Large magnetoresistance and high spin-transfer torque obtained in CPP-GMR devices with Heusler alloy electrodes through high-throughput compositional optimization

Vineet Barwal, Hirofumi Suto, Ryo Toyama, Kodchakorn Simalaotao, Taisuke Sasaki, Yoshio Miura, and Yuya Sakuraba

Research Center for Magnetic and Spintronic Materials, National Institute for Materials Science (NIMS), Tsukuba, 305-0047, Japan

Half-metallic Heusler alloys having high spin polarization are crucial for future high-density hard disk drives and energy-efficient memory technologies, as they enhance spin-transfer torque (STT) and magnetoresistance (MR) in spintronic devices. While compositional tuning improves performance, conventional methods for optimizing composition are time-consuming. To address this, we developed a high-throughput compositional optimization method using combinatorial sputtering, microfabrication, and automated measurements, enabling efficient exploration of MR and STT responses in current-perpendicular-to-plane giant magnetoresistance devices. Focusing on $Co_2Mn_xFe_{1-x}Ge$ Heusler alloys, we observed a composition-dependent MR with the maximum of MR ratio ~ 38% in the Mn-rich region of $x = 0.85$ at 250°C annealing. The MR ratio further increased to ~ 45% at 350°C annealing along with high STT efficiency (~ 0.6) over a broad composition range ($0.2 \leq x \leq 0.7$). These results represent record-high MR values for all-metal CPP-GMR devices, achieved at relatively low annealing temperatures (250°C-350°C).

***Index Terms*—Spintronics, Heusler alloys, Giant magnetoresistance, Spin transfer torque**

I. Introduction

Giant magnetoresistance (GMR) and spin-transfer torque (STT) effects are technologically significant and serve as the operating principle in magnetoresistive random access memory [1], hard-disk-drive (HDD) read heads [2], spin-torque oscillators [3], and other spintronic devices. Enhancing device performance relies on materials with high spin polarization(P). Among these, half-metallic Heusler alloys are the promising candidates owing to their high P and high Curie temperature.

Compositional tuning of Heusler thin films used as ferromagnetic (FM) electrodes in current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) devices is carried out to enhance the device performance[4]–[6]. Traditionally, this involves depositing and fabricating devices for each selected composition, a process that is both time- and resource-consuming. The combinatorial sputtering can break through the limitation by enabling efficient and systematic investigation of a wide range of material compositions on a single library sample [7], [8]. We recently developed a high throughput composition optimization method to achieve large magnetoresistance (MR) and high STT in CPP-GMR devices incorporating composition-gradient Co_xFe_{1-x} ($0 \leq x \leq 1$) as FM electrodes [4].

Here, we report the large MR ratio and high STT efficiency in CPP-GMR devices containing epitaxial $Co_2Mn_xFe_{1-x}Ge$ ($0 \leq x \leq 1$) Heusler alloy thin films achieved through the high throughput and detailed composition optimization method. $Co_2Mn_xFe_{1-x}Ge$ (CMFG), a mixture of the intermetallic compounds, Co_2MnGe (CMG) and Co_2FeGe (CFG), is expected to exhibit strong chemical ordering behavior at low-temperature annealing[9]. Such alloys exhibit a strong driving force for chemical ordering from a disordered state in thin-film form. High atomic ordering in Heusler alloys is critical for achieving high P, thereby enhancing device performance. For several potential applications such as the read head of HDDs, high atomic order in Heusler film is required to be obtained at lower process temperature below 350°C [2]. CMFG is particularly well suited for such applications.

II. Experimental Details

Epitaxial thin film stacks incorporating CMFG composition-gradient layers were deposited on 2×2 cm^2 MgO (001) substrate using combinatorial sputtering system. Figure 1 (a) shows the combinatorial sputtering process. Two types of CPP-GMR sample stacks, shown in figure 1(b), were prepared. The Type-I sample consisted of a CPP-GMR stack with both magnetic layers being $Co_2Mn_xFe_{1-x}Ge$, used to evaluate the MR output of the devices. The Type-II sample was comprised of CPP-GMR stack with magnetic layers being $Co_2Mn_xFe_{1-x}Ge$ and $Ni_{80}Fe_{20}$, used for STT measurement. The red arrows in figure 1(b) indicate the layer after which in-situ post annealing was done. The top 8 nm Ru cap layer was deposited after cooling down the sample to room temperature. CPP-GMR devices were fabricated for the Type-I and Type-II samples' stacks for the MR and STT measurements.

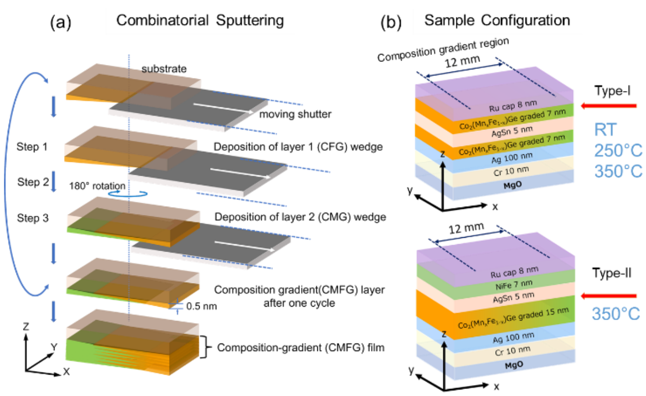

Fig. 1. Schematic showing combinatorial sputtering process. Sample configuration for the Type-I and Type-II sample structures with arrows indicating temperature and layer after which in-situ annealing was done.

