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Full-Heusler Co_2FeSi alloy thin films with perpendicular magnetic anisotropy induced by MgO-interfaces

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A 100-nm-thick $L2_1$-ordered full-Heusler Co_2FeSi (CFS) alloy film was fabricated using the facing targets sputtering (FTS) method at a substrate temperature T_S of 300 °C. The degrees of $L2_1$- and $B2$-order for the film were 37% and 96%, respectively. In addition, full-Heusler CFS alloy thin films with perpendicular magnetic anisotropy (PMA) induced by the magnetic anisotropy of MgO-interfaces were also successfully fabricated using the FTS method. The CFS/MgO stacked layers exhibited PMA when the CFS layer had a thickness of $0.6\,\text{nm} \leq d_{CFS} \leq 1.0\,\text{nm}$. The PMA in these structures resulted from the CFS/MgO interfacial perpendicular magnetic anisotropy.
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I. INTRODUCTION

Perpendicular magnetic tunnel junctions (p-MTJs)[1–3] consisting of ferromagnetic electrodes with perpendicular magnetic anisotropy (PMA) have attracted considerable attention, since they can achieve a low critical current density for current-induced magnetization switching (CIMS). To achieve large tunnel magnetoresistance (TMR) ratio in p-MTJs, highly spin polarized ferromagnetic materials with PMA are required.

Half-metallic ferromagnets (HMFs)[4] are perfectly spin polarized ferromagnetic materials with a spin polarization of 100%. Most Co-based full-Heusler alloys, such as Co_2FeSi (CFS),[5–10] Co_2FeGe,[6,11–13] Co_2MnSi,[14] and $Co_2FeSi_{1-x}Al_x$,[15] are predicted to be HMFs with Curie temperatures above room temperature (RT). Thus, these full–Heusler alloys are expected as HMF materials to have potential use in spintronics devices. However, because the half-metallicity of full-Heusler alloys depends on their atomic ordered crystal structure, the formation of highly ordered crystal structures is necessary.[16]

Recently, interfacial perpendicular magnetic anisotropy induced by an MgO interface[17] was realized in full-Heusler Co_2FeAl (CFA) alloy thin films;[18] however, full-Heusler CFA alloys are not HMFs according to band calculations. In the CFA/MgO system, the interactions between Fe and O atoms play an important role in establishing the PMA.[19,20] Half-metallic full-Heusler CFS alloys also include Fe atoms, and therefore MgO-induced PMA is expected in CFS-MgO systems.

In this paper, we report the fabrication of $L2_1$-ordered full- Heusler CFS alloy thin films using the facing targets sputtering (FTS) method. Stacked structures consisting of a CFS ultrathin layer and an MgO layer were also fabricated using this method and PMA induced by interfacial magnetic anisotropy between the CFS and MgO layers was observed.

II. SAMPLE PREPARATION

Multilayer stacks with a CFS/MgO structure were fabricated on a Pd-buffered MgO (100) single-crystal substrate. All the samples were prepared using the FTS method at a substrate temperature T_S of 300 °C. The base pressure was 1×10^{-4} Pa. The Pd buffer layers were sputtered in a Kr atmosphere at 0.13 Pa, while the CFS and Ta layers were sputtered in an Ar atmosphere at 0.13 Pa. The MgO layers were formed via reactive sputtering of Mg targets in an Ar and O_2 gas mixture (O_2 partial pressure was 1.4×10^{-3} Pa.). The deposition rates for the Pd, CFS, MgO, and Ta layers were 0.03 nm/s, 0.05 nm/s, 0.005 nm/s, and 0.06 nm/s, respectively.

