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Improved tunnel magnetoresistance characteristics of magnetic tunnel junctions with a Heusler alloy thin film of Co$_2$MnGe and a MgO tunnel barrier

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We fabricated magnetic tunnel junctions (MTJs) with a Co-based full-Heusler alloy thin film of Co$_2$MnGe (CMG) and a MgO tunnel barrier. The microfabricated MTJs with a Co-rich CMG film showed relatively high tunnel magnetoresistance ratios of 83% at room temperature and 185% at 4.2 K. These values are much higher than those previously obtained for CMG/MgO MTJs with a Co-deficient CMG film. © 2007 American Institute of Physics.

Cobalt-based full-Heusler alloy (Co$_{2YZ}$) thin films have recently attracted much interest as highly promising ferromagnetic electrodes for spintronic devices.$^{1-7}$ This is because of the half-metallic ferromagnetic nature$^8$ theoretically predicted for some of these alloys,$^9,10$ and because of their high Curie temperatures, which are well above room temperature (RT).$^{11,12}$ Relatively high tunnel magnetoresistance (TMR) ratios have been demonstrated at RT for magnetic tunnel junctions (MTJs) with a Co$_{2YZ}$ thin film.$^{13-23}$ We developed fully epitaxial MTJs with a Co$_{2YZ}$ thin film and a MgO tunnel barrier,$^{16-19,22-24}$ and showed a relatively high TMR ratio of 109% at RT (317% at 4.2 K) for Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al/MgO/Co$_9$Ge$_{50}$ MTJs,$^{23}$ and a TMR ratio of 90% at RT (192% at 4.2 K) for Co$_2$MnSi (CMS)/MgO/Co$_9$Ge$_{50}$ MTJs (CMS-MTJs).$^{22}$ Compared with these MTJs, previously reported fully epitaxial Co$_2$MnGe (CMG)/MgO/Co$_9$Ge$_{50}$ MTJs (CMG-MTJs)$^{22}$ showed a much lower TMR ratio of 14% at RT (70% at 7 K).$^{17,18}$ The CMG film used in the previous study was a Co-deficient CMG film (the film composition was Co$_2$Mn$_{0.53}$Ge$_{1.7}$),$^{17,18}$ while the CMS film used in the previous study was a Co-rich CMS film (the film composition was Co$_2$Mn$_{0.54}$Si$_{0.46}$).$^{22}$ Hereafter, we refer to this Co-deficient CMG film as the CMG$_d$ film. Our purpose in the present study was to improve the TMR characteristics of CMG-MTJs. For this purpose, we fabricated CMG-MTJs with a Co-rich CMG film and investigated their TMR characteristics.

We fabricated epitaxial MTJs that consisted of a CMG thin film and a wedge-shaped MgO tunnel barrier. The composition of the CMG film used in this study was determined to be Co$_2$Mn$_{0.73}$Ge$_{0.43}$ (Co-rich CMG), with an accuracy of 2%–3% for each element, through inductively coupled plasma analysis. This Co-rich film composition was obtained by using a sputtering target with a composition of Co$_2$Mn$_{0.5}$Ge$_{0.5}$. Hereafter, we refer to this Co-rich CMG film as the CMG$_r$ film. The fabricated epitaxial MTJ layer structure (from the substrate side) consisted of a MgO buffer layer (10 nm), a CMG lower electrode (50 nm), a MgO tunnel barrier (1.6–2.8 nm), a Co$_9$Fe$_{50}$ upper electrode (3 nm), a layer of Ru (0.8 nm), a layer of Co$_9$Fe$_{50}$ (2 nm), a layer of IrMn (10 nm), and a Ru cap (5 nm); this structure was grown on a MgO(001) substrate. Each layer in the MTJ layer structure was successively deposited in an ultrahigh vacuum chamber (with a base pressure of around 6 × 10$^{-10}$ Pa) through the combined use of magnetron sputtering and electron beam (EB) evaporation. The CMG layer was deposited at RT using magnetron sputtering and subsequently annealed in situ at 500–600 °C for 15 min. The MgO tunnel barrier was deposited by EB evaporation at RT. The pressure during the deposition of the MgO tunnel barrier was around 6 × 10$^{-7}$ Pa. The nominal thickness of the MgO tunnel barrier ($t_{MgO}$) was varied from 1.6 to 2.8 nm on each 20 × 20 mm$^2$ substrate by using a linearly moving shutter during fabrication. All the layers in the MTJ layer structure were deposited with a magnetic field applied. We fabricated MTJs with the layer structure described above by using photolithography and Ar ion milling. The fabricated junction sizes were from 8 × 8 to 10 × 10 μm$^2$. The magnetoresistance was measured with a magnetic field applied along the [110] axis of the Co$_{2YZ}$ at temperatures from 4.2 K to RT using a dc four-probe method. We defined the TMR ratio as $R_{AP}/R_A - R_P/R_A$, where $R_{AP}$ and $R_P$ are the respective resistance-area products for the antiparallel and parallel magnetization configurations between the upper and lower electrodes. As shown below, as-fabricated (i.e., not ex situ annealed) MTJs showed exchange-biased TMR characteristics. Transport properties of as-fabricated MTJs are described below.

