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Influence of Mn composition in Co_2MnSi films on magnetoresistance characteristics of Co_2MnSi-based current-perpendicular-to-plane spin valves

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Abstract

The influence of Mn composition in Co_2MnSi (CMS) films on the giant magnetoresistance characteristics was investigated for CMS-based current-perpendicular-to-plane (CPP) spin valves prepared with various Mn compositions α in $Co_2Mn_\alpha Si_{0.82}$ electrodes. The magnetoresistance ratio of the prepared CMS/$Co_{50}Fe_{50}$ (CoFe) (1.1 nm)/Ag/CoFe (1.1)/CMS spin valves systematically increased with increasing α from 9.8 % for Mn-deficient α = 0.62 to 17.8 % for Mn-rich α = 1.45 at 290 K. Our finding suggests that suppressing the harmful Co_{Mn} antisites by a Mn-rich composition is highly effective for enhancing the half-metallicity of CMS spin sources in CPP spin valves.

1. Introduction

Co-based Heusler alloy thin films have been extensively studied as a promising ferromagnetic electrode material for spintronic devices, including magnetic tunnel junctions (MTJs) [1-5] and current-perpendicular-to-plane (CPP) giant magnetoresistance (GMR) devices [6,7], and for spin injection into semiconductors [8,9]. We have shown that harmful defects in Heusler alloy thin films of Co_2MnSi (CMS), $Co_2(Mn,Fe)Si$ (CMFS), and Co_2MnGe 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 [1-5], and have demonstrated high tunneling magnetoresistance (TMR) ratios of up to 1995% (354%) at 4.2 K (290 K) in CMS/MgO/CMS MTJs having Mn-rich CMS electrodes [2], and up to 2610% (429%) at 4.2 K (290 K) in CMFS/MgO/CMFS MTJs having (Mn+Fe)-rich CMFS electrodes [3,5]. For CPP-GMR devices, Li *et al.* reported an enhancement of the MR ratio in Mn-rich $Co_2Mn(Ge, Ga)$ (CMGG) [7]. They compared the MR characteristics of two CMGG-based CPP-GMR devices, one having a Co-rich composition of $Co_2Mn_{0.97}(Ge_{0.63}Ga_{0.24})$, the other having a Mn-rich composition of $Co_2Mn_{1.24}(Ge_{0.76}Ga_{0.31})$. In their samples, the Mn composition α and the (Ge + Ga) compositions β' with respect to Co in the composition expression of $Co_2Mn_\alpha(Ge,Ga)_{\beta'}$ were changed simultaneously, although the degree of Co_{Mn} antisites depends on both α and β'. Thus, systematic investigations of the Mn-composition dependence of the MR characteristics are necessary to clarify the influence of off-stoichiometry in CMS films on the MR characteristics of CPP spin valves. In this study, we fabricated CPP-GMR devices with exchange-biased spin-valve structure having CMS electrodes with various Mn compositions, α, and an Ag spacer, and systematically investigated the influence of α on the MR characteristics.

2. Experimental Methods

The layer structures of the fabricated CPP spin valves were as follows: (from the substrate side) MgO buffer (10 nm)/$Co_{50}Fe_{50}$ (CoFe) seed (10)/Ag buffer (100)/CoFe buffer (10)/CMS lower electrode (3)/CoFe (1.1)/Ag spacer (5)/CoFe (1.1)/CMS upper electrode (3)/CoFe (1.1)/IrMn (10)/Ru cap (5) with $Co_2Mn_\alpha Si_{0.82}$ electrodes, grown on MgO(001) substrates. Ultrathin 1.1-nm-thick CoFe layers were inserted at both interfaces of the Ag spacer with the lower and upper CMS electrodes, aiming to enhancing the MR ratio. We prepared $Co_2Mn_\alpha Si_\beta$ electrodes with various values of α ranging from 0.62 to 1.45 while fixing the value of β (= 0.82) for both the lower and upper CMS electrodes. The preparation procedure of CMS electrodes with various values of α was the same as for the CMS/MgO MTJs [1-4]. Just after the deposition of CMS upper electrodes, the layer structure was *in-situ* annealed at 550 °C. We fabricated CPP spin valves with the nominal junction sizes ranging from 70 × 120 nm to 400 × 640 nm by using EB lithography and Ar ion milling. The MR characteristics were measured using a dc four-probe method from 4.2 K to 290 K. We defined the MR ratio as $(R_{AP} - R_P)/R_P$, where R_P and R_{AP} are the junction resistances for the parallel and antiparallel magnetization configurations.

