Fabrication of Fully Epitaxial $Co_2Cr_{0.6}Fe_{0.4}Al$/MgO/$Co_2Cr_{0.6}Fe_{0.4}Al$ Magnetic Tunnel Junctions

T. Marukame, T. Ishikawa, S. Hakamata, K.-I. Matsuda, T. Uemura, and M. Yamamoto

Division of Electronics for Informatics, Graduate School of Information Science and Technology, Hokkaido University, Sapporo 060-0814, Japan

Fully epitaxial magnetic tunnel junctions (MTJs) of $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA)/MgO/CCFA with exchange biasing were fabricated. The fabricated MTJs showed clear exchange-biased tunnel magnetoresistance (TMR) characteristics due to the CCFA/Ru/$Co_{90}Fe_{10}$/IrMn exchange-biased synthetic ferrimagnetic layer. The TMR characteristics were investigated as a function of *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, 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 $Co_2Cr_{0.6}Fe_{0.4}Al$ (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_2MnGe, or Co_2MnSi as a bottom electrode, and a MgO tunnel barrier [7]–[12]. We demonstrated high TMR ratios of 109% at RT and 317% at 4.2 K for CCFA/MgO/$Co_{50}Fe_{50}$ MTJs with exchange biasing [12]. From the TMR ratios, a high tunneling spin polarization of 0.88 at 4.2 K was obtained for the epitaxial CCFA films with the B2 structure [12].

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. Our approach for this was to use an upper CCFA electrode in a CCFA/Ru/$Co_{90}Fe_{10}$ trilayer, which was exchange-biased with an IrMn antiferromagnetic layer through the $Co_{90}Fe_{10}$/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)/$Co_{90}Fe_{10}$ (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, $Co_{90}Fe_{10}$, 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 $Co_{2.0}Cr_{0.55}Fe_{0.35}Al_{1.00}$ 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

Digital Object Identifier 10.1109/TMAG.2007.893698

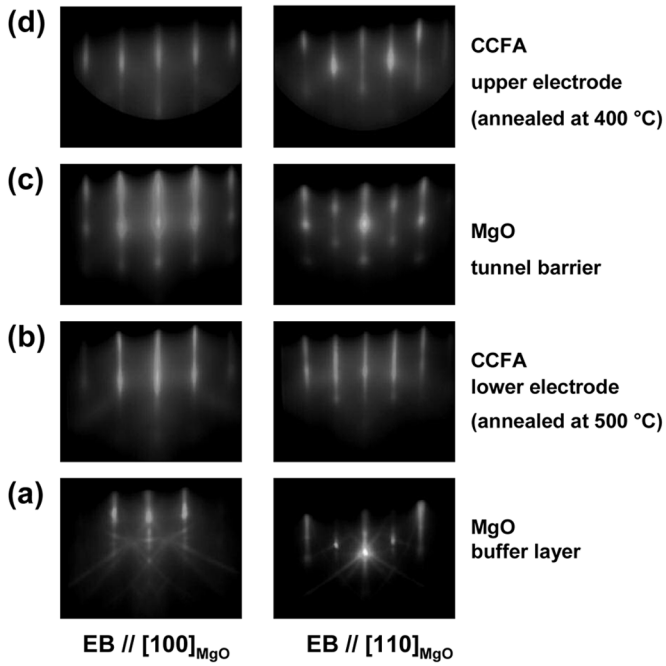

Fig. 1. RHEED patterns, along the azimuths of $[100]_{\mathrm{MgO}}$ and $[110]_{\mathrm{MgO}}$ (corresponding to $[110]_{\mathrm{CCFA}}$ and $[100]_{\mathrm{CCFA}}$, respectively), obtained *in situ* for each successive layer in the $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA)/MgO/CCFA MTJ layer structure during fabrication: (a) a MgO(001) buffer layer (10 nm), (b) a lower CCFA electrode (50 nm) deposited at RT and annealed *in situ* at 500 °C, (c) a MgO tunnel barrier (2.0 nm), (d) an upper CCFA electrode annealed *in situ* at 400 °C after deposition.

