Development of Heusler-alloy-based Current-Perpendicular-to-Plane Giant Magnetoresistive Devices

著者	CHEN Jiamin
内容記述	この博士論文は内容の要約のみの公開（または一部非公開）になっています
year	2017
その他のタイトル	ホイスラー合金を使った面直電流磁気抵抗素子の開発
学位授与大学	筑波大学 (University of Tsukuba)
学位授与年度	2017
報告番号	12102甲第8411号
URL	http://hdl.handle.net/2241/00152348

Graduate School of Pure and Applied Sciences

Development of Heusler-alloy-based Current-Perpendicular-to-Plane Giant Magnetoresistive Devices

(ホイスラー合金を使った面直電流磁気抵抗素子の開発)

Jiamin CHEN
Doctoral Program in Materials Science and Engineering
Student ID Number: 201436004
Doctor of Philosophy in Engineering
Advised by Prof. Kazuhiro Hono

Summary

Spintronics is an exciting and rapidly expanding new field of microelectronics and nanoelectronics, which is based on exploiting the fact that electrons have spin as well as charge. The development of spintronics in the past several decades has led to the birth of various new applications. One of the most distinct applications of spintronics is magnetoresistive (MR) devices such as giant magnetoresistive (GMR) and tunneling magnetoresistive (TMR) devices.[1–3] The attainment of large current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) output is of paramount importance towards the realization of ultrahigh density magnetic recording, specifically, hard disk drive (HDD) read sensors. Large bulk and interfacial spin-dependent scattering are key factors according to the Valet and Fert theory.[4] Up to now, highly spin polarized Heusler alloys such as Co_2MnSi (CMS)[5], $Co_2Mn(Ga_{0.25}Ge_{0.75})$ (CMGG)[6], $Co_2(Fe_{0.4}Mn_{0.6})Si$ (CFMS)[7] and $Co_2Fe(Ga_{0.5}Ge_{0.5})$ (CFGG)[8] have been utilized to provide large bulk spin polarization, and substantially large magnetoresistance (MR) ratios have been realized. However, a room-temperature (RT) ΔRA of at least 15 mΩ μm^2 is required for using CPP-GMR as read sensors for areal density over 2 $Tbit/in^2$ while the current density is approaching to 1 $Tbit/in^2$;[9] therefore, further improvement of the MR output is essential. For instance, finding a new Heusler alloy which has both high spin polarization and high driving force for $L2_1$-ordering even at low annealing temperature is strongly desired to attain compatibility with the current fabrication process. On the other hand, from a practical point of view, the study on industrially viable high performance epitaxial CPP-GMR devices on a Si single crystalline substrate is also necessary. Proper choice of crystallographic orientation in ferromagnetic (FM) and non-magnetic (NM) materials may affect the thermal stability of the CPP-GMR stack against interlayer atomic diffusion and multilayer delamination. Multilayer engineering of the CPP-GMR stack is in strong need to satisfy rapid development of next generation read head sensors. Last but not least, new concept of processing is strongly required to solve the problem of annealing temperature limit, so that more freedom of material selection can be obtained to achieve higher CPP-(G)MR output.

In short, the motivation of the thesis is as below:

(1) To establish a fabrication process for (110)-oriented epitaxial CPP-GMR devices and investigate its crystallographic orientation dependence of MR output.

(2) To search for new highly spin-polarized Heusler alloy with high driving force for $L2_1$-order even at low annealing temperature for CPP-GMR device application.

(3) To grow high quality epitaxial CPP-GMR device on Si(001) wafer by selecting appropriate buffer materials.

(4) To fabricate single crystalline CPP-GMR device on polycrystalline electrode using wafer bonding technology.

