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

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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 0.1Ωμm^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 Co_2MnSi, $Co_2FeGa_{0.5}Ge_{0.5}$(CFGG) and $Co_2Fe_{0.4}Mn_{0.6}Si$[2-4], which is one order of magnitude larger than CPP-GMR with general 3*d* 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.

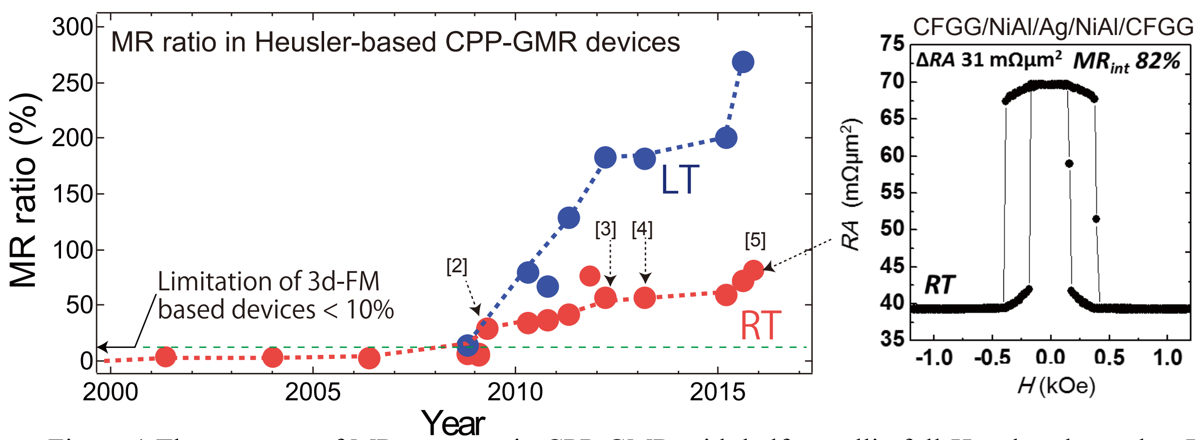

Figure 1.The progress of MR property in CPP-GMR with half-metallic full-Heusler electrodes. Left figure shows MR curve at RT in CFGG/NiAl/Ag/NiAl/CFGG CPP-GMR device[5].

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

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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 example[1)]. 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 future[2)] (Fig.4).

Acknowledgment

This work was supported by S-Innovation program, Japan Science and Technology Agency (JST).

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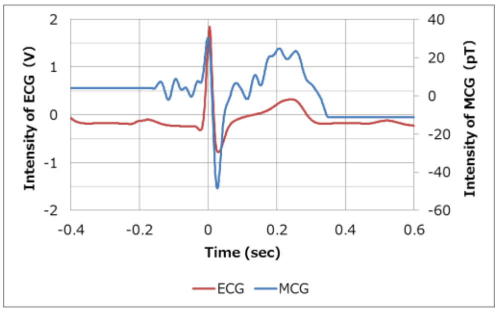

