# Origin of bi-quadratic interlayer exchange coupling in Co<sub>2</sub>MnSi-based pseudo spin valves

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### Abstract

We fabricated current-perpendicular-to-plane pseudo spin valves (CPP-PSVs) having Co<sub>2</sub>MnSi electrodes with various Mn compositions,  $\alpha$ , and an Ag spacer, and investigated the strength of bi-quadratic interlayer exchange coupling (90° coupling) in the CPP-PSVs by comparing magnetoresistance characteristics and spin-transfertorque induced magnetization switching characteristics. We experimentally found that (a) the strength of 90° coupling increased as  $\alpha$  increased, (b) it was weakened by insertion of CoFe between CMS and the Ag spacer, resulting in the suppression of the 90° coupling at 290 K, and (c) it strongly depended on temperature. These results can be understood by the interpretation that loosely coupled spins due to Mn atoms diffused into an Ag spacer are the origin of 90° coupling.

## 1. Introduction

Current-perpendicular-to-plane (CPP) giant magnetoresistance (GMR) devices using Co-based Heusler alloys have attracted much interest as read sensors for next-generation hard disk drives [1–5]. To take full advantage of the half-metallic character of Co-based Heusler alloys, it is important to suppress structural defects such as antisite and/or disorder of constituent atoms [6,7]. We have recently demonstrated that the MR ratio at 290 K increased as the Mn composition of Co<sub>2</sub>MnSi (CMS) in CMS-based CPP spin valves increased and have shown that Mn-rich CMS is highly effective in suppressing Co<sub>Mn</sub> antisite, which is detrimental to the half-metallicity [5]. However, the MR ratio decreased as the temperature (T) decreased in a certain low T region below a characteristic temperature  $T_{\rm M}$ . Moreover, the abnormal T regions extended as Mn composition increased, i.e.,  $T_{\rm M}$  increased with an increase in Mn composition, suggesting that the Mn concentration plays a significant role in the abnormal T dependence.

A similar decrease in the MR ratio at low *T* was previously reported for other CPP-GMR devices [1–3], and its origin is still an open question. Recently, Goripati et al. reported that the decrease in the MR ratio at low *T* was alleviated by spin transfer torque (STT) switching and suggested that the origin of the reduction in the MR ratio is the presence of a bi-quadratic interlayer exchange coupling (90° coupling) between the upper and lower ferromagnetic layers [3]. Taking the strong *T*-dependence of the coupling strength into consideration, they suggested that a possible origin of 90° coupling is related to the loose spin model. In the loose spin model proposed by Slonczewski [8], spins loosely coupled to each other are assumed to exist in a spacer or at ferromagnet (F)/nonmagnet (N) interfaces, predicting a strong *T*-dependence of the coupling strength because of the relatively strong *T*-dependence of the paramagnetic character of the loose spins. However, the origin of the loose spins in the CMS-based GMR is unclear.

The purpose in the present study was to clarify the origin of 90° coupling observed in CMS-based CPP GMR devices in terms of the loose spin model. To do this, we systematically investigated the influences of the Mn composition of CMS, the insertion of CoFe, and the operating temperature on the MR characteristics and STT characteristics for CMS-based CPP pseudo spin valves (PSVs).

## 2. Experimental Methods

We prepared two series of CMS-based CPP-PSVs without a CoFe insertion layer at the interfaces (series A) and with an ultrathin CoFe insertion layer at the interfaces (series B) both with various Mn compositions,  $\alpha$ , in Co<sub>2</sub>Mn<sub> $\alpha$ </sub>Si<sub>0.82</sub> electrodes ranging from  $\alpha = 0.62$  to 1.40. The layer structures of series-APSVs were as follows: (from the substrate side) MgO buffer (10 nm)/Co50Fe50 (CoFe) seed (10)/Ag buffer (100)/CoFe buffer (10)/CMS lower electrode (10)/Ag spacer (5)/CMS upper electrode (3)/Ru cap (5), grown on MgO (001) substrates. For series-B PSVs, ultrathin 1.8-nm-thick CoFe layers were inserted at both interfaces of the Ag spacer with the lower and upper CMS electrodes, with the aim of forming an identical interface among series-B PSVs. Just after deposition of the upper electrode, the layer structure was in-situ annealed at 550°C. We fabricated CPP-PSVs with junction sizes ranging from  $26 \times 50$  nm to  $164 \times 240$  nm by electron beam lithography and Ar ion milling. The MR curve and STT curve were measured using a dc four-probe method by sweeping the magnetic field and bias current, respectively. The bias voltage was defined with respect to the lower CMS electrode (fixed layer), and positive (negative) current stabilizes parallel (antiparallel) configuration of magnetization in measuring STT characteristics. Both measurements were done at 290 K and 4.2 K.

## 3. Results and Discussion

Figures 1(a) and 1(b) show the comparison between an MR curve and an STT curve at 290 K for series-A PSVs (w/o CoFe insertion) with Mn compositions of (a)  $\alpha = 0.62$  and (b)

 $\alpha = 1.40$ . In each figure, the red and black curve indicate the MR curve and STT curve, respectively, and the bottom and top axis indicate the magnetic field applied in measuring the MR characteristics and the current density supplied through the junction in measuring the STT characteristics, respectively. We observed clear resistance change in both the MR curve and the STT curve for both samples. We found that intermediate states appeared between the parallel (P) state and antiparallel (AP) state in the MR curve for a Mn-rich sample of  $\alpha = 1.40$  (see Fig. 1(b)), which indicates the presence of 90° coupling. Moreover, the AP resistance obtained by the STT  $(R_{AP}^{(STT)})$  is larger than that obtained by the magnetic field sweep  $(R_{AP}^{(Mag)})$ . This is also evidence of the presence of 90° coupling because this result can be understood by the interpretation that the AP state was not perfectly formed by sweeping the magnetic field because of the presence of the 90° coupling and that the STT brought the magnetization configuration more closely to the perfect AP state.

