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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

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Fully epitaxial magnetic tunnel junctions (MTJs) with exchange biasing were fabricated with a full-Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) thin film and a MgO tunnel barrier, where a $Co_{50}Fe_{50}$ upper electrode was used in a synthetic ferrimagnetic $Co_{50}Fe_{50}/Ru/Co_{90}Fe_{10}$ trilayer exchange-biased with an IrMn layer through the $Co_{90}Fe_{10}$/IrMn interface. The fabricated MTJs exhibited clear exchange-biased tunnel magnetoresistance (TMR) characteristics with high TMR ratios of 109% at room temperature and 317% at 4.2 K. A high tunneling spin polarization of 0.88 at 4.2 K was estimated for epitaxial CCFA films with the *B*2 structure. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2428412]

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.[1]

Cobalt-based full-Heusler alloys, whose composition is represented by Co_2YZ, 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_2YZ) thin film, where they used a polycrystalline $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) thin film as a lower 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_2MnSi (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_2YZ thin film of CCFA,[9–12] Co_2MnGe,[11,13] or Co_2MnSi (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 $Co_2MnSi/MgO/Co_{50}Fe_{50}$ MTJs.[14] 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 *B*2 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 ferrimagnetic) $Co_{50}Fe_{50}/Ru/Co_{90}Fe_{10}$ 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_{ex}) 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 8×10^{-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 $Co_2Cr_{0.57}Fe_{0.39}Al_{1.12}$, 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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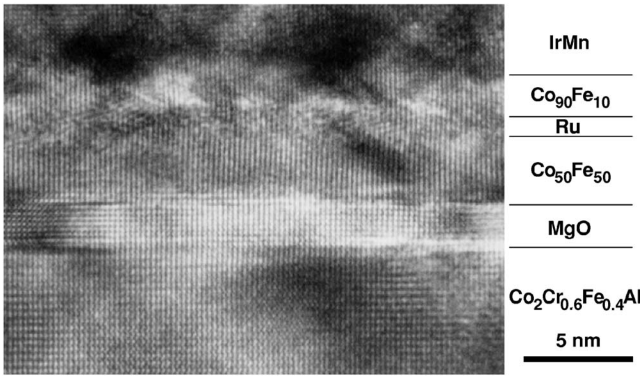

FIG. 1. Cross-sectional high-resolution transmission electron microscope image of a MTJ layer structure that consisted of $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) (50 nm)/MgO (2 nm)/$Co_{50}Fe_{50}$ (3 nm)/Ru (0.8 nm)/$Co_{90}Fe_{10}$ (2 nm)/IrMn (10 nm)/Ru cap (5 nm), along the [110] direction of the CCFA.

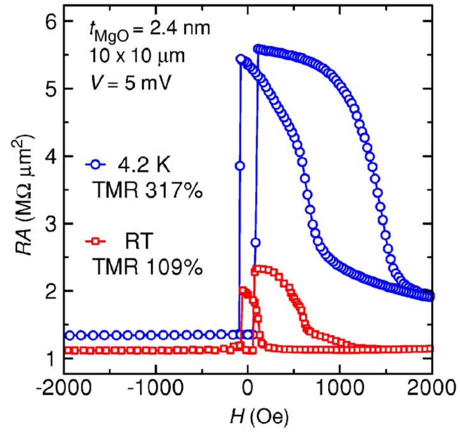

FIG. 2. (Color online) Typical magnetoresistance curves at RT and 4.2 K for a fully epitaxial, exchange-biased $Co_2Cr_{0.6}Fe_{0.4}Al$/MgO (2.4 nm)/$Co_{50}Fe_{50}$ MTJ. The junction size was 10×10 μm^2. The bias voltage was 5 mV.

was 10×10 μm^2. After the microfabrication, the MTJs were annealed at 175 °C for 1 h in a vacuum of 5×10^{-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 $(RA_{AP}-RA_P)/RA_P$, where RA_{AP} and RA_P are the respective resistance-area products for the antiparallel and parallel magnetization configurations between the upper and lower electrodes.

We will now describe the structural characterization of the fabricated layer structures. First, we will describe the structural properties of the CCFA/MgO/$Co_{50}Fe_{50}$ tunnel junction trilayer within the MTJ layer structure. RHEED patterns observed *in situ* for each layer during fabrication clearly indicated that the CCFA lower electrode, MgO tunnel barrier, and $Co_{50}Fe_{50}$ upper electrode grew epitaxially. X-ray diffraction measurement of the 50-nm-thick CCFA thin film annealed *in situ* at 500 °C showed that the film grew epitaxially and crystallized into the *B*2 structure. Figure 1 shows a cross-sectional high-resolution transmission electron microscope (HRTEM) image of the fabricated MTJ layer structure from the CCFA layer to the IrMn layer. This image clearly reveals that all the layers of the CCFA/MgO/$Co_{50}Fe_{50}$ basic tunnel junction trilayer were grown epitaxially and were single crystalline. It also confirmed that extremely smooth and abrupt interfaces were formed. All these structural properties agreed with our previous results.[9–12] Next, we will describe the structural properties of the $Co_{50}Fe_{50}$/Ru/$Co_{90}Fe_{10}$/IrMn quadrilayer which was part of the MTJ layer structure. We also observed streak patterns in RHEED patterns that were dependent on the incident direction of the electron beam for layers of Ru, $Co_{90}Fe_{10}$, and IrMn, indicating that the layers grew epitaxially on the single-crystal $Co_{50}Fe_{50}$ electrode. Furthermore, cross-sectional HRTEM lattice images (Fig. 1) clearly showed that all the layers of Ru, $Co_{90}Fe_{10}$, and IrMn were grown epitaxially on the single-crystal $Co_{50}Fe_{50}$ electrode and were single crystalline.