The thesis consists of seven chapters. Chapter 1 introduces the general background on spintronics, giant magnetoresistance (GMR) effect, Co-based Heusler alloy and the issues that motivate the work in the thesis. Chapter 2 provides experimental procedures including sample preparation, device fabrication and analysis methods. Chapter 3 describes the work on CPP-GMR epitaxial spin-valves with (110)-oriented Heusler alloy $Co_2Fe(Ga_{0.5}Ge_{0.5})$ (short as CFGG) and different spacers (Ag, Cu and NiAl), in which orientation dependence of MR output has been investigated. Chapter 4 describes the development of a new series of Heusler alloys prepared by the co-sputtering technique, $Co_2(Fe_{1-x}Ti_x)Si$, which have a strong merit of high spin-polarization together with high driving force for $L2_1$-ordering even at low annealing temperature. Chapter 5 shows the work of realizing high quality epitaxial CPP-GMR pseudo spin-valves on Si(001) single crystalline wafer using NiAl buffer layer. It is the first demonstration of epitaxial spintronic devices with NiAl template for various practical applications in the field of ultra-high density hard disk drive (HDDs), magnetic sensor and so on. Chapter 6 describes the work of fabricating single crystalline magnetoresistive sensors on polycrystalline electrode using a three-dimensional integration technology, which enables the integration of Heusler-alloy-based epitaxial CPP-GMR devices to high sensitive magnetic sensors regardless of the limitation of high temperature processing. Chapter 7 provides summary of the thesis. The main results are summarized as below:

1. **Crystal orientation dependence of CPP-GMR pseudo spin-valves (PSVs) using $Co_2Fe(Ga_{0.5}Ge_{0.5})$ Heusler alloy and different spacers (Ag, Cu and NiAl)**

a) **Ag and Cu spacer**

Although there are many investigations on CPP-GMR, epitaxial CPP-GMR devices with crystal orientations other than (001) have not yet been examined. We consider the spin-dependent transport may have crystallographic orientation dependence; hence, we prepared CPP-GMR pseudo-spin-valves (PSVs) consisting of epitaxial layers of the Heusler alloy $Co_2Fe(Ga_{0.5}Ge_{0.5})$ (CFGG) with the (110) orientation (see **Fig. 1**). Two nonmagnetic metals, Ag and Cu, were used for the spacer layer, and the effects of the crystal orientation upon the MR properties were examined by comparing these devices with those comprising of (001) epitaxial layers. For substrates with an Ag spacer, the PSV with the $(001)[110]_{CFGG}//(001)[010]_{Ag}$ interface grown on MgO(001) exhibits a higher MR output compared with the $(110)[001]_{CFGG}//(111)[1\bar{1}0]_{Ag}$ interface grown on sapphire $(11\bar{2}0)$. In contrast, a higher MR output is obtained using a Cu spacer with the $(110)[001]_{CFGG}//(111)[1\bar{1}0]_{Cu}$ interface (see **Fig. 2**). These results demonstrate that the MR outputs depend upon the crystal orientation at the interface, and that interfaces with a small misfit tend to exhibit less magnetic dead layer and a larger MR output (see **Fig. 3**). This study indicates the influence of crystal orientation as well as lattice mismatch upon the interfacial spin scattering asymmetry.

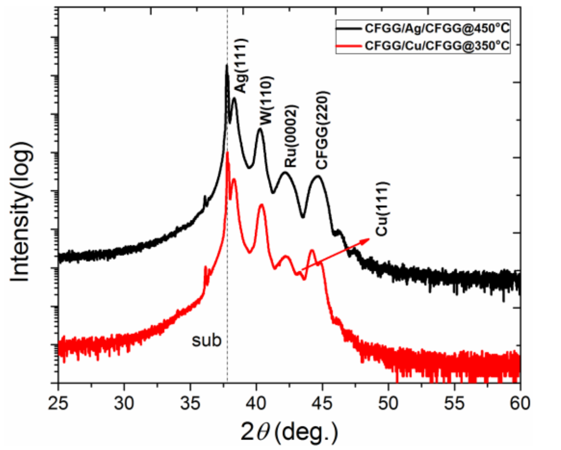

Fig. 1 The 2θ-□ XRD profiles of the film stacks Ta(20 nm)/W(20 nm)/Ag(100 nm)/CFGG(10 nm)/Ag annealed at 450°C and Cu(5 nm)/CFGG(10 nm)/Ag(5 nm)/Ru(8 nm) annealed at 350°C .

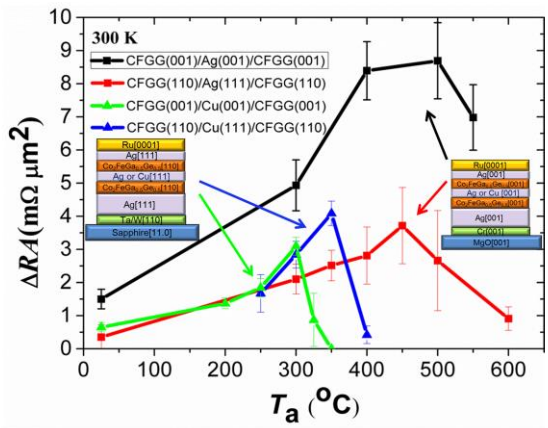

Fig. 2 The MR curve of the optimally annealed (a) CFGG(110)/Cu(111)/CFGG(110), (b) CFGG(110)/Ag(111)/CFGG(110) PSVs at 10K (open square) and 300K (solid square).

