Half-metallic Heusler compounds: Spin-dependent transport properties in thin films and magnetoresistive devices

Y. Sakuraba, T. M. Nakatani, Y. K. Takahashi, T. Furubayashi, T. T. Sasaki, Y. Miura, and K. Hono National Institute for Materials Sciece (NIMS), Sengen 1-2-1, Tsukuba, Ibaraki, Japan

History of half-metallic materials started more than 30 years ago from the first prediction of half-metallicity in NiMnSb by Groot et al.[1] Although nearly 100% spin-polarization at room temperature (RT) has never been observed so far in magnetoresistive devices via spin-dependent transport measurements, there is still large expectation in various spintronic applications with half-metals because large spin-polarization without using tunnelling spin-filter effect is beneficial to realize high performance spintronic devices with very low device resistance. Recent extensive studies on current-perpendicular-to-plane giant magnetoresistive (CPP-GMR) devices using half-metallic Co-based full Heusler compounds successfully demonstrated large enhancement of magnetoresistance at RT with small RA below $0.1\Omega\mu m^2$ due to high spin-polarization of Co-based full Heusler compounds. Large MR ratio over 30% at RT has been reported in fully-epitaxial CPP-GMR devices with Co-based Heusler such as Co₂MnSi, Co₂FeGa_{0.5}Ge_{0.5}(CFGG) and Co₂Fe_{0.4}Mn_{0.6}Si[2-4], which is one order of magnitude larger than CPP-GMR with general 3d transition metals (Figure 1). We have recently fabricated fully-epitaxial CPP-GMR devices CFGG/Ag/CFGG with very thin NiAl insertion to CFGG/Ag interfaces and observed surprisingly large MR ratios of 82% at RT and 285% at 10 K[5]. This enhancement by inserting thin NiAl seems to be related with good electronic band matching between NiAl and CFGG electrode, but careful analysis beyond the framework of diffusive transport model is necessary because the thickness of inserted NiAl is just 0.21 nm which is shorter than mean free path. On the other hand, the effect of chemical disordering of Heusler on spin-polarization of conduction electron have been carefully analysed in our recent study via AMR effect and anomalous XRD measurement in SPring-8.[6,7] Our studies clearly confirmed the importance to suppress Co antisite by optimizing the composition ratio in Heusler film/electrode for obtaining large spin-polarization. The progress of recent study and future prospect of half-metallic Heusler compounds will be presented.



Figure 1.The progress of MR property in CPP-GMR with half-metallic full-Heusler electrodes. Left figure shows MR curve at RT in CFGG/NiAl/Ag/NiAl/CFGG CPP-GMR device[5].

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Development of High-Resolution TMR Sensor Device for Application of Bio-Magnetic Field Measurement

J. Jono², K. Fujiwara¹, M. Oogane¹, M. Tsuchida² and Y. Ando¹ ¹Department of Applied Physics, Graduate School of Engineering, Tohoku University, Sendai, Japan ²KONICA MINOLTA, INC., Tokyo, Japan

Bio-magnetic field applications have been expected for many years to the functional diagnosis of a human body. A diagnostic device with superconducting coil to diagnose epilepsy is an application example¹⁾. High-sensitive magnetic sensors which operate at room temperature without liquid Helium have been expected as next-generation magnetic sensors in order to expand the scope of application of bio-magnetic field measurement devices.

We are developing Tunneling Magneto Resistance (TMR) sensors with the aim of bio-magnetic field measurement and have developed a sensor device which can detect tens of pico Tesla of magnetic cardiac field (MCG) in low frequency band (Fig.1)

Noise reduction of TMR sensor device is important as same as improvement of the response to the magnetic field of the TMR sensor to obtain high magnetic field resolution. Therefore, Magnetic tunnel junctions (MTJs) coupled to soft magnetic layer for high magnetic sensitivity were connected series and parallel to reduce noise of the MTJs. 150% TMR ratio and about 5 Oe anisotropy field (Hk) near saturation range of magnetic property were observed with the arrayed MTJs (Fig.2). In addition, low noise analog amplifier especially in low frequency band was developed to reduce system noise. (Fig.3)

We are studying to apply more high magnetic responsive materials to the sensor device for detecting bio-magnetic field of brain in the near future²⁾ (Fig.4).

