Effect of off-stoichiometry on half-metallicity of quaternary Heusler alloy $Co_2(Mn,Fe)Si$ 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 $Co_2(Mn,Fe)Si$ (CMFS)-based magnetic tunnel junctions (MTJs) and showed that the (Mn+Fe)-rich composition is critical to suppressing harmful $Co_{Mn/Fe}$ 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, μ_s, of CMFS films with various compositions of α' and β' in $Co_2(Mn_{\alpha'}Fe_{\beta'})Si_{0.84}$.

Figure 1 shows the film composition dependence of the experimental μ_s of $Co_2(Mn_{\alpha'}Fe_{\beta'})Si_{0.84}$ and $Co_2Mn_{1.40}Si_{0.84}$ films along with the half-metallic Slater-Pauling values (Z_t-24) and the theoretical total spin magnetic moment/f.u., m_{spin}, calculated using the antisite-based site-specific formula unit (SSFU) composition model [1,2]. Although the experimental μ_s was lower than both Z_t-24 and theoretical m_{spin} for Mn-rich $Co_2Mn_{1.40}Si_{0.84}$, its value for $Co_2Mn_{1.24}Fe_{0.16}Si_{0.84}$ in which a small amount of Mn was replaced by Fe for $Co_2Mn_{1.40}Si_{0.84}$ got almost close to the half-metallic Z_t-24. Figure 2 shows how the TMR ratio at 4.2 K of MTJs with Mn-rich, lightly Fe-doped $Co_2Mn_{\alpha'}Fe_{0.16}Si_{0.84}$ electrodes depends on α' 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 $Co_2Mn_\alpha Si_{0.84}$ electrodes on the Mn composition α 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 δ from $\alpha = 1.30$ to 1.40 with a small amount of β' of 0.16 was consistent with the dependence of μ_s shown in Fig. 1. The theoretical m_{spin} values well explained the experimental μ_s values except Mn-rich $Co_2Mn_{1.40}Si_{0.84}$ ($\alpha = 1.40$ 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 $Co_{Mn/Fe}$ antisites were further reduced by (Mn+Fe)-rich composition.

References

[1]. H.-x. Liu et al., J. Phys. D: Appl. Phys. 48, 164001 (2015). [2]. G.-f. Li, et al., Phys. Rev. B 89, 014428 (2014).

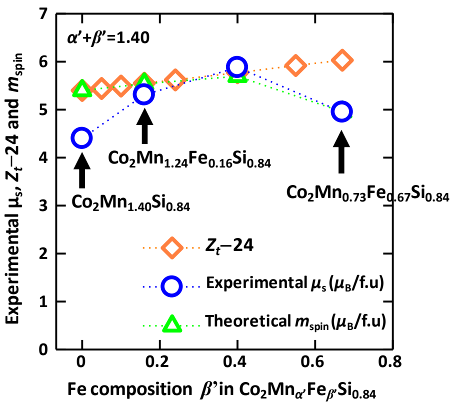

Fig.1. Saturation magnetization per formula unit of $Co_2Mn_{1.40}Si_{0.84}$ and $Co_2Mn_{\alpha'}Fe_{\beta'}Si_{0.84}$ films with $\alpha'+\beta' = 1.40$ in comparison with Slater-Pauling value (Z_t-24) and theoretical m_{spin}.

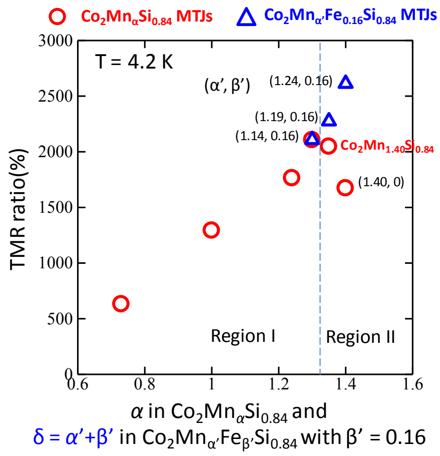

Fig. 2. TMR ratios of CMFS-based MTJs as a function of $\delta=\alpha'+\beta'$ in $Co_2Mn_{\alpha'}Fe_{\beta'}Si_{0.84}$ electrodes and that of identically fabricated CMS-based MTJs as a function of α in $Co_2Mn_\alpha Si_{0.84}$ electrodes.