Effect of off-stoichiometry on half-metallicity of quaternary Heusler alloy \( \text{Co}_2(\text{Mn,Fe})\text{Si} \) investigated through saturation magnetization and tunneling magnetoresistance

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We have recently investigated the effect of off-stoichiometry on the tunneling magnetoresistance (TMR) of the quaternary Heusler alloy \( \text{Co}_2(\text{Mn,Fe})\text{Si} \) (CMFS)-based magnetic tunnel junctions (MTJs) and showed that the (Mn+Fe)-rich composition is critical to suppressing harmful \( \text{Co}_{\text{Mn/Fe}} \) antisites and obtaining half-metallicity [1]. Furthermore, we demonstrated giant TMR ratios of 2610% at 4.2 K and 429% at 290 K for MTJs having Mn-rich, lightly Fe-doped CMFS electrodes [1]. The purpose of the present study was to clarify the origin of the giant TMR ratio of MTJs with Mn-rich, lightly Fe-doped CMFS electrodes. To do this, we experimentally investigated the film composition dependence of the saturation magnetization per formula unit, \( \mu_s \), of CMFS films with various compositions of \( \alpha' \) and \( \beta' \) in \( \text{Co}_2(\text{Mn}_{\alpha'},\text{Fe}_{\beta'})\text{Si}_{0.84} \).

Figure 1 shows the film composition dependence of the experimental \( \mu_s \) of \( \text{Co}_2(\text{Mn}_{\alpha'},\text{Fe}_{\beta'})\text{Si}_{0.84} \) and \( \text{Co}_2\text{Mn}_{1.40}\text{Si}_{0.84} \) films along with the half-metallic Slater-Pauling values \( (Z-24) \) and the theoretical total spin magnetic moment/f.u., \( m_{\text{spin}} \), calculated using the antisite-based site-specific formula unit (SSFU) composition model [1,2]. Although the experimental \( \mu_s \) was lower than both \( Z-24 \) and theoretical \( m_{\text{spin}} \) for Mn-rich \( \text{Co}_2\text{Mn}_{1.40}\text{Si}_{0.84} \), its value for \( \text{Co}_2\text{Mn}_{1.40}\text{Fe}_{0.16}\text{Si}_{0.84} \) in which a small amount of Mn was replaced by Fe for \( \text{Co}_2\text{Mn}_{1.40}\text{Si}_{0.84} \) got almost close to the half-metallic \( Z-24 \). Figure 2 shows how the TMR ratio at 4.2 K of MTJs with Mn-rich, lightly Fe-doped \( \text{Co}_2\text{Mn}_{\alpha'}\text{Fe}_{\beta'}\text{Si}_{0.84} \) electrodes depends on \( \alpha' \) ranging from \( \alpha' = 1.14 \) (\( \delta = \alpha' + \beta' = 1.30 \)) to \( \alpha' = 1.24 \) (\( \delta = 1.40 \)) along with the dependence of the TMR ratio for CMS MTJs with \( \text{Co}_2\text{Mn}_{\alpha}\text{Si}_{0.84} \) electrodes on the Mn composition \( \alpha \) ranging from \( \alpha = 0.73 \) to 1.40. The drop in the TMR of the CMS MTJ with Mn-rich \( \alpha = 1.40 \) and the contrasted further increase in the TMR of CMFS MTJs with increasing \( \delta \) from \( \alpha = 1.30 \) to 1.40 with a small amount of \( \beta' \) of 0.16 was consistent with the dependence of \( \mu_s \) shown in Fig. 1. The theoretical \( m_{\text{spin}} \) values well explained the experimental \( \mu_s \) values except Mn-rich \( \text{Co}_2\text{Mn}_{1.24}\text{Si}_{0.84} \) (\( \alpha = 1.40 \) CMS). This discrepancy can be attributed to the assumed nominal half-metallic SSFU composition for Mn-rich \( \alpha = 1.40 \) CMS. Thus, the origin of the giant TMR for MTJs with Mn-rich, lightly Fe-doped CMFS electrodes was attributed to that (1) the nominal half-metallic SSFU composition was recovered by replacing a small amount of Mn by Fe for \( \alpha = 1.40 \) CMS and (2) the residual \( \text{Co}_{\text{Mn/Fe}} \) antisites were further reduced by (Mn+Fe)-rich composition.

References