Fig. 1. Waveform of MCG and ECG

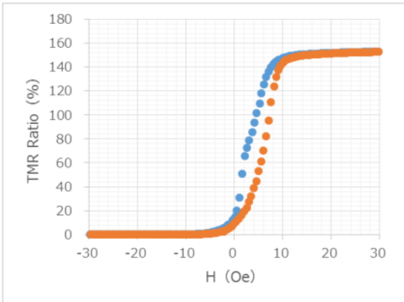

Fig. 2. R-H curve of MTJs with NiFe

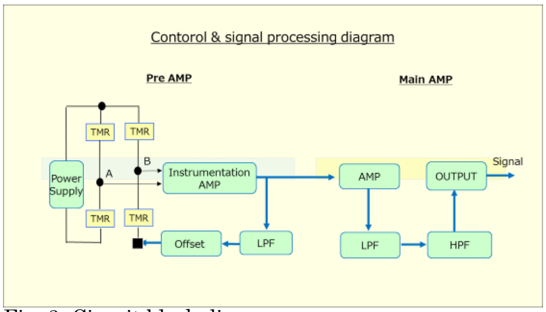

Fig. 3. Circuit block diagram

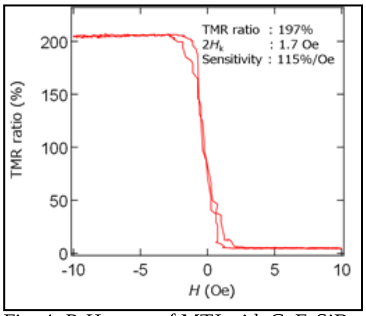

Fig. 4. *R-H* curve of MTJ with CoFeSiB

Evolution of synchronization in spin torque oscillators

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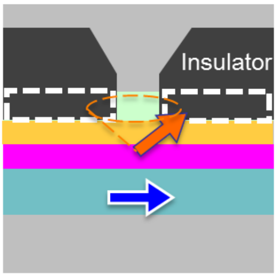
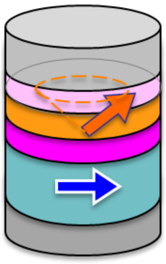
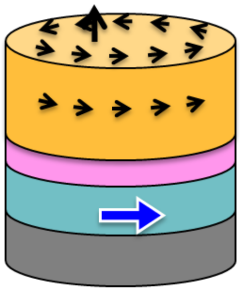
Spin-torque oscillator (STO)[1] has attracted much interest from a viewpoint of application to a nano-scale oscillator because of the wide range frequency tunability and high compatibility with semiconductor CMOS circuits. For the practical use, a lot of work has been done to improve emission power and 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 μW.[2] The power increase was a dramatic leap from a few pW reported in the initial GMR based STO[1]. Recent studies have revealed that STOs having perpendicularly magnetized free layer[3] and sombrero-type free layer[4] exhibit further increased emission power up to a few μW (Table I). Furthermore, quite recently, an emission power over 10 μW has been achieved in a vortex-type STO with a narrow linewidth of 100 kHz. [5,6] The value of 10 μW is as large as that of commercial crystal oscillators.

In the second approach, S. Kaka[7] and F. B. Mancoff[8] 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 I Several types of STOs and its features.

Type	Sombrero[4] 		Perpendicularly Magnetized free layer[3] 	Vortex[5,6] 	
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	

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.

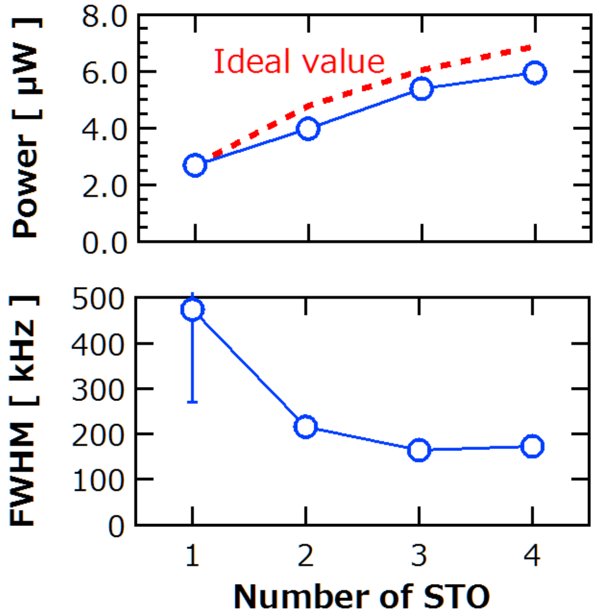


Figure 1 The STO number dependence of emission power and linewidth.

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Spin Torque Oscillations in Giant Magnetoresistance Devices with Heusler Alloys

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A spin-polarized current flowing through a ferromagnet exerts torque on the magnetization. This quantum-mechanical 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, $Co_2(Fe,Mn)Si$ (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 Co_2MnSi 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.

