

# NIMS WEEK 2018

## Abstracts

**Day 1**

**NIMS Award 2018**

**Academic Symposium**

— Magnetic Materials Innovations —

**October 15, 2018 Tokyo International Forum, Japan**

Organized by National Institute for Materials Science (NIMS)

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MEGA EVOLUTION of MATERIALS

NIMS Award Symposium

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## Greetings from the President



The National Institute for Materials Science (NIMS) is one of the three designated national research and development agencies in Japan. One of the missions of NIMS is to carry out fundamental research on materials science and nanotechnology, thereby developing human resources in the area. As a core national institute, NIMS is also expected to contribute to strengthening Japan's industrial competitiveness toward the realization of Society 5.0 (Super Smart Society), proposed in the 5th Science and Technology Basic Plan of Japan.

In order to meet the expectation, NIMS encourages its researchers to carry out fundamental research based on their diverse and challenging ideas in a world-class research environment. Moreover, NIMS intends to play a major role as a platform for industry-academia collaboration, where "human resources", "funds", and "knowledge and technology" are exchangeable between academia and industry. NIMS attempts to strengthen Japan's competitiveness in materials industry through the "Materials Open Platform (MOP)", which is an open innovation research system for the horizontal collaborations among competing companies in the same industrial area.

The NIMS WEEK is the largest and most important event that NIMS organizes every year. It is comprised of an academic symposium to honor the NIMS Award winners, a research showcase and an open house. NIMS Award is an international award, which is given to distinguished researchers who have made innovative research in materials science. This year, NIMS wishes to honor distinguished achievements in the field of "magnetic and spintronic materials" by selecting the following two winners who have made tremendous contributions to the creation of a new industry through innovations in magnetic materials: Dr. Masato Sagawa (Daido Steel Co. Ltd., Adviser) and Dr. Terunobu Miyazaki (Tohoku University, Professor Emeritus). The award ceremony and their award-winning lectures will be held on the first day of the NIMS WEEK. To explain the impact of the winners' innovations, world-renowned scientists in the relevant technical field will also give overview talks on the current status of the science and technologies in permanent magnets, magnetic recording and magnetoresistive memories. The detailed citations for the NIMS Award winners are described in the abstract booklet. In addition, we have scheduled a poster session for young researchers who will be presenting their research results in the award winners' relevant area.

I would like you to see the front line of evolving materials research throughout this NIMS WEEK.

A handwritten signature in black ink, reading "K. Hashimoto". The signature is fluid and stylized, with a long horizontal stroke extending to the right.

Kazuhito Hashimoto

President, National Institute for Materials Science (NIMS)

## Aim and Scope

Electric vehicles (EV) are rapidly replacing existing cars, hard disk drives (HDD) can store big data in the IoT society, and magnetoresistive random access memories (MRAM) are being used as nonvolatile memory for computing. These applications were made possible by the innovation of magnetic materials. NIMS will present the 2018 NIMS AWARD to two material scientists who have made epoch-making inventions in magnetic and spintronic materials. Dr. Masato Sagawa invented the world's strongest neodymium magnet that is now used for motors for EVs and HDDs, and Dr. Terunobu Miyazaki invented the room temperature tunneling magneto-resistance device that led to real spintronic applications such as read sensors in HDDs and memory cells in MRAMs. At the 2018 NIMS AWARD lecture, the award recipients will present overview lectures on how each material invention was made and how it was developed for practical use. Following the awards lectures, other distinguished scientists in relevant areas will give lectures on recent advances in permanent magnets, magnetic data storage, and magnetoresistive random access memory (MRAM). This academic symposium will provide an overview of the current status and future prospects of the state-of-the-art material research in magnetic and spintronics applications. Everyone is welcome to participate in the 2018 NIMS AWARD Lectures and Symposium. All the lectures will be presented in both Japanese and English through simultaneous interpretation and is intended for participants with various backgrounds in science and engineering.

# Program

- 10:00~10:05 Opening Address by **Dr. Kazuhito Hashimoto** (President)
- 10:05~10:25 Lecture by **Dr. Kazuhito Hashimoto** (President)  
"Curiosity driven research and organization mission research at NIMS"
- 10:25~10:40 NIMS Award Ceremony (Chair: Dr. Hono)
- 10:40~11:25 [NIMS Award Winning Lecture 1] (Chair: Dr. Hono)  
**Dr. Masato Sagawa** (Adviser, Daido Steel Co., Ltd.)  
"Developments of R-Fe-X Permanent Magnets (X=B,C,N)"
- 11:25~12:10 [NIMS Award Winning Lecture 2] (Chair: Dr. Mitani)  
**Prof. Terunobu Miyazaki** (Professor Emeritus, Tohoku University)  
"Development of Spintronics -focusing on my personal experience-"
- 12:10~13:30 **Poster Session / Lunch time**

## Forefront of Permanent Magnetic Materials (Chair: Dr. Hirohata)

- 13:30~14:00 [NIMS Talk 1]  
**Dr. Kazuhiro Hono** (Director, Research Center for Magnetic and Spintronic Materials, NIMS)  
"Permanent magnet research for the era of mass usage"
- 14:00~14:30 [Invited Talk 1]  
**Prof. Oliver Gutfleisch** (Technical University of Darmstadt)  
"Towards deciphering the DNA of hysteresis in magnetic materials"

## Next-generation of Magnetic Recording and Data Storage (Chair: Dr. Takahashi)

- 14:30~15:00 [Invited Talk 2]  
**Prof. Jimmy Zhu** (Director, Data Storage Systems Center, Carnegie Mellon University)  
"Microwave assisted magnetic recording for next generation of hard disk drives"
- 15:00~15:30 [Invited Talk 3]  
**Dr. Jan-Ulrich Thiele** (Seagate Technology)  
"Heat Assisted Magnetic Recording (HAMR) - next generation hard disk drive technology -"
- 15:30~15:45 [NIMS Talk 2]  
**Dr. Yuya Sakuraba** (Group Leader, Magnetic Materials Group, NIMS)  
"Development of advanced materials for high density magnetic recording"
- 15:45~16:30 **Poster Session / Coffee Break**

## Forefront of MRAM Research (Chair: Dr. Uchida)

- 16:30~17:00 [Invited Talk 4]  
**Dr. Shinji Yuasa** (Director Spintronics Research Center, AIST)  
"Giant tunnel magnetoresistance effect in MgO-based magnetic tunnel junctions and its device applications"
- 17:00~17:30 [Invited Talk 5]  
**Dr. Daniel Worledge** (Distinguished Research Staff Member, Senior Manager,  
MRAM & PCM, IBM Almaden Research Center)  
"Spin-Transfer-Torque MRAM"
- 17:30~17:45 [NIMS Talk 3]  
**Dr. Seiji Mitani** (Group Leader, Spintronic Materials Group, NIMS)  
"Development of materials for next generation MRAM"
- 17:45~18:00 Poster Award Ceremony and  
Closing by **Dr. Kazuhiro Hono** (Executive Vice President)

## Poster Presentation List

### I. Permanent Magnet and other Magnetic Materials (I-01~I-38)

Presen No.	Title	First name	Last name	Affiliation	Presentation Title
I-01 (Invited)	Mr.	Yasuhiro	Une	Daido Steel Co., Ltd.	Development of the Highest Performance Dy-free Nd-Fe-B sintered Magnet
I-02 (Invited)	Dr.	Keiko	Hioki	Daido Corporate Research & Development Center, Daido Steel Co., Ltd.	Heavy-Rare-Earth-Free Hot-deformed Nd-Fe-B Magnets for Traction Motors
I-03	Dr.	Taisuke	Sasaki	NIMS	Development of ultimate Nd-Fe-B based permanent magnet by grain boundary and interface engineering
I-04	Dr.	Hossein	Sepehri-Amin	NIMS	Development of high coercivity Nd-Fe-B based permanent magnets with improved thermal stability by microstructure modifications
I-05	Prof.	Toshiyuki	Koyama	Nagoya University	Discussion on Nd composition in grain boundary phase based on phase-field approaches – What determines the composition of grain boundary phase ? –
I-06	Mr.	Daiki	Kato	Graduate School of Engineering, Nagoya University	Phase-field simulation of Dy-shell formation in Nd-Fe-B hard magnets
I-07	Dr.	Xin	Tang	NIMS	Development of high performance Ce-substituted hot-deformed Nd-Fe-B magnets
I-08	Dr.	Takafumi	Hawai	High energy accelerator research organization (KEK)	Neutron powder diffraction on rare earth permanent magnets (Nd <sub>1-y</sub> Ce <sub>x</sub> La <sub>y</sub> ) <sub>2</sub> Fe <sub>14</sub> B
I-09	Dr.	Takafumi	Hawai	High energy accelerator research organization (KEK)	Inelastic neutron scattering study for RE <sub>2</sub> Fe <sub>14</sub> B and REFe <sub>11</sub> Ti (RE = Y, Nd)
I-10	Dr.	Hiroyuki	Okazaki	National Institutes for Quantum and Radiological Science and Technology(QST)	Phase analysis of Nd-Fe-B sintered magnet during heating process studied by X-ray diffraction
I-11	Dr.	Ikuro	Ohnuma	NIMS	Heat Capacity Measurement of (Nd <sub>x</sub> Dy <sub>1-x</sub> ) <sub>2</sub> Fe <sub>14</sub> B alloys
I-12	Dr.	Takuya	Yoshioka	Department of Applied Physics, Tohoku University	Theoretical Revisit of the Magnetic Properties in the 2-14-1 Type Compounds
I-13	Assoc. Prof.	Yasutomi	Tatetsu	Meio University	Role of typical elements in Nd <sub>2</sub> Fe <sub>14</sub> X (X = B, C, N, O, F)
I-14	Dr.	Munehisa	Matsumoto	Institute for Solid State Physics, The University of Tokyo	<i>Ab initio</i> trail toward the champion magnet Nd <sub>2</sub> Fe <sub>14</sub> B
I-15	Assoc. Prof.	Yasutomi	Tatetsu	Meio University	First-principles study on searching for the stable subphase structures in Ga-doped Nd-Fe-B sintered magnets
I-16	Dr.	Asako	Terasawa	Tokyo Institute of Technology	First-principles structural and magnetic analyses of amorphous Nd-Fe alloys
I-17	Prof.	Satoshi	Okamoto	Tohoku University	Detection of single magnetization reversal events in Nd-Fe-B hot-deformed magnet
I-18	Dr.	Kentaro	Toyoki	Japan Synchrotron Radiation Research Institute (JASRI)	Applications of Soft X-ray Magnetic Circular Dichroism Spectromicroscope to Analyze Local Magnetism

