Extended Abstracts of the 2015 International Conference on Solid State Devices and Materials, Sapporo, 2015, pp408-409

# Bias voltage dependence of the spin-dependent tunneling conductance of $Co_2$(Mn,Fe)Si-based magnetic tunnel junctions exhibiting giant tunneling magnetoresistances

Kidist Moges, Bing Hu, Hongxi Liu, Tetsuya Uemura, and Masafumi Yamamtoto
Division of Electronics for Informatics, Graduate School of Information Science and Technology,
Hokkaido University, Sapporo 060-0814, Japan
Phone: +81-11-706-6442,  E-mail: kidist@nsed.ist.hokudai.ac.jp

**Abstract**

**We systematically investigated the d*I*/d*V* versus *V* characteristics (= *G* spectra) for the antiparallel magnetization alignment (AP) of magnetic tunnel junctions (MTJs) having $Co_2$(Mn,Fe)Si electrodes that showed giant tunneling magnetoresistance (TMR) ratios. The degree of the half-metallicity of $Co_2$(Mn,Fe)Si electrodes was varied by varying their film compositions. Clear dip structures were found in the *G* spectra for AP in a small bias region of up to ±70 mV. We found that the d*I*/d*V* value for AP at a characteristic voltage, $V_s$ = ±70 mV, normalized by the value at $V = 0$ increased linearly with the TMR ratio at 4.2 K. This behavior was explained by spin-flip tunneling via magnon excited by hot electrons.**

## 1. Introduction

Co-based Heulser alloys have been extensively studied as one of the most suitable spin source materials for spintronics devices because of their half-metallicity that provides complete spin polarization at the Fermi level ($E_F$) and their Curie temperatures being well above room temperature. We recently showed that the suppression of harmful antisites is essential to retain the half-metallicity of Heusler alloy thin films of both ternary alloy $Co_2MnSi$ (CMS) [1-3] and quaternary alloy $Co_2$(Mn,Fe)Si (CMFS) [4]. As a result, we demonstrated giant TMR ratios of 1995% at 4.2 K (354% at 290 K) for CMS/MgO/CMS MTJs (CMS MTJs) with Mn-rich CMS electrodes [2] and 2610% at 4.2 K (429% at 290 K) for CMFS/MgO/CMFS MTJs (CMFS MTJs) with lightly Fe-doped, Mn-rich CMFS electrodes [4]. Furthermore, we showed that the experimental saturation magnetization of $Co_2Mn_{1.24}Fe_{0.16}Si_{0.84}$ electrode was very close to the half-metallic Slater-Pauling value of $Z_t$−24 [5]. Moreover, through tunneling spectroscopy analysis of the differential conductance of CMFS MTJs, we found out that the Fermi energy was located around the middle of the half-metal energy gap for this material [6]. All these results clearly indicated the enhanced half-metallicity of the quaternary Heulser alloy $Co_2$(Mn,Fe)Si having Mn-rich and lightly Fe-doped composition.

To take full advantage of the half-metallic character of CMS and CMFS thin films for spintronic device applications, it is important to clarify the voltage and temperature dependence of spin-dependent tunneling conductance and their origins. Therefore, the purpose of this study was to investigate the bias voltage dependence of spin-dependent tunneling conductance of CMFS MTJs exhibiting giant TMR ratios and clarify the origin of this dependence. To do this, we systematically investigated the *G* spectra of CMFS MTJs, in particular for AP, by varying the degree of half-metallicity through the film composition of CMFS electrodes.

## 2. Experimental methods

The preparation of fully epitaxial CMFS MTJs on CoFe-buffered MgO (001) substrates having various Mn compositions $\alpha$' and $\beta$' in $Co_2Mn_{\alpha'}Fe_{\beta'}Si_{0.84}$ electrodes has been reported in Ref. 4. The layer structure of the MTJs prepared for this study was as follows: (from the substrate side): MgO buffer/CoFe buffer/CMFS lower electrode (3 nm)/MgO barrier (1.4−3.2nm)/CMFS upper electrode (3 nm)/CoFe (1.1 nm)/IrMn/Ru cap. All the layers were epitaxially grown on single-crystal MgO substrates in an ultrahigh vacuum chamber. The quaternary alloy CMFS electrodes were prepared by RF magnetron cosputtering from nearly stoichiometric CMS, Mn and Fe targets. $\alpha$' and $\beta$' in $Co_2Mn_{\alpha'}Fe_{\beta'}Si_{0.84}$ were varied by adjusting the relative amounts of sputtered Mn and Fe. We measured the *G* spectra of the fabricated MTJs with a conventional lock-in method at 4.2 K. The bias voltage (*V*) was defined with respect to the lower CMFS electrode. The tunneling resistances $R_P$ and $R_{AP}$ were measured by a dc four-probe method. The TMR ratio was defined as TMR = $(R_{AP}- R_P)/R_P$.