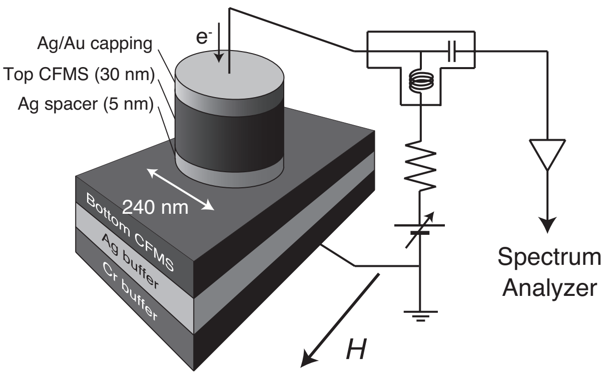

Fig. 1 Schematic illustration of the micro fabricated GMR-STO with a CFMS vortex along with the measurement circuit used for the microwave measurement.

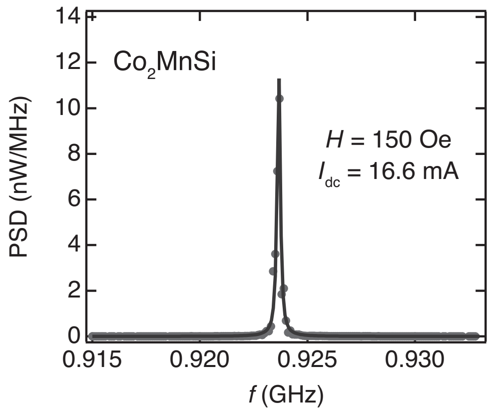

Fig. 2 Power spectrum obtained from a GMR-STO with a Co_2MnSi vortex.

Acknowledgments

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

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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 Co_2FeSi/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^{o}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

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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-MOSFETs [1)] 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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微小磁場下におけるアモルファス CoFeSiB 電極 強磁性トンネル接合の磁気抵抗特性

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Tunnel Magneto-resistance Properties in Magnetic Tunnel Junctions with Amorphous CoFeSiB Electrode in Low Magnetic Field

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背景

微小な生体磁場 (< 10^{-6} Oe) を計測することにより、病気の早期診断や高次機能解明が可能になる。近年、強磁性トンネル接合 (MTJ) 素子の高感度化により、室温で生体磁場計測が可能な MTJ センサの開発が積極的に行われている [1]。これまでにアモルファス CoFeSiB をフリー層に用いた MTJ 素子において、115%/Oe の世界最高の磁場感度(=TMR 比/2H_k ,H_k : 異方性磁場)が得られているが [2]、詳細なセンサ特性評価は行われていなかった。本研究では、CoFeSiB 電極 MTJ 素子の微小磁場下における磁気抵抗特性の評価を行った。

実験方法

超高真空マグネトロンスパッタ装置を用い、熱酸化膜付シリコン基板上に MTJ 薄膜を作製した。MTJ の膜構成は Si, SiO_2 subs./Buffer/$Co_{70.5}Fe_{4.5}Si_{15}B_{10}$ (100)/Ru (0.4)/$Co_{40}Fe_{40}B_{20}$ (3)/MgO (1.45, 2.5) /$Co_{40}Fe_{40}B_{20}$ (3)/Pin/Cap (in nm)である。フォトリソグラフィ法により 4 端子 MTJ 素子を形成した。二度の磁場中熱処理により磁化容易軸が直交した磁場センサ型 MTJ を作製した。直流磁場下における磁気抵抗特性を四端子法により測定した。3.3Hz の交流磁場下におけるシグナル電圧、および、0.1 – 10 Hz の低周波領域のノイズ特性評価をブリッジ回路により測定した。

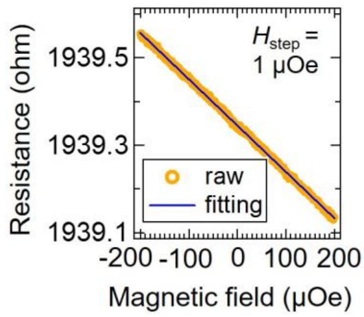

Fig.1 Magneto-resistance curve in low field region

実験結果

Fig. 1 に直流磁場範囲 200 μOe の磁気抵抗曲線を示す。ヒステリシスは観測されず、外部磁場に対してリニアな特性を有していることが分かる。Fig. 2 にシグナル電圧の交流磁場振幅依存性を示す。振幅に対してリニアに出力が変化している領域が磁場を検出できていることを示している。直線とノイズ電圧の交点である、最小検出可能磁場は 2×10^{-4} Oe であった。この値は 100×100 個程度の MTJ 素子の集積化により、心臓磁場を検出可能な性能である。本講演では、シグナル電圧およびノイズ特性の外部磁場依存性についても議論する予定である。