In order to compare the strength of the  $90^{\circ}$  coupling, we introduced enhancement factor, r, defined an by  $R_{\rm AP}^{\rm (STT)}/R_{\rm AP}^{\rm (Mag)}$ , as a measure of the coupling strength. Figure 2(a) shows the enhancement factor r at 290 K as a function of Mn composition for series-A PSVs and series-B PSVs. The value of r increased as  $\alpha$  increased for series-APSVs, indicating that the strength of 90° coupling increased with an increase in Mn composition in CMS electrodes. On the other hand, the value of r was almost constant for series-B PSVs at 290 K, indicating that the insertion of CoFe suppressed the 90° coupling at 290 K. Figure 2(b) shows the enhancement factor as a function of Mn composition for series B PSVs (w/ CoFe) at 290 K and 4.2 K. The values of r at 4.2 K increased as T decreased, reaching up to 1.3 at 4.2 K. This means that the 90° coupling appeared at 4.2 K for series-B PSVs even though it was suppressed at 290 K. Such a strong T dependence of the coupling strength suggests the relation of the loose spin model. Interestingly, the value of r at 4.2 K increased as the Mn composition  $\alpha$  increased.

Now we will discuss the origin of loose spins. In the loose spin model, two possible origins were assumed for the loose spins: paramagnetic impurities in the spacer or loosely-coupled spins located at the F/N interface [8]. For series-B PSVs, the interfacial structure between the ferromagnet and the Ag spacer was made identical by inserting an ultrathin CoFe layer at both interfaces. Thus, the loose interfacial spins cannot explain the Mn-composition-dependent  $R_{AP}^{(STT)}/R_{AP}^{(Mag)}$ observed at 4.2 K in series-B PSVs, and it is concluded that the origin of loose spins is Mn atoms diffused into the Ag spacer. Moreover, this model can explain our experimental findings that (a) the strength of 90° coupling increases as  $\alpha$ increases, (b) it is suppressed at 290 K by CoFe insertion, and (c) it depends strongly on T, as follows: The findings (a) and (b) are explained by the interpretation that the diffusion of Mn atoms into an Ag spacer increases as  $\alpha$  increases, and that it is weakened by CoFe insertion, respectively. The finding (c) is ascribed to the strong T dependence of the paramagnetic character of the Mn atoms diffused into an Ag spacer.

#### 4. Summary

We experimentally found that the strength of 90° coupling in CMS-based CPP PSVs increases as the Mn composition of CMS increases, it is suppressed at 290 K by CoFe insertion between CMS and the Ag spacer, and it depends strongly on *T*. These experimental results can be understood in the framework of the loose spin model. It is concluded that the loose spins due to Mn atoms diffused into the Ag spacer are the origin of 90° coupling. Furthermore, it was demonstrated that the insertion of an ultrathin CoFe layer at interfaces resulted in the suppression of the 90° coupling at 290 K through a possible reduction of Mn diffusion.

## Acknowledgements

This work was partly supported by the Japan Society for the Promotion of Science (KAKENHI: Grant Nos. 16K04872 and 17H03225).

#### References

- [1] T. Furubayashi et al., J. Appl. Phys. 107, 113917 (2010).
- [2] Y. Sakuraba et al., J. Phys. D: Appl. Phys. 44, 064009 (2011).
- [3] H. S. Goripati et al., J. Appl. Phys. 110, 123914 (2011).
- [4] J. W. Jung et al., Appl. Phys. Lett. 108, 102408 (2016).
- [5] M. Inoue et al., Appl. Phys. Lett. **111**, 082403 (2017).
- [6] S. Picozzi et al., Phys. Rev. B 69, 094423 (2004).
- [7] M. Yamamoto et al., JPCM 22, 164212 (2010).
- [8] J.C. Slonczewski, J. Appl. Phys. 73, 5957 (1993).



Fig. 1. Comparison between an MR curve and an STT curve at 290 K for series-A PSVs (w/o CoFe insertion) with Mn compositions of (a)  $\alpha = 0.62$  and (b)  $\alpha = 1.40$ . The red and black curve indicate the MR curve and STT curve, respectively.



Fig. 2. Enhancement factor *r* as a function of Mn composition  $\alpha$  in Co<sub>2</sub>Mn<sub> $\alpha$ </sub>Si<sub>0.82</sub> Fig. 2. Enhancement factor *r* as a function of Mn composition (a) for series A PSVs (w/o CoFe) and series B PSVs (w/ CoFe) at 290 K and (b) for series B PSVs (w/ CoFe) at 290 K and 4.2 K. The enhancement factor, which is a measure of the strength of 90° coupling, is defined as  $r = R_{AP}^{(STT)}/R_{AP}^{(Mag)}$ , where  $R_{AP}^{(STT)}$  and  $R_{AP}^{(Mag)}$  are antiparallel resistance obtained by the STT and by the magnetic field sweep, respectively.