X-ray pole figure scans of a CMG film deposited at RT and subsequently annealed at 600 °C showed fourfold symmetry of the CMG (111) peaks at $\chi$=54.7°, which gives direct evidence that the 600 °C annealed film is epitaxial and crystallized in the L2$_1$ structure. Because the $\varphi$ values for the CMG (111) peaks were shifted by 45° with respect to those of the MgO (111) peaks, the crystallographic relationship was CMG (001)[100] // MgO (001)[110] on a 45° in-plane rotation. These structural properties are similar to those previously observed for the CMG$_d$ films.$^6,17$

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We observed the surface morphologies of the 45-nm-thick CMG, films deposited on MgO buffer layers (10 nm) using atomic force microscopy. The root mean square (rms) values of the surface roughness increased with postdeposition annealing, from a rms roughness of 0.16 nm for the as-deposited film to 0.41 nm for the 600 °C annealed film. This dependence on postdeposition annealing temperature was in contrast to that previously observed for the CMG, film, i.e., the rms values of the surface roughness decreased with postdeposition annealing, from a rms value of roughness of 0.40 nm for the as-deposited film to 0.26 nm for the 600 °C annealed film.\(^1\)

Figure 1 shows reflection high-energy electron diffraction (RHEED) patterns, along the azimuth of [110] of MgO corresponding to [110] of CMG, obtained in situ for a Co-rich Co\(_{2}\)MnGe (CMG) film during fabrication. The CMG film was deposited at RT on a MgO buffer layer (10 nm) and subsequently in situ annealed step by step at 200–600 °C. The substrate was a MgO (001) single crystal. The arrows indicate streak patterns corresponding to CMG (111) reflection. (c) A RHEED pattern for comparison, along the azimuth of [110] of MgO corresponding to [110] of CMG, obtained previously for an as-deposited Co-deficient CMG film during fabrication (Ref. 17). Streak patterns corresponding to the CMG (111) reflection were not observed.

Next, we will describe the spin-dependent tunneling characteristics of fabricated epitaxial MTJs. Figure 2(a) shows typical magnetoresistance curves at a bias voltage (V) of 5 mV at RT and 4.2 K for an as-fabricated CMG/MgO/Co\(_{50}\)Fe\(_{50}\) MTJ with a Co-rich CMG film where the CMG lower electrode was in situ annealed at 600 °C after deposition (t\(_{MGO}\)=2.4 nm). The junction size was 8×8 \(\mu\)m\(^2\). TMR ratios were 83% at RT and 185% at 4.2 K. (b) TMR ratio, as well as RA\(_{AP}\) and RA\(_P\), at V=5 mV for the same MTJ shown in Fig. 2(a) as a function of temperature from 4.2 K to RT, 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.

FIG. 1. [(a)–(d)] RHEED patterns, along the azimuth of [100] of MgO corresponding to [110] of CMG, obtained in situ for a Co-rich Co\(_{2}\)MnGe (CMG) film during fabrication. The CMG film was deposited at RT on a MgO buffer layer (10 nm) and subsequently in situ annealed step by step at 200–600 °C. The substrate was a MgO (001) single crystal. The arrows indicate streak patterns corresponding to CMG (111) reflection. (e) A RHEED pattern for comparison, along the azimuth of [100] of MgO corresponding to [110] of CMG, obtained previously for an as-deposited Co-deficient CMG film during fabrication (Ref. 17). Streak patterns corresponding to the CMG (111) reflection were not observed.

FIG. 2. (a) Typical magnetoresistance curves at a bias voltage (V) of 5 mV at RT and 4.2 K for a fabricated Co\(_{2}\)MnGe (CMG)/MgO/Co\(_{50}\)Fe\(_{50}\) MTJ with a Co-rich CMG film where the CMG lower electrode was in situ annealed at 600 °C after deposition (t\(_{MGO}\)=2.4 nm). The junction size was 8×8 \(\mu\)m\(^2\). TMR ratios were 83% at RT and 185% at 4.2 K. (b) TMR ratio, as well as RA\(_{AP}\) and RA\(_P\), at V=5 mV for the same MTJ shown in Fig. 2(a) as a function of temperature from 4.2 K to RT, 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.