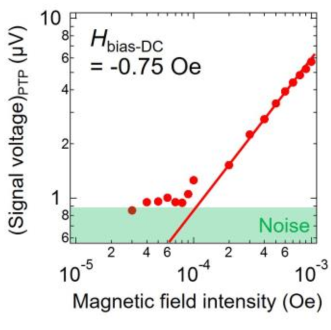

Fig. 2 External magnetic field dependence of signal voltage

謝辞

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強磁性トンネル接合における室温巨大磁気キャパシタンス効果

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Room temperature large magnetocapacitance effect in magnetic tunnel junctions

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はじめに

近年、スピントロニクス材料・デバイスにおける磁気キャパシタンス（MC）効果は、交流スピンダイナミクスに関する新たな学術的知見を与えられる一方、高感度磁気センサー、高周波磁気インピーダンス素子への応用も期待されていることから国内外で大きな注目を集めている[1-5]。中でも、強磁性トンネル接合（MTJ）は、興味深い交流スピンダイナミクスを示すと同時に、室温にて 50%程度の大きなトンネル磁気キャパシタンス（TMC）効果を示す。しかしながら、TMC 効果のメカニズムには不明な点が多く、また、TMC 比は、トンネル磁気抵抗（TMR）比の最大値（~600%）と比較しても、一桁程度小さい。そこで、本研究では、TMC 効果のメカニズムを明らかにするとともに、TMC 比の向上を目指すことを目的とした。

実験方法

超高真空マグネトロンスパッタ装置を用いて、熱酸化 Si 基板上に Ta/$Co_{50}Fe_{50}$/IrMn/$Co_{50}Fe_{50}$/Ru/$Co_{40}Fe_{40}B_{20}$/MgO/$Co_{40}Fe_{40}B_{20}$/Ta/Ru から構成される MTJ を作製した。強磁性層 $Co_{40}Fe_{40}B_{20}$ の膜厚は 3 nm、絶縁層 MgO の膜厚は 2 nm とした。微細加工にはフォトリソグラフィーとイオンミリング法を用いた。接合面積は 1800 μm^2 とした。TMC および TMR 効果の測定には、室温磁場中交流 4 端子法を用いた。測定周波数帯域は 80–1MHz、交流振幅電圧は 0.26 mV$_{rms}$、最大印加磁場は 1.4 kOe とした。

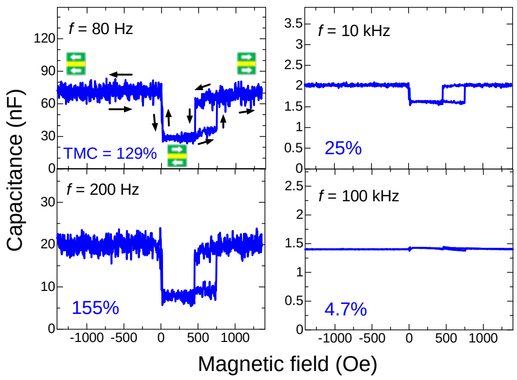

図 1: MTJ における TMC 効果

実験結果

図 1 に TMC 効果の周波数依存性を示す。200 Hz 付近で TMC 比が最大値（=155%）を示す。これは TMC 比の従来値（~50%）を大きく超える。また、TMR 比は周波数に依存せず 108%であったため、TMC 比は TMR 比よりも大きくなることも明らかになった。図 2 に TMC 比と磁化平行・反平行状態でのキャパシタンス $C_{P(AP)}$の周波数特性を示す。実験結果は Debye-Fröhlich モデルを用いた計算結果（実線）と良い一致を示した。すなわち、磁場により MTJ の磁化配置が変化すると、絶縁層をトンネルするキャリアの緩和時間が変化し、これにより動的誘電分極が変化する。この誘電分極の変化がキャパシタンスの変化となる[6]。講演ではより詳細な実験・計算結果を報告する。