(001)[100] || MgO (001) [110]. Fig. 1 shows RHEED patterns, along the azimuths of $[100]_{\mathrm{MgO}}$ and $[110]_{\mathrm{MgO}}$ (corresponding to $[110]_{\mathrm{CCFA}}$ and $[100]_{\mathrm{CCFA}}$, respectively), obtained *in situ* for each successive layer in the CCFA/MgO/CCFA MTJ layer structure during fabrication. We confirmed from the *in situ* RHEED observation that an as-deposited lower CCFA layer on a MgO buffer layer 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 tunnel barrier layer had grown epitaxially. The streak patterns of a CCFA film annealed *in situ* at 400 °C [Fig. 1(d)] became sharper and more distinct compared with those of an as-deposited film. This indicates that the surface flatness of the CCFA thin film deposited at RT was improved by the post-deposition annealing at 400 °C. We also observed streak patterns for layers of Ru, $Co_{90}Fe_{10}$, and IrMn that were dependent on the incident direction of the electron beam, indicating 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 annealed *in situ* at 400 °C. The applied bias voltage was 5 mV. The MgO tunnel barrier thickness was 2.3 nm. The MTJs showed clear exchange-biased TMR characteristics due to the CCFA/Ru/$Co_{90}Fe_{10}$/IrMn exchange-biased synthetic ferrimagnetic layer [13]. Typical TMR ratios for the MTJ with the upper CCFA electrode annealed *in situ* at 400 °C were 60% at RT and 238% at 4.2 K.

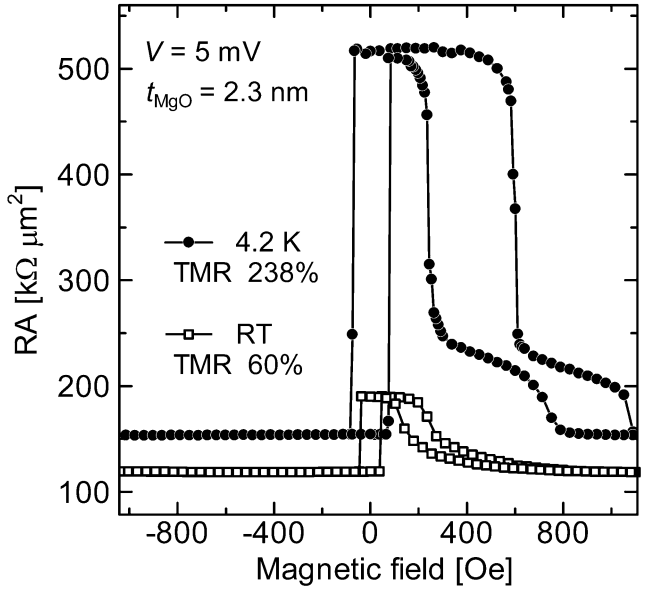

Fig. 2. Magnetoresistance curves at RT and 4.2 K for a fabricated fully epitaxial CCFA/MgO/CCFA MTJ with an upper CCFA electrode annealed *in situ* at 400 °C. The junction size was 10 × 10 μm. The applied bias voltage was 5 mV. The MgO tunnel barrier thickness was 2.3 nm.

