





Temperature dependence of spin-dependent tunneling conductance of magnetic tunnel junctions with half-metallic Co<sub>2</sub>MnSi electrodes

## $Co<sub>2</sub>MnSi$

Spintronic devices, which utilize the spin degree of freedom in addition to the charge of the electron, have attracted much interest as future-generation electron devices. This is because of their potential advantages of nonvolatility, decreased power consumption, and reconfigurable logic function capabilities. Half-metallic ferromagnets (HMFs) are one of the most suitable spin source materials because of their complete spin polarization  $(P)$  at the Fermi level  $(E_F)$  arising from an energy gap for one spin direction. Co-based Heusler alloys (Co2*YZ*, where *Y* is usually a transition metal and *Z* is a main group element) are among the most extensively applied to spintronic devices. This is because of the HMF nature theoretically predicted for many of these alloys and because of their high Curie temperatures, which are well above room temperature.

Co2MnSi (CMS) is one of the most extensively investigated ferromagnetic electrode materials among the Co2*YZ* family. This is because of its theoretically predicted half-metallic nature with a large halfmetal gap of 0.81 eV for its minority-spin band and its high Curie temperature of 985 K. Liu *et al*. from our research group demonstrated giant tunneling magnetoresistance (TMR) ratios of up to 1995% at 4.2 K and up to 354% at 290 K for CMS/MgO/CMS (CMS MTJs) having Mn-rich CMS electrodes and up to 2610% at 4.2 K and 429% at 290 K for  $Co_2(Mn,Fe)Si$  (CMFS)/MgO/CMFS MTJs (CMFS) MTJs) with Mn-rich, lightly Fe-doped CMFS electrodes.

To take full advantage of the half-metallic nature of Co-based Heusler alloys as spin source materials at room temperature, it is important to clarify the origin of the temperature (*T*) dependence of spindependent tunneling conductance (*G*) of MTJs. The *T* dependence of *G* for the parallel and antiparallel magnetization alignments,  $G_P$  and  $G_{AP}$ , of MTJs has been discussed on the basis of models proposed by Zhang *et al.* and by Shang *et al..* Zhang *et al.* accounted for the *T* dependence of  $G_P$  and  $G_{AP}$ by spin-flip inelastic tunneling via a thermally excited magnon while assuming a *T*-independent *P*, and predicted that both  $G_P$  and  $G_{AP}$  increase with increasing *T*. On the other hand, Shang *et al.* took into consideration only spin-conserving elastic tunneling with a decaying *P* with increasing *T*. The Shang model predicts that  $G_{AP}$  increases with *T*, of which the effect on  $G_{AP}(T)$  is apparently in agreement with that of the Zhang model. However, note that the tunneling mechanisms that determine the *T* dependence are intrinsically different between these two models. On the other hand, the Shang model predicts that  $G_P$  decreases, which is opposed to the prediction of the Zhang model. Thus, a more systematic experimental study is essential to get a deeper understanding of the origin of the *T* dependence of spin-dependent  $G_P$  and  $G_{AP}$ .

A promising approach would be to investigate half-metal-based MTJs with systematically varied *P* values at 0 K, as MTJs with highly spin-polarized electrodes arising from half-metallicity would show most typical *T* dependence of  $G_P$  and  $G_{AP}$ . In addition, it is a particularly desirable approach to extract how the *T* dependence of  $G_P$  and  $G_{AP}$  changes with *P* at 0 K by investigating a series of high-quality MTJs with systematically varied *P* at 0 K. Furthermore, development of a more comprehensive model for the *T* dependence of  $G_P$  and  $G_{AP}$  is requisite as the predictions of the Zhang and Shang models for  $G_P(T)$  are opposite. This difference in the predictions also suggests that an analysis of the experimental  $G_P(T)$  would be critical to revealing the origin of the *T* dependence of spin-dependent *G*.