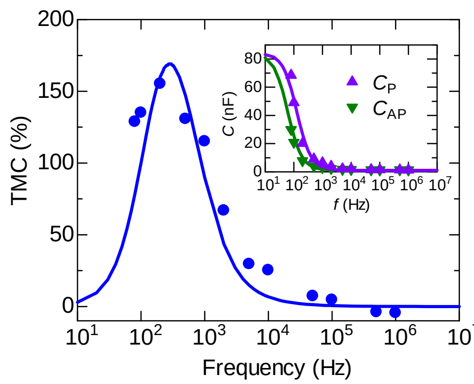

図 2: TMC 比と $C_{P(AP)}$の周波数特性

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Fe_3O_4 電極と MgO バリアを用いたトンネル磁気抵抗素子の作製

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Fabrication of tunnel magnetoresistance devices using Fe_3O_4 electrode and MgO barrier

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はじめに

Fe_3O_4 は、理論的にフェルミ面近傍で－100%のスピン分極率を示すハーフメタルという性質を持つと予測されており、Fe_3O_4 を電極材料として用いたトンネル接合において大きな負の TMR 効果を得ることが期待されている。しかし、今のところ、期待されたほど大きな TMR 効果が得られていない。過去に Fe_3O_4 電極と Al_2O_3 バリアを用いた接合において－12%の TMR 比が得られたという報告[1)]があるが、Fe_3O_4 のスピン分極率から考えると、十分大きな値とは言えない。一方で Fe(100)電極を用いた接合においてバリアを Al_2O_3 から MgO に変えることで飛躍的な TMR 比の増大がなされている。そこで本研究では、Fe_3O_4/MgO/Fe のトンネル磁気抵抗素子を作製し、電気的、磁気的評価を試みた。

実験方法

本研究では、到達真空度 1.0×10^{-7}Pa の超高真空中で MBE 法を用いて製膜を行った。作製した接合の構造は、MgO(100)基板/MgO/NiO/Fe_3O_4/MgO/Fe/Au とした。製膜後の結晶性の評価には RHEED を用いた。電気的、磁気的特性の評価をするために、フォトリソグラフィー、Ar イオンミリング、スパッタを用いて微細加工を行い、素子を作製した。作製した素子を用いて I-V 測定や磁気抵抗効果の測定を行った。

実験結果

Fe_3O_4 層は基板温度 300℃、O_2 雰囲気下で反応性蒸着を行い、600℃、O_2 雰囲気下で 30 分間アニールを行った。MgO バリア層は室温、O_2 雰囲気下で反応性蒸着を行い、その後、150℃で 30 分間アニールを行った。RHEED からは、Fe_3O_4、MgO ともにストリークを示しており、平坦性の良い膜が得られた。微細加工後の素子の磁気抵抗効果の測定から 80K において－47%の TMR 効果が得られた（Fig.1）。また、TMR 比の温度依存性から低温になるにつれて、TMR 比の増大が確認された(Fig.2)。

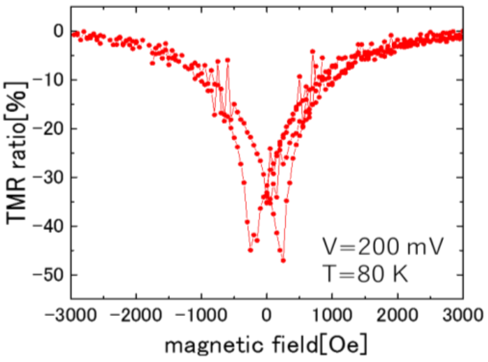

Fig.1 TMR observed for Fe_3O_4 MTJs

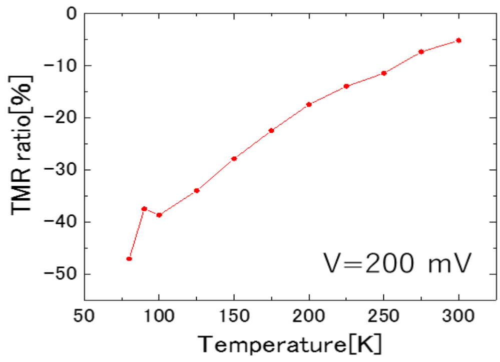

Fig.2 Temperature dependence of TMR ratio

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強磁性絶縁体を用いた MTJ における MR 効果

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MR effect in MTJ with ferromagnetic insulator