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Fig. 1. Waveform of MCG and ECG



Fig. 3. Circuit block diagram



Fig. 2. R-H curve of MTJs with NiFe



Evolution of synchronization in spin torque oscillators

S. Tsunegi, K Yakushiji, A. Fukushima, S. Yuasa, and H. Kubota (AIST, Spintronics Research Center, Tsukuba, Japan)

Spin-torque oscillator (STO)¹⁾ has attracted much interest from a viewpoint of application to a nano-scale oscillator because of the wide range frequency tunability and high compatibility with semiconductor CMOS circuits. For the practical use, a lot of work has been done to improve emission power and narrow linewidth of emission spectrum. The studies so far have been conducted from mainly two aspects. The one is material development for STO devices such as MgO barrier and suitable free layers. The other is system development using a magnetic or electric interaction between multiple STOs or between an STO and external rf signals.

In the first approach, we carried out serial studies with STOs developed using MgO-based magnetic tunnel junctions (MTJs), resulted in the emission power increase up to 0.1 μ W.²⁾ The power increase was a dramatic leap from a few pW reported in the initial GMR based STO¹⁾. Recent studies have revealed that STOs having perpendicularly magnetized free layer³⁾ and sombrero-type free layer⁴⁾ exhibit further increased emission power up to a few μ W (Table I). Furthermore, quite recently, an emission power over 10 μ W has been achieved in a vortex-type STO with a narrow linewidth of 100 kHz. ^{5,6)} The value of 10 μ W is as large as that of commercial crystal oscillators.

In the second approach, S. Kaka⁷⁾ and F. B. Mancoff⁸⁾ demonstrated the reduction of linewidth in two-point-contact STOs in 2005. They realized the synchronization between STOs' precession, where precession frequencies drew each other through spin-wave and dipole-diple interactions. W. H. Rippard demonstrated electrical synchronization of STO precession to a large rf current injected from an external signal source.⁹⁾ In this case the synchronization was induced by rf spin torque. However, in the early stage of synchronization investigation, it was impossible to realize the electrical synchronization among STOs because of very low emission powers generated by the STOs.

Table 1 Several types of S10s and its features.					
	Sombrero ⁴⁾		Perpendicularly Magnetized free layer ³⁾	Vortex ^{5,6)}	
Туре	Insulator				
Size	4 µm		120 nm (250 nm)	300 nm	
Power µW	2.4	0.1	0.5 (2.0)	1.4	3.4
Frequency GHz	4	11	6 (7.0)	0.23	0.48
Q factor (freq./linewidth)	330	3200	130 (2300)	6400	210
Features	High power, high Q		Small, high power	High Q, Low frequency	

Table I Several types of STOs and its features.

It is at the very moment that the two approaches are merged. As mentioned, the recent progress of the STO performance has enabled the electrical synchronization among STOs. Indeed, we demonstrated the self-synchronization in a vortex-STO,¹⁰ where the vortex gyration was synchronized to rf currents generated by the STO itself.^{11,12} In such system, the phase difference between the STO and the reinjected rf current gives remarkable influence on the gyrotropic motion of the vortex. The fact indicates that the phase difference is essential to the electrical mutual synchronization as theory predicted.¹³ By taking account of the effect of the phase difference, we have finally demonstrated the electrical mutual synchronization among STOs. The emission power and linewidth were successfully improved as shown in Fig. 1.^{14,15} In the presentation, we report our latest results on the vortex-STOs as well as their electrical mutual synchronization.



Figure 1 The STO number dependence of emission power and linewidth.