Presen No.	Title	First name	Last name	Affiliation	Presentation Title
I-19	Prof.	Satoshi	Okamoto	Tohoku University	Study on magnetization reversal process of a Ga-doped Nd-Fe-B sintered magnet using FORC analysis
I-20	Dr.	Motohiro	Suzuki	Japan Synchrotron Radiation Research Institute (JASRI)	Three-Dimensional Visualization of Magnetic Domain Structure Using Scanning Hard X-ray Microtomography
I-21	Dr.	Yuta	Toga	NIMS	Free energy landscape of magnetization reversal in permanent magnets
I-22	Dr.	Masamichi	Nishino	NIMS	Dynamical features of an atomistic model for Nd-Fe-B permanent magnets
I-23	Dr.	Shotaro	Doi	Institute for Solid State Physics (ISSP), The University of Tokyo	First-principles based calculation of magnetic properties at finite temperature
I-24	Mr.	Fumiya	Saito	Department of Applied Physics, Tohoku University	Theoretical study of Gilbert damping in Rare-Earth Permanent Magnets
I-25	Dr.	Ruma	Mandal	NIMS	Magnetization Dynamics Study of High PMA thin films by Time-resolved MOKE Microscopy
I-26	Dr.	Taichi	Hinokihara	The University of Tokyo	Constructing Macro Spin Model from Atomistic Spin Model
I-27	Dr.	Takashi	Miyake	AIST	First-principles study of chemical substitution effects in NdFe <sub>12</sub> and NdFe <sub>12</sub> N
I-28	Assoc. Prof.	Hiroki	Tsuchiura	Tohoku University	Theoretical Study on Finite-Temperature Magnetic Properties in Sm(Fe <sub>1-x</sub> M <sub>x</sub> ) systems
I-29	Dr.	Arkapol	Saengdeejing	Tohoku University	Thermodynamic, Electronic and Dynamic Stability Study of Potential New Permanent Magnet from First-principles Calculations
I-30	Prof.	Trinh	Thang Thuy	Institute for Chemical Research, Kyoto University	Crystallographic and Micromagnetic Properties of Sm(Fe <sub>0.8</sub> Co <sub>0.2</sub> ) <sub>12-x</sub> Ti <sub>x</sub> Micropowders
I-31	Dr.	Jungryang	Kim	Institute for Chemical Research of Kyoto University	Chemical synthesis and magnetic properties of Sm(Fe <sub>1-x</sub> Co <sub>x</sub> ) <sub>12-y</sub> Ti <sub>y</sub> particles
I-32	Dr.	Daisuke	Ogawa	NIMS	Microstructure and magnetic properties of highly textured Sm(Fe <sub>0.8</sub> Co <sub>0.2</sub> ) <sub>12</sub> thin films with ThMn <sub>12</sub> structure
I-33	Dr.	Imants	Dirba	NIMS	SmFe <sub>12</sub> -based permanent magnets, intrinsic magnetic properties and toward development of bulk high-performance permanent magnet
I-34	Dr.	Taro	Fukazawa	AIST	Search for magnet compounds using Bayesian optimization
I-35	Dr.	Masashi	Matsuura	Tohoku University	Preparation of high performance Sm-Fe-N bulk magnets
I-36 (Invited)	Mr.	Junichi	Nagaoka	TDK Corporation	High-Performance Ferrite Magnets without Irreversible Low Temperature Demagnetization
I-37	Dr.	Keita	Ito	Tohoku University	Formation of L1 <sub>0</sub> -ordered FeNi Films by Extracting Nitrogen from FeNiN Films



Presen No.	Title	First name	Last name	Affiliation	Presentation Title
I-38	Mr.	Seiji	Yoshikawa	Department of Physics, The University of Tokyo	Crystal structure simulation by assimilating incomplete powder diffraction data: First application to magnetic material

## II. Materials for Magnetic Recording and Data Storage (II-01~II-09)

Presen No.	Title	First name	Last name	Affiliation	Presentation Title
II-01	Dr.	Zhenchao	Wen	Tohoku University	Large CPP-GMR effect in dual-spacer $\text{Co}_2\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si}/\text{AgMg}$ nanojunctions
II-02	Dr.	Takahide	Kubota	Tohoku University	Heusler Alloy-based CPP-GMR Junctions with an $\text{L}_{12}$ - $\text{Ag}_3\text{Mg}$ Ordered Alloy Spacer
II-03	Dr.	Yukiko K.	Takahashi	NIMS	Development of FePt-based granular film for HAMR media
II-04	Dr.	Jian	Wang	NIMS, ICYS	Next generation magnetic recording technology -- FePt based heat assisted magnetic recording
II-05	Prof.	Satoshi	Okamoto	Tohoku University	Microwave-assisted switching behavior of CoCrPt based granular media
II-06	Dr.	Ippei	Suzuki	NIMS	Modification of grain density in the FePt-C granular films
II-07	Dr.	Hossein	Sepehri-Amin	NIMS	Development of spin-torque-oscillator for microwave-assisted-magnetic-recording
II-08	Dr.	Hiroo	Tajiri	Japan Synchrotron Radiation Research Institute	Evaluation of Structural Order in Magnetic Films by Anomalous X-ray Diffraction using Synchrotron X-rays
II-09	Dr.	Weinan	Zhou	NIMS	Inducing out-of-plane precession of magnetization for microwave assisted magnetic recording using an oscillating spin polarizer in spin torque oscillator

## III. Spintronic Materials for Memory (III-01~III-31)

Presen No.	Title	First name	Last name	Affiliation	Presentation Title
III-01	Ms.	Altansargai	Buyandalai	Toshiba Corporation	Reduction of write current with ultra-thin Spin-Hall-effect electrode in Voltage-Control Spintronics Memory (VoCSM)
III-02	Dr.	Tatsuya	Yamamoto	AIST	Thermally Induced Write Errors in Voltage-Driven Magnetization Switching
III-03	Dr.	Hiroshi	Imamura	AIST	Pulse shape control of write error rate of a voltage-torque MRAM
III-04	Dr.	Takayuki	Nozaki	AIST	Materials engineering for voltage-controlled magnetic anisotropy effect
III-05	Dr.	Qingyi	Xiang	NIMS	Voltage controlled perpendicular magnetic anisotropy in quantum wells

Presen No.	Title	First name	Last name	Affiliation	Presentation Title
III-06	Mr.	Yuki	Iida	NIMS	Perpendicular magnetic anisotropy in W-doped epitaxial Fe/MgO heterostructures
III-07	Mr.	Kazuma	Kunimatsu	Tohoku University	Analysis of lattice distortion of $\angle 10^\circ$ -MnGa ultrathin films grown on CoGa
III-08	Dr.	Muftah	Al-Mahdawi	Center for Spintronics Research Network, Tohoku University	Tunneling anisotropic magnetoresistance (TAMR) in spin-dependent Fe quantum well
III-09	Dr.	Butsurin	Jinnai	Tohoku University	Spin-orbit torque switching in in-plane nanomagnets characterized by planar Hall effect
III-10	Dr.	Butsurin	Jinnai	Tohoku University	Spin-orbit torque switching in perpendicular-magnetized Co/Pt multilayers
III-11	Dr.	Lakhan	Bainsla	AIMR, Tohoku University	Equiatomic quaternary CoFeMnSi Heusler alloy for spintronic applications
III-12	Dr.	Kazuya	Suzuki	WPI-AIMR, Tohoku University	Development of Mn-based nano-layer films toward spintronic applications
III-13	Mr.	Shintaro	Kon	Spintronics Research Center, AIST Chiba Institute of Technology	Fabrication and Magnetoresistance of Fully Epitaxial Magnetic Tunnel Junctions with a Rock-Salt Type ZnO/MgO bilayer tunnel barrier
III-14	Dr.	Tomoki	Tsuchiya	Tohoku University	Tunnel magnetoresistance of equiatomic quaternary CoFeCrAl Heusler alloy films
III-15	Prof.	Shoji	Ikeda	Tohoku University	High Performance CoFeB-MgO Magnetic Tunnel Junctions for Nonvolatile VLSI
III-16	Dr.	Thomas	Scheike	NIMS	Perpendicular Magnetic Anisotropy in Epitaxial $\text{Fe}_{80}\text{Al}_{20}/\text{MgAl}_2\text{O}_4$ Heterostructures
III-17 (Invited)	Mr.	Katsuyuki	Nakada	TDK Corporation	Reactive deposition technology for a uniform spinel $\text{MgAl}_2\text{O}_4$ barrier toward high quality magnetic tunnel junctions
III-18	Dr.	Kay	Yakushiji	AIST	Fabrication of an $\text{MgAl}_2\text{O}_4$ -based Fully-Epitaxial Magnetic Tunnel Junction on an 8" Si Wafer
III-19	Dr.	Hitoshi	Kubota	AIST	Magneto-static coupling in magnetic tunnel junctions
III-20	Prof.	Yoshishige	Suzuki	Graduate School of Engineering Science, Osaka University	X-ray absorption spectroscopy for voltage-controlled magnetic anisotropy effect
III-21	Dr.	Tomohiro	Nozaki	AIST	Manipulation of antiferromagnetic spin using tunable parasitic magnetization in doped $\text{Cr}_2\text{O}_3$ film
III-22	Dr.	Yu	Shiratsuchi	Osaka University	Magnetoelectric control of antiferromagnetic domain state
III-23	Dr.	Songtian	Li	National Institutes for Quantum and Radiological Science and Technology, QST	Preparation and characterization of a novel heterostructure of graphene/ $\text{Co}_2\text{Fe}(\text{Ge}_{0.5}\text{Ga}_{0.5})$ for high-performance spintronic device application
III-24	Dr.	Shinya	Kasai	NIMS	Magnetic tunnel junctions with semiconductor $\text{Cu}(\text{In,Ga})\text{Se}_2$ spacers

Presen No.	Title	First name	Last name	Affiliation	Presentation Title
III-25	Dr.	Keisuke	Masuda	NIMS	Theoretical study for tunnel magnetoresistance and magnetic anisotropy in magnetic tunnel junctions with semiconductor $\text{CuIn}_{1-x}\text{Ga}_x\text{Se}_2$ spacers
III-26	Dr.	Yoshio	Miura	NIMS	A first-principles study on perpendicular magnetic anisotropy in W-inserted Fe/MgO and Fe/MgAl <sub>2</sub> O <sub>4</sub> interfaces
III-27	Dr.	Masahito	Tsujikawa	Research Institute of Electrical Communication, Tohoku University	Huge voltage-controlled magnetic anisotropy effect of monoatomic 5d-transition-metal layer inserted into MgO/Fe interface
III-28	Dr.	Shutaro	Karube	Tohoku University	Anomalous field-like torque on Py layer via Edelstein effect at W/Pt interface
III-29	Mr.	Ryoto	Enoki	Tohoku University	Observation of strong spin orbit interaction in Cu by doping nitrogen impurities
III-30	Dr.	Yong Chang	Lau	Department of Physics, The University of Tokyo NIMS(Guest researcher)	Spin Hall effect from hybridized 3d-4p orbitals
III-31	Prof.	Ko	Mibu	Nagoya Institute of Technology	Electronic Tunneling through Epitaxial $\text{Co}_x\text{Fe}_{3-x}\text{O}_{4+\delta}$ (001) films with Perpendicular Magnetic Anisotropy

#### IV. Others (IV-01~IV09)

Presen No.	Title	First name	Last name	Affiliation	Presentation Title
IV-01	Dr.	Tomohiro	Taniguchi	AIST	Spin-current induced synchronization between spin-Hall nano-oscillators
IV-02	Prof.	Yoshishige	Suzuki	Graduate school of Engineering Science, Osaka University	Radio-frequency amplification induced by Joule heating induced magnetization dynamics
IV-03	Dr.	Sumito	Tsunegi	AIST	Large emission power in vortex spin torque oscillator
IV-04	Dr.	Tomohiro	Ichinose	AIMR, Tohoku University	Tunnel magnetoresistance properties of magnetic tunnel junctions sputtered with the off-axis cathode
IV-05	Mr.	Hiroshige	Onoda	Department of Applied Physics, University of Tsukuba	Tuning of magnetic anisotropy in cobalt ferrite thin films by epitaxial strain
IV-06	Prof.	Jun	Okabayashi	The University of Tokyo	Microscopic Origin of Interfacial Perpendicular Magnetic Anisotropy Studied by X-ray Magnetic Circular Dichroism
IV-07	Dr.	Shinji	Isogami	NIMS	Potential spintronic devices based on anti-perovskite transition metal nitrides
IV-08	Dr.	Kosuke	Fujiwara	Department of Applied Physics, Tohoku University	Room Temperature Measurement of Bio-Magnetic Field using Tunnel Magneto-Resistance Sensor
IV-09	Dr.	Yusuke	Kozuka	NIMS	Controlling the all-in-all-out magnetic domains in pyrochlore iridate thin films and heterostructures



# NIMS Award



# NIMS Award Winner 1

## Masato Sagawa (Dr.)

Adviser, Daido Steel Co., Ltd.