3. Results and Discussion

Figure 1(a) shows a typical MR curve at 290 K for a CMS/CoFe/Ag/CoFe/CMS spin valve with Mn-rich (α = 1.45) CMS electrodes. The nominal junction area A was 70 × 120 nm^2. It showed clear MR characteristics with a MR ratio of approximately 18%. The values of $R_P \cdot A$ and $\Delta R \cdot A$ were 52 mΩ·μm^2 and 8.4 mΩ·μm^2, respectively, where $\Delta R = R_{AP} - R_P$. Figure 1(b) shows the MR ratio of the CoFe-inserted CMS spin valves at 290 K as a function of the Mn composition, α. Most importantly, the MR ratio significantly increased from 9.8 % for the Mn-deficient composition (α = 0.62) to 17.8 % for the Mn-rich (α = 1.45). This result suggests a continuous increase in the spin polarization of the CMS electrodes working as spin sources in the CoFe-inserted CMS spin valves with increasing α from a Mn-deficient to a Mn-rich composition. Furthermore, the increase in the spin polarization can be understood in terms of the suppression of Co_{Mn} antisites by Mn-rich compositions.

Figure 2(a) shows typical temperature (T) dependence of R_{AP} and R_P for a CMS/CoFe/Ag/CoFe/CMS spin valve with Mn-rich (α = 1.45) CMS electrodes. R_P decreased almost monotonically with decreasing T from 290 K to 4.2 K. On the other hand, R_{AP} decreased with decreasing T for a T range from 290 K to a certain temperature, T_M, of about 220 K, and then it decreased rapidly for a T range below T_M. Because R_{AP} decreased more slowly with decreasing T than R_P for a T range from 290 K to T_M of 220 K, the MR ratio increased with decreasing T for this T range and took the maximum value at T = 220 K. While the MR ratio of the α = 1.45 spin valve quickly decreased with decreasing T for a T range from T_M of 220 K to about 100 K (Fig. 2(b)) due to the significant decrease in R_{AP}. Such anomalous behaviors were also reported previously in CMS or CMFS-based pseudo spin valves [10-12], and two possible origins, both of which are related with Mn diffusion from CMS into an Ag spacer, were proposed: One is a presence of a bi-quadratic interlayer exchange coupling between upper and lower CMS via diffused Mn [10], and the second one is a formation of spin-glass state of diffused Mn, resulting in the reduction of spin diffusion length of the Ag spacer [11]. Figure 2(b) shows the T dependence of MR ratio for α = 0.62, 1.00, and 1.45. The characteristic temperature T_M (indicated by arrows in Fig. 2(b)) increased with increasing α, resulting in the extension of the T range, in which the anomalous T dependence of the MR ratio was observed, as α increased. This result suggests possible origins related to Mn diffusion into an Ag spacer as discussed in Refs. 10 and 11.

Figure 3 compares the MR ratios of the spin valves as a function of $\alpha + \beta$ with the TMR ratios of $Co_2Mn_\alpha Si_\beta$/MgO/$Co_2Mn_\alpha Si_\beta$ MTJs as a function of $\alpha + \beta$. The MR ratios of the CPP spin valves showed a tendency similar to the TMR ratios of MTJs reported in Refs. 2 and 3. Thus, it is demonstrated that the suppression of Co_{Mn} antisites by using Mn-rich compositions is critical for CPP spin valves as it is in CMS-based and CMFS-based MTJs.

