Effect of off-stoichiometry on the half-metallic character of Heusler alloy \( \text{Co}_2(\text{Mn,Fe})\text{Si} \) thin films

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1. Introduction

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 MTJs and demonstrated giant TMR ratios of 2610\% at 4.2K and 429\% at 290K [1]. The purpose of the present study was to clarify the effect of off-stoichiometry on the half-metallicity of CMFS films. To do this, we experimentally investigated the saturation magnetization/f.u. \( \mu_s \) of CMFS thin films having various Mn and Fe compositions (\( \alpha \) and \( \beta \)) in \( \text{Co}_2\text{Mn}_x\text{Fe}_y\text{Si}_{0.84} \) films along with first-principles calculations.

2. Experimental and Theoretical Methods

We fabricated samples having a layer structure of (from the substrate side) MgO-buffer/\( \text{Co}_2\text{Mn}_x\text{Fe}_y\text{Si}_{0.84}/\text{MgO} \) barrier grown on a MgO (001) substrate. The saturation magnetization \( (M_s) \) was measured at 10 K using a SQUID magnetometer. Determining \( \mu_s \) from the \( M_s \) value requires precise determination of the film thicknesses, which we measured using low angle x-ray reflectivity. First-principles density functional calculations were performed on the basis of the Korringa-Kohn-Rostoker (KKR) Green’s function method with the coherent potential approximation. The antisite-based site specific formula unit (SSFU) composition model proposed for off-stoichiometric ternary Heusler alloy \( \text{Co}_2\text{MnSi} \) [2] was extended for quaternary Heusler alloy CMFS and implemented as a basis of the theoretical calculations.

3. Results and Discussion

Figure 1 shows how the experimental \( \mu_s \) varied with the Fe composition \( \beta \) ranging from 0 to 0.67 for \( \text{Co}_2\text{Mn}_x\text{Fe}_y\text{Si}_{0.84} \) films with a fixed Mn composition \( \alpha \) of 0.73. Also, the half-metallic Slater-Pauling values, \( Z_r \sim 24 \), and the theoretical total spin magnetic moments per f.u., \( m_{\text{spin}} \), are plotted in the same figure. \( Z_r \) is the total number of valence electrons per f.u. provided by the antisite-based SSFU composition model [2]. The drop of \( \mu_s \) from \( Z_r \sim 24 \) for \( \beta = 0 \) has been explained by the existence of \( \text{Co}_{0.6\alpha} \) antisites [2]. \( \mu_s \) increased with an increase in \( \beta \) from 0 to 0.57 growing closer to \( Z_r \sim 24 \) and was very close to \( Z_r \sim 24 \) for \( \beta = 0.57 \). Furthermore, the experimental \( \mu_s \) values and the theoretical \( m_{\text{spin}} \) values were in good agreement, indicating the validity of the antisite-based SSFU model for the quaternary CMFS. Note that as \( \mu_s \) got closer to the half-metallic \( Z_r \sim 24 \) value, the TMR ratio increased, thus indicating the improvement in the half-metallicity of these films. In summary, it was shown that for (\( \text{Mn+Fe} \))-rich films having higher TMR ratios the \( \mu_s \) values are closer to \( Z_r \sim 24 \). These results indicate that the suppression of Co antisites at the nominal Mn/Fe sites is critical for obtaining half-metallic quaternary \( \text{Co}_2(\text{Mn,Fe})\text{Si} \) in a similar way as ternary alloy \( \text{Co}_2\text{MnSi} \). These findings and the giant TMR ratios for CMFS MTJs show the promise of Co-based Heusler alloy thin films as spin source materials by virtue of their half-metallicity and diversity arising from ternary and quaternary material systems.

Reference


Fig. 1. Experimental saturation magnetization per f.u. at 10 K (\( \mu_s \)) and theoretical spin magnetic moment (\( m_{\text{spin}} \)) of CMFS thin films as a function of Fe composition \( \beta \) in \( \text{Co}_2\text{Mn}_x\text{Fe}_y\text{Si}_{0.84} \) films with \( \alpha = 0.73 \).