[Date of Birth] June 15th, 1943 (Age: 75)

[Research Field] Permanent magnet materials



### History

1966	B.S. in Electric Engineering from Kobe University
1968	M.S. in Electric Engineering from Kobe University
1972	Doctoral degree in materials engineering from Tohoku University
1972 - 1982	Fujitsu Limited
1982 - 1988	Sumitomo Special Metals (SSM)
1988	Intermetallics Co.
2013	NDFEB Co.
2016	Adviser, Daido Steel Co., Ltd.

### Awards

1984	Osaka Science Prize
1986	AIP, International Prize for New Materials
1988	The Japan Institute of Metals and Materials Award
1990	Asahi Prize
1991	MSJ Society Award
1993	Okochi Memorial Prize
1998	Acta Metallurgica J. Hollomon Award
2003	Honda Memorial Award
2003	American Physical Society Prize
2006	Kato Memorial Award
2012	Japan Prize
2018	NIMS Award

## Research Achievement Title

Invention and practical application of neodymium magnets

## Research Summary

Dr. Sagawa invented neodymium (Nd) magnets—the world’s strongest magnets—and led the industrialization of anisotropic neodymium magnets through a powder metallurgy process. The use of these magnets enabled the development of various electronic devices, such as hard disk drives, hybrid and electric vehicles. Dr. Sagawa independently discovered Nd-Fe-B-based magnetic alloys as a promising permanent magnet material. Immediately after joining Sumitomo Special Metals Inc., Dr. Sagawa developed the world’s strongest Nd-Fe-B-based magnet. He then applied for a patent under the name of Sumitomo Special Metals in 1982 and published the approved patent in 1983. The development of Nd sintered magnets was revolutionary not only because their magnetic fields were stronger than those of the previously strongest Sm-Co-based permanent magnets, but also because they are composed mainly of Fe, which is abundant in nature, and Nd, which is a relatively abundant rare-earth element. The discovery of neodymium magnets was also groundbreaking because it defied the common belief that cobalt is a requisite ingredient in permanent magnets and completely resolved the issue of the limited availability of Co resources. Nd magnets remain the world’s strongest 35 years after their invention, and their applications, which now include electric vehicles and robotics, continue to grow.

## Impact on the Academic and Industrial Sectors

Dr. Sagawa not only invented the anisotropic neodymium magnet, but also promoted basic research on the neodymium magnets by supplying samples to academic researchers around the world. This effort greatly contributed to advances in materials science of permanent magnets. Sumitomo Special Metals began commercial production of the Nd-Fe-B sintered magnet product NEOMAX in 1985. Production of NEOMAX—the most powerful permanent magnet in the world which can be manufactured using inexpensive raw materials—rapidly increased as the range of its applications expanded. Approximately 70,000 tons of NEOMAX were estimated to have been produced worldwide in 2015. Used in hard disk spindle motors and head actuators, neodymium magnets have become indispensable components of modern data storage technologies. More recently, Nd magnets have been used in the motors and generators of hybrid and electric vehicles, wind power generation, energy-efficient air conditioners and the drive units of robots. The use of these magnets for these purposes is expected to continue growing. Thus, society has benefited immensely from the neodymium magnets. For this contribution to the society, Dr. Sagawa received many prestigious awards like Osaka Science Award, James C. McGroddy Prize for New Materials, Asahi Prize, Honda Memorial Award and Japan Prize.



## Development of the R-Fe-X permanent magnets

**Masato Sagawa**

Advisor, Daido Steel Co. Ltd.

### Abstract

Industrialized R-Fe-X (R=Rare earth and X=3<sup>rd</sup> element) magnets are Nd-Fe-B magnets and Sm-Fe-N magnets. Either magnet was started to research from an idea of a beginner in the field of permanent magnets, industrialized fairly soon after the invention, and followed by overwhelming developments of basic researches. It proved that either magnet was not the one that should have been derived from the existing knowledge and technologies at the time of invention. The both magnets surely nucleated a new research field of R-Fe-X magnets.

Now the basic researches on R-Fe-X magnets have been carried out successfully by the project ESICMM (Element Strategy Initiative Center for Magnetic Materials). The development of the basic researches enhances the overall technical ability of the society and cultivates human resources. We can expect another nucleation of research field someday. However, it is impossible to predict when and what kind of nucleation may occur.

My thought is that administration of research activities should not require the basic research scientist an exit to application. Such requirement causes only irresponsible result. Basic research itself is valuable for us all.



## NIMS Award Winner 2

### Terunobu Miyazaki (Prof.)

Professor Emeritus, Tohoku University

[Date of Birth] August 3rd, 1943 (Age: 75)

[Research Field] Spintronics, magnetic materials



#### History

1967	B.S. in Applied Physics, Graduate School of Engineering, Tohoku University
1969	M.S. in Applied Physics, Graduate School of Engineering, Tohoku University
1972	Dr. degree in Applied Physics, Tohoku University
1972 - 1973	Research associate at Tohoku University
1973 – 1975	Research associate and a Humboldt research fellowship at university Regensburg, West Germany
1975	Research associate at Tohoku University
1985	Associate Professor at Tohoku University
1991 – 2007	Professor at Tohoku University
2007 – 2013	Professor at WPI Advanced Institute for Materials Research Tohoku University(WPI-AIMR)
2013 - 2016	Advisor at WPI-AIMR, Advisor of Impact project (JST) and Scientific Research on Innovative Area (JSPS)

#### Awards

2003	MSJ Society Award
2003	Yazaki Scientific Prize
2005	Yamazaki-Teiichi Prize
2006	Prize for Science and Technology by Minister of Education, Culture, Sports and Technology of Japan (MEXT)
2007	Fellow of Applied Physics Japan
2008	JSPS Outstanding Achievement Award
2008	Asahi Prize
2009	American Physical Society Oliver E. Buckley Condensed Matter Physics Prize
2018	NIMS Award

## Research Achievement Title

Development of tunneling magnetoresistance devices capable of generating giant magnetoresistance at room temperature and application thereof to spintronics devices.

## Research Summary

A magnetic tunnel junction (MTJ) is a tri-layer structure composed of two ferromagnetic layers sandwiching an insulating layer. Tunnel magnetoresistance in MTJ changes in response to differences in magnetic orientation between the two ferromagnetic layers, i.e., the tunneling magnetoresistance (TMR) effect. Although the TMR effect was first reported as early as the 1960s, observations prior to Dr. Miyazaki's pioneering research on TMR devices at Tohoku University were limited to the detection of subtle TMR at extremely low temperatures. In 1995, Dr. Miyazaki observed a giant TMR effect at room temperature for the first time using tri-layer (Fe/alumina/Fe) magnetic tunnel junction. This success triggered the interest in the practical application of TMR devices. After demonstrating the successful operation of TMR device at room temperature, Dr. Miyazaki continued his research by collaborating with worldwide academic and industrial researchers. His contributions were crucial in the development of spintronics. TMR is now used as read sensors of high capacity hard disk drives, magnetic random access memories and other magnetic sensors. Dr. Miyazaki's greatly contributed to society by advancing the applications of TMR devices in data storage, nonvolatile magnetic memories and magnetic sensors.

## Impact on the Academic and Industrial Sectors

Dr. Miyazaki's observation of a magnetoresistance effect at room temperature using TMR devices had a profound impact on the academic and industrial sectors. TMR devices were put into practical use in high-sensitivity read heads for hard disk drives (HDDs), which were commercialized in 2001 and still in use today. The improved performance of TMR read heads enabled an increase in HDD recording density, from tens of Gbit/in<sup>2</sup> before 2001 to 1 Tbit/in<sup>2</sup> today, greatly increasing the storage capacity of HDDs, which now serve as vital infrastructure for our advanced information society. TMR elements are also used as memory cells in MRAM devices, a promising candidate for next-generation non-volatile memory. The global R&D effort to develop these technologies has its roots in Dr. Miyazaki's discovery. The room temperature TMR effect has been applied in a wide variety of engineering and scientific research projects, such as exploration of high spin polarization materials and magnetically anisotropic materials and spin torque generated by spin transport. These research projects were highly valued by academic and industrial groups and won many awards, including the Asahi Prize, the JSAP (Japan Society of Applied Physics) Award and the American Physical Society Award. As a longtime faculty member at Tohoku University, Dr. Miyazaki has educated many students and young researchers in the magnetism and spintronics field. He has contributed greatly to maintaining Japan's high research standards in this field.

## Development of Spintronics - focusing on my personal experience -

Terunobu Miyazaki

Professor Emeritus, Tohoku University

### Abstract

As is well known, the discovery of giant magnetoresistance (GMR) and large tunnel magnetoresistance (TMR) effects became a trigger for the research field called “spintronics”. These discoveries were reported in 1988 and 1995, respectively. Therefore, I look at first **(1) the activities of magnetic thin film research between 1980 and 1990**, just before these findings. Here I show two review articles of the Magnetism Society of Japan in 1982. The research topics carried out at that time will be compared with those of this year. Also I show some examples of International Conference Talk which are closely related to the study of magnetoresistance. Then I show **(2) the research subject titles related to magnetoresistance in our group between 1987 and 1993**. Here, I show one example discussed with bachelor student just before the start of the tunnel magnetoresistance research. After finding a large TMR effect, although our paper was not yet published in journal, I talked our experimental results at several symposium and/or workshop held inside and outside Japan. Our paper was accepted to the journal of Magnetism and Magnetic Materials in 1995 march. The project of magnetoresistive random access memory (MRAM) started within this year at IBM and Motorola. I will summarize the reports related to the tunnel magnetoresistance research in an early stage. Meanwhile, it shows a strong tendency to develop MRAM inside Japan. **(3) Several planning and/or specialized committees were set up in Research & Development Association for Future Electron Devices (FED)**. I will introduce several activities carried out through FED. Although it took a long time to set up national MRAM project, the effort paid at that time contributed to setting up several projects related MRAM in Japan. Next **(4) activities at the Japan Society of Applied Physics (JSAP)** will be explained. Before 2000, we discussed mostly the spintronics research at Magnetic Society of Japan (MSJ). Suzuki san (Osaka University) proposed to shift the meeting society from MSJ to JSAP, where there are so many semiconductor researchers historically, since to develop MRAM the technologies and related knowledge of semiconductor are indispensable. I will show briefly the activities done at JSAP. Of course, the development of spintronics research field depends on excellent and outstanding findings reported by many researchers. **(5) Here, I will show personally several remarkable contributions and outputs of spintronics**. Finally, **(6) I will give my opinion ( “tsubuyaki”) on scientific research in general**.



