Highly spin-polarized tunneling in fully epitaxial Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al/MgO/Co$_{50}$Fe$_{50}$ magnetic tunnel junctions with exchange biasing

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Half-metallic ferromagnets (HMFs) are expected to be a key material for ferromagnetic electrodes that can provide highly spin-polarized currents. This is because HMFs are characterized by an energy gap at the Fermi level ($E_F$) for the minority-spin band, leading to complete spin polarization at $E_F$.

Cobalt-based full-Heusler alloys, whose composition is represented by Co$_2$YZ, have attracted much interest due to the half-metallic nature theoretically predicted for some of these alloys$^{2,3}$ and their high Curie temperatures, which are well above room temperature (RT).$^{4,5}$ The potentially high spin polarization of Co-based full-Heusler alloys is very advantageous for obtaining high tunnel magnetoresistance (TMR) ratios in magnetic tunnel junctions (MTJs) according to Jullière’s model.$^6$ Inomata et al. first demonstrated a relatively high TMR ratio of 16% at RT for MTJs using a Co-based full-Heusler alloy (Co$_2$YZ) thin film, where they used a polycrystalline Co$_{2}$Cr$_{0.6}$Fe$_{0.4}$Al (CCFA) thin film as a low-electrode and an amorphous AlO$_x$ tunnel barrier.$^7$ Sakuraba et al. reported a high TMR ratio of 570% at 2 K (67% at RT) for MTJs consisting of a lower electrode made of epitaxially grown Co$_2$MnSi (CMS) (which is a full-Heusler alloy), an amorphous AlO$_x$ tunnel barrier, and a highly oriented CMS upper electrode.$^8$

We recently developed fully epitaxial MTJs that have a Co$_2$YZ thin film of CCFA,$^9$ or Co$_2$MnGe,$^{10,11}$ or Co$_2$MnSi (Ref. 14) as a lower electrode, and a MgO tunnel barrier, and have demonstrated a relatively high TMR ratio of 90% at RT (240% at 4.2 K) for CCFA/MgO/Co$_50$Fe$_{50}$ MTJs (Ref. 12) and a TMR ratio of 90% at RT (192% at 4.2 K) for Co$_2$MnSi/MgO/Co$_50$Fe$_{50}$ MTJs.$^{12}$ A high tunneling spin polarization of 0.79 at 4.2 K was estimated from the TMR ratios for the epitaxial CCFA films with the B2 structure.$^{12}$ For these CCFA/MgO/Co$_50$Fe$_{50}$ MTJs, however, the parallel and antiparallel magnetization configurations were controlled by using the difference in the coercive forces between the CCFA lower electrode and the Co$_50$Fe$_{50}$ upper electrode. This resulted in peaked magnetoresistance versus magnetic field characteristics,$^{12}$ which probably led to TMR ratios lower than they potentially could be. Exchange biasing is favorable for realizing high degrees of the parallel and antiparallel magnetization configurations.$^{15}$ Our purpose in the present study was to demonstrate the potentially high tunneling spin polarization of a Co-based full-Heusler alloy of CCFA. To do this, we fabricated fully epitaxial MTJs with exchange biasing that consisted of a CCFA thin film and a MgO tunnel barrier, and then investigated the TMR characteristics of the fabricated MTJs. Our approach was to use an upper electrode of Co$_50$Fe$_{50}$ film in an antiferromagnetically coupled (i.e., synthetic ferromagnetic) Co$_50$Fe$_{50}$/Ru/Co$_{90}$Fe$_{10}$ trilayer exchange-biased by an IrMn antiferromagnetic layer through the Co$_{90}$Fe$_{10}$/IrMn interface to obtain a high exchange-bias field value ($H_{eb}$) for epitaxial Co$_50$Fe$_{50}$ electrodes.