III. RESULTS AND DISCUSSION

The crystal structures of the 100-nm-thick CFS films formed via FTS were analyzed using x-ray diffraction (XRD) with out-of-plane configuration measurements (Cu $K\alpha$ source). The definitions of the multi-axis goniometer geometries of the out-of-plane configuration are shown in detail in Ref. 12, where ϕ and ψ are the in-plane rotation and the tilt angles, respectively. Figure 1(a) shows a θ–2θ scan pattern for the CFS(100 nm)/Ta-cap(10 nm) multilayer structure on a Pd-buffered MgO(100) substrate. The CFS(200) and CFS(400) diffraction peaks were clearly observed, indicating that the CFS film was (100)-oriented and had at least the $B2$ structure. Figures 1(b) and 1(c) show ϕ scan patterns for the CFS film obtained at (b) $2\theta = 45.3°$ and $\psi = 45°$ and (c) $2\theta = 27.4°$ and $\psi = 54.7°$, respectively. In these figures, $\phi = 0°$ was defined as the direction of the MgO(220). The (220), (202), $(2\bar{2}0)$, and $(20\bar{2})$ diffraction peaks for CFS were clearly observed, and were periodically separated from one another with an angle difference of 90°, indicating fourfold symmetry in the sample plane. The CFS(111), $(1\bar{1}1)$, $(1\bar{1}\bar{1})$, and $(11\bar{1})$ diffraction peaks also exhibited fourfold symmetry (see Fig. 1(c)), indicating that the CFS film had the $L2_1$ structure. Furthermore, the (220) and (111) diffraction peaks in the ϕ scan patterns were separated by 45°, which corresponded to the crystal structure of CFS. These results indicate that the CFS film was epitaxially grown.

Next, the order parameters for the CFS film were evaluated using the extended Webster model.[7] The degrees of long-range order for the $L2_1$ and $B2$ structures, S_{L21}, and S_{B2},

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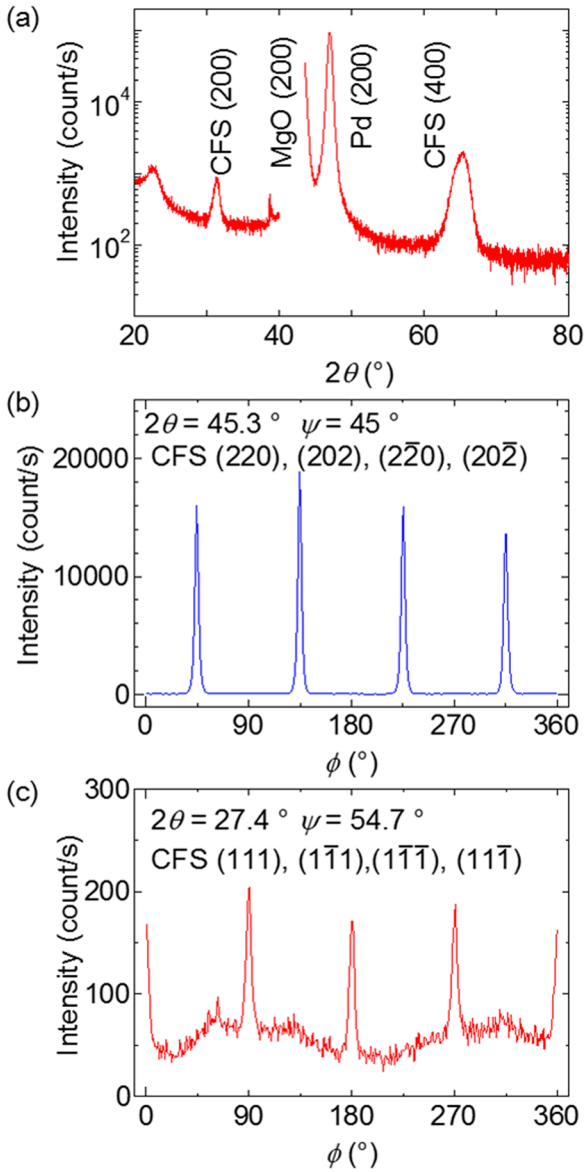

FIG. 1. Out-of-plane XRD patterns of an FTS-formed CFS film for the (a) θ–2θ scan, (b) ϕ scan for the CFS(220) diffraction ($2\theta = 45.33°$, $\psi = 45°$), and (c) ϕ scan for CFS(111) diffraction ($2\theta = 27.36°$, $\psi = 54.7°$).

