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


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Fully epitaxial magnetic tunnel junctions (MTJs) of Co$_2$Cr$_{0.6}$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$_2$Cr$_{0.6}$Fe$_{0.4}$IrMn exchange-biased synthetic ferrimagnetic layer. The TMR characteristics were investigated as a function of in situ annealing temperature ($T_a$) for 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$_2$Cr$_{0.6}$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$_2$MnGe, or Co$_2$MnSi as a bottom electrode, and a MgO tunnel barrier, and a Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al/MgO/Co$_2$Cr$_{0.6}$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].

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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$_2$Cr$_{0.6}$Fe$_{0.4}$Al (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$_2$Cr$_{0.6}$Fe$_{0.4}$Al, 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$_2$Cr$_{0.6}$Fe$_{0.4}$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...
Fig. 1. RHEED patterns, along the azimuths of [001]_{MgO} and [110]_{MgO} (corresponding to [110]_{CCFA} and [100]_{CCFA}, respectively), obtained in situ for each successive layer in the Co_{2-CuFeAl} (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), and (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]_{MgO} and [110]_{MgO} (corresponding to [110]_{CCFA} and [100]_{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_{80}Fe_{20}, 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_{80}Fe_{20}/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.

We next investigated TMR ratios of the fully epitaxial CCFA/MgO/CCFA MTJs as a function of in situ annealing temperature for the upper CCFA electrode.

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.
dependence of the TMR of FeAl, Cr% on MgO/Co% C-annealed C. It was in contrast to a more moderate value MTJs (% and MnSi/MgO/Co% C to the of 500 value was in 400 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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