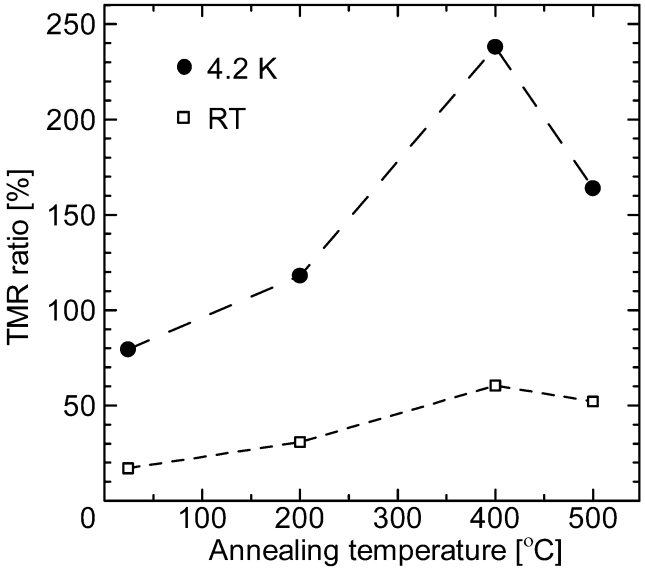

Fig. 3. Tunnel magnetoresistance (TMR) ratios at 4.2 K and RT for CCFA/MgO/CCFA MTJs as a function of *in situ* annealing temperature for the upper CCFA electrode.

We next investigated TMR ratios of the fully epitaxial CCFA/MgO/CCFA MTJs as a function of *in situ* annealing temperature (T_{a}) for the upper CCFA layer. Fig. 3 shows the T_{a} dependence of TMR ratios at 4.2 K and RT for the MTJs. As shown in Fig. 3, TMR ratios at both 4.2 K and RT increased with increasing T_{a} up to 400 °C and then decreased for T_{a} of 500 °C. 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} up to 400 °C, we investigated X-ray diffraction peak intensities (I_{P}) for 002 and 004 peaks of CCFA films

(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 $I_{\rm P}(002)/I_{\rm P}(004)$ for as-deposited CCFA films, that of 300°C- and 500°C-annealed films rose with increasing $T_{\rm a}$. This means the ratio of the B2 structure region to the A2 structure region (the B2 ratio) increased with increasing $T_{\rm a}$ up to 500 °C. It was theoretically predicted that A2-type disorder would significantly decrease the spin polarization in Co_2CrAl 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 $T_{\rm a}$ 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 $T_{\rm a}$ of 500 °C showed lower TMR ratios of 164% at 4.2 K (52% at RT) than the case for MTJs with $T_{\rm a}$ of 400 °C. The cause of the lower TMR ratio for MTJs with $T_{\rm a}$ 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 $\gamma = \alpha(4.2\,{\rm K})/\alpha({\rm RT})$, where α is the TMR ratio, to represent the degree of T dependence of the TMR ratio, γ for the fully epitaxial CCFA/MgO/CCFA MTJs was 3.9. This γ value was in contrast to a more moderate value of $\gamma = 1.5\,(\alpha({\rm RT}) = 125\%$ and $\alpha(4.2{\rm K}) = 185\%)$ for the reference fully epitaxial $Co_{50}Fe_{50}$/MgO/$Co_{50}Fe_{50}$ MTJs [12], and even higher than a value of 2.9 previously reported for fully epitaxial CCFA/MgO/$Co_{50}Fe_{50}$ MTJs ($\alpha({\rm RT}) = 109\%$ and $\alpha(4.2{\rm K}) = 317\%$) [12] and that of 2.1 for fully epitaxial Co_2MnSi/MgO/$Co_{50}Fe_{50}$ MTJs ($\alpha({\rm RT}) = 90\%$ and $\alpha(4.2{\rm K}) = 192\%$) [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 $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA)/MgO/CCFA and investigated tunnel magnetoresistance (TMR) as a function of the *in situ* annealing temperature ($T_{\rm a}$) for the upper CCFA layer. We obtained TMR ratios of 60% at room temperature (RT) and 238% at 4.2 K for MTJs with $T_{\rm a}$ of 400 °C; those for MTJs with $T_{\rm a}$ 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.

Acknowledgment

This work was supported in part by a Grant-in-Aid for Scientific Research (B) (Grant No. 18360143), a Grant-in-Aid for Creative Scientific Research (Grant No. 14GS0301), and a Grant-in-Aid for Young Scientists (B) (Grant No. 17760267) from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

References

[1] R. A. de Groot, F. M. Mueller, P. G. van Engen, and K. H. J. Buschow, "New class of materials: Half-metallic ferromagnets," *Phys. Rev. Lett.*, vol. 50, pp. 2024–2027, 1983.