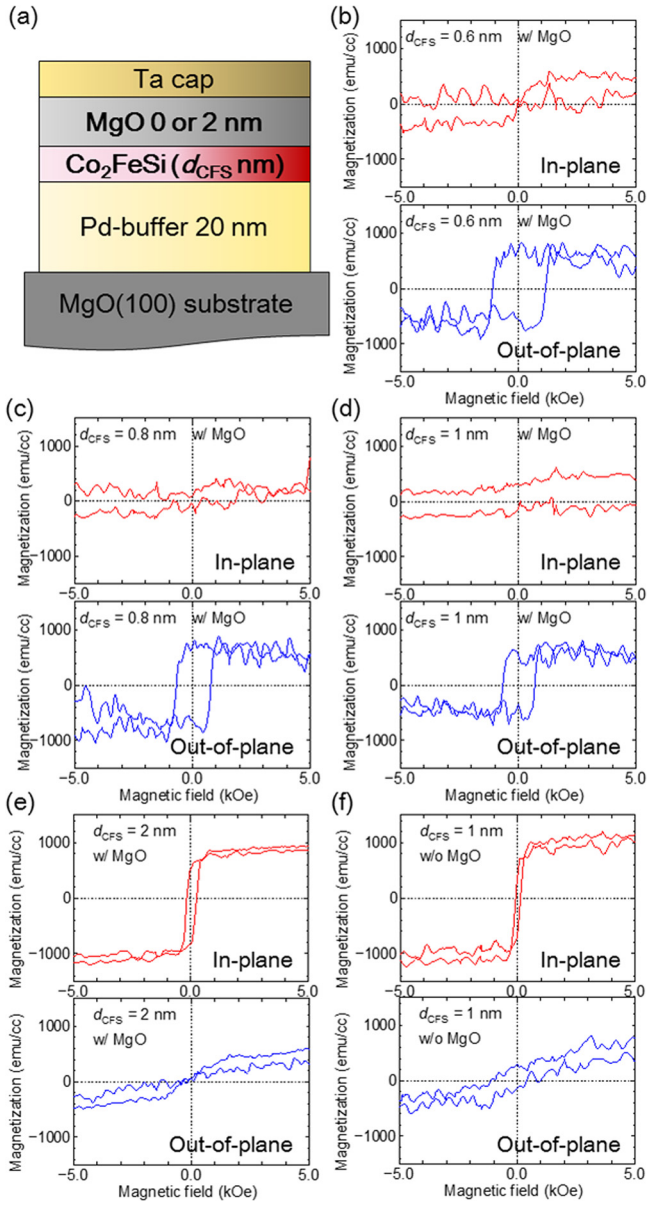

FIG. 2. (a) Schematic of stacked sample structures. In-plane and out-of-plane magnetic hysteresis loops of the stacked layers for (b) $d_{CFS} = 0.6$ nm, (c) 0.8 nm, (d) 1.0 nm, and (e) 2.0 nm with an MgO layer, and (f) 1.0 nm without an MgO layer.

can be evaluated using the following formulas: $I_{200}/I_{400} = S_{B2}^2 I_{200}^{\text{full-order}}/I_{400}^{\text{full-order}}$ and $I_{111}/I_{220} = (S_{L2_1}(3-S_{B2})/2)^2 I_{111}^{\text{full-order}}/I_{220}^{\text{full-order}}$ where I_{hkl}, and $I_{hkl}^{\text{full-order}}$ are the experimentally obtained diffraction intensity for the CFS(hkl) plane and its reference intensity[21,22] calculated for fully $L2_1$-ordered CFS alloys, respectively. The values for S_{L21} and S_{B2} were found to be 37% and 96%, respectively. The value of S_{B2} was very high in spite of the low process temperature of 300 °C and comparable to that for S_{B2} of CFS films[7] formed *via* rapid thermal annealing at 800 °C. On the other hand, the S_{L21} value was small and insufficient for realizing half-metallicity for the CFS film.[23] Formation at higher substrate temperatures and the post annealing process would likely be effective for achieving a higher S_{L21} value.[24] However, it can be concluded that an $L2_1$-phase CFS film was successfully formed using the FTS method.