# Oral Presentations

## Permanent magnet research for the era of mass usage

**Kazuhiro Hono**

Elements Strategy Initiative Center for Magnetic Materials (ESICMM)  
National Institute for Materials Science (NIMS), Tsukuba, Japan

### Abstract

Since the rare earth crisis in 2010, there has been worldwide interest to attain high coercivity in Nd-Fe-B magnets without using heavy rare earth elements. Although the supply of rare earth elements has been stabilized recently, the increasing demands of high performance permanent magnets for emerging applications such as vehicle electrification, robotics, electric airplanes and wind turbines require long term research on the development of high performance magnets without relying on critical elements. In this talk, we will present an overview on our recent advances in Dy-free or Dy-saving Nd-Fe-B magnets that were carried out at NIMS in collaboration with industrial collaborators. To obtain better understandings of the microstructure-coercivity relationships, we have carried out thorough microstructure investigations of experimental magnets that were processed with different routes and chemical compositions using aberration-corrected STEM, atom probe tomography (APT), magneto-optical Kerr microscopy and finite element micromagnetic simulations. Based on new experimental findings on microstructure-property relationships, we propose a method to increase the coercivity of Nd-Fe-B magnets while maintaining high remanence. Lastly, we will discuss the possibility of industrially viable high-performance magnets other than the Nd-Fe-B system, *i.e.*, Sm(Fe,Co)<sub>12</sub> based permanent magnets based on our recent experimental investigations.

This talk is based on our collaborative researches with industrial partners including TOYOTA, Toyota Central Research Lab., Daido Steel Co. Ltd., and YSM under JST's Collaborative Research Based on Industrial Demand.



**Kazuhiro Hono** is a fellow and the director of the Research Center for Magnetic and Spintronic Materials at the National Institute for Materials Science (NIMS), Japan, and is a professor in materials science and engineering at the University of Tsukuba, Japan. He received his BS and MS degrees in materials science from Tohoku University, Japan, and his PhD degree in metals science and engineering from The Pennsylvania State University in 1988. After completing postdoctoral research at Carnegie Mellon University, and as a research associate at the Institute for Materials Research, Tohoku University, he moved to NIMS as a staff scientist in 1995. His research interests include microstructure-property relationships of metallic materials, in particular, magnetic and spintronic materials and devices. Hono can be reached by email at [kazuhiro.hono@nims.go.jp](mailto:kazuhiro.hono@nims.go.jp).



## Towards deciphering the DNA of hysteresis in magnetic materials

**Prof. Oliver Gutfleisch**

Professor, Technical University of Darmstadt, Material Science, Germany

### Abstract

Magnets are key components of energy-related technologies, such as direct drive wind turbines and e-mobility. They are also important in robotics and automation, sensors, actuators, and information technology. The magnetocaloric effect (MCE) is of strong interest for new and disruptive solid state-based refrigeration. Magnetic hysteresis – and its inherent energy product – characterises the performance of all magnetic materials. Despite considerable progress in the modelling, characterisation and synthesis of magnetic materials, hysteresis is a long-studied phenomenon that is still far from being completely understood. Discrepancies between intrinsic and extrinsic magnetic properties remain an open challenge and magnets do not operate yet at their physical limits. The design of hysteresis for the magnets for the above applications requires an expanded detailed knowledge on different length scales. Ultimately, new strategies for effective magnetic hardening mechanisms of permanent magnets resisting high external magnetic fields and temperatures and for strong thermomagnetic responses in low fields of magnetocaloric materials will need to be derived.

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**Prof. Oliver Gutfleisch** is a full Professor (W3) for Functional Materials at TU Darmstadt and a scientific manager at Fraunhofer IWKS Materials Recycling and Resource Strategies. He studied Material Science at TU Berlin, did his PhD in Birmingham, UK, and was a group leader at Leibniz Institute IFW Dresden. 2012 he joined TU Darmstadt. His scientific interests span from new permanent magnets for power applications to solid state energy efficient magnetic cooling, ferromagnetic shape memory alloys, magnetic nanoparticles for biomedical applications, and to solid state hydrogen storage materials with a particular emphasis on tailoring structural and chemical properties on the nanoscale. Resource efficiency on element, process and product levels as well as recycling of rare earth containing materials are also in the focus of his work.

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<http://www.mawi.tu-darmstadt.de/fm>

## Microwave assisted magnetic recording for next generation of hard disk drives

**Jian-Gang (Jimmy) Zhu**

ABB Professor of Engineering

Director, Data Storage Systems Center

Department of Electrical and Computer Engineering

Carnegie Mellon University

### Abstract

As conventional perpendicular magnetic recording technology approaches its physical limits, increasing area storage density capability of a hard disk drive becomes a challenge. Microwave assisted magnetic recording (MAMR) is one of the main stream alternative technologies for future hard disk drive applications, along with heat assisted magnetic recording (HAMR). In MAMR, a two-terminal spin torque oscillator is embedded in the write-gap of a current recording head, generating high frequency ( $\sim 30\text{GHz}$ ) ac magnetic field in the media with a dc current injection. The localized high frequency ac magnetic field can significantly enhance the writability and write field gradient, enabling the use of small grain high anisotropy media and substantial increase of the area density capability. This talk will cover the underlying physical mechanism of the recording in MAMR technology along with micromagnetic simulation-based modeling study. The use of dual-side spin torque oscillator and segmented media will be discussed.



**Dr. Jian-Gang (Jimmy) Zhu**, an IEEE Fellow, is the ABB Professor of Electrical and Computer Engineering and Director of the Data Storage Systems Center at Carnegie Mellon University. Dr. Zhu received his Ph.D. in Physics from University of California at San Diego in 1989. Prior coming to Carnegie Mellon in 1997, he had been an Assistant Professor and later an Associate Professor in the Department of Electrical Engineering at University of Minnesota from 1990-1996. Some of the awards that Dr. Zhu has received include the McKnight Land Grant Professorship from University of Minnesota in 1992, the NSF Presidential Young Investigator Award in 1993, the R&D Magazine Top 100 Invention Award in 1996, IEEE Magnetic Society Distinguished Lecturer in 2004, Carnegie Mellon University Outstanding Research Award in 2010, IEEE Magnetic Society Achievement Award, the highest award of the IEEE Magnetic Society, in 2011 and ETA KAPPA NU Excellent Teaching Award from Carnegie Mellon University in 2012. He has authored and co-authored over 300 refereed papers in major international journals along with eight book chapters and has given over 95 invited papers at various major international conferences. He holds 22 U.S. patents.

## Heat Assisted Magnetic Recording (HAMR) - next generation hard disk drive technology -

**Jan-Ulrich Thiele**

Seagate Technology, Fremont Research Center

### Abstract

Magnetic hard disk drives (HDDs) store over 90% of the world's digital data enabling the internet and economical access to data to power everything from social media to self-driving cars. Heat assisted magnetic recording (HAMR) is being developed as the next recording technology for HDDs. HAMR will bring profound changes to the HDD components and architecture. Critical components of this technology, such as the plasmonic near field transducer and high anisotropy granular FePt media, as well as recording demonstrations and fully integrated drives have been reported. One of the ongoing challenges of magnetic recording in general and HAMR in particular has been the demonstration of high linear density recording approaching the grain-size limit of the recording media, and a clear pathway to smaller grain sizes while maintaining good magnetic properties and distributions. In this presentation we will demonstrate the extensibility of FePt-based media, and using a head with high thermal gradient we will show a high linear recording density approaching the grain size limit of the media. Assuming continued progress in growth, microstructure and architecture of the media stack and the head architecture, and based on simplistic geometrical scaling, these demonstrations provide visibility out to the 3-4 Tb/in<sup>2</sup> areal density range, enabling high capacity HAMR HDDs that will serve the demand for future economical storage solutions for the world's ever growing data. As announced in a recent blog (<http://blog.seagate.com/intelligent/hamr-next-leap-forward-now/>), Seagate is now shipping HAMR units to select customers for integration tests, and will start shipping commercial HAMR products to key customers by the end of 2018, with full-scale mass production of industry leading high-capacity drives slated for 2020.



**Jan-Ulrich Thiele** holds a MSc in Physics from the University of Hannover in Germany and a PhD in Physics from the University of Basel in Switzerland. In 1996 he was awarded a Feodor-Lynen-Fellowship by the Alexander-von-Humboldt Foundation, and joined the IBM Almaden Research Center in San Jose as a Postdoctoral Researcher. After 12 years at IBM and Hitachi GST as a Research Staff Member and Research Manager working on many aspects of magnetic recording technology, he joined Seagate Technology in 2008 as a Managing Technologist (Senior Director) for Media R&D. Dr. Thiele currently leads a team of Scientists and Engineers developing Heat Assisted Magnetic Recording (HAMR) media technology. Significant milestones of this program include the first 1 Tb/in<sup>2</sup> recording demonstration in 2012, the demonstration of a fully functioning HAMR drive lasting for 1000 hours of continuous writing in 2014, and the first HAMR drives

shipped to an external customer for extensive real-world testing in 2016. The technology is currently being staged for the start of commercial production at the end of 2018. Dr. Thiele has authored or co-authored over 70 peer-reviewed publications and holds 24 US patents. He has served in a number of roles for the IEEE Magnetics Society, the Intermag conference series and the Advanced Storage Research Consortium (ASRC, formerly ASTC/INSIC/NSIC).

## Development of Advanced materials for high density magnetic recording -

Y. Sakuraba, T. Nakatani, Y. K. Takahashi, S. Kasai, H. Sepheri-Amin, T. Sasaki,  
T. Ohkubo and K. Hono

Reserch Center for Magnetic and Spintronics Materials, NIMS

### Abstract

For realizing HDD having higher areal recording density over 2Tbit/inch<sup>2</sup>, a breakthrough of advanced material and device is desired for next generation read head, energy assisted writing technologies such as microwave/heat assisted magnetic recording(MAMR and HAMR), and recording media. In our research center, we have been working on all these elemental technologies comprehensively.

