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Fabrication of Fully Epitaxial $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}/\text{MgO}/\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ Magnetic Tunnel Junctions


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Fully epitaxial magnetic tunnel junctions (MTJs) of $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ (CCFA)/MgO/CCFA with exchange biasing were fabricated. The fabricated MTJs showed clear exchange-biased tunnel magnetoresistance (TMR) characteristics due to the CCFA/Ru/Co$_{90}\text{Fe}_{10}$/IrMn exchange-biased synthetic ferrimagnetic layer. The TMR characteristics were investigated as a function of in situ annealing temperature ($T_a$) for MTJs with $T_a$ of 400 °C, while those for MTJs with $T_a$ of RT (i.e., having an as-deposited upper CCFA layer) were 17% at RT and 80% at 4.2 K. These results clearly suggest that the spin polarization of the as-deposited upper CCFA layer was significantly increased by in situ annealing.

Index Terms—Co-based full-Heusler alloy, epitaxial growth, half-metallic ferromagnet, magnetic tunnel junction, tunnel magnetoresistance.

I. INTRODUCTION

COBALT-BASED full-Heusler alloy thin films have been intensively studied as attractive ferromagnetic electrodes in spintronic devices. This is because of the half-metallic ferromagnetic nature [1] theoretically predicted for some of these alloys [2] and because of their high Curie temperatures, which are well above room temperature (RT) [3]. The Co-based full-Heusler alloy $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ (CCFA) features high spin polarizations theoretically predicted for both the ordered L2$_1$ structure and the disordered B2 one [4], along with a relatively high Curie temperature of 750 K [5]. Inomata et al. first demonstrated a relatively high tunnel magnetoresistance (TMR) ratio of 16% at RT for MTJs with a Co-based full-Heusler alloy thin film, where they used a CCFA thin film [6].

We recently fabricated fully epitaxial MTJs with a Co-based full-Heusler alloy thin film of CCFA, Co$_2$MnGe, or Co$_2$MnSi as a bottom electrode, and a MgO tunnel barrier, and a Co$_2$Cr$_{1-x}$Fe$_x$Al (CCFA) features high spin polarizations theoretically predicted for both the ordered L2$_1$ structure and the disordered B2 one [4], along with a relatively high Curie temperature of 750 K [5]. Inomata et al. first demonstrated a relatively high tunnel magnetoresistance (TMR) ratio of 16% at RT for MTJs with a Co-based full-Heusler alloy thin film, where they used a CCFA thin film [6].

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A promising approach for further enhancing TMR ratios in such fully epitaxial MTJs is to use Co-based full-Heusler alloy thin films as both the lower and upper electrodes. Our purpose in the present study has been to develop fully epitaxial MTJs consisting of Co-based full-Heusler alloy CCFA thin films as the lower and upper electrodes and a MgO tunnel barrier. For this type of MTJ, one of the electrodes must be exchange-biased by an antiferromagnetic layer to form parallel and antiparallel magnetization configurations. For this approach we chose to use a MgO buffer electrode in a CCFA/Ru/Co$_{90}\text{Fe}_{10}$ trilayer, which was exchange-biased with an IrMn antiferromagnetic layer through the Co$_{90}\text{Fe}_{10}$/IrMn interface [13]. In this paper, we report on the fabrication of fully epitaxial MTJs of CCFA/MgO/CCFA with exchange biasing. We then show their TMR characteristics as a function of in situ annealing temperature for the upper CCFA layer and an estimation of the tunneling spin polarization of the upper CCFA layer.

II. EXPERIMENTAL METHODS

The fabricated MTJ layer structure was as follows: (from the substrate side) MgO buffer (10 nm)/lower CCFA (50)/MgO tunnel barrier/upper CCFA (10)/Ru (0.8)/Co$_{90}\text{Fe}_{10}$ (2)/IrMn (10)/Ru cap (5), grown on MgO(001) substrates. Each layer in the MTJ layer structure was successively deposited in an ultrahigh vacuum chamber through the combined use of magnetron sputtering and electron beam evaporation. The lower CCFA layer was deposited at RT using magnetron sputtering and subsequently annealed in situ at 500 °C for 15 min. The MgO tunnel barrier was deposited at RT by electron beam evaporation. The upper CCFA layer was also deposited at RT and subsequently annealed in situ at up to 500 °C. The layers of Ru, Co$_{90}\text{Fe}_{10}$, and IrMn were all deposited by magnetron sputtering at RT. We investigated the structural properties of the fabricated MTJ layer structures through in situ reflection high-energy electron diffraction (RHEED) observations for each successive layer during the deposition. The composition of the fabricated CCFA film was determined as Co$_{2.01}\text{Cr}_{0.58}\text{Fe}_{0.35}\text{Al}_{1.00}$ through inductively coupled plasma analysis with an accuracy of 2%–3% for the composition of each element. We fabricated MTJs with the fully epitaxial layer structure through photolithography and Ar ion milling. The fabricated junction size was 10 × 10 μm. The magnetoresistance was measured with a magnetic field applied along the [110] axis of the CCFA at temperatures from 4.2 K to RT using a dc four-probe method. We defined the TMR ratio as $(R_{AP} - R_P)/R_P$, where $R_{AP}$ and $R_P$ are the respective resistances for the antiparallel and parallel magnetization configurations between the upper and lower electrodes.

