMS, and it was 0.6 nm for CFS. The interface anisotropy energies ($K_i$) were 1.5, 1.5, and 1.2 erg/cm$^2$ for CMS, CFMS, and CFS, respectively. The difference in $K_s$ probably originated from the different alloying conditions at the bottom interface between Pd and Co$_2$Fe$_x$Mn$_{1-x}$Si layers.

[doi:10.2320/matertrans.ME2015050]

(Received December 25, 2015; Accepted March 1, 2016; Published April 15, 2016)

Keywords: Heusler alloy, interface anisotropy, perpendicular magnetization

1. Introduction

Perpendicularly magnetized thin films are of great interest to the spintronics field, and the spin-transfer torque magnetoresistive random access memory (STT-MRAM) is one of the major device applications$^{1,2}$). Various kinds of thin films with perpendicular magnetic anisotropy (PMA) have been studied for magnetic tunnel junctions (MTJs); e.g. CoPt, FePt, MnGa, Mn$_3$Ge ordered alloys$^{3,6}$), rare-earth transition metal alloys$^7$), Co-Pt(Pd) superlattices$^{8–10}$), and ultra-thin-films with interfacial anisotropies$^{11,12}$). Recent progress as shown in Refs. 10) and 11), has accelerated STT-MRAMs development significantly, however, it is still necessary to investigate PMA materials with high spin polarization for giga-bit-class MRAM developments. Heusler alloys are a class of half-metallic compounds$^{13}$), which are promising for generating highly spin polarized conduction electrons for MTJs. Tunnel magnetoresistance (TMR) effects with half-metallic electrodes were reported for MTJs using some Cobalt (Co)-based full-Heusler alloys such as Co$_2$MnSi$^{14,15}$, Co$_2$Fe(Mn,Sn)$^{16,17}$, and Co$_2$Fe(AlSi)$^{18}$). Recently, the number of reports of PMA films using Co-based full-Heusler alloys further increases. Layered structures including a Co$_2$FeAl/MgO interface have been typical samples studied as PMA-Heusler alloys$^{19–22}$), and TMR effects using PMA-Co$_2$FeAl have also been reported$^{21,23}$). Although a number of studies have been reported for PMA-Co$_2$FeAl, or Co$_2$FeZ (Z = Al$_{1-x}$Si$_x$, Si) compounds$^{24,25}$), PMA of other Heusler alloys which contain Mn, such as Co$_2$MnSi and relatives have not been studied so much. Our group previously reported the composition dependence of PMA for Co$_2$(Fe-Mn)Si thin films$^{26}$), however, quantitative discussion for the composition dependence of the interfacial magnetic anisotropy energy was lacking. In this work, we have systematically studied magnetic properties of Co$_2$Fe$_x$Mn$_{1-x}$Si thin films deposited onto MgO (100) substrates with an epitaxially grown Pd under layer, and estimated interfacial magnetic anisotropy energy at Pd/Co$_2$Fe$_x$Mn$_{1-x}$Si/MgO interfaces.

$^*$Corresponding author, E-mail: tkubota@imr.tohoku.ac.jp

2. Experimental

The stacking structure of the films was MgO (100) substrate / Cr (40 nm) / Pd (10 nm) / Co$_2$Fe$_x$Mn$_{1-x}$Si (t) / MgO (2 nm) / Ta (5 nm). All metallic layers were deposited using an ultra-high vacuum magnetron sputtering system with a base pressure less than $3 \times 10^{-7}$ Pa, and the MgO layer was deposited using electron-beam evaporation technique. The deposition temperature for the Pd layer was 350°C, and it was an ambient temperature for other layers. Post-annealing was carried out after the deposition of Cr and Co$_2$Fe$_x$Mn$_{1-x}$Si layers at 700°C and 500°C, respectively. The compositions, x of the Co$_2$Fe$_x$Mn$_{1-x}$Si layers were 0.25 (CMS), 0.5 (CFMS), and 1 (CFS). The thickness, t was changed from 0.6 to 1.4 nm in 0.2 nm increments. Magnetization curves were measured by superconducting quantum interference device-vibrating sample magnetometer (SQUID-VSM, MPMS®3, Quantum Design, Inc.) at 300 K. Applied magnetic field was parallel direction or perpendicular direction to the film plane. In the parallel case, the magnetic field was applied to the [100] direction of the MgO substrate, which corresponds to the [110] direction of CFMS layers. The maximum magnetic field was 30 kOe.