Since present read head based on MgO-MTJ is expected to reach a limitation of usage for over 2Tbit/inch because of its large device resistance, current-perpendicular-to-plane giant magnetoresistive (CPP-GMR) devices having small device resistance is the most promising candidate for the alternative read head. We have been developing CPP-GMR using half-metallic Heusler to improve MR ratio in CPP-GMR for long years and achieved to obtain giant MR ratio over 50% in both fundamental epitaxial and practical poly-crystalline devices. Recently we demonstrated MR ratio of about 50% with RA of about 0.1  $\Omega\mu\text{m}^2$  in the poly-crystalline CPP-GMR devices with AgInZnO spacer, which satisfies the required performance for 5Tbit/inch as shown in Figure 1. For MAMR, it is essential to develop spin torque oscillator (STO) which is able to generate strong ac magnetic field with a small threshold electric current for excitation of magnetization oscillation. We also utilized highly spin-polarized Heusler as a spin injection layer of STO and successfully reduced the threshold current density for out-of-plane mode oscillation. To achieve 4 Tbit/in<sup>2</sup>, the development of the recording media is also important. We demonstrated the uniform microstructure with the grain size of 6 nm and high coercivity in FePt-C granular film in 2011. In this presentation, we will show our recent progress on the development of CPP-GMR for read head, STO for MAMR, and FePt-based HAMR media.

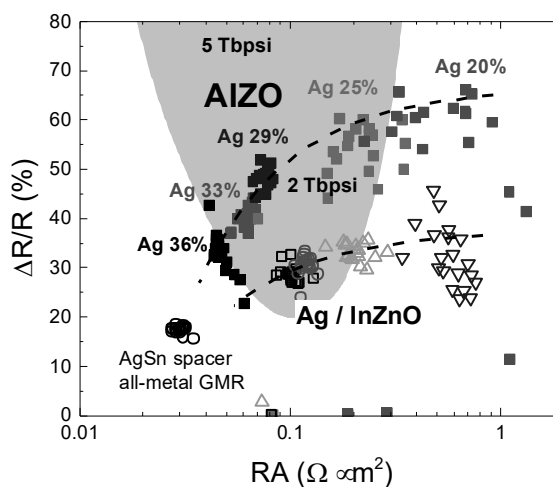


Figure 1 Required MR ratio and RA in next generation read head for 2 and 5Tbit/inch. Our recent data in poly-crystalline CPP-GMR with Heusler electrode is plotted.



**Yuya Sakuraba** is a group leader of the Magnetic Materials Group in Research Center for Magnetic and Spintronic Materials at the National Institute for Materials Science (NIMS), Japan. He received his BS and MS, PhD degrees in applied physics from Tohoku University, Japan from Prof. Terunobu Miyazaki. After finishing PhD course, he became an assistant professor at the Institute for Materials Research, Tohoku University, and he moved to NIMS as a senior researcher in 2013. From 2018, he became a group leader of magnetic materials group. Sakuraba can be reached by email at [Sakuraba.Yuya@nims.go.jp](mailto:Sakuraba.Yuya@nims.go.jp)

## Giant tunnel magnetoresistance effect in MgO-based magnetic tunnel junctions and its device applications

**Shinji Yuasa**, Kay Yakushiji, Takayuki Nozaki, Hitoshi Kubota, and Akio Fukushima  
National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan

### Abstract

This paper describes review and future prospects of basic technologies for spintronics and their applications to electronic devices. As shown in Fig. 1, the giant TMR effect in MgO-based magnetic tunnel junction (MTJ) [1,2] is the main stream technology for read heads of hard disk drives (HDDs) and magneto-resistive random-access-memory (MRAM) [3]. Concerning spin manipulation, spin-transfer torque (STT) [4-6] is the current main stream for device applications such as STT-MRAM [7,8], spin-torque oscillator (STO) [9,10], spin-torque diode (STD) [11] and physical random number generator named Spin Dice [12]. Moreover, novel voltage-induced torque [13-15] is expected to be an ultimate technology for spin-manipulation due to its ultra-low power consumption and ultra-fast dynamics. Perspectives and major challenges for next-generation devices will be discussed. This work was supported by the ImpACT Program of the Council for Science, Technology, and Innovation.

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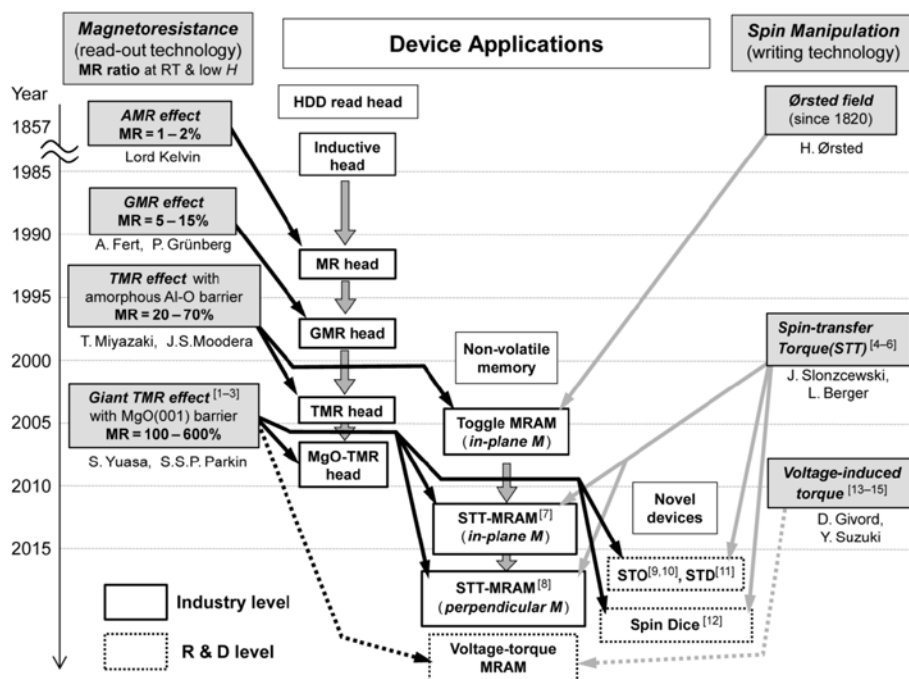


Figure 1 Basic phenomena for spintronics and their device applications.



**Shinji Yuasa** is the director of the Spintronics Research Center at the National Institute of Advanced Industrial Science and Technology, Japan. He received his BS degree in 1991, and his PhD degree in 1996, both in physics, from Keio University, Japan. His research focuses on developments related to tunnel magnetoresistance (TMR), spin-transfer torque, and spin dynamics in magnetic tunnel junctions (MTJs). He has been awarded or co-awarded more than 20 prizes for his achievement of the giant TMR effect in MgO-based MTJs, including the Asahi Award in 2007. Yuasa can be reached by email at [yuasa-s@aist.go.jp](mailto:yuasa-s@aist.go.jp).

## Spin-Transfer-Torque MRAM

Daniel C. Worledge

IBM Research Division, Almaden Research Center

### Abstract

Spin-Transfer-Torque MRAM was invented at IBM by John Slonczewski in the early 1990s. By using a spin-polarized current, instead of a magnetic field, to write a magnetic free layer in a magnetic tunnel junction, the required write current naturally decreases with area, providing attractive technology scaling. The discovery of perpendicular magnetic anisotropy in thin CoFeB/MgO layers at IBM in 2009 and independently at Tohoku University enabled a dramatic reduction in the switching current, and opened the way to practical perpendicular magnetic tunnel junctions for dense Spin-Transfer-Torque MRAM.

This talk will provide an overview of Spin-Transfer-Torque MRAM, including an introduction to the basic physics of spin-transfer torque and applications of Spin-Transfer-Torque MRAM. Then I will review why perpendicular anisotropy is advantageous for MRAM compared to in-plane anisotropy, and the materials challenges of perpendicular anisotropy. I will discuss the research at IBM in 2009 that led to our discovery of perpendicular magnetic anisotropy in thin CoFeB/MgO layers, and our use of these layers to make the first practical perpendicular magnetic tunnel junctions and the first demonstration of reliable writing in Spin-Transfer-Torque MRAM. Finally I will review our recent results on methods to lower the switching current of Spin-Transfer-Torque MRAM by using optimized magnetic materials and double magnetic tunnel junctions.



**Dr. Worledge** received a BA with a double major in Physics and Applied Mathematics from UC Berkeley in 1995, receiving the Department Scholar Award in physics and the Dorothea Klumpke Roberts Prize in mathematics. He then received a PhD in Applied Physics from Stanford University in 2000, with a thesis on spin-polarized tunneling in oxide ferromagnets, measuring the largest tunneling spin-polarization in (LaSr)MnO<sub>3</sub> and the first negative tunneling spin-polarization in SrRuO<sub>3</sub>. After joining the Physical Sciences Department at the IBM T. J. Watson Research Center as a Post-doc in 2000, he became a Research Staff Member in 2001, inventing and developing Current-in-Plane Tunneling as a fast turn-around measurement method for magnetic tunnel junctions. In 2003, Dr. Worledge became the manager of the MRAM Materials and Devices group, and in 2013 he became Senior Manager of MRAM. In 2014 he was promoted to Principle Research Staff

Member and in 2015 to Distinguished Research Staff Member. He has worked on developing Toggle and then Spin Torque MRAM, including discovering perpendicular magnetic anisotropy in Ta/CoFeB/MgO, and leading the IBM team that developed perpendicular magnetic tunnel junctions and demonstrated the first integrated perpendicular spin torque MRAM, with ultra-low bit error rate. His current research interests include magnetic devices and their behavior at small dimensions, and new magnetic devices for logic applications. Dr. Worledge has received three IBM Outstanding Technical Achievement Awards and the IBM Research Client Award.



## Development of materials for next generation MRAM

Seiji Mitani

National Institute for Materials Science (NIMS)

### Abstract

Magnetic random access memories (MRAMs) are of growing interest as one of the next generation non-volatile memory technologies. Magnetic tunnel junctions (MTJs) are the heart of MRAMs, and developing new materials has improved the performance of MTJs. A typical example is the MgO tunnel barrier [1,2], which dramatically enhances tunnel magnetoresistance (TMR) of the MTJs. In this talk, I would like to introduce the research activities at NIMS in developing materials and magnetic nanostructures useful for MRAM technologies.

#### (1) Spinel tunnel barriers

Lattice-matched tunnel barriers were developed by using spinel oxides such as  $\text{MgAl}_2\text{O}_4$  (Fig.1). The lattice matching in MTJs improves the bias-voltage dependence of TMR [3]. Furthermore, by choosing appropriate constituent elements (e.g.,  $\text{MgGa}_2\text{O}_4$ ), the MTJ device resistance can be reduced efficiently through the band gap engineering [4].

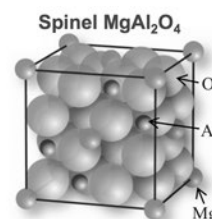


Fig. 1. Spinel structure [3].

#### (2) Heusler alloys

Heusler alloys such as  $\text{Co}_2\text{FeAl}$  and  $\text{Co}_2\text{MnSi}$  possess high spin polarizations that cause large TMR in the MTJs. In addition, many of Heusler alloys show a small magnetic damping constant. Thus, the family of Heusler alloy is considered as a candidate material for ferromagnetic electrodes of MTJ. In fact, large TMR (>300%) has been demonstrated using  $\text{Co}_2\text{FeAl}$  etc [5].