Figure 2 shows typical magnetoresistance curves at RT and 4.2 K for a fabricated fully epitaxial, exchange-biased CCFA/MgO/$Co_{50}Fe_{50}$ MTJ. The applied bias voltage was 5 mV. The MTJ exhibited clear exchange-biased TMR characteristics with high TMR ratios of 109% at RT and 317% at 4.2 K. These values are significantly higher than our previously reported values of 90% at RT and 240% at 4.2 K for fully epitaxial CCFA/MgO/$Co_{50}Fe_{50}$ MTJs, in which we used the difference in the coercive forces to form the antiparallel magnetization configurations between the CCFA lower electrode and the $Co_{50}Fe_{50}$ upper electrode.[12] We 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 $Co_{50}Fe_{50}$ electrode.

Figure 3 shows the TMR ratio for a fabricated fully epitaxial, exchange-biased CCFA/MgO/$Co_{50}Fe_{50}$ 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 $Co_{50}Fe_{50}$/MgO/$Co_{50}Fe_{50}$ MTJ (a reference $Co_{50}Fe_{50}$-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 $Co_{50}Fe_{50}$. The $Co_{50}Fe_{50}$-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 $Co_{50}Fe_{50}$ (50 nm)/MgO (2.2 nm)/$Co_{50}Fe_{50}$ (3 nm)/Ru

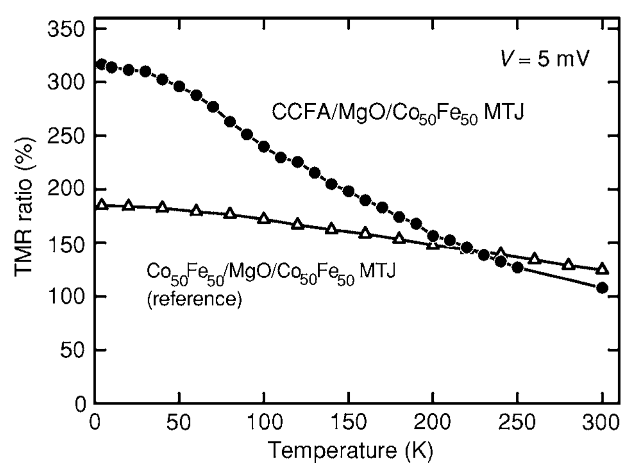

FIG. 3. Temperature dependence of the TMR ratio of a fully epitaxial, exchange-biased $Co_2Cr_{0.6}Fe_{0.4}Al$/MgO/$Co_{50}Fe_{50}$ MTJ (the same MTJ shown in Fig. 2) at V=5 mV compared with that of a fully epitaxial, exchange-biased $Co_{50}Fe_{50}$/MgO (2.2 nm)/$Co_{50}Fe_{50}$ MTJ (the reference $Co_{50}Fe_{50}$-MTJ).

(0.8 nm)/$Co_{90}Fe_{10}$ (2 nm)/IrMn (10 nm)/Ru cap (5 nm), and the structure was grown on a MgO-buffered MgO substrate. The $Co_{50}Fe_{50}$-MTJs showed TMR ratios of 185% at 4.2 K and 125% at RT. As shown in Fig. 3, the TMR ratio of the CCFA-MTJ was definitely higher than that of the $Co_{50}Fe_{50}$-MTJ below about 220 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., $TMR = 2P_1P_2/(1-P_1P_2)$. 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).[16] 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,[17] 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_{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_{CoFe} value of 0.69 at 4.2 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.[17] 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_{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_{CoFe} value of 0.69 at 4.2 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_{CCFA} value of 0.88 is larger than the theoretically predicted P_{CCFA} value of 0.78 (Ref. 18) even though we assumed an effective spin polarization of 0.69 at 4.2 K (0.62 at RT) for the epitaxial $Co_{50}Fe_{50}$ electrodes in the estimation rather than 0.50 at 4.2 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 $\gamma = \alpha(4.2\ \mathrm{K})/\alpha(\mathrm{RT})$, where α is the TMR ratio, to represent the degree of T dependence of the TMR ratio, γ for the CCFA-MTJs was 2.9. This γ value was higher than the value of 2.1 previously reported for CMS/MgO/$Co_{50}Fe_{50}$ MTJs [$\alpha(\mathrm{RT})=90\%$ and $\alpha(4.2\ \mathrm{K})=192\%$] (Ref. 14) and in contrast to a more moderate value of $\gamma=1.5$ [$\alpha(\mathrm{RT})=125\%$ and $\alpha(4.2\ \mathrm{K})=185\%$] for the reference $Co_{50}Fe_{50}$-MTJs. Regarding the T dependences of RA_{AP} and RA_P, RA_{AP} also decreased with increasing T, while RA_P was almost independent of T for the CCFA-MTJs (not shown). [These T dependences of RA_{AP} and RA_P were similar to those observed for $Co_{70}Fe_{30}$/MgO/$Co_{84}Fe_{16}$ MTJs (Ref. 19) and CMS/MgO/$Co_{50}Fe_{50}$ MTJs.[14]] These results indicate that the decreasing TMR ratio with increasing T for the CCFA-MTJs was mainly due to the RA_{AP} decrease. To clarify the reason for the strong T dependence of the TMR ratio, or equivalently that of the RA_{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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