III. Results and Discussion

A. X-ray Diffraction

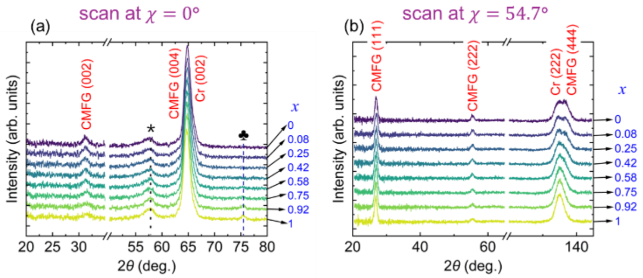

Fig. 2. θ-2θ XRD profiles for the 350ºC annealed Type-I sample along (a) (002) and (b) (111) plane. The * symbol marks the peak position for the Ru 102 plane and ♣ marks the peak position for the hcp $Co_{1.75}Ge$ 004, or fcc Co 220, or hcp Co 110 planes.

The XRD measurements were conducted at several positions with different compositions on the Type-I samples. The atomic ordering was found to increase with annealing temperature. Figure 2(a) and 2(b) shows the XRD scans at χ = 0° and χ = 54.7°, respectively, for the 350ºC annealed Type-I sample. The presence of a 111 superlattice diffraction peaks in the 350°C annealed sample indicates the presence of $L2_1$ ordered phase (atomic order between (Fe, Mn) and Ge sites.

B. Magnetoresistance and spin tranfer torque

Figure 3(a) shows the change in intrinsic MR ratio with Mn content for the CPP-GMR devices from the Type-I sample. In the as-deposited sample, the MR ratio exhibited gradual change with Mn content. The CFG side showed lower MR ratio ~ 2.5% and the CMG side showed slightly higher MR ratio ~ 5%. The CPP-GMR devices fabricated from the stacks annealed at 250°C exhibit clear composition dependence of MR with a maximum MR ratio ~ 38 % in the Mn-rich region of x = 0.85. The MR ratio was further enhanced with increasing annealing temperature to 350°C and we observed maximum MR ratio close to ~ 45% over a broad composition range $0.2 \leq x \leq 0.7$. An overall improvement in MR ratio with increasing annealing temperature can be attributed to the enhanced atomic order with increasing annealing temperature.

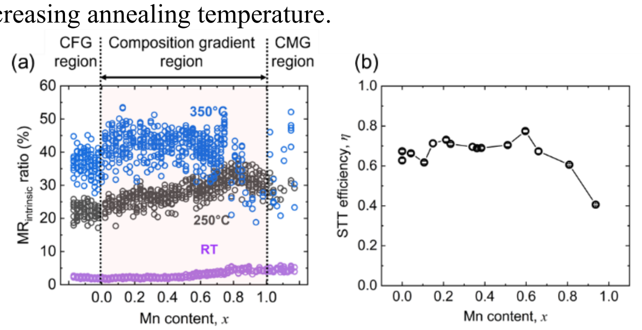

Fig. 3. Change in (a) intrinsic magnetoresistance (MR) ratio and (b) STT efficiency with Mn content for the Type-I and Type-II CPP-GMR sample stack , respectively.

We performed the STT induced magnetization reversal measurements to evaluate the composition dependent STT efficiency for the 350°C annealedType-II sample using devices with circular pillar geometry having designed diameter of 80 nm. Figure 3(b) shows the change in STT efficiency parameter (η) of CPP-GMR devices with Mn content for the Type-II sample. The η exhibits high value ~ 0.6 across the composition range $0 \leq x \leq 0.8$, decline slightly with value reaching close to 0.3 towards the Mn rich (x = 0.9) composition. The high value of η indicates high spin polarization in epitaxially grown CMFG Heusler thin films over a broad composition range.

IV. Conclusion

We fabricated high performance CPP-GMR devices from the GMR stacks containing composition-gradient $Co_2Mn_xFe_{1-x}Ge$ ($0 \leq x \leq 1$) film by high throughput and detailed composition optimization method. The combinatorially sputtered composition-gradient film and local measurements enabled efficient compositional optimization at fine composition interval. The CPP-GMR devices fabricated for the 250°C annelaed sample exhibit clear composition dependence of MR with a maximum of MR ratio ~ 38 % in the Mn-rich region of x = 0.85. The MR ratio was further enhanced with increasing annealing temperature to 350°C and we observed maximum MR ratio close to ~ 45% (ΔRA ~ 8 mΩ μm^2) along with high STT efficiency ~ 0.6 over a broad composition range $0.2 \leq x \leq 0.7$. We achieved record high MR for the all-metal CPP-GMR devices at low annealing temperature of 250°C by the present composition optimization method. The results provide comprehensive guidance on the composition optimization to obtain large MR ratio and high STT efficiency in the CPP-GMR devices using $Co_2Mn_xFe_{1-x}Ge$ at lower process temperature.

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