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Exchange Bias Effect in Full-Heusler Alloy
Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al Epitaxial Thin Films

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We fabricated trilayer structures consisting of a Co-based full-Heusler alloy Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al (CCFA) layer, a Ru ultrathin film, and a Co$_{30}$Fe$_{10}$ layer, and demonstrated well-established antiferromagnetic coupling in the fabricated structures. Furthermore, we observed a clear exchange bias effect in a CCFA/Ru/Co$_{30}$Fe$_{10}$/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], 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 Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al (CCFA) or Co$_3$MnGe as a bottom electrode, a MgO tunnel barrier, and a Co$_{30}$Fe$_{10}$ 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/Co$_{30}$Fe$_{10}$ layer structure and investigated their magnetic properties. After that, we fabricated CCFA/Ru/Co$_{30}$Fe$_{10}$ trilayers exchange-biased with an IrMn layer through the Co$_{30}$Fe$_{10}$/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/Co$_{30}$Fe$_{10}$ 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)/Co$_{30}$Fe$_{10}$ (2.0)/Ru cap (5.0). The second set was CCFA/Ru/Co$_{30}$Fe$_{10}$ trilayer structures exchange-biased with an IrMn layer through the Co$_{30}$Fe$_{10}$/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)/Co$_{30}$Fe$_{10}$ (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, Co$_{30}$Fe$_{10}$, 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

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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 Co\(_{30}\)Cr\(_{0.6}\)Fe\(_{0.4}\)Al (CCFA)/Ru/Co\(_{30}\)Fe\(_{10}\) trilayer structures with CCFA layer thicknesses \( (t_{\text{CCFA}}) \) 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/Co\(_{30}\)Fe\(_{10}\) trilayer. (b) \( M-H \) curves for trilayer structures with \( t_{\text{CCFA}} \) 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).

Fig. 2 shows a typical magnetization curve at 300 K for a quadrilayer structure of CCFA/Ru/Co\(_{30}\)Fe\(_{10}\)/IrMn with \( t_{\text{CCFA}} \) of 3.0 nm. For comparison, a magnetization curve for a bilayer structure of CCFA/IrMn with the same CCFA layer thickness is also shown. We observed a clear exchange bias effect in the CCFA/IrMn bilayer structure with a typical \( H_{\text{ex}} \) 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 \( H_{\text{ex}} \) value was about 430 Oe. For ferromagnetic/antiferromagnetic bilayers, \( H_{\text{ex}} \) is given by

\[
H_{\text{ex}} = \frac{E_{\text{ex}}}{M_s t}
\]  

(1)

where \( E_{\text{ex}} \) 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, \( M_{\text{ex}} \) in the denominator in (1) is replaced by the total saturation magnetization of the AF-coupled trilayer, \( M_{\text{ex}} = M_{\text{ex}1} - M_{\text{ex}2} \), where the subscripts 1 and 2 denote the two ferromagnetic layers in the trilayer structure coupled through the nonmagnetic metal. This results in

\[
H_{\text{ex}} = \frac{E_{\text{ex}}}{(M_{\text{ex}1} - M_{\text{ex}2}) t}
\]  

(2)

Therefore, \( H_{\text{ex}} \) 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 \( H_{\text{ex}} \) obtained for the CCFA/Ru/Co\(_{30}\)Fe\(_{10}\)/IrMn quadrilayer structure to this mechanism. This enhancement of \( H_{\text{ex}} \) indicates that use of a CCFA thin film in AF-coupled trilayers is advantageous for obtaining a higher \( H_{\text{ex}} \) value.

Fig. 3 shows the dependence of \( H_{\text{ex}} \) on \( t_{\text{CCFA}} \) for the exchange-biased CCFA/Ru/Co\(_{30}\)Fe\(_{10}\)/IrMn quadrilayer structures. The layer structures with \( t_{\text{CCFA}} \) ranging from 2.5 to 5.0 nm showed a clear exchange bias effect and \( H_{\text{ex}} \) values of 300 Oe to 430 Oe were obtained. As shown in Fig. 3, \( H_{\text{ex}} \) increased slightly with increasing \( t_{\text{CCFA}} \). The increase of \( H_{\text{ex}} \) with increasing \( t_{\text{CCFA}} \) was in qualitative agreement with what is expected from (2). This dependence, though, was much

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**Fig. 1.** Magnetization curves up to a magnetic field \( (H) \) of 2500 Oe at 300 K for Co\(_{30}\)Cr\(_{0.6}\)Fe\(_{0.4}\)Al (CCFA)/Ru/Co\(_{30}\)Fe\(_{10}\) trilayer structures with CCFA layer thicknesses \( (t_{\text{CCFA}}) \) 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/Co\(_{30}\)Fe\(_{10}\) trilayer. (b) \( M-H \) curves for trilayer structures with \( t_{\text{CCFA}} \) 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).

**Fig. 2.** Magnetization curve at 300 K for a Co\(_{30}\)Cr\(_{0.6}\)Fe\(_{0.4}\)Al (CCFA)/Ru/Co\(_{30}\)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 Co\(_{30}\)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. The magnetization curve for a CCFA/IrMn bilayer with the same CCFA layer thickness is also shown.

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**Fig. 3.** Dependance of \( H_{\text{ex}} \) on \( t_{\text{CCFA}} \) for the exchange-biased CCFA/Ru/Co\(_{30}\)Fe\(_{10}\)/IrMn quadrilayer structures. The layer structures with \( t_{\text{CCFA}} \) ranging from 2.5 to 5.0 nm showed a clear exchange bias effect and \( H_{\text{ex}} \) values of 300 Oe to 430 Oe were obtained. As shown in Fig. 3, \( H_{\text{ex}} \) increased slightly with increasing \( t_{\text{CCFA}} \). The increase of \( H_{\text{ex}} \) with increasing \( t_{\text{CCFA}} \) was in qualitative agreement with what is expected from (2). This dependence, though, was much
trilayers and a clear exchange bias effect in CCFA/Ru/CoFe/FeIrMn layer structures with typical exchange fields of $\sim 430$ 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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