## 3. Experimental results and discussion

Figure 1 shows typical *G* spectra of CMFS MTJ with $Co_2Mn_{0.73}Fe_{0.62}Si_{0.84}$ electrodes for the parallel (P) and antiparallel (AP) magnetization alignments at 4.2 K for *V* of up to ±0.60V. $G_N$ for P ($G_{N,P}$) and AP ($G_{N,AP}$) represent the *G* values normalized by their respective values at $V = 0$. A marked linear increase in $G_{N,AP}$ with increasing *V* was observed in the small bias voltage region of $|V| \leq 70$ mV at 4.2 K. The increase in $G_{N,AP}$ was weakened for applied bias voltages of above $V$ = ±70 mV. These features are similar to those observed in CMS MTJs [7]. In contrast, the *V* dependence of $G_{N,P}$ was very weak. Figure 2 shows the $G_{N,AP}$ spectra in a small bias voltage region of up to ±0.20 V for CMFS MTJs having various $\beta$' in $Co_2Mn_{0.73}Fe_{\beta'}Si_{0.84}$ electrodes ($\beta$' = 0.0, 0.27, 0.57 and 0.62). Note that the TMR ratio increased with increasing (Mn+Fe) composition through $\beta$' from 570% at 4.2 K for $\beta$' = 0 (Mn-deficient $Co_2Mn_{0.73}Si_{0.84}$) to 1765% for $\beta$' = 0.62 ((Mn+Fe)-rich $Co_2Mn_{0.73}Fe_{0.62}Si_{0.84}$). The following features were observed for the $G_{N,AP}$ spectra. First, the values of the characteristic voltage, $V_s$, up to which $G_{N,AP}$ showed a marked linear increase with $V$ were almost the same for all the MTJs (this value was ≈ 70 mV for all the MTJs). On the other hand, the slope of $G_{N,AP}$ spectra in the voltage region of $|V| \leq 70$ mV was larger for MTJs having higher TMR ratios. Thus, the magnitude of $G_{N,AP}$ at $V = V_s$ was larger for MTJs having higher TMR ratios. Figure 3 shows the values of $G_{N,AP}$ at $V = V_s$ (=70mV) of CMFS MTJs as a function of (TMR(4.2K) + 1). The plot shows the almost linear dependence of $G_{N,AP}(V_s)$ on (TMR(4.2K) + 1). This behavior is consistent with the model proposed by Zhang et al. [8] where the tunneling conductance as a function of bias voltage in a small bias voltage region is explained by a spin-flip tunneling process via a magnon excited by hot electrons. This model predicts that $G_{N,AP}$ increases with $1/\boldsymbol{\xi} = [(\rho_M{}^2+\rho_m{}^2)/(2\rho_M\rho_m)] = (1+P^2)/(1-P^2)$ while $G_{N,P}$ increases with $\boldsymbol{\xi}$, where $\rho_M$ and $\rho_m$ represent the density of states of majority- and minority- spin bands at $E_F$ and $P$ is the spin polarization at $E_F$. Here, $1/\boldsymbol{\xi}$ is equal to $G_P/G_{AP}$ (at T = 0) = (TMR (0 K) + 1), which was approximated by (TMR (4.2 K) + 1) in Fig. 3. Thus, according to this model, the $V$ dependence up to $V_s$, where $eV_s$ corresponds to the maximum magnon energy of $3kT_c/(S+1)$ ($T_c$ is the Curie temperature of the electrode and $S$ is the spin parameter) [8], is significantly stronger for AP compared with that for P and becomes stronger for the higher spin polarization. Thus, the experimentally observed features, in particular, the linear increase in $G_{N,AP}(V_s)$ with (TMR (4.2 K) + 1), can be explained by the model of Zhang et at. Moreover, the prefactor obtained from the $V$ dependence of $G_{N,AP}$ shown in Fig.3 was in good agreement with that obtained from fitting of the temperature dependence of the tunneling conductance for AP.