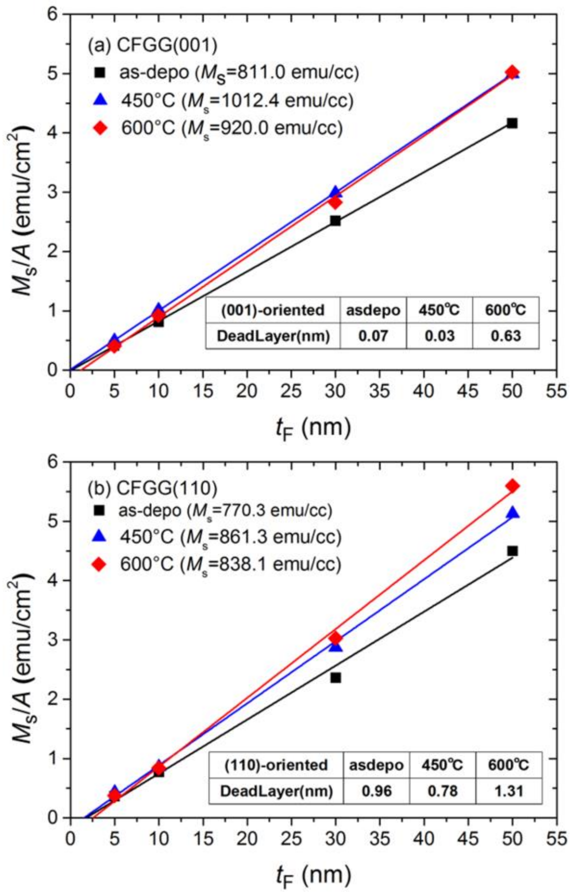

Fig. 3 The MR curve of the optimally annealed (a) CFGG(110)/Cu(111)/CFGG(110), (b) CFGG(110)/Ag(111)/CFGG(110) PSVs at 10K (open square) and 300K (solid square).

b) NiAl spacer

In our previous study using epitaxial CPP-GMR devices, we found the change of crystal orientation in $Co_2FeGa_{0.5}Ge_{0.5}$(CFGG)/Ag system had introduced a different lattice mismatch at CFGG/Ag interface resulting in various MR outputs, i.e., the CPP-PSV with $(001)_{CFGG}/(001)_{Ag}$ interface showed higher MR output than that with $(011)_{CFGG}/(111)_{Ag}$ interface.[10] The influence of lattice matching on the interfacial spin scattering asymmetry is considered to be the main reason for the *ΔRA* value difference. However, how the band matching changes according to different crystal orientation is still unclear, because both lattice matching and band matching change when we alter the crystal orientation in the CFGG(bcc)/Ag(fcc) system. To focus only on the band matching influence, in this work, high quality epitaxial CFGG/NiAl/CFGG all-B2-trilayers structure devices were fabricated on both MgO and sapphire single crystal substrates to create (001) and (110) crystal orientations (see **Fig. 4**). Same magneto-transport properties were observed from these two differently orientated devices indicating that there is no or little orientation dependence of band matching on MR output (see **Fig. 5**). We also found that all-B2-trilayer structure was free of lattice matching influence depending on the crystal orientation, which made it a good candidate for CPP-GMR device.

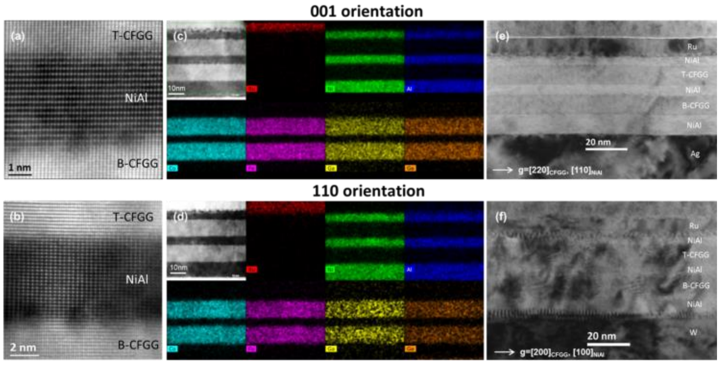

Fig. 4 High resolution HAADF-STEM images (a), EDS mapping images (c) and two-beam condition bright field image (e) of CFGG(001)/NiAl(001)/CFGG(001) film and High resolution HAADF-STEM images (b), EDS mapping images (d) and two-beam condition bright field image (f) of CFGG(110)/NiAl(110)/CFGG(110) film.

