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Author(s)	Ishikawa, T.; Marukame, T.; Matsuda, K.-I. et al.
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Exchange Bias Effect in Full-Heusler Alloy Co₂Cr_{0.6}Fe_{0.4}Al Epitaxial Thin Films

T. Ishikawa, T. Marukame, 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

We fabricated trilayer structures consisting of a Co-based full-Heusler alloy Co₂Cr_{0.6}Fe_{0.4}Al (CCFA) layer, a Ru ultrathin film, and a Co₉₀Fe₁₀ layer, and demonstrated well-established antiferromagnetic coupling in the fabricated structures. Furthermore, we observed a clear exchange bias effect in a CCFA/Ru/Co₉₀Fe₁₀/IrMn layer structure with a typical exchange bias field of about 430 Oe at room temperature. These results indicate that the use of a CCFA thin film in antiferromagnetically coupled trilayers is advantageous for obtaining a strong exchange bias field.

Index Terms—Co-based full-Heusler alloy, exchange bias, half-metallic ferromagnet, synthetic ferrimagnet.

I. INTRODUCTION

HALF-METALLIC ferromagnets (HMFs) are characterized by an energy gap for one spin direction at the Fermi level (E_F) leading to a complete spin polarization at E_F [1]. This feature of HMFs is highly favorable for ferromagnetic electrodes used in spintronic devices. Epitaxially grown cobalt-based full-Heusler alloy thin films have been intensively studied as attractive ferromagnetic electrode materials for spintronic devices [2]–[9]. This is because of the half-metallic nature theoretically predicted for some of these alloys [10], [11] and because of their high Curie temperatures, which are well above room temperature (RT) [12].

We recently fabricated fully epitaxial magnetic tunnel junctions (MTJs) that had a Co-based full-Heusler alloy thin film of either Co₂Cr_{0.6}Fe_{0.4}Al (CCFA) or Co₂MnGe as a bottom electrode, a MgO tunnel barrier, and a Co₅₀Fe₅₀ top electrode [3], [6], [8], and obtained relatively high tunnel magnetoresistance (TMR) ratios of 42% at RT and 74% at 55 K for the CCFA-MTJs. However, there is much room for further enhancing the TMR ratio of these MTJs. A promising approach is to use the Co-based full-Heusler alloy thin films as both the bottom and top electrodes. 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 purpose in this study has been to realize exchange bias in CCFA thin films epitaxially grown on MgO buffer layers. Such a CCFA film would be a prototype of a CCFA upper electrode that could be grown on a MgO tunnel barrier in fully epitaxial MTJs with a Co-based full-Heusler alloy thin film as a lower electrode. Our approach was to use a CCFA film in an antiferromagnetically (AF) coupled trilayer exchange-biased by an IrMn layer to obtain a large value of exchange bias field (H_{ex}) for epitaxial CCFA thin films.

We first fabricated trilayers consisting of a CCFA/Ru/Co₉₀Fe₁₀ layer structure and investigated their magnetic properties. After that, we fabricated

CCFA/Ru/Co₉₀Fe₁₀ trilayers exchange-biased with an IrMn layer through the Co₉₀Fe₁₀/IrMn interface and investigated the exchange bias effect in the fabricated layer structures.

II. EXPERIMENTAL METHODS

We fabricated two sets of layer structures. The first set was CCFA/Ru/Co₉₀Fe₁₀ trilayer structures without an antiferromagnetic layer. The layers were structured as follows: MgO buffer layer (10 nm)/CCFA (2.5–4.0)/Ru spacer (0.8)/Co₉₀Fe₁₀ (2.0)/Ru cap (5.0). The second set was CCFA/Ru/Co₉₀Fe₁₀ trilayer structures exchange-biased with an IrMn layer through the Co₉₀Fe₁₀/IrMn interface. The layers in this case were structured as follows: MgO buffer layer (10 nm)/CCFA (2.5–5.0)/Ru spacer (0.8)/Co₉₀Fe₁₀ (2.0)/IrMn(10)/Ru cap (5.0). As reference samples, we also fabricated bilayer structures, with a Ru cap, of CCFA (3 nm)/IrMn (10)/Ru cap (5.0). The 5-nm-thick Ru cap layer was used to protect the IrMn layer from corrosion.