4. Summary

We fabricated CPP spin valves having CMS electrodes and investigated the influence of the Mn composition α on the magnetoresistance characteristics. The MR ratio significantly increased from 9.8 % to 17.8 % at 290 K with increasing α from 0.62 to 1.45. This result suggests a continuous increase in the spin polarization with increasing α from a Mn-deficient to a Mn-rich one. We thus demonstrated that enhancing the half-metallicity of CMS by appropriately controlling the film composition toward a Mn-rich one is highly effective in CMS-based spin valves as it is in MTJs with CMS electrodes.

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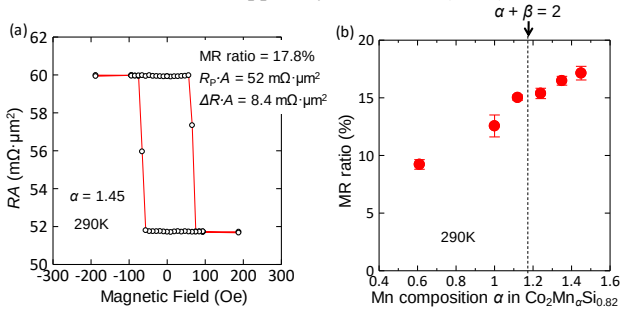

Fig. 1. (a) Typical MR curve at 290 K for a Co_2MnSi (CMS)/$Co_{50}Fe_{50}$ (CoFe) (1.1 nm)/Ag/CoFe (1.1)/CMS spin valve with Mn-rich (α = 1.45) CMS electrodes, and (b) MR ratio of CMS spin valves at 290 K as a function of Mn composition, α. We defined ΔR and the MR ratio as $R_{AP} - R_P$ and $(R_{AP} - R_P)/R_P$, respectively, where R_P and R_{AP} are the junction resistances for the parallel and antiparallel magnetization configurations.

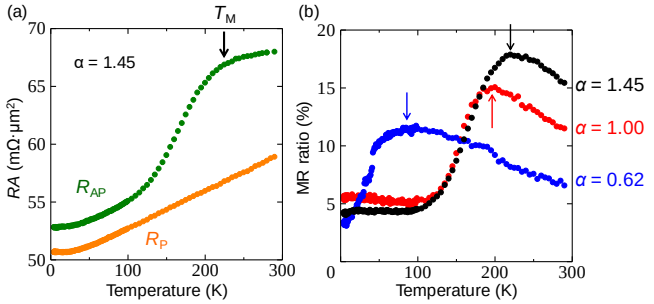

Fig. 2. (a) Temperature dependence of R_{AP} and R_P for a CMS/CoFe/Ag/CoFe/CMS spin valve with Mn-rich (α = 1.45) CMS electrodes, and (b) temperature dependence of MR ratio for α = 0.62, 1.00, and 1.45.

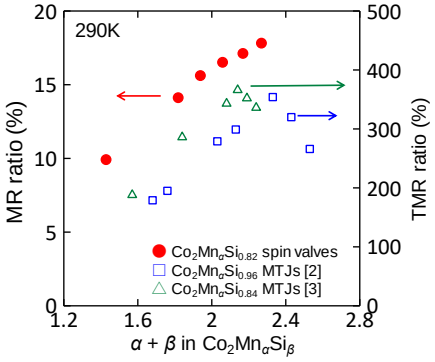

Fig. 3. Comparison of the MR ratios of CMS-based spin valves at 290 K as a function of $\alpha + \beta$ with the TMR ratios of $Co_2Mn_\alpha Si_\beta$/MgO/$Co_2Mn_\alpha Si_\beta$ MTJs at 290 K as a function of $\alpha + \beta$. The MR ratios of the spin valves showed a tendency similar to the MTJs.