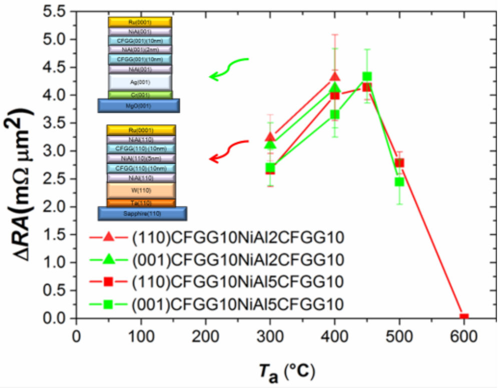

Fig. 5 Annealing temperature dependence of ΔRA of the film stack in (110) and (100) orientation with NiAl spacer.

2. Enhancement of $L2_1$ order and spin-polarization in Co_2FeSi thin film by substitution of Fe with Ti

According to the theoretical calculation reported by Miura et al.,[11] Co_2TiSi Heusler alloy has high spin polarizations in the $L2_1$ structure and high tolerance for the Co-related atomic disorder. On the other hand, Co_2FeSi Heusler alloy shows $L2_1$-order even in the as-deposited state and has large exchange stiffness.[12] By combining these two alloys, $Co_2(Fe_{1-x}Ti_x)Si$ (CFTS) could be a promising Heusler alloy which has both high spin polarization and high kinetics for $L2_1$-ordering. In this study, we investigated the effect of Ti substitution for Fe in Co_2FeSi Heusler alloy ($Co_2(Fe_{1-x}Ti_x)Si$) on their electronic structure, chemical ordering and spin-dependent transport properties. First-principles calculations of the density of states (DOS) indicated that the peak just above the Fermi level in the minority-spin DOS (the conduction band edge of half-metallic gap) shifts towards higher energies with increasing Ti, leading to the enhancement in the half-metallic gap and the spin-polarization (see **Fig. 6**). We found in epitaxial CFTS thin films that the required annealing temperature for long range $L2_1$-ordering can be substantially reduced from 650 ºC for Co_2FeSi to 400 ºC for $x \geq 0.2$ (see **Fig. 7**). The enhancement of spin-polarization by the substitution of Fe with Ti was experimentally confirmed from anisotropic magnetoresistance measurements (see **Fig. 8**) and spin-accumulation signals in non-local spin valve devices with the $Co_2Fe_{1-x}Ti_xSi$ films (see **Fig. 9**).

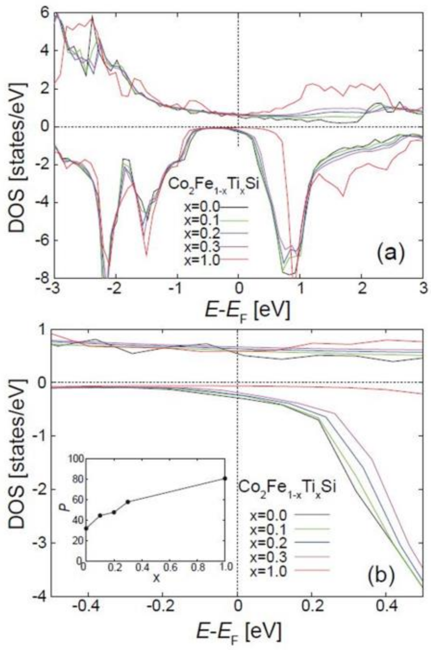

Fig. 6 (a) The energy dependences of the DOS for $Co_2(Fe_{1-x}Ti_x)Si$ Heusler alloys with different x. (b) The enlarged view of Fig. 1(a) around the Fermi level E_F. The inset shows the x dependence of the spin polarization P.