\(\gamma\) is the TMR ratio, to represent the degree of T dependence of the TMR ratio, \(\gamma\) for CMG/MgO/Co\(_{50}\)Fe\(_{50}\) MTJs was 2.2. This value is comparable to the previously obtained \(\gamma\) for CMS/MgO/Co\(_{50}\)Fe\(_{50}\) MTJs (a TMR ratio of 192% at 4.2 K and 90% at RT)\(^2\) and significantly lower than \(\gamma\) =5.0, which was previously obtained for CMG\(_d\)/MgO/Co\(_{50}\)Fe\(_{50}\) MTJs (a TMR ratio of 70% at 7 K
FIG. 3. RA<sub>P</sub> and TMR ratio at RT (measured at V = 5 mV) as a function of MgO tunnel barrier thickness t<sub>MgO</sub> for fabricated Co<sub>50</sub>MnGe (CMG)/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs with a Co-rich CMG film where the CMG lower electrode was in situ annealed at 500 °C after deposition. RA<sub>P</sub> represents the resistance-area product RA for the parallel magnetization configuration. The junction size was 10 × 10 µm<sup>2</sup>. The scale of the vertical axis for RA<sub>P</sub> is logarithmic.

and 14% at RT).<sup>17,18</sup> As shown in Fig. 2(b), RA<sub>AP</sub> also increased with decreasing T, while RA<sub>P</sub> was almost independent of T. These behaviors were similar to those previously observed for CCFAMA/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs (Ref. 24) and CMS/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs. These behaviors were also observed for CoO<sub>5</sub>Fe<sub>28</sub>/MgO/Co<sub>5</sub>Fe<sub>16</sub> MTJs. These behaviors were also observed for CoO<sub>5</sub>Fe<sub>28</sub>/MgO/Co<sub>5</sub>Fe<sub>16</sub> MTJs.<sup>25</sup>

Figure 3 plots RA<sub>P</sub> and the TMR ratio at RT (measured at V = 5 mV) as a function of t<sub>MgO</sub> for the fabricated CMG/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs where the CMG lower electrode was in situ annealed at 500 °C after deposition. The junction size was 10 × 10 µm<sup>2</sup>. A clear exponential dependence of RA<sub>P</sub> on t<sub>MgO</sub> was observed for the t<sub>MgO</sub> range of 2.0–2.8 nm, indicating typical tunnel junction behavior. Relatively high TMR ratios from 72% to 88% were obtained at RT for this wide range of t<sub>MgO</sub> from 2.0–2.8 nm.

We estimated the spin polarization for the CMG electrodes by using Jullière’s model for the TMR ratio;<sup>26</sup> TMR = 2P<sub>1</sub>P<sub>2</sub>/(1 – P<sub>1</sub>P<sub>2</sub>), where P<sub>1</sub> and P<sub>2</sub> are the spin polarizations at the Fermi level (E<sub>F</sub>) of the ferromagnetic electrodes in MTJs. We first estimated the effective spin polarization for the CoO<sub>5</sub>Fe<sub>28</sub> electrode from the TMR ratio of 146% at 4.2 K (96% at RT) obtained for the identically fabricated epitaxial CoO<sub>5</sub>Fe<sub>28</sub>/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs by using Jullière’s model. Thus, the effective spin polarization value obtained for the CoO<sub>5</sub>Fe<sub>28</sub> electrode (P<sub>CoFe</sub>) was 0.65 at 4.2 K (0.57 at RT). Then, we estimated the effective spin polarization of the CMG film (P<sub>CMG</sub>) from the TMR ratio of 185% at 4.2 K (83% at RT) for the epitaxial CMG/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs by using Jullière’s model with P<sub>CoFe</sub> of 0.65 at 4.2 K (0.57 at RT). The obtained effective spin polarization or tunneling spin polarization values of P<sub>CMG</sub> were 0.74 at 4.2 K and 0.51 at RT. These P<sub>CMG</sub> values are comparable to previously obtained values of 0.75 at 4.2 K and 0.54 at RT for the CMS films.<sup>22</sup>

The enhanced TMR ratios for the CMG/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs fabricated with a Co-rich CMG film demonstrated that the lower TMR ratios observed previously for the CMG/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs with a Co-deficient CMG film<sup>17,18</sup> were not due to an intrinsic property of the Co-based full-Heusler alloy of Co<sub>5</sub>MnGe. The improved TMR characteristics in terms of the TMR ratio or the effective spin polarization at E<sub>F</sub> are probably related to the improved structural properties of the CMG film in terms of the degree of structural order.

In summary, we fabricated epitaxial MTJs with a Co-based full-Heusler alloy thin film of CMG and a MgO tunnel barrier. The microfabricated MTJs with a Co-rich CMG film demonstrated relatively high tunnel magnetoresistance ratios of 83% at RT and 185% at 4.2 K. These values are much higher than those previously obtained for CMG/MgO/Co<sub>50</sub>Fe<sub>50</sub> MTJs with a Co-deficient CMG film.

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