3. Results and Discussion

Figures 1, 2, and 3 show hysteresis loops of the CMS, CFMS and CFS films, respectively. Perpendicularly magnetized films were achieved for 0.6 nm-thick CMS, CFMS, and CFS films, and 0.8 nm-thick CMS and CFMS films, as reported in a previous paper$^{26}$). Saturation magnetizations ($M_s$) obtained from the hysteresis loops are summarized in Fig. 4. $M_s$ shows no clear dependence on the layer thickness (t) for all compositions. The values of $M_s$ are close to bulk values for the CMS and CFMS films, on the other hand, those are about 20% larger than the bulk value for most of the CFS films. The reason is not clear for the enhancement of $M_s$ of the CFS films, however, a possibility is that the CFS films are not ordered but disordered showing larger magnetization values similarly to those of disordered Co-Fe alloys$^{27}$). According to
ternary phase diagrams of Co-Mn-Si\textsuperscript{28) and Co-Fe-Si\textsuperscript{29)}, the area of the solid-solution phases for the body-centered-structure is larger for the Co-Fe-Si alloys than that for the Co-Mn-Si ones; which may cause larger amount of the chemical disordering in the Co\textsubscript{2}FeSi films. However it is unclear that the border line of the Fe composition for the easiness of the disordering, the disordering was probably less occurred in the CFMS films than the CFS ones because of the small amount of the Fe composition.

Figure 5 shows magnetization per unit area ($M_s/\text{unit} \times t$) as a function of magnetic field ($H$). Figure 6 shows the thickness dependence of saturation magnetizations ($M_s$) for Co\textsubscript{2}MnSi (\textsuperscript{\textregistered}), Co\textsubscript{2}Fe\textsubscript{0.5}Mn\textsubscript{0.5}Si (\textsuperscript{\textbullet}), and Co\textsubscript{2}FeSi (\textsuperscript{\texttriangle}). The arrows inside the graph point bulk values of $M_s$ for the three compounds.
function of the layer thickness, \( t \). Linear fitting was performed for estimating the dead-layer thickness and the \( M_s \) values, which are summarized in Table 1. The data point of the 1.4 nm-thick CMS sample was neglected for the fitting, because it was deviated remarkably from the linear dependence. The deviation was probably from an extrinsic factor, such as a poor reproducibility. The thicknesses of dead layers were about 0.1 nm or less for all three compounds.

Figure 6 shows the thickness dependence of \( K_u \times t \). Here, the effective anisotropy energy, \( K_u^{\text{eff}} \), was calculated from the area enclosed by out-of-plane and in-plane magnetization curves. Linear fitting was performed using data points of the thickness, \( t \), ranging from 0.8 nm to 1.4 nm. Here, the data point of the 1.4 nm-thick CMS sample was neglected for the same reason as the thickness dependence of \( M_s \times t \). The anisotropy energy density including both bulk and interface contributions is expressed as follows: \( K_u^{\text{eff}} \times t = K_u \times t + K_s \), where \( K_u^{\text{eff}}, K_u, \) and \( K_s \) stand for the effective uniaxial, volume, and interface anisotropy energies, respectively. Here, \( K_s = K_{\text{crystal}} - 2\pi M_s^2 \), which is a balance of magnetocrystalline anisotropy energy (\( K_{\text{crystal}} \)) and the demagnetization energy. From the intercept of the linear fitting in Fig. 6, \( K_u^{\text{eff}} \) was calculated for the Pd/CoFe0.5Mn0.5Si/MgO interfaces. The values of \( K_u \) and \( K_s \) are summarized in Table 2, showing a difference; CMS and CFMS films exhibited larger values than that of CFS. In addition, \( K_s \) values for the present CMS and CFMS films are higher than that of the Pt/CoFeAl/MgO films reported by Wen et al.\(^{20}\). Concerning the absolute value of \( K_u \), the magnitude is larger than that of the demagnetization energy which is of the order of \( 7-9 \times 10^6 \) erg/cm\(^3\). The large magnitude of \( K_u \) probably originates from a tetragonal distortion of the CoFe0.5Mn0.5Si lattice due to a lattice mismatch between the Pd under layer (or the MgO capping) and CoFe0.5Mn0.5Si alloy. The values of \( K_s/t \) are also listed in Table 2, which are about \( 19 \times 10^6 \) erg/cm\(^3\) and are comparable values with the previously reported PMA-CoFeAl films\(^{20,23}\).