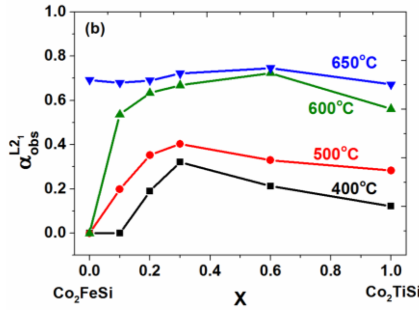

Fig. 7 Dependence of $L2_1$ ordering of $Co_2Fe_{1-x}Ti_xSi$ films on annealing temperature and Ti composition.

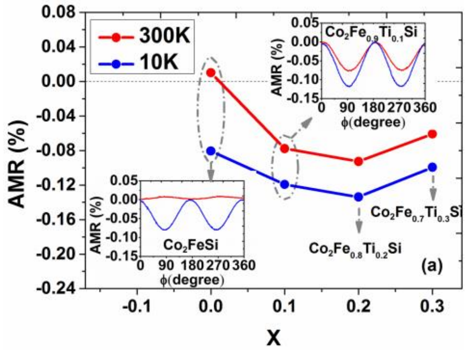

Fig. 8 Dependence of AMR ratio on Ti composition in $Co_2Fe_{1-x}Ti_xSi$ films.

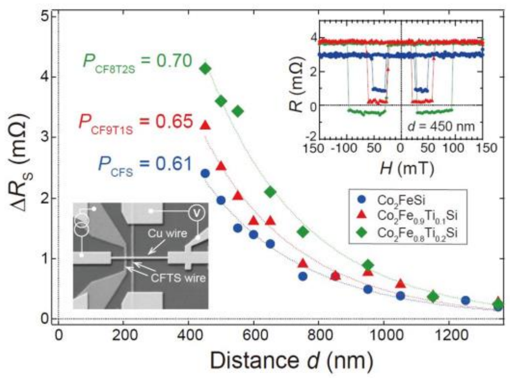

Fig. 9 Spin signal ΔR_s as a function of wire distance d for each $Co_2Fe_{1-x}Ti_xSi$ device. Dashed line indicates the fitting result by the one dimensional spin diffusion model. The insets show SEM image of typical LSV device and the R-H curves for the devices with d = 450 nm.

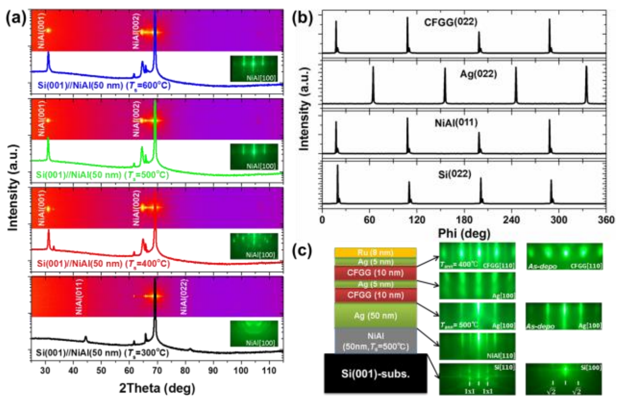

Fig. 10 (a) and (b) XRD profiles and corresponding two-dimensional diffraction images of 50 nm thick NiAl layer on Si(001) substrate with deposition temperature ranging from 300 to 600°C. (c) Structure illustration of whole CPP-GMR film stack and corresponding RHEED patterns for each layer..

3. Realization of high quality epitaxial CPP-GMR pseudo spin-valves on Si(001) wafer using NiAl buffer layer

It is undisputed that spintronics is one of the research fields that have rapidly developed in these two decades. However, only a few applications reached to the practical level so far, *i.e.* there is still large distance between fundamentals and applications in spintronics field. Therefore, further efforts to build up a "bridge" between fundamental studies and practical applications are strongly desired. Many previous fundamental studies focused only on magnetic tunnel junctions (MTJs) and CPP-GMR devices epitaxially grown on unpractical MgO single crystalline substrate. Therefore, finding an optimum fabrication process for developing high quality single crystalline spintronic devices on practical Si substrate is one of valuable challenges to make a shortcut to various potential applications based on single crystalline devices. In this work, we reported NiAl buffer layer as a template for the integration of epitaxial CPP-GMR devices on a Si(001) single crystalline substrate. By depositing NiAl on Si at an elevated temperature of 500°C, a smooth and epitaxial *B2*-type NiAl(001) layer was obtained (see **Fig. 10**). The surface roughness was further improved by depositing Ag on the NiAl layer and applying subsequent annealing process (see **Fig. 11**). The epitaxial CPP-GMR devices grown on the buffered Si(001) substrate present large MR outputs comparable with the devices grown on an MgO(001) substrate, demonstrating the possibility of epitaxial spintronic devices with the NiAl template for practical applications (see **Fig. 12**). This study demonstrated the feasibility of growing epitaxial CPP-GMR on Si wafers, which can exhibit much higher MR output than polycrystalline devices that are deposited on amorphous buffered industrially standard chemical-mechanical-polished substrates. It gives a potential for developing industrially viable single crystalline CPP-GMR devices on Si wafer for allowing higher temperature annealing in combination with the wafer bonding technology.