The fabricated MTJ layer structure was as follows: (from the substrate side) MgO buffer (10 nm)/CCFA (50 nm)/MgO barrier (2.4 nm)/Co$_50$Fe$_{50}$ (3.4 nm)/Ru (0.8 nm)/Co$_{90}$Fe$_{10}$ (2 nm)/IrMn (10 nm)/Ru cap (5 nm). All layers in these MTJs were successively deposited on MgO(001) single-crystal substrates in an ultrahigh vacuum chamber (with a base pressure of about $8 \times 10^{-8}$ Pa) through the combined use of magnetron sputtering and electron beam evaporation. The CCFA lower electrode was deposited by rf magnetron sputtering at RT and subsequently annealed in situ at 500 °C. The CCFA film composition was determined to be Co$_{2.64}$Cr$_{0.39}$Fe$_{0.39}$Al$_{1.13}$, with an accuracy of 2%–3% for each element, through inductively coupled plasma analysis. The MgO tunnel barrier was deposited by electron beam evaporation at RT. The layers of Co$_50$Fe$_{50}$, Ru, Co$_{90}$Fe$_{10}$, and IrMn were all deposited by magnetron sputtering at RT. We carried out in situ reflection high-energy electron diffraction (RHEED) observations for each successive layer during fabrication. Because RHEED observation and deposition of the ferromagnetic and antiferromagnetic layers under a magnetic field were not compatible, all the layers were deposited with no magnetic field applied. We fabricated fully epitaxial MTJs with the layer structure described above by using photolithography and Ar ion milling. The fabricated junction size

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**References**

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was $10 \times 10 \, \mu m^2$. After the microfabrication, the MTJs were annealed at 175 °C for 1 h in a vacuum of $5 \times 10^{-2}$ Pa under a magnetic field of 5 kOe. The magnetoresistance was then measured through a dc four-probe method at temperatures from RT to 4.2 K. We defined the TMR ratio as a magnetic field of 5 kOe. The magnetoresistance was then obtained relatively high $H_{ex}$ values of about 350 Oe at RT and about 1000 Oe at 4.2 K as shown in Fig. 2. We can reasonably attribute the high $H_{ex}$ values obtained for the fabricated MTJs to a lower net saturation magnetization of the synthetic ferrimagnetic trilayer compared with a saturation magnetization of the Co50Fe50 electrode.

Figure 3 shows the TMR ratio for a fabricated fully epitaxial, exchange-biased Co2Cr0.6Fe0.4Al/MgO/Co50Fe50 MTJ (CCFA-MTJ) as a function of temperature ($T$) from 4.2 K to RT (this is the same MTJ as shown in Fig. 2). For comparison, the TMR ratio as a function of $T$ is also plotted for a fully epitaxial, exchange-biased Co50Fe50/MgO/Co50Fe50 MTJ (a reference Co50Fe50-MTJ) identically fabricated with the same layer structure as that of the exchange-biased CCFA-MTJ except that the lower electrode CCFA was replaced with Co50Fe50. The Co50Fe50-MTJs were postfabrication annealed under the same annealing conditions as for the CCFA-MTJs (i.e., at 175 °C under a magnetic field of 5 kOe). The layer structure (from the substrate side) was Co50Fe50 (50 nm)/MgO (2.2 nm)/Co50Fe50 (3 nm)/Ru.
superconductor/AlO of the TMR ratio by a coherent tunneling contribution for epitaxial CCFA electrode. This result indicates enhancement to a more moderate value of $P$ at 42 K, and it reached 317% at 4.2 K (although it was slightly lower at RT).

We next estimated the tunneling spin polarization of the epitaxial CCFA electrode from the obtained TMR ratios. The TMR ratios for MTJs have been traditionally related to the spin polarizations at $E_F$, $P_1$, and $P_2$, of the ferromagnetic electrodes through Jullière's model (Ref. 6), i.e.,

