Giant tunneling magnetoresistance in fully epitaxial Co₂(Mn,Fe)Si/MgO/Co₂(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^{1,2} and GMR devices,^{3,4} and for spin injection into semiconductors.⁵ Previously, we have shown that harmful defects in Heusler alloy thin films of Co₂MnSi (CMS) and Co₂MnGe can be suppressed by appropriately controlling the film composition, *i.e.*, Co_{Mn} antisites detrimental to the half-metallicity can be suppressed by a Mn-rich composition.¹ 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-buffered CMS/MgO/CMS MTJs (CMS MTJs) having Mn-rich CMS electrodes.² 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 Co₂(Mn,Fe)Si (CMFS) electrodes and a MgO barrier. The quaternary CMFS system has been applied to GMR devices,^{3,4} demonstrating a high

device performance, and AlO_x-barrier MTJs.⁶

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The fabricated MTJ layer structures were as follows: (from the substrate side) MgO buffer (10 nm)/Co₅₀Fe₅₀ (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 having film compositions of $Co_2Mn_{\alpha}Fe_{\beta}Si_{0.84}$ with various α and β values were 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 Co₂Mn_{0.73}Si_{0.84}. Aberration-corrected atomic number (Z)-contrast scanning TEM images clearly showed that all the layers of a CMFS MTJ having Co₂Mn_{1.24}Fe_{0.16}Si_{0.84} electrode were grown epitaxially and single-crystalline. It also demonstrated that both the lower and upper CMFS electrodes consisted of regions of the L2₁ 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 β in combination with Mn composition α for fixed $\alpha + \beta$ (= δ) values ranging from δ = 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 $Co_2Mn_{1.24}Fe_{0.16}Si_{0.84}$ (or $Co_2(Mn_{0.89}Fe_{0.11})_{1.40}Si_{0.84}$) 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 Co₂Mn_{1.30}Si_{0.84} electrodes. The increased TMR ratios at 4.2 K can be explained by the enhanced half-metallicity evidenced by the increased half-metal gap of Co₂Mn_{1.24}Fe_{0.16}Si_{0.84}, obtained by dI/dV-V characteristics at 4.2 K, compared with Co₂Mn_{1.40}Si_{0.84} electrodes and by the reduced strain in the 3-nm-thick Co2Mn1.24Fe0.16Si0.84 electrodes suggested by the increased in-plane lattice constant of a Co₂Mn_{1,24}Fe_{0,16}Si_{0.84} film compared with a Mn-rich CMS film.



Fig. 1. TMR ratios at 290K for CMFS/MgO/ CMFS MTJs having various values of Fe composition β in combination with Mn composition α for fixed $\alpha + \beta$ (= δ) values of δ = 1.30, 1.35 and 1.40.



Fig. 2. Typical TMR curves at 4.2 K and 290 K for a CMFS/MgO/CMFS MTJ having $Co_2Mn_{1,24}Fe_{0,16}Si_{0.84}$ electrodes.

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