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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(Received 25 September 2006; accepted 29 November 2006; published online 4 January 2007)

Fully epitaxial magnetic tunnel junctions (MTJs) with exchange biasing were fabricated with a full-Heusler alloy Co$_2$Cr$_{0.6}$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$_{50}$Fe$_{10}$ trilayer exchange-biased with an IrMn layer through the Co$_{50}$Fe$_{10}$/IrMn interface. The fabricated MTJs exhibited high tunnel-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 $E_F$. Cobalt-based full-Heusler alloys, whose composition is represented by Co$_3$YZ, have attracted much interest due to the half-metallic nature theoretically predicted for some of these alloys and their high Curie temperatures, which are well above room temperature (RT). 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. Inomata et al. first demonstrated a relatively high TMR ratio of 16% at RT for MTJs using a Co-based full-Heusler alloy (Co$_3$YZ) thin film, where they used a polycrystalline Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al (CCFA) thin film as a lower electrode and an amorphous AlO$_x$ tunnel barrier. 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.

We recently developed fully epitaxial MTJs that have a Co$_3$YZ thin film of CCFA, Co$_3$MnGe, or Co$_3$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. 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. 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, 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. 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$_{50}$Fe$_{10}$ trilayer exchange-biased by an IrMn antiferromagnetic layer through the Co$_{50}$Fe$_{50}$/IrMn interface to obtain a high exchange-bias field value ($H_e$) 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$_{50}$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$_{2.5}$Cr$_{0.5}$Fe$_{0.9}$Al$_{1.2}$, 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$_{50}$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...
was 10 × 10 μm². 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 measured through a dc four-probe method at temperatures.

Next, we will describe the structural properties of the CCFA/MgO/Co50Fe50 tunnel junction trilayer which was part of 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 Co50Fe50 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 B2 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/Co50Fe50 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 characterization of the fabricated layer structures. First, we will describe the structural properties of the Co50Fe50/Ru/Co90Fe10/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, Co90Fe10, and IrMn, indicating that the layers grew epitaxially on the single-crystal Co50Fe50 electrode. Furthermore, cross-sectional HRTEM lattice images (Fig. 1) clearly showed that all the layers of Ru, Co90Fe10, and IrMn were grown epitaxially on the single-crystal Co50Fe50 electrode and were single crystalline.

Figure 2 shows typical magnetoresistance curves at RT and 4.2 K for a fabricated fully epitaxial, exchange-biased Co50Fe50/MgO/Co50Fe50 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/Co50Fe50 MTJs, in which we used the difference in the coercive forces to form the antiparallel magnetization configurations between the CCFA lower electrode and the Co50Fe50 upper electrode.12 We obtained relatively high Hex 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 Hex 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 Co50Fe50/MgO/Co50Fe50 MTJ (CCFA-CCFA-MTJ) except that the lower electrode CCFA was replaced with Co50Fe50. The Co90Fe50-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 (0.8 nm)/Co50Fe50 (2 nm)/IrMn (10 nm)/Ru cap (5 nm), along the [110] direction of the CCFA.
superconductor/AlO of the TMR ratio by a coherent tunneling contribution for epitaxial CCFA electrode. This result indicates enhancement of the reference Co50Fe50 tunneling spin polarization of 0.50, derived from superconductor/AlO,

Similarly, we estimated the tunneling spin polarization or effective spin polarization for the epitaxial Co50Fe50 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 Co50Fe50-MTJs. We obtained a \( P_{\text{CoFe}} \) value of 0.69 at 4.2 K (0.62 at RT), which was higher than the above value of 0.50 derived from superconductor/AlO,

However, a straightforward application of Jullière’s model for a TMR ratio of 317% at 4.2 K for fully epitaxial CCFA/MgO/Co50Fe50 MTJs with a Co50Fe50 electrode spin polarization of 0.50, derived from \( dI/dV \) curves of superconductor/AlO,

Therefore, we estimated the tunneling spin polarization or effective spin polarization for the epitaxial Co50Fe50 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 Co50Fe50-MTJs. We obtained a \( P_{\text{CoFe}} \) value of 0.69 at 4.2 K (0.62 at RT), which was higher than the above value of 0.50 derived from superconductor/AlO,

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.

This work was partly supported by a Grant-in-Aid for Scientific Research (B) (Grant No. 18360143), a Grant-in-Aid for Creative Scientific Research (Grant No. 14GS0301), and a Grant-in-Aid for Young Scientists (B) (Grant No. 17760267) from the Ministry of Education, Culture, Sports, Science and Technology, Japan. One of the authors (T.M.) was also supported by a Research Fellowship for Young Scientists from the Japan Society for the Promotion of Science.