There are two different interfaces which can provide anisotropy energy for the Heusler alloy layer, namely, CoFe0.5Mn0.5Si/MgO interface (top interface) and Pd/CoFe0.5Mn0.5Si interface (bottom interface). It is expected that the contribution from the bottom interface is larger than that from the top interface for the present samples, according to the previous report on the buffer layer dependence of the hysteresis loops for CFS films\(^{26}\). For the bottom interface, hybridization of \( d \)-bands for Co and Pd atoms promotes the enhancement of the orbital moment of Co, which may enhance the anisotropy at the bottom interface\(^{30,31}\). In addition to the interfacial contribution, the alloying effect should be also considered. For the present CMS, and CFMS films, there is a thin dead-layer which can be attributed to an alloying region and may enhance the anisotropy of the C(F)MS films. Concerning the CFS films, there are nearly no dead-layer around the bottom interface, which implies that the alloying effect is less than the other two films. Such different conditions for the bottom interface probably result in the difference in the \( K_u \) values for the CoFe0.5Mn0.5Si films.

4. Summary

Epitaxially grown CoFe0.5Mn0.5Si (CMS), CoFe0.5Mn0.5Si (CFMS), and CoFe0.5Si (CFS) ultra-thin films with a Pd-buffer layer and an MgO-capping were fabricated, and the interface anisotropy was quantitatively studied using hysteresis loops measured by SQUID-VSM at 300 K. Saturation magnetization, \( M_s \) values were comparable to those in bulk samples for CMS and CFMS films, while those were larger than the bulk value for CFS films. The enhanced \( M_s \) might be caused by the

---

**Table 1** A summary of the dead-layer thickness and saturation magnetization (\( M_s \)) estimated from the linear fitting in Fig. 5.

<table>
<thead>
<tr>
<th>Compound</th>
<th>Dead-layer thickness (nm)</th>
<th>( M_s ) from fitting (emu/cm(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>CoFe0.5Mn0.5Si</td>
<td>0.14</td>
<td>1100 ± 290</td>
</tr>
<tr>
<td>CoFe0.5Mn0.5Si</td>
<td>0.10</td>
<td>1200 ± 170</td>
</tr>
<tr>
<td>CoFeSi</td>
<td>-0</td>
<td>1540 ± 240</td>
</tr>
</tbody>
</table>

**Table 2** A summary of the volume anisotropy energy (\( K_u \)), the surface anisotropy energy (\( K_s \)) from the fitting in Fig. 6, and \( K_s/t \) for a typical layer thickness, \( t \).

<table>
<thead>
<tr>
<th>Compound</th>
<th>( K_u \times 10^6 ) erg/cm(^3)</th>
<th>( K_s ) (erg/cm(^3))</th>
<th>( K_u/t \times 10^6 ) erg/cm(^3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CoMnSi</td>
<td>-15</td>
<td>1.5 ± 0.4</td>
<td>19, ( t ) = 0.8 nm</td>
</tr>
<tr>
<td>CoFe0.5Mn0.5Si</td>
<td>-17</td>
<td>1.5 ± 0.4</td>
<td>19, ( t ) = 0.8 nm</td>
</tr>
<tr>
<td>CoFeSi</td>
<td>-16</td>
<td>1.2 ± 0.2</td>
<td>19, ( t ) = 0.6 nm</td>
</tr>
</tbody>
</table>

---

Fig. 5 The thickness, \( t \), dependence of the magnetization per unit area (\( M_s \times t \)) for CoFe0.5Mn0.5Si (○), CoFe0.5Mn0.5Si (□), and CoFeSi (△), which is a balance of magnetocrystalline anisotropy, was calculated from the data about 0.1 nm or less for all three compounds.

Fig. 6 The thickness, \( t \), dependence of \( K_u \times t \) for CoFe0.5Mn0.5Si (○), CoFe0.5Mn0.5Si (□), and CoFeSi (△) films, where \( K_u \) is the effective uniaxial anisotropy energy.
disordered structure of the CFS films. The values of $K_s$ were 1.5, 1.5, and 1.2 erg/cm$^2$ for the CMS, CFMS, and CFS films, respectively. The difference in $K_s$ is probably caused by different alloying conditions at the bottom interface between the Pd and Co$_{2}$Fe$_{M_{1-x}}$Mn$_{x}$Si layers, considering the difference in the dead-layer thickness among the films.

Acknowledgment

This work was partially supported by MEXT-Supported Program for the Strategic Research Foundation at Private Universities, 2013–2017.

REFERENCES