Giant tunneling magnetoresistance in fully epitaxial Co$_2$(Mn,Fe)Si/MgO/Co$_2$(Mn,Fe)Si magnetic tunnel junctions

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Co-based Heusler alloy thin films have been extensively studied as promising ferromagnetic electrode materials for spintronic devices, including MTJs and GMR devices, and for spin injection into semiconductors. Previously, we have shown that harmful defects in Heusler alloy thin films of Co$_2$MnSi (CMS) and Co$_2$MnGe can be suppressed by appropriately controlling the film composition, i.e., Co$_{1-x}$Mn$_x$ antisites detrimental to the half-metallicity can be suppressed by a Mn-rich composition. As a result, we have recently demonstrated high TMR ratios of up to 1995% at 4.2 K and up to 354% at 290 K for fully epitaxial CoFe$_{1-y}$Mn$_y$Si/MgO/CoFe$_{1-y}$Mn$_y$Si MTJs (CMS MTJs) having Mn-rich CMS electrodes. Half-metallic Heusler alloys feature a lot of variety of materials, including quaternary alloys. The purpose of the present study was to clarify how the substitution of Fe for Mn in CMS changes the half-metallicity by investigating the spin-dependent tunneling characteristics of epitaxial MTJs having (Mn+Fe)-rich Co$_2$(Mn,Fe)Si (CMFS) electrodes and a MgO barrier. The quaternary CMS system has been applied to GMR devices, demonstrating a high device performance, and AlO$_x$-barrier MTJs.

The fabricated MTJ layer structures were as follows: (from the substrate side) MgO buffer (10 nm)/Co$_9$Fe$_{20}$ (CoFe) buffer (30 nm)/CMFS lower electrode (3 nm)/MgO barrier (1.4-3.2 nm)/CMFS upper electrode (3 nm)/CoFe (1.1 nm)/IrMn (10 nm)/Ru cap (5 nm), (CMFS MTJ), grown on MgO(001) substrates. CMFS films have film compositions of Co$_{2-x}$Mn$_x$Fe$_{2.1}$Si$_{1.84}$ with various $\alpha$ and $\beta$ values prepared by co-sputtering from an almost stoichiometric CMS target, a Mn target and an Fe target. The film composition of the original CMS film sputtered from only the CMS target was Mn- and Si-deficient Co$_{1.77}$Mn$_{0.84}$Si. Aberration-corrected atomic number ($Z$)-contrast scanning TEM images clearly showed that all the layers of a CMFS MTJ having Co$_{2-x}$Mn$_{1.24}$Fe$_{0.16}$Si$_{1.84}$ electrode were grown epitaxially and single-crystalline. It also demonstrated that both the lower and upper CMFS electrodes consisted of regions of the L2$_1$ structure and those of the B2 structure. Furthermore, atomically flat lower and upper interfaces were revealed.

Figure 1 shows the TMR ratios of (Mn+Fe)-rich CMFS MTJs having various values of Fe composition $\beta$ in combination with Mn composition $\alpha$ for fixed $\alpha+\beta$ ($=\delta$) values ranging from $\delta = 1.30$ to 1.40. As shown in Fig. 1, these MTJs showed high TMR ratios ranging from around 350% up to 429% at 290 K for a wide range of Fe substitution. The highest TMR ratios obtained by Fe substitution were 2611% at 4.2 K and 429% at 290 K (Fig. 2) for a MTJ having Co$_{2}$Mn$_{1.24}$Fe$_{0.16}$Si$_{1.84}$ electrodes. These values are significantly higher than the TMR ratios of 2114% at 4.2 K and 370% at 290 K obtained for an identically fabricated CMS MTJ having Co$_{2}$Mn$_{1.30}$Si$_{1.84}$ electrodes. The increased TMR ratios at 4.2 K can be explained by the enhanced half-metallicity evidenced by the increased half-metallic gap of Co$_{2}$Mn$_{1.24}$Fe$_{0.16}$Si$_{1.84}$ obtained by $dI/dV$-V characteristics at 4.2 K, compared with Co$_{2}$Mn$_{1.30}$Si$_{1.84}$ electrodes and by the reduced strain in the 3-nm-thick Co$_{2}$Mn$_{1.24}$Fe$_{0.16}$Si$_{1.84}$ electrodes suggested by the increased in-plane lattice constant of a Co$_{2}$Mn$_{1.24}$Fe$_{0.16}$Si$_{1.84}$ film compared with a Mn-rich CMS film.