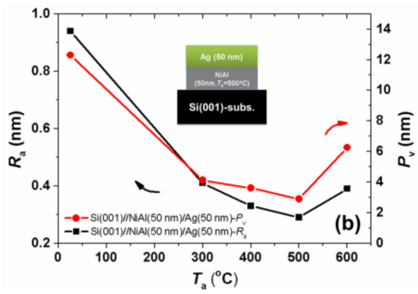

Fig. 11 Surface roughness measured by AFM of the film stacks Si(001)//NiAl(50 nm)/Ag(50 nm).

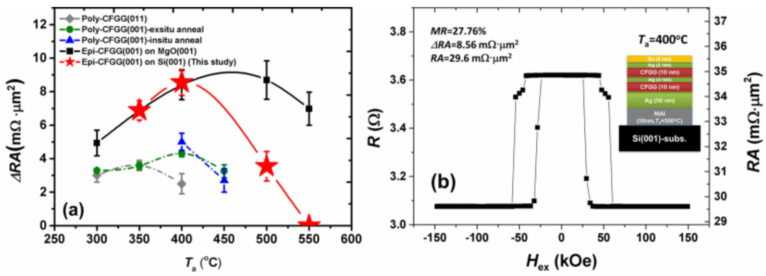

Fig. 12 Annealing temperature dependence of Δ*RA* for epitaxial CPP-GMR devices on Si(001) substrate (red star), epitaxial CPP-GMR devices on MgO(001) substrate (black square) and polycrystalline CPP-GMR devices on Si/SiO_2 substrate (grey square, green circle and blue triangle). (b) Typical MR curve of CPP-GMR devices annealed at optimal temperature 400°C.

4. Fabrication of Single Crystalline Magnetoresistive Sensors on Polycrystalline Electrode using Three-Dimensional Integration Technology

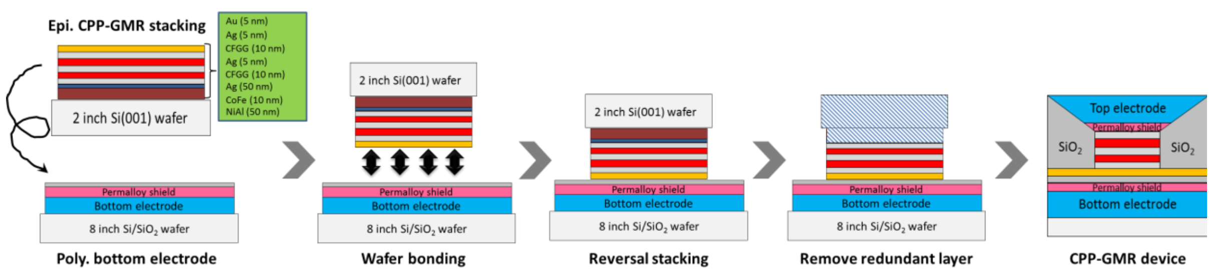

Fig. 13 Wafer bonding process in this study. Epitaxial CPP-GMR film on 2inch Si(001) substrate was bonded on poly-crystalline permalloy film deposited on 8 inch Si/SiO_2 wafer.