All layers in these samples were successively deposited on MgO (001) single-crystal substrates in an ultrahigh vacuum chamber (with a base pressure of about 8×10^{-8} Pa) through the combined use of magnetron sputtering and electron beam (EB) evaporation. Before depositing the CCFA layer, we deposited a 10-nm-thick MgO buffer layer on the MgO substrate through EB evaporation at 400 °C to reduce or eliminate surface defects and microscopic roughness. The CCFA layer was then deposited on the MgO buffer layer at RT by magnetron sputtering. We carried out *in situ* reflection high-energy electron diffraction (RHEED) observations for each successive layer during fabrication. We observed clear streak patterns dependent on the incident directions of the electron beam, which indicated the CCFA film grew epitaxially on a MgO buffered MgO substrate. The layers of Ru, Co₉₀Fe₁₀, and IrMn were all deposited by magnetron sputtering at RT. We also observed streak patterns for these layers, indicating that the layers grew epitaxially. Because RHEED observation and deposition of the ferromagnetic and antiferromagnetic layers under a magnetic field were not compatible, all the layers, including the IrMn layer, were deposited with no magnetic field applied. After deposition, these layer structures were annealed *ex situ* at 375 °C for one hour in a vacuum of 5×10^{-2} Pa under a magnetic

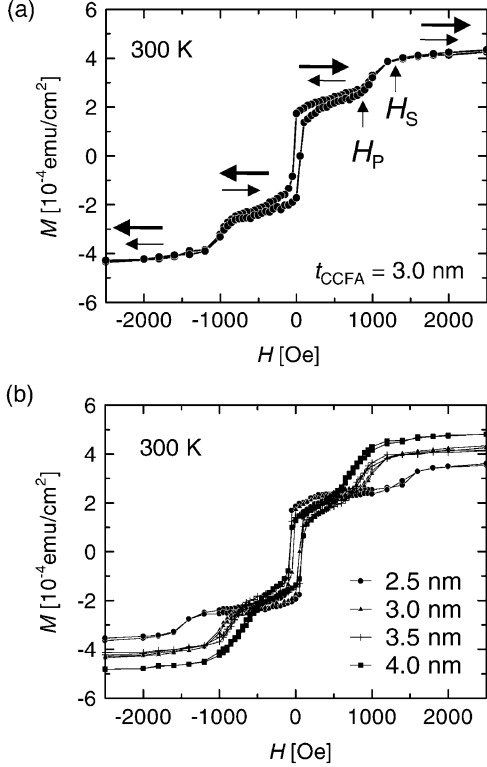


Fig. 1. Magnetization curves up to a magnetic field (H) of 2500 Oe at 300 K for $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ (CCFA)/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ trilayer structures with CCFA layer thicknesses (t_{CCFA}) from 2.5 to 4.0 nm: The magnetic field was applied in the film plane along the $[110]_{\text{CCFA}}$ axis. (a) M - H curve of a CCFA (3 nm)/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ trilayer. (b) M - H curves for trilayer structures with t_{CCFA} from 2.5 to 4.0 nm. The magnetic field was applied along the $[110]_{\text{CCFA}}$ axis. Magnetization effects due to the substrate have been subtracted. The saturation field (H_s) and the plateau field (H_p) are indicated in Fig. 1(a).