#### (3) Fe/oxide interfaces with large PMA

Perpendicular magnetic anisotropy (PMA) is indispensable for high density MRAMs. A large PMA up to  $1.4 \text{ MJ/m}^3$  (Fig.2), which is comparable to the theoretical predictions, has been demonstrated in monocrystalline magnetic nanostructures of  $\text{Cr/Fe/MgO(001)}$  etc. [6,7].

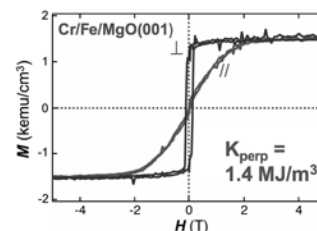


Fig. 2. PMA in  $\text{Cr/Fe/MgO}$  [6].

In addition, other materials useful for MRAMs, such as Mn-based alloys and spin Hall materials, will also be discussed.

These studies were partly supported by the ImpACT program of the Council for Science, Technology and Innovation (Cabinet office, Government of Japan) and JSPS KAKENHI Grant Number 16H06332.

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**Seiji Mitani** is a group leader (spintronics group) of the Research Center for Magnetic and Spintronic Materials at National Institute for Materials Science (NIMS), and is a professor in materials science and engineering at University of Tsukuba. He received his Ph.D. degree in engineering from Nagoya University. After working at the Institute for Materials Research, Tohoku University as a research associate and an associate professor, he moved to NIMS in 2008. His research interests include magnetic thin films and nanostructures, spin-dependent transport and spintronic materials and devices.





# Poster Presentations

- I. Permanent Magnet and other Magnetic Materials
- II. Materials for Magnetic Recording and Data Storage
- III. Spintronic Materials for Memory
- IV. Others

**I-01 (Invited)****Development of the Highest Performance Dy-free Nd-Fe-B sintered Magnet**

Y. Une<sup>(a)</sup>, T. Mizoguchi<sup>(a)</sup>, M. Nakamura<sup>(a)</sup>, T. Iriyama<sup>(b)</sup>, H. Hashino<sup>(a)</sup> and M. Sagawa<sup>(a)</sup>

<sup>(a)</sup>*Daido Steel Co., Ltd., Nagoya, Aichi, Japan*

<sup>(b)</sup>*Intermetallics Co., Ltd., Nakatsugawa, Gifu, Japan*

We have realized the highest performance Dy-free Nd-Fe-B sintered magnet by developing both the new press less process (PLP), of which one of the authors, M. Sagawa, has already reported these basic ideas [1] and our original grain boundary diffusion (GBD) technology. Refinement of the starting alloy powders, optimization of the chemical composition of the magnet and the improvement of Tb-GBD process to the Dy-free Nd-Fe-B sintered bodies make it possible to obtain the following magnetic properties: remanence ( $B_r$ ) of 1.49 T, coercivity ( $H_{cJ}$ ) of 1,840 kA/m and the squareness (SQ) of the demagnetization curve of around 97 % with about 6 mm thickness magnet.

These remarkable properties mentioned above can be obtained from very low content of impurities in the sintered bodies and no deterioration of the orientation degree, which corresponds to  $B_r$ , through the refinement of the alloy powders down to  $D_{50}$  of 1  $\mu\text{m}$  as the base material made by PLP.

Content of impurities, especially carbon, has been found to have a great influence on the increase of  $H_{cJ}$  during GBD [2]. If the base material contains larger than 1100 ppm carbon in weight, then the  $H_{cJ}$  increase after Tb-GBD becomes less than 800 kA/m, but that of less than 1000 ppm,  $H_{cJ}$  increase larger than 1000 kA/m, respectively.

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## Heavy-Rare-Earth-Free Hot-deformed Nd-Fe-B Magnets for Traction Motors

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Nd-Fe-B magnets are widely used in clean energy applications such as hybrid electric vehicles (HEVs), consumer electronics and various other electronic devices, and wind turbines, because they exhibit the highest maximum energy product,  $(BH)_{\max}$ , of all the permanent magnet materials.<sup>(1)</sup> Especially, the demand of Nd-Fe-B magnets with high remanence and high heat resistance for traction motors have been increasing. On the other hand, their coercivity shows only around 20 % of the full potential of the anisotropy field, and its coercivity mechanism has still not clarified completely. Therefore, heavy rare earth elements (HREEs) with higher anisotropy fields (typically Dy and Tb) are commonly added by around 10 % in order to improve coercivity instead of remanence degradation for high temperature applications such as HEVs. However, HREEs' prices are unstable because they are not abundant and are distributed unevenly. Taking the conditions that the production of HEVs has been increasing into consideration, it is the most important problem to reduce HREE consumption in Nd-Fe-B magnets without significant reduction of coercivity.

Hot-deformed Nd-Fe-B magnets are attractive material to eliminate HREEs. They have fine crystal grain microstructure (the lateral size of platelet grains is around 300 nm which is almost the same as the single domain grain size of Nd<sub>2</sub>Fe<sub>14</sub>B magnet phase) due to its unique production process. It was proved that the fine microstructure brings both the reduction of HREEs content and better heat resistance.<sup>(2)</sup> However, their coercivities are still lower than expected from their grain size. <sup>(3)</sup> Recent researches showed that magnetic isolation of Nd<sub>2</sub>Fe<sub>14</sub>B grains was effective for the coercivity improvement.<sup>(4)(5)(6)</sup>

In order to realize the traction motor with HREE-free magnet, we developed high performance hot-deformed magnet as well as modifying the motor design.<sup>(7)</sup> Excellent magnetic properties of the hot-deformed magnets with HREEs free were achieved by optimization of chemical composition and hot-deformation process. We will present these development results mainly in the symposium.

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**I-03****Development of ultimate Nd-Fe-B based permanent magnet by grain boundary and interface engineering**

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Due to the limited natural resources and high cost of Dy, finding ways to enhance the coercivity of Nd-Fe-B sintered magnets without the use, or with the minimum use of Dy has received intense research interest. The coercivity of the Nd-Fe-B magnet is influenced by various microstructural features such as size and alignment of the Nd<sub>2</sub>Fe<sub>14</sub>B grains, constitution of Nd-rich phases at triple junctions, and presence of thin Nd-rich grain boundary (GB) phases and their magnetism. This work presents some strategies to achieve high coercivity over 2.0 T in Nd-Fe-B sintered magnets by the microstructure modification.

One effective way to achieve the high coercivity is to form non-ferromagnetic Nd-rich phases along the grain boundaries. A trace addition of 0.5%Ga into a Nd-Fe-B sintered magnet with Nd-rich composition resulted in the substantial coercivity increase to  $\mu_0 H_c = 1.8 \text{ T}$  due to the formation of non-ferromagnetic phase throughout the grain boundaries. However, the formation of such non-ferromagnetic grain boundary phase has lead to the deterioration of the squareness,  $H_{k90}/H_c$ , to only 0.75, which is much lower than that of the commercial sintered magnet, 0.95. The high coercivity of 2.0 T and high squareness of 0.95 were achieved by decreasing the Ga content to 0.1% as well as refining the grain size to 3  $\mu\text{m}$ . The poster will exhibit other promising approaches to achieve the high coercivity such as grain boundary diffusion etc.

**I-04****Development of high coercivity Nd-Fe-B based permanent magnets with improved thermal stability by microstructure modifications**

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Development of high coercivity Nd-Fe-B based permanent magnets without reliance on heavy-rare-earth elements have been the center of research interest in the past decade. In order to realize a high coercivity magnet, we have combined advanced multi-scale microstructure characterizations using scanning transmission electron microscopy and atom probe tomography with magnetic domain observations and micromagnetic simulations to have a better understanding on microstructure-coercivity relationship. In this work, we will first show how a high room temperature coercivity of 2.2 T with remanent magnetization of 1.30 T is achieved in Dy-free permanent magnets by control of grain size and shape, and engineering of the grain boundary phase. We will discuss how the reduction of aspect ratio in Nd<sub>2</sub>Fe<sub>14</sub>B grains and modification of the grain boundary/interface chemistry can result in improvement of thermal stability of coercivity mainly due to the reduction of local demagnetizing factor. Thereafter, we will show by infiltration of low melting point Nd-Tb-Cu alloy in the hot-deformed Nd-Fe-B magnets, ultimate permanent magnet with a coercivity of 2.5 T and remanent magnetization of 1.4 T, and excellent thermal stability of coercivity of  $-0.32 \text{ }^\circ\text{C}^{-1}$  can be achieved. The microstructure origin for this excellent magnetic properties will be addressed based on the microstructure analysis and micromagnetic simulations.

## Discussion on Nd composition in grain boundary phase based on phase-field approaches — What determines the composition of grain boundary phase ? —

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Nd composition of the grain boundary phase (GBP) in Nd<sub>2</sub>Fe<sub>14</sub>B hard magnet is a key factor to achieve high coercivity. Recently, it has been experimentally elucidated that the Nd composition of GBP in contact with the c-plane of Nd<sub>2</sub>Fe<sub>14</sub>B grain is higher than that contacts with a-plane. The enrichment of Nd content of GBP in contact with the a-plane of Nd<sub>2</sub>Fe<sub>14</sub>B phase is a major issue for attaining high coercivity of the hot-deformed magnet. In this study, we discuss the several factors which will have an influence on the Nd composition of GBP, such as an anisotropy of interfacial energy, the effect of phase separation in liquid phase, the local equilibrium composition which is constrained due to the local volume fraction of GBP phase, and kinetics of the formation process of Nd-rich solid phase.

As a consequence, it is suggested that the difference of Nd composition in GBPs, which depends on the location of GBP in the microstructure, can't be explained by the anisotropy of interfacial energy of Nd<sub>2</sub>Fe<sub>14</sub>B phase, the local volume fraction effect calculated by Hillert's GBP model, and two-phase separation in liquid phase. The possible process which can explain the experimental evidence is the Nd segregation during solidification process followed by Nd-rich crystalline phase formation.

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## Phase-field simulation of Dy-shell formation in Nd-Fe-B hard magnets

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Nd-Fe-B based permanent magnets exhibit not only high remanence but also high coercivity, but have the disadvantage of large thermal demagnetization. When Dy is added to a Nd-Fe-B magnet by means of a grain boundary diffusion process, Dy atoms are localized selectively around the grain boundary region by replacing Nd atoms, which provides Dy-rich shell structure, and it has also been reported that the coercivity is greatly improved by covering the Nd<sub>2</sub>Fe<sub>14</sub>B grains with Dy-rich shell [1].

In this study, the non-equilibrium phase-field method [2] is applied to the formation process of Dy-rich shell, where the moving liquid thin film at Nd<sub>2</sub>Fe<sub>14</sub>B grain boundary region is considered. The results obtained are as follows. It is demonstrated that the liquid phase movement is greatly affected by the surface curvature of Nd<sub>2</sub>Fe<sub>14</sub>B grains. The Dy concentration in Dy-rich shell is influenced by the Dy content inside the moving liquid film and the Gibbs energy difference between Nd<sub>2</sub>Fe<sub>14</sub>B and Dy<sub>2</sub>Fe<sub>14</sub>B.

Acknowledgment: This work is partly supported by the Elements Strategy Initiative Center for Magnetic Materials (ESICMM) under the outsourcing project of MEXT.