A current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) device inherently possesses low *RA* due to its all-metallic structure; however, the MR ratio used to be too small for any applications. Recently, substantially large MR ratios (>80%) and ultralow *RA* of 0.05 $\Omega\mu m^2$, which satisfy the requirements for >2 Tbit/in^2,[9] have been realized in CPP-GMR devices with Co-based Heusler alloys such as Co_2MnSi and $Co_2Fe(Ga_{0.5}Ge_{0.5})$ (CFGG)[13,14] that exhibit half-metallic behavior. However, the following two critical issues still hinder practical applications of the CPP-GMR devices. First, MR ratios > 30–50% have been achieved only in (001)-oriented epitaxial CPP-GMR devices grown on unpractical MgO(001) substrates. Secondly, high-temperature annealing over 500°C is indispensable for attaining the high MR ratios. Such high-temperature annealing is not compatible with the conventional fabrication processes of read heads because it damages the permalloy magnetic shield and destroy layered structure of polycrystalline devices. To overcome these critical issues, we develop the following processes (see **Fig. 13**): (i) epitaxial growth and high-temperature annealing of CPP-GMR multilayer on Si(001) substrate, (ii) bond a sensor film stack onto a wafer with electrode by a direct wafer bonding technique, [15] (iii) removal of backside Si and (iv) micro-fabrication to make devices. These processes enable integration of the high-performance epitaxial CPP-GMR devices for sensor applications. High MR output (ΔRA of 8.6 mΩ • μm^2 and MR ratio of 27.8%) are achieved using the CFGG Heusler alloy as ferromagnetic layers on Si(001) substrate. It should be noted that by inserting CoFe as a diffusion barrier, MR output is further enhanced to 9.5 mΩ • μm^2 and 37% after annealing at 500ºC. These MR outputs are comparable with those for the multilayer grown on an MgO(001) substrate (see **Fig. 14**). CoFe insertion layer can acted as a diffusion barrier and suppressed the formation of silicide at NiAl/Si interface during the high-temperature annealing (see **Fig. 15**). The improvement of the annealing stability by inserting CoFe layer resulted in the enhancement of MR output as shown in Fig. 14 because the high-temperature annealing without inter-diffusion promoted $L2_1$ ordering in CFGG layers, which is essential for the half-metallic band structure.

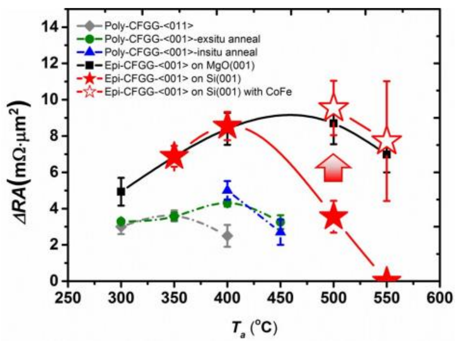

Fig. 14 Annealing temperature dependence of ΔRA for various CPP-GMR devices.

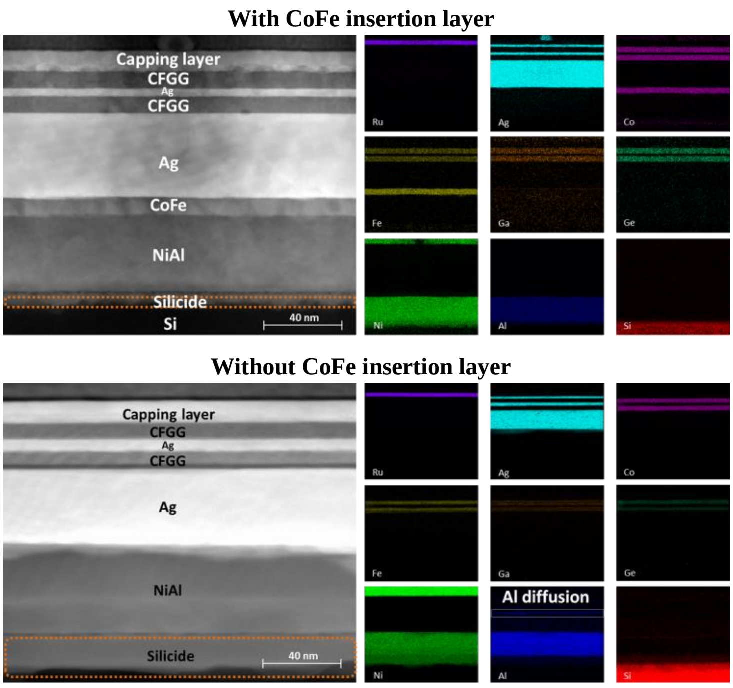

Fig. 15 Cross-sectional HAADF-STEM and EDS mapping image for CFGG/Ag/CFGG based CPP-GMR device annealed at 500 ^{o}C w/wo CoFe insertion layer.