The purpose of this research is to elucidate the origin of the  $T$  dependence of  $G_P$  and  $G_{AP}$  of MTJs

through analyzing the experimental  $G_P(T)$  and  $G_{AP}(T)$  of high-quality MTJs showing giant TMR ratios and to provide a unified picture of how the  $T$  dependence of  $G<sub>P</sub>$  and  $G<sub>AP</sub>$  changes with  $P$ . To do so, we experimentally investigated the  $T$  dependences of  $G<sub>P</sub>$  and  $G<sub>AP</sub>$  of CMS MTJs having systematically varied spin polarizations at 4.2 K by varying the Mn composition  $\alpha$  in Co<sub>2</sub>Mn<sub>α</sub>Si electrodes. Notable features in  $G_P(T)$  were found; i.e., it decreased with increasing *T* from  $T_1$  of about 30 K to  $T_2$  ranging from about 162 K to 237 K depending on  $\alpha$ ; then it increased for *T* over  $T_2$ . Furthermore, as *P* at 4.2 K increased,  $T_2$  increased and the maximum decrease in the normalized  $G_P$  [=  $G_P(T)/G_P(4.2 \text{ K})$ ] at  $T_2$  increased. To clarify the origin of the characteristic *T* dependence of  $G_P$ , we developed a model that took into account both spin-conserving elastic tunneling in which *P* decays with increasing *T* and spin-flip inelastic tunneling via a magnon by extending the original Zhang model.

This dissertation consists of five chapters and the main content of each chapter is as follows:

Chapter 1 introduces the background, purpose and approach of the present study.

Chapter 2 describes our experimental methods, including the preparation of MTJs, the estimation of the barrier height, and the measurement of the *T* dependence of spin-dependent tunneling conductance. Chapter 3 presents the results and discussion for the *T* dependence of spin-dependent tunneling conductance of CMS MTJs. At first, it describes the overall features of  $G_P(T)$  and  $G_{AP}(T)$  of CMS MTJs with various  $\alpha$  values that showed a high TMR ratio. Then, it describes the notable, nonmonotonic  $T$ dependence of  $G_P(T)$  of CMS MTJs that exhibited a giant TMR ratio. It also describes how the  $G_P(T)$ changed with *P* at 4.2 K. To explain these features of  $G<sub>P</sub>(T)$ , we devised an extension of the original Zhang model, in which only spin-flip inelastic tunneling was considered as the origin of the *T* dependence of  $G_P$  and  $G_{AP}$ , by introducing a *T*-dependent *P*. By analyzing these features of the  $G_P(T)$  with the extended Zhang model, it was revealed that both spin-flip inelastic tunneling via a magnon and spin-conserving elastic tunneling wherein *P* decreases with increasing *T* play key roles. Finally, the experimental  $G_{AP}(T)$ , including its stronger *T* dependence for higher *P* at 4.2 K, was also consistently explained with the extended Zhang model.

Chapter 4 describes the results of the *T* dependence of  $G_P$  and  $G_{AP}$  of CMFS MTJs with  $Co_2Mn_{\alpha'}Fe_{\beta'}Si_{0.84}$  electrodes having a fixed Mn composition of  $\alpha' = 0.73$  and various Fe compositions of  $\beta'$  ranging from 0 to 0.62. We found that the magnon lower cut off energy,  $E_c^{\ P}$  and  $E_c^{\ AP}$ , of (Mn + Fe)-rich CMFS MTJs with the Fe compositions,  $\beta'$ , being comparable to  $\alpha'$ , i.e., CMFS with ( $\beta' = 0.57$ ,  $\alpha' + \beta' = 1.30$ ) and ( $\beta' = 0.62$ ,  $\alpha' + \beta' = 1.35$ ), were larger than those of Mn-rich CMS MTJs with  $Co_2Mn_{\alpha}Si_{0.84}$  electrodes with  $\alpha = 1.30$ . Thus, it was revealed that the magnon lower cut off energy, which influences the *T* dependence of the TMR ratio, depends on the constituent transition-metal element *Y* in half-metallic  $Co<sub>2</sub>YSi$  Heusler alloys.

Chapter 5 summarizes and concludes this dissertation.

In summary, it was clarified that both spin-flip inelastic tunneling via a thermally excited magnon and spin-conserving elastic tunneling in which *P* decays with increasing *T* play key roles as the origins of the *T* dependence of the spin-dependent tunneling conductance of MTJs. Our findings provide a unified picture for understanding the origin of the *T* dependence of *G* of MTJs with a wide range of *P*, including MTJs with high *P* close to a half-metallic value.