$$\text{TMR} = 2P_1 P_2 (1-P_1 P_2).$$

Jullière's model was derived by assuming a loss of coherence in tunneling (i.e., nonconservation of the electron's wave vector component parallel to the interface). However, a straightforward application of Jullière's model for a TMR ratio of 317% at 4.2 K for fully epitaxial CCFA/MgO/Co$_{50}$Fe$_{50}$ MTJs with a Co$_{50}$Fe$_{50}$ electrode spin polarization of 0.50, derived from $dI/dV$ curves of superconductor/AlO$_x$/Co$_{50}$Fe$_{50}$ tunnel structures, corresponding to the originally defined spin polarization using majority- and minority-spin band density of states at $E_F$, results in an unrealistically high $P$ value exceeding 1.0 for the epitaxial CCFA electrode. This result indicates enhancement of the TMR ratio by a coherent tunneling contribution for fully epitaxial CCFA/MgO/Co$_{50}$Fe$_{50}$ MTJs. Furthermore, the obtained TMR ratios of 185% at 4.2 K and 125% at RT clearly indicate enhancement of the TMR ratio by a coherent tunneling contribution for the reference Co$_{50}$Fe$_{50}$-MTJs. Therefore, we estimated the tunneling spin polarization or effective spin polarization for the epitaxial Co$_{50}$Fe$_{50}$ electrode, $P_{\text{CoFe}}$, by applying Jullière's model for the TMR ratio of 185% at 4.2 K (125% at RT) of the reference Co$_{50}$Fe$_{50}$-MTJs. We obtained a $P_{\text{CoFe}}$ value of 0.69 at 42 K (0.62 at RT), which was higher than the above $P$ value of 0.50 derived from superconductor/AlO$_x$/Co$_{50}$Fe$_{50}$ tunnel structures. Similarly, we estimated the tunneling spin polarization or effective spin polarization for an epitaxial CCFA electrode in fully epitaxial CCFA/MgO/Co$_{50}$Fe$_{50}$ MTJs, $P_{\text{CCFA}}$, by applying Jullière's model for the TMR ratio of 317% at 4.2 K (109% at RT) of the CCFA-MTJs, along with a $P_{\text{CoFe}}$ value of 0.69 at 42 K (0.62 at RT) derived from the TMR ratio for the reference Co$_{50}$Fe$_{50}$ MTJs; in this case, we obtained a high tunneling spin polarization of 0.88 at 4.2 K (0.57 at RT) for the epitaxial CCFA thin film with the B2 structure. Although a rigorous comparison is not justified, the thus obtained $P_{\text{CCFA}}$ value of 0.88 is larger than the theoretically predicted $P_{\text{CCFA}}$ value of 0.78 (Ref. 18) even though we assumed an effective spin polarization of 0.69 at 42 K (0.62 at RT) for the epitaxial Co$_{50}$Fe$_{50}$ electrodes in the estimation rather than 0.50 at 42 K as was derived from superconductor/AlO$_x$/Co$_{50}$Fe$_{50}$ tunnel structures. This result also indicates a coherent tunneling contribution for fully epitaxial CCFA/MgO/Co$_{50}$Fe$_{50}$ MTJs.

Last, we will discuss the $T$ dependence of the TMR ratio of the fabricated CCFA-MTJs. If we use parameter $\gamma = \alpha(4.2 K)/\alpha(RT)$, where $\alpha$ is the TMR ratio, to represent the degree of $T$ dependence of the TMR ratio, $\gamma$ for the CCFA-MTJs was 2.9. This $\gamma$ value was higher than the value of 2.1 previously reported for CMS/MgO/Co$_{50}$Fe$_{50}$ MTJs [\(\alpha(\text{RT})=90\% \text{ and } \alpha(4.2 \text{ K})=192\%\)] (Ref. 14) and in contrast to a more moderate value of $\gamma=1.5$ [\(\alpha(\text{RT})=125\% \text{ and } \alpha(4.2 \text{ K})=185\%\)] for the reference Co$_{50}$Fe$_{50}$-MTJs. Regarding the $T$ dependences of $RA_{\text{AP}}$ and $RA_{\text{P}}$, $RA_{\text{AP}}$ also decreased with increasing $T$, while $RA_{\text{P}}$ was almost independent of $T$ for the CCFA-MTJs (not shown). [These $T$ dependences of $RA_{\text{AP}}$ and $RA_{\text{P}}$ were similar to those observed for Co$_{50}$Fe$_{50}$/MgO/Co$_{50}$Fe$_{16}$ MTJs (Ref. 19) and CMS/MgO/Co$_{50}$Fe$_{50}$ MTJs.11] These results indicate that the decreasing TMR ratio with increasing $T$ for the CCFA-MTJs was mainly due to the $RA_{\text{AP}}$ decrease. To clarify the reason for the strong $T$ dependence of the TMR ratio, or equivalently that of the $RA_{\text{AP}}$ observed for the CCFA/MgO/Co$_{50}$Fe$_{50}$ MTJs, further systematic study is needed.

In summary, we fabricated fully epitaxial, exchange-biased MTJs with a CCFA lower electrode and a MgO tunnel barrier. These MTJs exhibited high TMR ratios of 109% at RT and 317% at 4.2 K. A high tunneling spin polarization of 0.88 at 4.2 K was estimated for the epitaxial CCFA film with the B2 structure. The demonstrated high TMR ratios confirmed that fully epitaxial MTJs with a MgO tunnel barrier are promising as a key device structure for fully utilizing the potentially high spin polarization of Co-based full-Heusler alloy thin films.

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