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**I-07****Development of high performance Ce-substituted hot-deformed Nd-Fe-B magnets**Xin Tang<sup>1</sup>, H. Sepehri-Amin<sup>1</sup>, M. Matsumoto<sup>2</sup>, T. Ohkubo<sup>1</sup> and K. Hono<sup>1</sup><sup>1</sup>Elements Strategy Initiative Center for Magnetic Materials, National Institute for Materials Science (NIMS)<sup>2</sup>Institute for Solid State Physics, University of Tokyo

The Ce-substituted  $(\text{Ce}_x\text{Nd}_{1-x})_2(\text{FeCo})_{14}\text{B}$  hot-deformed magnets have been successfully fabricated to investigate the influence of Ce substitution on magnetic properties in the Co-containing and Co-free magnets. In the Co-containing sample, with increase of  $x$  to 10 at.% Ce substitution, the coercivity was enhanced from 1.36 to 1.44 T and the remanent magnetization was retained to be  $\sim 1.49$  T, while the thermal stabilities of coercivity were comparable. Thereafter, further increase of Ce content decreases the remanent magnetization and coercivity. Microstructure observations by scanning electron microscope (SEM) and scanning transmission electron microscope (STEM) in Ce-free and 10 at.% Ce substituted Co-containing hot-deformed magnets showed that grain size and chemistry in the grain boundary phase are similar to each other. Contrary to conventional believes, our microstructure investigations and *ab initio* electronic structure calculations showed that the enhanced magnetic properties may originate from improved intrinsic properties due to a particular volume effect caused by the presence of Ce and Co. In the Co-free Ce-substituted sample,  $(\text{Ce}_x\text{Nd}_{1-x})_2\text{Fe}_{14}\text{B}$ , a small increase in coercivity at  $x=0.2$  was ascribed to suppression of  $(\text{CeNd})_6\text{Fe}_{13}\text{Ga}$  formation that results in an increased rare earth concentration in the grain boundary phases strengthening the pinning force against reversed magnetic domain wall motion. Besides, we also produced hot-deformed  $(\text{La,Ce,Nd})-(\text{FeCo})-\text{B}$  magnets to examine how La doping influences the extrinsic properties. The preliminary results showed that to reduce the cost, Ce is much more competent candidate to replace Nd compared to La.

**I-08****Neutron powder diffraction on rare earth permanent magnets  $(\text{Nd}_{1-y}\text{Ce}_x\text{La}_y)_2\text{Fe}_{14}\text{B}$** 

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Substitution effects for permanent magnets  $(\text{Nd}_{1-x-y}\text{Ce}_x\text{La}_y)_2\text{Fe}_{14}\text{B}$  have theoretical and experimental importance. However, a quantitative evaluation of the effects is not trivial due to site-preference of rare earth atoms; the structure of  $\text{RE}_2\text{Fe}_{14}\text{B}$  (space group:  $P4_2/mnm$ ) has two Wyckoff site  $4f$  and  $4g$  for rare earth atoms. To investigate the site-preference, neutron scattering is suitable because both nuclear and magnetic scatterings can be observed at the same time. Therefore, neutron powder diffraction experiments were performed on  $(\text{Nd}_{1-x-y}\text{Ce}_x\text{La}_y)_2\text{Fe}_{14}\text{B}$  ( $(x, y) = (0.225, 0.075)$  and  $(0.375, 0.125)$ ) in a temperature range from 20 K to 600 K.

As a result, the moment size of the  $4g$  site seems to reduce with reducing a Nd content. Especially,  $(\text{Nd}_{0.5}\text{Ce}_{0.375}\text{La}_{0.125})_2\text{Fe}_{14}\text{B}$  shows quite small magnetic moment of the  $4g$  site above room temperature. In addition, as the temperature is lowered, increase of the moment size of the  $4g$  site is observed. These results indicate that nonmagnetic  $\text{Ce}^{4+}$  atoms transition to magnetic  $\text{Ce}^{3+}$  at low temperature, and suggest that Ce atoms prefer the  $4g$  site.

This work was partly supported by the ESICMM under the outsourcing project of MEXT. The sample fabrication was partly performed under the MagHEM project.

## Inelastic neutron scattering study for $RE_2Fe_{14}B$ and $REFe_{11}Ti$ ( $RE = Y, Nd$ )

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The magnetic interactions among magnetic ions are important quantities in a first-principles calculation. Spin wave measurement by inelastic neutron scattering are one of the ways to obtain quantities of the interactions experimentally. In this study, inelastic neutron scattering experiment was performed on  $RE_2Fe_{14}B$  and  $REFe_{11}Ti$  ( $RE = Y, Nd$ ) to investigate a relation between a magnetic interaction and crystal structure/composition.

A dispersion of ferromagnetic spin wave can be approximated by quadratic function  $E_g + Dq^2$  ( $E_g$ : spin wave gap,  $D$ : spin wave velocity,  $q$ : a length of a scattering vector) in small  $q$  region. The  $E_g$  and  $D$  correspond to magneto-crystalline anisotropy and exchange stiffness constant, respectively. The  $REFe_{11}Ti$  has larger  $D$  than  $RE_2Fe_{14}B$  and this result implies that the  $REFe_{11}Ti$  has larger stiffness constant. A Nd substitution increases both  $D$  and  $E_g$ . The  $NdFe_{11}Ti$  shows small  $E_g$  compared to  $Nd_2Fe_{14}B$ . By comparing our results to analytic solutions using continuum approximation, we found out that rare earth has a significant influence on an exchange stiffness constant. Our results and the calculation also indicate that the small spin wave gap in  $NdFe_{11}Ti$  can be originate from small rare-earth-iron exchange interactions.

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## Phase analysis of Nd-Fe-B sintered magnet during heating process studied by X-ray diffraction

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Improvement of the coercivity on Nd-Fe-B magnets is required for modern technological applications. In order to enhance the coercivity, control of the microstructure constructed by secondary-phases in the magnet is essential but there has been some confusion about the constituent phases in Nd-Fe-B sintered magnets. Thus, we performed *in situ* high-temperature synchrotron XRD experiments on a Nd-Fe-B-Cu as-sintered magnet and observed the change of secondary-phases at elevated temperature. The dhcp-Nd,  $NdO_x$ ,  $Nd_2O_3$ , fcc-Nd, and  $Nd_{1.1}Fe_4B_4$  have been identified as the secondary-phases. The  $c$ -axis of the dhcp-Nd phase is larger than that of pure dhcp-Nd, possibly due to partial oxygen occupation at the 4f-site. Additionally, we have visualized the electron density of  $Nd_2Fe_{14}B$  main-phase by the MEM/Rietveld analysis. The differences between the electron density distributions above and below the spin reorientation temperature were found, especially around the Nd atoms, suggesting a change due to a redistribution of the Nd 4f and 5d electron density.



## I-11

### Heat Capacity Measurement of $(\text{Nd}_x\text{Dy}_{1-x})_2\text{Fe}_{14}\text{B}$ alloys

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Heat capacity data of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ ,  $\text{Dy}_2\text{Fe}_{14}\text{B}$  and solutions of these compounds,  $(\text{Nd}_x\text{Dy}_{1-x})_2\text{Fe}_{14}\text{B}$  are indispensable for development of the thermodynamic database of Nd-Dy-Fe-B base magnet alloys. In this study,  $(\text{Nd}_x\text{Dy}_{1-x})_2\text{Fe}_{14}\text{B}$  alloys were prepared by induction melting and the heat capacity of prepared alloys were measured by DSC (Netzsch STA 449 F3 Jupiter) at the temperature ranges between 200K and 700K using liquid nitrogen, and between 400K and 1300K using a high temperature furnace. Consistent results were obtained between two different temperature ranges, which could be applied for the evaluation of thermodynamic parameters of the complex compound alloys.

## I-12

### Theoretical Revisit of the Magnetic Properties in the 2-14-1 Type Compounds

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In connection with the champion magnet Nd-Fe-B, the rare-earth intermetallic compounds  $R_2\text{Fe}_{14}\text{B}$  ( $R$ : rare-earth) were intensively studied around three decades ago. The temperature dependent magnetic properties on single crystal samples of  $R_2\text{Fe}_{14}\text{B}$  were subjected to many experiments, which were systematically analyzed by Yamada *et al.* by using a single ion model for rare-earth [1]. However, the parameters of the model such as crystal field parameters were determined by a multi parameter fit to the experimental data in which there is an inevitable ambiguity. To theoretically predict the magnetic properties, it is highly desirable to establish a method to construct a reliable microscopic model based on first-principles.

Quite recently, we have developed the theoretical formalism to analyze the finite temperature magnetic properties of rare-earth intermetallic compounds taking account of their electronic structure in a first principles way [2]. According to this, we re-examine the magnetization curves in  $R_2\text{Fe}_{14}\text{B}$  systems. The results are consistent with the experimental data [1]. We also show the finite temperature magnetic properties in series of 2-14-1 type compounds, and validate our theoretical approach.

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## Role of typical elements in $\text{Nd}_2\text{Fe}_{14}X$ ( $X = \text{B, C, N, O, F}$ )

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We study the magnetic properties and structural stability of  $\text{Nd}_2\text{Fe}_{14}X$  ( $X = \text{B, C, N, O, F}$ ) in terms of first-principles calculations focusing on the role of  $X$  [1]. We analyze the chemical and magnetovolume effects caused by B, and find that B reduces the magnetic moment and magnetization in  $\text{Nd}_2\text{Fe}_{14}\text{B}$ . Moreover, the crystal-field parameter  $A_{02}\langle r^2 \rangle$  of Nd is not enhanced by B either, indicating that B has minor roles in improving the magnetic properties of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ . These findings are in contrast to the long-held belief that B works effectively for enhancing the magnetic properties of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  [2, 3]. The magnetic properties are diverse when  $X$  changes from B to C, N, O and F. We compare the formation energies of  $\text{Nd}_2\text{Fe}_{14}X$  with that of  $\text{Nd}_2\text{Fe}_{17}X$ . The former becomes negative when  $X = \text{B}$  and C, whereas it becomes positive when  $X = \text{N, O}$  and F. This shows that B plays a significant role in stabilizing the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  phase.

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## *Ab initio* trail toward the champion magnet $\text{Nd}_2\text{Fe}_{14}\text{B}$

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The peak performance of the champion magnet compound  $\text{Nd}_2\text{Fe}_{14}\text{B}$  is analyzed on the basis of electronic structure calculations from first principles, putting  $\text{Nd}_2\text{Fe}_{14}\text{B}$  in a comprehensive scaling curve for  $4f$ - $3d$  intermetallic compounds. Sagawa's original intuition to enlarge Fe-Fe interatomic distance starting from  $\text{R}_2\text{Fe}_{17}$  ( $\text{R} = \text{rare earth}$ ) materials to enhance the intrinsic ferromagnetism is not only quantified *ab initio*, but also a marginal region in the scaling curve to further exploit the  $4f$ - $3d$  intermetallic compounds is found out. The recently developed candidate compounds for the next-generation champion magnet  $\text{RFe}_{12}$  are identified to fall into that marginal area. Even though we sometimes encounter a trade-off between structure stability and ferromagnetism as is the case for the  $\text{RFe}_{12}$  series, *ab initio* insight into the subtle interplay from various contributions therein helps us to find a way out. Several approaches to utilize a bunch of microscopic *ab initio* data at thermal equilibrium to address the off-equilibrium properties at mesoscopic scale and beyond are briefly discussed.