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Spin Torque Oscillations in Giant Magnetoresistance Devices with Heusler Alloys

T. Yamamoto¹, T. Seki^{1,2,3}, and K. Takanashi^{1,3}

¹ Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

² PRESTO, Japan Science and Technology Agency, Saitama 322-0012, Japan

³ Center for Spintronics Research Network, Tohoku University, Sendai 980-8577, Japan

A spin-polarized current flowing through a ferromagnet exerts torque on the magnetization. This quantummechanical torque, called spin-transfer torque (STT), offers novel methodologies to manipulate the static direction and/or the dynamical motion by an electric current. A spin torque oscillator (STO) [1] is a nano-sized oscillator which utilizes the magnetization dynamics excited by the STT as a source of the microwave emission. According to the device structure and/or the underlying phenomena of the conversion process from the magnetization dynamics to microwave, STOs can be categorized into several types, *e.g.* current-perpendicular-to-plane (CPP) giant magnetoresistance (GMR) STOs, magnetic tunnel junction (MTJ) STOs, and spin-Hall STOs. Since the output power of an STO scales with the magnetoresistance of the device, MgO-based MTJ-STOs have a great advantage in the achievement of large output power owing to the huge tunnel magnetoresistance. Nevertheless, the absence of the tunneling current in GMR-STOs would contribute to a reduction of the shot noise, and thus would be potentially advantageous to realize extremely high oscillation quality factor.

In order to overcome the disadvantage of GMR-STOs, namely, the low output power of the spin torque oscillation, we have developed Heusler alloy-based GMR-STOs including nanopillar STOs [2] and point-contact STOs [3]. Through these studies we demonstrated that the utilization of highly spin-polarized Heusler alloys is promising to realize high power in GMR-STOs. Also, these studies reminded us of the importance of the control of the magnetization dynamics with dealing with the non-uniform effective field in the oscillating layer arising from the magnetocrytstalline anisotropy and the Oersted field; the existence of non-uniform effective field prevents the excitation of coherent magnetization dynamics, and that results in low oscillation quality factor.

As the application of magnetic vortex dynamics has been intensively studied by several groups, it is useful to enhance the oscillation quality factor of STOs [4]. In addition to the high spin-polarization, Co₂(Fe,Mn)Si (CFMS) alloys exhibit soft magnetic properties, and that allows us to effectively control the magnetization configuration via microfabrication. Indeed, the direct observation of magnetic vortex formation was reported for epitaxially-grown CFMS discs [5]. Then we fabricated GMR-STOs using CFMS vortices as shown in Fig. 1. The GMR-STO consists of a 30-nm-thick and 240-nm-diameter CFMS vortex oscillating layer and a 20-nm-thick CFMS reference layer separated by a 5-nm-thick Ag spacer layer. Figure 2 shows a representative frequency-domain power spectrum obtained from our GMR-STO with a Co₂MnSi vortex. Here output power of 3.5 nW as well as high oscillation quality factor of 5400 were achieved. The output power was further improved by optimizing the Fe-Mn composition of CFMS, and the output power exceeding 10 nW was achieved even in the all-metallic STOs. Moreover, the estimated radii of the vortex core trajectories reached about 75% of the actual radii of the CFMS oscillating layers. These experimental results indicate the potential of the highly spin-polarized Heusler alloys for the development of high performance GMR-STOs.



Fig. 1 Schematic illustration of the micro fabricated GMR-STO with a CFMS vortex along with the measurement circuit used for the microwave measurement.



Fig. 2 Power spectrum obtained from a GMR-STO with a Co_2MnSi vortex.

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Spin injection, transport and detection technology in ferromagnet/MgO/Si devices

Yoshiaki Saito, Mizue Ishikawa, Tiwari Ajay, Hideyuki Sugiyama and Tomoaki Inokuchi (Toshiba Corporation, 1, Komukai-Toshiba, Kawasaki, 212-8582, Japan)