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はじめに

現在 MR 素子に使われている強磁性金属(FM)/非磁性絶縁体(NI)/強磁性金属(FM)の 3 層構造では、高密度化すなわち微細化の際に書き込み電流が増加してしまうという問題点を抱えている。この解決策の一つとして強磁性絶縁体によるスピンフィルター効果を利用した素子が提案されている。本研究では強磁性金属(FM)/非磁性金属(NM)/強磁性絶縁体(FI)/非磁性金属(NM)の 4 層構造を考える。この素子においても MR 効果が発現することを示し、応用上有利となる材料や構造の条件を明らかにすることを本研究の目的とする。

モデルと計算方法

本研究ではエピタキシャルに接合されている、すなわち乱れが無い 4 層構造 FM/NM/FI/NM を考える。この構造に対して 3 次元量子井戸型ポテンシャルのモデルを適応する。今回は FM と NM 部分が Fe/Cr の特徴を再現するようにポテンシャルを設定した。このモデルにおいて 3 次元シュレディンガー方程式を解析的に解くことで透過率に対する表式を求め、ランダウアー公式を用いて FM と FI の磁化が平行と反平行の場合のコンダクタンス G_p,G_{ap} を求めた。この際必要となる波数についての積分はコンピュータで数値計算した。また、MR 比を MR=1-G_{ap}/G_p と定義した。

計算結果

NM 膜厚と MR 比の関係を Fig.1 に示す。MR 効果が発現し、NM 膜厚の増加とともに MR 比が振動している。その振動周期は NM のフェルミ波長の 2 分の 1 になっており、MR 比の振動は NM 内での干渉効果によって生じていると考えられる。次に従来の 3 層構造 FM/NI/FM 及び本研究の 4 層構造における絶縁体膜厚と MR 比の関係を Fig.2 に示す。ここで 4 層構造の NM 膜厚は各 FI 膜厚に対して MR 比が最大となる膜厚を選択した。4 層構造においては FI の膜厚の増加とともに MR 比の最大値が増加した。また、FI 膜厚が 1nm 以上では MR 比の最大値は従来の 3 層構造 FM/NI/FM の MR 比より大きくなることが明らかとなった。

以上の結果から、今回の 4 層構造で FI および NM の膜厚を適切に選択することで、従来の 3 層構造を超える MR 比が得られると結論付けることが出来る。さらに、NM の材料としてフェルミ波長の長い物質を用いることで、NM 膜厚のばらつきによる MR 比のばらつきを抑制できると考えられる。

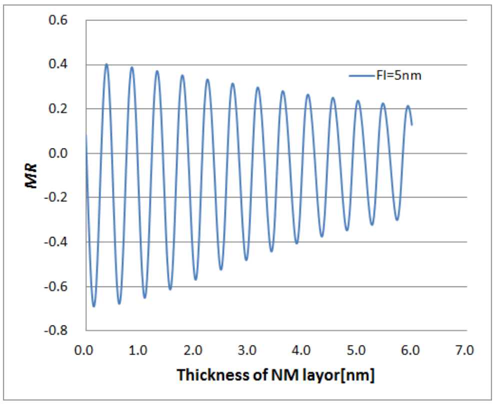

Fig.1 MR ratio vs NM thickness

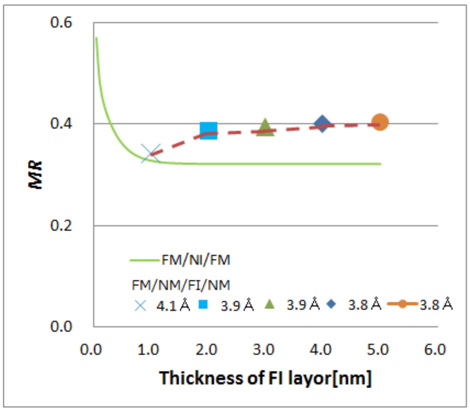

Fig.2 MR ratio vs FI thickness calculated for various NM thickness