field of 5 kOe. The magnetic properties of the fabricated layer structures were measured using a SQUID magnetometer at RT. The contribution of the MgO substrate was subtracted.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Fig. 1 shows the magnetization curves up to a magnetic field (H) of 2500 Oe at 300 K for CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ trilayer structures with CCFA layer thicknesses (t_{CCFA}) from 2.5 to 4.0 nm. Fig. 1(a) shows the M - H curve of the CCFA (3 nm)/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ and Fig. 1(b) shows the curves for the trilayer structures with t_{CCFA} from 2.5 to 4.0 nm. The magnetic field was applied in the film plane along the $[110]_{\text{CCFA}}$ axis. The M - H curves in Fig. 1 show that antiferromagnetic coupling, due to the Ruderman–Kittel–Kasuya–Yoshida (RKKY) indirect exchange interaction [13], [14], was well established for the CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ trilayers with t_{CCFA} from 2.5 to 4.0 nm, with a typical plateau field (H_p) of 900 Oe and a saturation field (H_s) of 1200 Oe for a CCFA (3.0 nm)/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ trilayer. Thus, it was confirmed that the CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ trilayers with an ultrathin Ru layer of 0.8 nm behaved magnetically as synthetic ferrimagnets. Note that the magnetization direction of the $\text{Co}_{90}\text{Fe}_{10}$ upper layer was parallel to the magnetic field direction in the synthetic ferrimagnetic state for these trilayers with t_{CCFA} from 2.5 to 4.0 nm. This was because the saturation magnetization (M_s) of $\text{Co}_{90}\text{Fe}_{10}$ was about three times that of

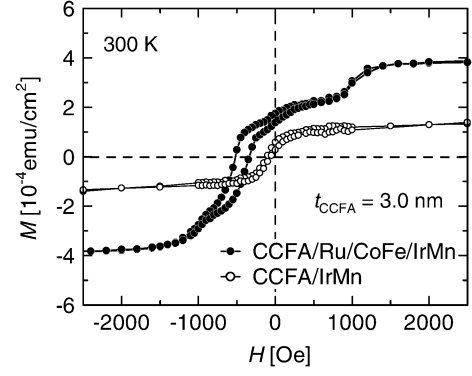


Fig. 2. Magnetization curve at 300 K for a $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ (CCFA)/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn quadrilayer structure with a CCFA layer thickness of 3.0 nm. For comparison, the magnetization curve for a CCFA/IrMn bilayer with the same CCFA layer thickness is also shown.

CCFA (the measured M_s for 50-nm-thick $\text{Co}_{90}\text{Fe}_{10}$ thin films was about 1500 emu/cm^3 , while that for 100-nm-thick CCFA films deposited at RT was about 500 emu/cm^3 [8], both at RT).

Fig. 2 shows a typical magnetization curve at 300 K for a quadrilayer structure of CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn with t_{CCFA} of 3.0 nm. For comparison, a magnetization curve for a bilayer structure of CCFA/IrMn with the same t_{CCFA} is also shown. We observed a clear exchange bias effect in the CCFA/IrMn bilayer structure with a typical H_{ex} value of about 100 Oe. We also observed a significantly enhanced exchange bias effect for the AF-coupled trilayer with an IrMn layer, and in this case the obtained H_{ex} value was about 430 Oe. For ferromagnetic/anti-ferromagnetic bilayers, H_{ex} is given by

$$H_{\text{ex}} = E_{\text{ex}}/M_s t \quad (1)$$

where E_{ex} is the exchange coupling energy per unit area, M_s is the saturation magnetization for the ferromagnet, and t is the ferromagnetic layer thickness. On the other hand, for exchange-biased, AF-coupled trilayers, $M_s t$ in the denominator in (1) is replaced by the total saturation magnetization of the AF-coupled trilayer, $M_{s1}t_1 - M_{s2}t_2$, where the subscripts 1 and 2 denote the two ferromagnetic layers in the trilayer structure coupled through the nonmagnetic metal. This results in

$$H_{\text{ex}} = E_{\text{ex}}/(M_{s1}t_1 - M_{s2}t_2). \quad (2)$$

Therefore, H_{ex} for the exchange biased AF-coupled trilayer should be higher than that for the bilayers because of the lower total saturation magnetization. We can reasonably attribute the significantly enhanced H_{ex} obtained for the CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn quadrilayer structure to this mechanism. This enhancement of H_{ex} indicates that use of a CCFA thin film in AF-coupled trilayers is advantageous for obtaining a higher H_{ex} value.

