Effect of off-stoichiometry on the half-metallic character of Heusler alloy $Co_2(Mn,Fe)Si$ 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 $Co_2(Mn,Fe)Si$ (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 (μ_s) of CMFS thin films having various Mn and Fe compositions (α and β) in $Co_2Mn_\alpha Fe_\beta Si_{0.84}$ 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/$Co_2Mn_\alpha Fe_\beta Si$/MgO barrier grown on a MgO (001) substrate. The saturation magnetization (M_s) was measured at 10 K using a SQUID magnetometer. Determining μ_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 Heulser alloy Co_2MnSi [2] was extended for quaternary Heulser alloy CMFS and implemented as a basis of the theoretical calculations.

3. Results and Discussion

Figure 1 shows how the experimental μ_s varied with the Fe composition β ranging from 0 to 0.67 for $Co_2Mn_\alpha Fe_\beta Si_{0.84}$ films with a fixed Mn composition α of 0.73. Also, the half-metallic Slater-Pauling values, Z_t–24, and the theoretical total spin magnetic moments per f.u., m_{spin} are plotted in the same figure. Z_t is the total number of valence electrons per f.u. provided by the antisite-based SSFU composition model [2]. The drop of μ_s from Z_t–24 for β = 0 has been explained by the existence of Co_{Mn} antisites [2]. μ_s increased with an increase in β from 0 to 0.57 growing closer to Z_t–24 and was very close to Z_t–24 for β = 0.57. Furthermore, the experimental μ_s values and the theoretical m_{spin} values were in good agreement, indicating the validity of the antisite-based SSFU model for the quaternary CMFS. Note that as μ_s got closer to the half-metallic Z_t–24 value, the TMR ratio increased, thus indicating the improvement in the half-metallicity of these films. In summary, it was shown that for (Mn+Fe)-rich films having higher TMR ratios the μ_s values are closer to Z_t–24. These results indicate that the suppression of Co antisites at the nominal Mn/Fe sites is critical for obtaining half-metallic quaternary $Co_2(Mn,Fe)Si$ in a similar way as ternary alloy Co_2MnSi. 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.

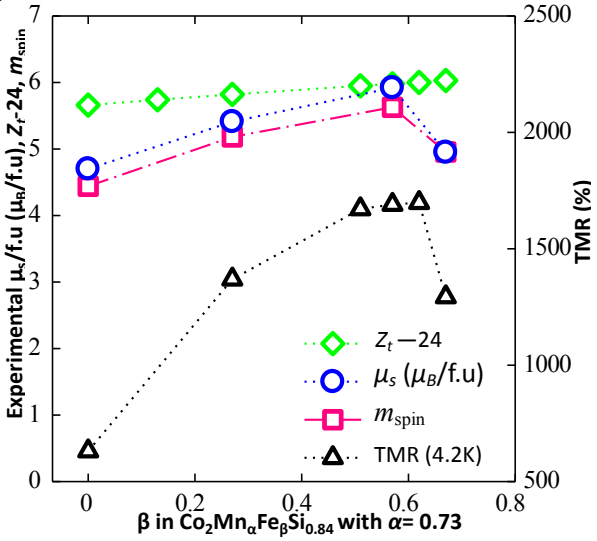

Fig. 1. Experimental saturation magnetization per f.u. at 10 K (μ_s) and theoretical spin magnetic moment (m_{spin}) of CMFS thin films as a function of Fe composition β in $Co_2Mn_\alpha Fe_\beta Si_{0.84}$ films with α = 0.73.

Reference

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