[2] S. Ishida, S. Fujii, S. Kashiwagi, and S. Asano, "Search for half-metallic compounds in Co_2MnZ (Z=IIIb, IVb, Vb element)," *J. Phys. Soc. Jpn.*, vol. 64, pp. 2152–2157, 1995.

[3] P. J. Webster, "Magnetic and chemical order in Heusler alloys containing cobalt and manganese," *J. Phys. Chem. Solids*, vol. 32, pp. 0221–1231, 1970.

[4] Y. Miura, K. Nagao, and M. Shirai, "Atomic disorder effects on half-metallicity of the full-Heusler alloys $Co_2(Cr_{1-x}Fe_x)Al$: A first-principles study," *Phys. Rev. B*, vol. 69, pp. 144413-1–144413-7, 2004.

[5] T. Block, C. Felser, G. Jakob, J. Ensling, B. Mühling, P. Gütlich, and R. J. Cava, "Large negative magnetoresistance effects in $Co_2Cr_{0.6}Fe_{0.4}Al$," *J. Solid State Chem.*, vol. 176, pp. 646–651, 2003.

[6] K. Inomata, S. Okamura, R. Goto, and N. Tezuka, "Large tunneling magnetoresistance at room temperature using a Heusler alloy with the B2 structure," *Jpn. J. Appl. Phys*, vol. 42, pp. L419–L422, 2003.

[7] T. Marukame, T. Kasahara, K.-I. Matsuda, T. Uemura, and M. Yamamoto, "Fabrication of fully epitaxial magnetic tunnel junctions using full-heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ thin film and MgO tunnel barrier," *Jpn. J. Appl. Phys.*, vol. 44, pt. 2, pp. L521–L524, 2005.

[8] M. Yamamoto, T. Marukame, T. Ishikawa, K.-I. Matsuda, T. Uemura, and M. Arita, "Fabrication of fully epitaxial magnetic tunnel junctions using cobalt-based full-Heusler alloy thin film and their tunnel magnetoresistance characteristics," *J. Phys. D: Appl. Phys.*, vol. 39, pp. 824–833, 2006.

[9] T. Marukame, T. Ishikawa, K.-I. Matsuda, T. Uemura, and M. Yamamoto, "High tunnel magnetoresistance in fully epitaxial magnetic tunnel junctions using a full-Heusler Alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ thin Film," *Appl. Phys. Lett*, vol. 88, pp. 262503-1–262503-3, 2006.

[10] T. Marukame, T. Ishikawa, K.-I. Matsuda, T. Uemura, and M. Yamamoto, "Tunnel magnetoresistance in epitaxial magnetic tunnel junctions using full-Heusler alloy Co_2MnGe thin film and MgO tunnel barrier," *J. Appl. Phys.*, vol. 99, pp. 08A904-1–08A904-3, 2006.

[11] T. Ishikawa, T. Marukame, H. Kijima, K.-I. Matsuda, T. Uemura, M. Arita, and M. Yamamoto, "Spin-dependent tunneling characteristics of fully epitaxial magnetic tunneling junctions with a full-Heusler alloy Co_2MnSi thin film and a MgO tunnel barrier," *Appl. Phys. Lett.*, vol. 89, pp. 192505-1–192505-3, 2006.

[12] T. Marukame, T. Ishikawa, S. Hakamata, K.-I. Matsuda, T. Uemura, and M. Yamamoto, "Highly spin-polarized tunneling in fully epitaxial $Co_2Cr_{0.6}Fe_{0.4}Al$/MgO/$Co_{50}Fe_{50}$ magnetic tunnel junctions with exchange biasing," *Appl. Phys. Lett*, vol. 90, pp. 012508-1–012508-3, 2007.

[13] T. Ishikawa, T. Marukame, K.-I. Matsuda, T. Uemura, and M. Yamamoto, "Exchange bias effect in full-Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ epitaxial thin films," *IEEE Trans. Magn.*, vol. 42, no. 10, pp. 3002–3004, Oct. 2006.

Manuscript received October 31, 2006 (e-mail: yamamoto@nano.ist.hokudai.ac.jp).