Spin metal-oxide semiconductor field-effect transistors (spin-FETs) [1, 2], whose source and drain electrodes consist of ferromagnetic materials, are expected to lead to a new logic-in-memory architecture. Recently, many studies of silicon (Si) spintronics for realizing the spin-FETs have been reported because of the observation of room-temperature spin signals in Si. [3-9,12,13] However, in order to realize the spin-FETs, it is necessary to improve injection and detection efficiencies of electrical spin in semiconductors [10-13]. We have been observing spin accumulation signals in Si with relatively long spin relaxation time by measuring three-terminal and four-terminal Hanle signals for CoFe/MgO/n⁺-Si(100) and Heusler Co₂FeSi/MgO/n⁺-Si(100) devices [4, 7-13], and observing local magnetoresistance (MR) and nonlocal (NL)-MR signals up to room temperature. [9, 11-13] However, the estimated spin polarization using standard spin diffusion theory was a small value of ~ 0.16 . [8, 9, 11] The spin polarization in n^+ -Si estimated by other gropes using standard spin diffusion theory has also exhibited small values, for example: ~ 0.05 for Fe/MgO/ n^+ -Si at 8 K [6] and ~ 0.15 for CoFe/ n^+ -Si devices at room temperature. [5] It is therefore necessary to improve the spin polarization (spin injection and detection efficiency) in Si to achieve large spin signals in Si. Recently, we have been succeeded in improving the spin polarization in Si. [12, 13] The estimated spin polarization (P) and spin life time (τ) are P ~ 40% and τ ~ 1 nsec, respectively at room temperature. The large spin injection and detection efficiency into Si and relatively long spin relaxation time even at room temperature and spin signals at room temperature along with its robustness up to 400°C are observed.

In this invited talk, we review the resent progress and our current status of Ferromagnet/MgO/n⁺-Si junction technology for increasing the spin signals in Si. This work was partly supported by ImPACT Program of Council for Science, Technology and Innovation (Cabinet Office, Government of Japan) and Grant-in-Aid for Scientific Research from JSPS.

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Recent Progress in Silicon-based Spintronics Devices

Hayato Koike¹, Yuichiro Ando², Shinji Miwa³, Yoshishige Suzuki³, Masashi Shiraishi² ¹ Technology HQ, TDK Corporation, Chiba 272-8558, Japan ² Department of Electronic Science and Engineering, Kyoto University, Kyoto 615-8510, Japan ³ Graduate School of Engineering Science, Osaka University, Osaka 560-8531, Japan

Spin-dependent transport phenomena in semiconductor have been attracting much attention from both the fundamental and the practical points of view. The devices based on such phenomena have a possibility to archive continuous performance improvement of conventional ICT devices. Among them, spin-MOSFETs¹⁾ are expected as one of the promising candidates for beyond-CMOS devices. Even in the case of spin MOSFETs, silicon is attractive as a channel material because it has the advantage in terms of good spin coherence (= weak spin-orbit interaction).

To realize silicon-based spintronics devices, it is necessary to perform a series of processes consisting of "spin injection, spin transport (modulation), spin detection" in electrical method. For investigation of these spin related properties, multi-terminal lateral spin valve (LSV) devices are extremely useful. It is widely recognized it should be demonstrated completely by spin-valve effect measurement and Hanle effect measurement, using nonlocal (4-terminal) and local (2-terminal) geometries.

Especially in recent years, remarkable progresses have been made in the field of silicon-based spintronics. First example is the observation of spin output signal at room temperature. It has already been reported by several research groups $^{2), 3), 4}$ and most of them have utilized FM/MgO/*n*-Si system (= ferromagnetic metal electrode, MgO barrier and *n*-type degenerate silicon channel). Second example is the modulation of the spin output signal by the gate voltage application at room temperature. ^{5) 6} It has been achieved in FM/MgO/*n*-Si (*n*-type non-degenerate silicon channel) system with back-gate electrode of SOI structure. These progresses have led to the further understanding of fundamental spin related physics, such as spin drift effect, and the further improvement of spin output voltage (over 1 mV).⁷

Although silicon spintronics devices have advanced steadily for practical application, technological issues to be solved still are abound. Among them, improvement of spin injection efficiency is strongly desirable. In order to overcome these issues, even now research and development of silicon-based spintronics devices have been carried out energetically.

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