Microstructure of the bonding interface was analyzed by high-resolution HAADF-STEM and EDS mapping (see Fig. **16**). Smooth Ta/Au bonding interface without any defects is observed. In the magnified HAADF image of bonding interface region, a clear bonding is obtained while keeping the single-crystal structure of Au layer on top of amorphous Ta layer. Note that this is first demonstration for direct bonding epitaxial MR multilayer film on top of amorphous layer. The EDS mapping images show that the Au/Ta interface is sharp without any oxidation, which means the epitaxial CPP-GMR film is successfully bonded on top of the permalloy bottom electrode.

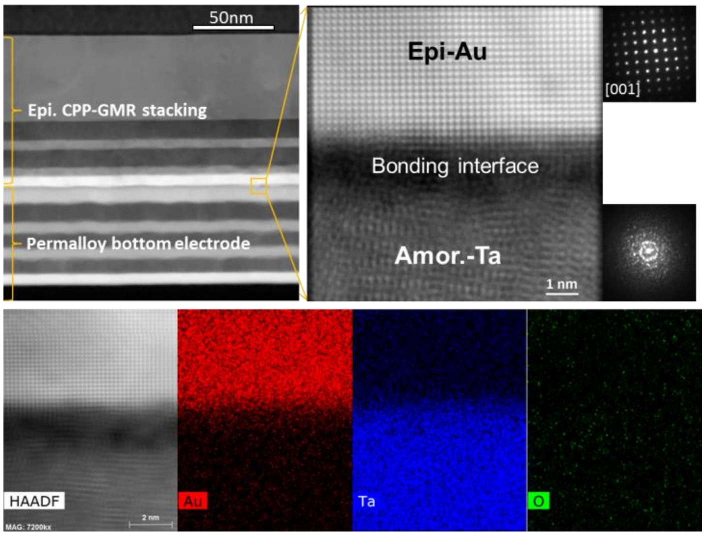

Fig. 16 Microstructure analysis of the stacking film after bonding and bonding interface by HR-HAADF STEM. Epitaxial CPP-GMR film is successfully bonded on Ta capping layer without any remarkable defects at the interface.

References

[1] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, Nat. Mater. **3**, 868 (2004).
[2] S.S.P. Parkin, C. Kaiser, A. Panchula, P.M. Rice, B. Hughes, M. Samant, and S.-H. Yang, Nat. Mater. **3**, 862 (2004).
[3] C. Chappert, A. Fert, and F.N. Van Dau, Nat. Mater. **6**, 813 (2007).
[4] T. Valet and A. Fert, Phys. Rev. B **48**, 7099 (1993).
[5] Y. Sakuraba, K. Izumi, T. Iwase, S. Bosu, K. Saito, K. Takanashi, Y. Miura, K. Futatsukawa, K. Abe, and M. Shirai, Phys. Rev. B **82**, (2010).
[6] Y.K. Takahashi, N. Hase, M. Kodzuka, A. Itoh, T. Koganezawa, T. Furubayashi, S. Li, B.C.S. Varaprasad, T. Ohkubo, and K. Hono, J. Appl. Phys. **113**, 223901 (2013).
[7] J. Sato, M. Oogane, H. Naganuma, and Y. Ando, Appl. Phys. Express **4**, 113005 (2011).
[8] Y.K. Takahashi, A. Srinivasan, B. Varaprasad, A. Rajanikanth, N. Hase, T.M. Nakatani, S. Kasai, T. Furubayashi, and K. Hono, Appl. Phys. Lett. **98**, 152501 (2011).
[9] M. Takagishi, K. Yamada, H. Iwasaki, H.N. Fuke, and S. Hashimoto, Magn. IEEE Trans. On **46**, 2086 (2010).
[10] J. Chen, S. Li, T. Furubayashi, Y.K. Takahashi, and K. Hono, J. Appl. Phys. **115**, 233905 (2014).
[11] Y. Miura, M. Shirai, and K. Nagao, J. Appl. Phys. **99**, 08J112 (2006).
[12] O. Gaier, J. Hamrle, S. Trudel, B. Hillebrands, H. Schneider, and G. Jakob, J. Phys. Appl. Phys. **42**, 232001 (2009).
[13] Y. Sakuraba, K. Izumi, T. Iwase, S. Bosu, K. Saito, K. Takanashi, Y. Miura, K. Futatsukawa, K. Abe, and M. Shirai, Phys. Rev. B **82**, (2010).
[14] S. Li, Y.K. Takahashi, T. Furubayashi, and K. Hono, Appl. Phys. Lett. **103**, 042405 (2013).
[15] H. Takagi and R. Maeda, J. Cryst. Growth **292**, 429 (2006).