Figure 2(a) shows a schematic of the stacked structure used to obtain MgO-induced magnetic anisotropy in the CFS films. Ultrathin CFS/MgO stacked layers were formed on Pd-buffered MgO (100) substrates using the FTS method. A CFS/MgO interface is expected to induce PMA when the CFS film is very thin, since the interfacial perpendicular magnetic anisotropy energy becomes relatively large for the in-plane magnetic anisotropy of the CFS layers.[17,18] Figures 2(b)–2(e) show magnetic hysteresis loops for the magnetization for the MgO(100)/Pd(20 nm)/CFS(d_{CFS} nm)/MgO (2 nm)/Ta(10 nm) stacks with nominal CFS layer thickness of $d_{CFS} = 0.6$ nm, 0.8 nm, 1.0 nm, and 2.0 nm. The *M-H* loops for the in-plane and out-of-plane magnetic field are presented in the upper and lower panels, respectively. Hysteresis loops for the out-of-plane magnetic field with good squareness were observed for the sample of $0.6\,\text{nm} \le d_{CFS} \le 1.0\,\text{nm}$. In addition, their in-lane *M-H* loops indicated that the in-plane direction was the axis of hard magnetization. These results

suggest that PMA was obtained for the stacked layers. The saturation magnetization M_S and coercivity H_C of the samples with $d_{CFS} = 1.0$ nm were 800 emu/cc and 710 Oe, respectively. The M_S value was slightly smaller than that of the CFS bulk value[10] of 1100 emu/cc. (This degradation is discussed in next paragraph.) Unfortunately, it was not possible to determine the magnetic anisotropy energy K_U, since these in-plane hysteresis loops contained large noises due to the low sensitivity of the vibrating sample magnetometer (VSM). The hysteresis loops for the sample with $d_{CFS} = 2.0$ nm indicated that the magnetic easy axis was in-plane, as shown in Fig. 2(e), suggesting that the PMA observed for the samples with thinner CF layers was caused by the interfaces. In this sample, the in-plane magnetic anisotropy energy of the bulk, which is proportional to the thickness (volume) of CFS layer, is expected to be larger than PMA energy of the CFS/MgO interface.

The origin of the observed PMA was then investigated by comparing samples with and an without MgO layer. Figure 2(f) shows the hysteresis loops for the sample with a 1.0-nm-thick CFS film, but without an MgO layer, and it can be seen that the sample without an MgO layer had in-plane magnetic anisotropy, even when CFS layer thickness was 1.0 nm. Based on these results, it was confirmed that the observed PMA resulted from the CFS/MgO interfacial magnetic anisotropy. Note that the M_S of the sample without an MgO layer was approximately 1000 emu/cc, which was larger than that that of the sample with an MgO interface. The reduction of the M_S in the sample with the MgO layer may be the result of processes that occurred during the formation of the MgO on the CFS, such as interface oxidation of the CFS layer. Further investigation for these ultrathin CFS films facing MgO layer requires cross-sectional transmission electron microscope (TEM) observations and *via* x-ray photoelectron spectroscopy (XPS).

In summary, we demonstrated the fabrication of a 100-nm-thick $L2_1$-ordered full-Heusler CFS alloy film using the facing targets sputtering (FTS) method at $T_S = 300\,^{\circ}$C. The degrees of $L2_1$- and $B2$-order for the film were 37% and 96%, respectively. Full-Heusler CFS alloy thin films with PMA induced by MgO-interface magnetic anisotropy were also successfully fabricated using the FTS method. The CFS/MgO stacked layers showed PMA when the CFS films had a thickness $0.6\,\text{nm} \leq d_{CFS} \leq 1.0\,\text{nm}$. The PMA in these structure resulted from the CFS/MgO interfacial magnetic anisotropy. This stacked structure with PMA should be directly applicable to p-MTJ structures.

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