## 4. Summary

We showed that the voltage dependence of the tunneling conductance for AP of CMFS-based MTJs in a small bias voltage region of up to about 70 mV was stronger for electrodes having higher spin polarization. This feature was consistently explained by a spin-flip tunneling via magnon excited by hot electrons along with strong temperature dependence of tunneling conductance for AP by spin-flip tunneling via a thermally excited magnon.

## References


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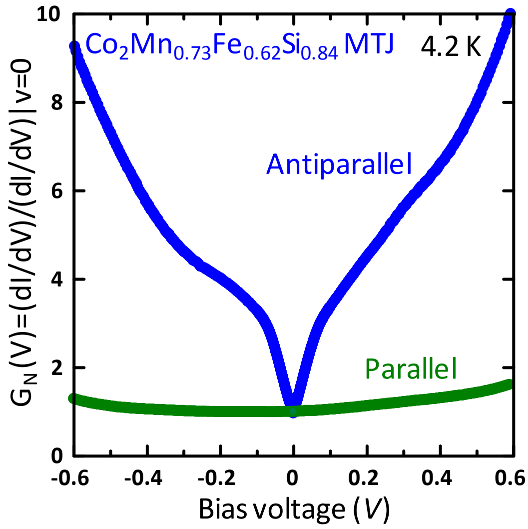


Fig. 1. Normalized d$I$/d$V$ (= $G$) vs. $V$ characteristics for P and AP of a CMFS MTJ with $Co_2Mn_{0.73}Fe_{0.62}Si_{0.84}$ electrodes that exhibited a TMR ratio of 1765% at 4.2K.

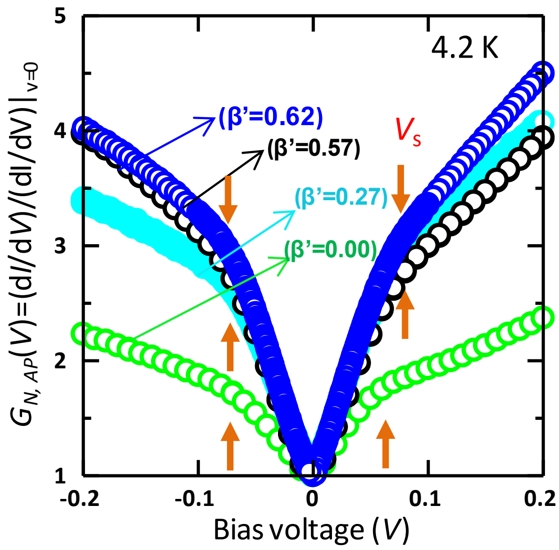


Fig. 2. Normalized d$I$/d$V$ vs. $V$ characteristics for AP in a small bias voltage region of up to ±0.20 V for CMFS MTJs having various $\beta$' in $Co_2Mn_{0.73}Fe_{\beta'}Si_{0.84}$ electrodes ($\beta$' = 0.0, 0.27, 0.57 and 0.62).

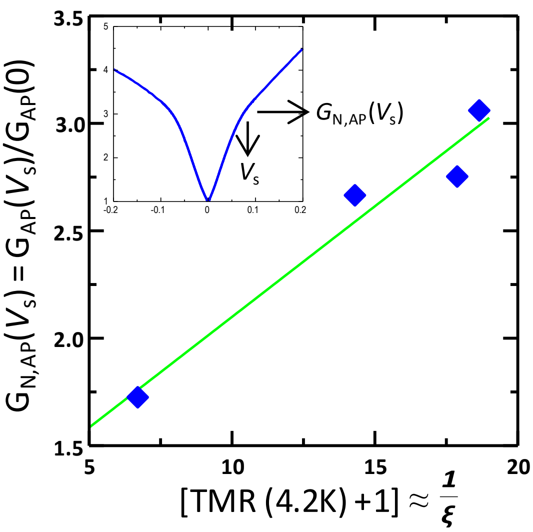


Fig. 3. Normalized $G$ (= d$I$/d$V$) values at $V = V_s$ (= 70 mV) for CMFS MTJs as a function of (TMR(4.2K) + 1).