Fig. 3 shows the dependence of H_{ex} on t_{CCFA} for the exchange-biased CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn quadrilayer structures. The layer structures with t_{CCFA} ranging from 2.5 to 5.0 nm showed a clear exchange bias effect and H_{ex} values of 300 Oe to 430 Oe were obtained. As shown in Fig. 3, H_{ex} increased slightly with increasing t_{CCFA} . The increase of H_{ex} with increasing t_{CCFA} was in qualitative agreement with what is expected from (2). This dependence, though, was much

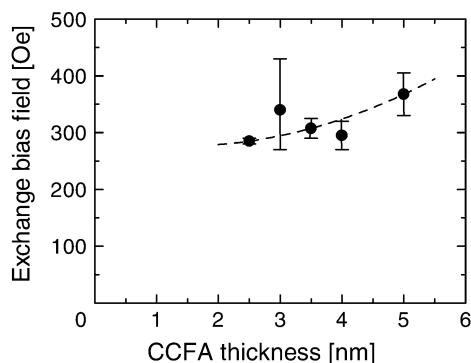


Fig. 3. Dependence of the exchange bias field on the $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ (CCFA) layer thickness in CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn layer structures. The dashed line is a guide to the eye.

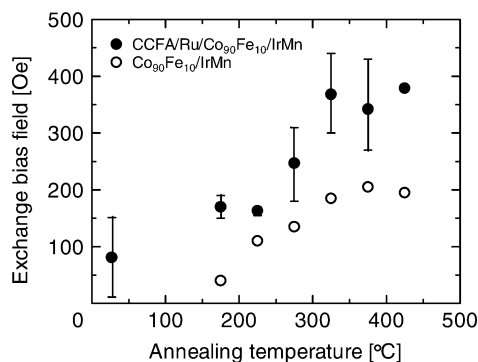


Fig. 4. Annealing temperature dependence of the exchange bias field for $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ (CCFA)/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn quadrilayer structures with a CCFA layer thickness of 3.0 nm. For comparison, the dependence of $\text{Co}_{90}\text{Fe}_{10}$ (2.0 nm)/IrMn bilayer structures is also shown.

weaker than that predicted from (2). To clarify the origin of this dependence, further investigation will be required.

Fig. 4 shows the annealing temperature dependence of H_{ex} for the CCFA (3 nm)/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn layer structures. As shown in Fig. 4, the H_{ex} increased significantly up to about 2.2 times as the annealing temperature increased from 225 °C to 325 °C. We attribute the significant increase of H_{ex} after annealing at 325 °C or higher to the annealing temperature exceeding the blocking temperature (T_B) of the antiferromagnetic IrMn layer ($T_B \sim 280$ °C). For comparison, we also investigated the annealing temperature dependence of H_{ex} for the $\text{Co}_{90}\text{Fe}_{10}$ (2 nm)/IrMn bilayer structures. The H_{ex} values we obtained for the bilayers are also shown in Fig. 4. Note that the $\text{Co}_{90}\text{Fe}_{10}$ /IrMn bilayers were deposited with no magnetic field applied as in the case of the CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn quadrilayer structures. As shown in Fig. 4, H_{ex} for the $\text{Co}_{90}\text{Fe}_{10}$ /IrMn bilayers showed a similar dependence on the annealing temperature. This result supports the above interpretation; i.e., that annealing at a temperature above T_B is essential.

IV. SUMMARY

We have demonstrated well-established antiferromagnetic coupling in fabricated $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ (CCFA)/Ru/ $\text{Co}_{90}\text{Fe}_{10}$

trilayers and a clear exchange bias effect in CCFA/Ru/ $\text{Co}_{90}\text{Fe}_{10}$ /IrMn layer structures with typical exchange fields of ~ 430 Oe at RT. These results indicate that the use of a CCFA thin film in antiferromagnetically coupled trilayers is advantageous for obtaining strong exchange bias field.

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