III. EXPERIMENTAL RESULTS AND DISCUSSION

X-ray diffraction pole figure measurements of a 50-nm-thick CCFA thin film annealed in situ at 500 °C showed that the film grew epitaxially on a MgO buffer layer and crystallized into the B2 structure. The crystallographic relationship was CCFA
The junction size was 10 nm. The CCFA electrode (50 nm) deposited at RT and annealed in situ at 500 °C grew epitaxially, as shown in Fig. 1(b). A 2.0-nm-thick MgO tunnel barrier, deposited at RT on a lower CCFA layer, also grew epitaxially [Fig. 1(c)]. These results were in agreement with our previous work [7]–[9]. We then confirmed from the in situ RHEED observation that an as-deposited upper CCFA layer on a MgO buffer layer grew epitaxially, as shown in Fig. 1(d). A 2.0-nm-thick MgO tunnel barrier, deposited at RT on a lower CCFA layer, also grew epitaxially [Fig. 1(c)]. This indicates that the layers grew epitaxially.

Fig. 2 shows typical magnetoresistance curves at RT and 4.2 K for a microfabricated CCFA/MgO/CCFA MTJ with the upper CCFA electrode and a MgO tunnel barrier deposited at RT. The applied bias voltage was 5 mV. The MgO tunnel barrier thickness was 2.3 nm. We observed a significant increase of the TMR ratio from 79% to 238% at 4.2 K (and from 17% to 60% at RT) for MTJs with $T_a$ of 400 °C compared to those with the as-deposited upper CCFA layer. To clarify why the TMR ratios rose with increasing $T_a$, we investigated X-ray diffraction peak intensities ($I_P$) for 002 and 004 peaks of CCFA films.

![Image](image.png)
(50 nm) deposited at RT on MgO buffer layers (10 nm) as a function of in situ annealing temperature, where the 002 peak is specific to the B2 structure. Compared to the peak intensity ratio of $I_p(002)/I_p(004)$ for as-deposited CCFA films, that of 300°C- and 500°C-annealed films rose with increasing $T_a$. This means the ratio of the B2 structure region to the A2 structure region (the B2 ratio) increased with increasing $T_a$ up to 500°C. It was theoretically predicted that A2-type disorder would significantly decrease the spin polarization in Co$_2$Cr$_2$Al while B2-type disorder would have little effect [4]. Consequently, the increase of the B2 ratio should lead to an increased spin polarization of the upper CCFA electrode. Note that a sufficiently high spin polarization for the lower CCFA electrode [9], [12], compared to that for the upper CCFA electrode, could be kept almost constant regardless of the in situ annealing after the deposition of the upper CCFA electrode, because the lower CCFA electrode was already annealed in situ at 500°C. Therefore, we can reasonably attribute the observed TMR increase as $T_a$ rose to 400°C to the increase of the B2 ratio in the upper CCFA electrode, leading to the increased spin polarization value of the upper CCFA films. MTJs with $T_a$ of 500°C showed lower TMR ratios of 164% at 4.2 K (52% at RT) than the case for MTJs with $T_a$ of 400°C. The cause of the lower TMR ratio for MTJs with $T_a$ of 500°C should be ascribed to another mechanism. To clarify this point, further studies are needed.

Finally, we will discuss the $T$ dependence of the TMR ratio of the fabricated fully epitaxial CCFA/MgO/CCFA MTJs. If we use parameter $\gamma = \alpha(4.2K)/\alpha(\text{RT})$, where $\alpha$ is the TMR ratio, to represent the degree of $T$ dependence of the TMR ratio, $\gamma$ for the fully epitaxial CCFA/MgO/CCFA MTJs was 3.9. This $\gamma$ value was in contrast to a more moderate value of $\gamma = 1.5(\alpha(\text{RT}) = 125\%$ and $\alpha(4.2K) = 185\%)$ for the reference fully epitaxial Co$_2$Fe$_2$MgO/Co$_2$Fe$_2$MgO MTJs [12], and even higher than a value of 2.9 previously reported for fully epitaxial CCFA/MgO/Co$_2$Fe$_2$Co$_2$MgO MTJs ($\alpha(\text{RT}) = 109\%$ and $\alpha(4.2K) = 317\%)$ [12] and that of 2.1 for fully epitaxial Co$_2$MnSi/MgO/Co$_2$Fe$_2$Co$_2$MgO MTJs ($\alpha(\text{RT}) = 90\%$ and $\alpha(4.2K) = 192\%)$ [11]. To clarify the reason for the strong $T$ dependence of the TMR ratio observed for fully epitaxial MTJs with a Co-based full-Heusler alloy thin film as at least one of the electrodes, including CCFA/MgO/CCFA MTJs, further systematic study is needed.

IV. SUMMARY

We fabricated fully epitaxial, exchange-biased magnetic tunnel junctions (MTJs) consisting of Co$_2$Cr$_{0.05}$Fe$_{0.4}$Al (CCFA)/MgO/CCFA and investigated TMR ratio (TMR) as a function of the in situ annealing temperature ($T_a$) for the upper CCFA layer. We obtained TMR ratios of 60% at room temperature (RT) and 238% at 4.2 K for MTJs with $T_a$ of 400°C; those for MTJs with $T_a$ of RT (i.e., an as-deposited upper CCFA layer) were 17% at RT and 80% at 4.2 K. These results clearly suggest that the spin polarization of the as-deposited upper CCFA layer was significantly increased by in situ annealing.

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