## First-principles study on searching for the stable subphase structures in Ga-doped Nd-Fe-B sintered magnets

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Improving magnetic properties of magnets, e.g., magnetic moments, coercivity, Curie temperatures, etc., is strongly related to control the microstructures of magnetic materials, especially grain boundaries (GB), where main phases and subphases compose complicated structures. So far, there have been many reports on the crystal structures of the subphases in Ga-doped Nd-Fe-B sintered magnets, for example, Nd-rich phases, Nd-poor amorphous-like phases, Ia-3 Nd-rich phases, and NdO<sub>x</sub> phases [1-3]. Generally speaking, it is not easy to determine the crystal structure of subphases by experiments alone, therefore, non-empirical simulations are helpful in determining detailed structures around GB. We performed first-principles calculations for Ga-doped Nd-Fe-B sintered magnets in order to explain what kind of subphase structures can exist stably at the grain boundaries in these sintered magnets by using OpenMX [4]. From the formation energy analysis on fcc-, dhcp-, and bcc-Nd-Fe alloys, we find that fcc-type Nd-Fe alloys are stable among these structures. Furthermore, we clarify the stable position for oxygen in NdO<sub>x</sub> systems by comparing the total energies of the systems.

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## First-principles structural and magnetic analyses of amorphous Nd-Fe alloys

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The details of structural and magnetic properties of grain boundary (GB) phases of Nd-Fe-B magnets has become one of the important issues in the field of application of permanent magnets. In particular, we targeted the relationship between the crystallinity of GB phases and the relative angles between the interfaces and the *c*-plane of neighboring grains [1] and the magnetism of the amorphous grain boundary phases. We performed analyses using the Delaunay [2] and Gabriel graphs [3] for the amorphous Nd-Fe alloys created by melt-quench molecular dynamics simulations [4] with OpenMX [5]. It was shown by the comparison of radial distribution functions (RDFs) that the Gabriel graphs depict the nearest neighbor networks in amorphous Nd-Fe alloys well [6]. In the presentation, we will also present the exchange coupling constant calculation to the amorphous systems using the Liechtenstein method [7].

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## Detection of single magnetization reversal events in Nd-Fe-B hot-deformed magnet

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Coercivity problem of Nd-Fe-B magnets is one of crucial issues for their application to traction motors of electric/hybrid vehicles. Magnetization reversal in Nd-Fe-B magnets takes places as multiple and simultaneous events of nucleation and domain wall propagation. On the other hand, the recent theoretical study on a large-scale spin system has evolved to reproduce a realistic magnetization reversal event under the thermal fluctuation of spins. However, it has been still limited to a single reversal event. Thus, there is a very large gap between the experiment and theoretical studies on the magnetization reversal in Nd-Fe-B magnets. Here, we have challenged to experimentally detect single magnetization reversal events in Nd-Fe-B magnets. For this purpose, a Nd-Fe-B hot-deformed magnet is used as a sample due to its unique microstructure, and anomalous Hall effect (AHE) measurement is adopted due to its very high-sensitivity. The sample is thinned up to several  $\mu\text{m}$  in thickness by a mechanical polishing, and then cross-shaped patterning with a cross center area of several hundreds  $\sim$  tens  $\mu\text{m}$  in side length is done by a focused ion beam lithography. We confirmed that the micropatterning of AHE cross area up to several hundreds  $\mu\text{m}$  gives a slight change in the AHE curve, indicating that the magnetic deterioration due to the process damage is very small in the Nd-Fe-B hot-deformed magnet. However, the AHE curve of the sample with AHE cross area of 13  $\mu\text{m}$  becomes stepwise, corresponding to single magnetization reversal events. We will investigate the reversal behavior of each single reversal event in detail.

## Applications of Soft X-ray Magnetic Circular Dichroism Spectromicroscope to Analyze Local Magnetism

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Overcoming the challenge of observing realistic magnetization reversal processes is crucial to understanding and improving permanent magnets. In light of this, we recently developed a soft X-ray magnetic circular dichroism microscope with about 100 nm spatial resolution that can operate under high magnetic fields up to 8 T [Y. Kotani *et al.*, J. Synchrotron Radiation **25**]. This equipment enables the observation of the fractured surface of Nd-Fe-B sintered magnets, which maintain high coercivities that are almost the same as the bulk, in contrast to other conventional magnetic imaging techniques that require sample polishing or/and thinning resulting in degradation of the magnetic properties.

With this magnetic microscope, local magnetization curves for the fractured surface of a Nd-Fe-B sintered magnet were extracted from magnetic images taken under various applied fields. The local magnetization curves of some pairs of grains were horizontally-shifted in opposite directions. These shifts come from: 1) the different order of magnetization reversal in these two grains for each branch due to thermally-agitated stochastic reversal and 2) the oppositely-magnetized state being stabilized by the magneto-static coupling between them. This implies that the magnetization reversal process depends not only upon microscopic effects, *e.g.* domain wall pinning and nucleation near defects, but also long-range interactions *i.e.* magneto-static coupling.

## Study on magnetization reversal process of a Ga-doped Nd-Fe-B sintered magnet using FORC analysis

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A Ga-added Nd-Fe-B sintered magnet has attracted much attention due to its very high coercivity  $H_c$ . In this study, we have investigated the coercivity mechanism and the magnetization reversal process of a Ga-added Nd-Fe-B sintered magnet mainly based on first-order reversal curve (FORC) analysis. FORC diagram, which is a powerful method to visualize how and where the magnetization reversal occurs on the field plane, strongly suggests that single-domain (SD) magnetization reversal is dominant in the Ga-added magnet at ambient temperature. This is different from that of a commercial magnet, in which multi-domain (MD) reversal is dominant, and the very high  $H_c$  of Ga-added magnet is attributed to this magnetization reversal process. The SD reversal process of Ga-added magnet is also well verified by soft X-ray magnetic circular dichroism (XMCD) microscopy observation. On the other hand, the FORC diagram pattern of the Ga-added magnet changes from that of SD reversal to that of MD reversal when the temperature is elevated up to 200 °C, accompanying by a large reduction of  $H_c$ . This change in the magnetization reversal process at high temperature is discussed in terms of the local demagnetization field.

## Three-Dimensional Visualization of Magnetic Domain Structure Using Scanning Hard X-ray Microtomography

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We developed an X-ray tomographic technique to directly observe the internal magnetic domain structure in a micrometer-sized ferromagnetic sample. The technique is based on a scanning hard X-ray nanoprobe using X-ray magnetic circular dichroism (XMCD). The three-dimensional (3D) distribution of a single component of the magnetic vector in a GdFeCo microdisc was successfully reconstructed with a spatial resolution of 360 nm. This technique is applicable to practical magnetic materials including permanent magnets with sintered microstructures. 3D visualization of the magnetic domain formation process under external magnetic fields will be feasible.

## Free energy landscape of magnetization reversal in permanent magnets

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A central issue of rare-earth permanent magnets research, particularly Nd<sub>2</sub>Fe<sub>14</sub>B magnet, is to increase the coercive field  $H_c$  and to improve its temperature dependency. On the other hands, recent first-principles calculations suggest that the electronic states ( $\text{\AA}$  scale) near the interfaces/surfaces change [1]. Therefore, there is a growing need to clarify the influence of the electronic states on the coercivity at finite temperature.

The effects of thermal fluctuation on coercivity can be broadly divided into the effect of entropy and the effect of thermal activation. To handle the two types of thermal effects consistently, we calculate the free energy landscape with atomistic classical Heisenberg model [2,3] by using a Wang-Landau Monte Carlo method from which the density of states is obtained. In the presentation, we show how the free energy barriers are affected by the electronic states and thermal fluctuations.

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## Dynamical features of an atomistic model for Nd-Fe-B permanent magnets

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We study thermal properties and dynamical features of Nd<sub>2</sub>Fe<sub>14</sub>B magnet [1] with the use of an atomistic model at finite temperatures [2-4]. We report the dynamics of the magnetization reversal in unfavorable magnetic field focusing on the temperature and size dependences of trigger phenomena of the reversal, studied by the stochastic LLG equation [5] and also by Monte Carlo methods. In particular, we investigate the dependence of the relaxation dynamics on temperature, the strength of magnetic field, and also system size by the stochastic LLG simulation. How the domain-wall propagates from the nucleated point, including the effects of the inhomogeneity of the lattice is also studied.

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## First-principles based calculation of magnetic properties at finite temperature

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Ferromagnetic Rare-Earth Transition Metal (RETM) compounds such as  $\text{Nd}_2\text{Fe}_{14}\text{B}$  are widely used and among the strongest permanent magnetic materials owing to the high Curie temperature  $T_C$  and the large magnetocrystalline anisotropy (MCA). A challenge is currently in progress to discover a new material that exceeds existing permanent magnetic materials. Understanding the microscopic origin of the high  $T_C$  and the large MCA for RETM's from the first-principles calculations would be a useful guideline to realize a new practical permanent magnetic material. Quantitative evaluations of magnetic properties from first-principles are highly desirable and a scheme along with the purpose was proposed to estimate magnetic properties of RETM's such as  $T_C$ , saturation magnetizations  $M(T)$ , and MCA constants  $K(T)$ , at finite temperature as well as at  $T=0$  based on first-principles via the construction of an effective spin model whose parameters are deduced from first-principles. The exchange interaction parameters between atoms,  $J_{ij}$ , are directly evaluated for ferromagnetic ground states, noticing that  $J_{ij}$ 's between TM's describe the high  $T_C$ . The large MCA of lanthanide RETM compounds originates from partially filled 4f states at RE site. The crystal-electric-field (CEF) theory accounts for the large uniaxial MCA, whose CEF parameters are also deduced from first-principles. In this poster, we describe a way to deduce parameters from first-principles in detail and show results of analysis at finite temperature with a few examples.

## Theoretical study of Gilbert damping in Rare-Earth Permanent Magnets

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Since the discovery of Nd-Fe-B magnets, many researchers have investigated static magnetic properties of rare-earth permanent magnets [1]. Meanwhile, few studies on dynamical properties have been reported since the magnets have very high resonance frequencies. Ogawa et al. have reported that a broad intensity appears in experimental ferromagnetic resonance spectra for Fe/ $\text{Nd}_2\text{Fe}_{14}\text{B}$  films [2], from which we can suppose that the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  magnets have a large Gilbert damping constant  $\alpha$ .

In the present study, we aim to investigate dominant factors in  $\alpha$  of  $R_2\text{Fe}_{14}\text{B}$  magnets ( $R$ : rare-earth element). For this purpose, we theoretically describe the temperature dependence of  $\alpha$  on the basis of a ligand field theory [3], in which the Gilbert damping arises as a result of the spin transfer from an Fe spin system to a phonon heat bath via a strong spin-orbit interaction within a localized 4f electron system in  $R$  ions, and an orbit-lattice interaction between the 4f electron system and the phonon system. In this process, it is significant that the Fe spin precession excites the 4f electron system, and then the  $\alpha$  strongly depends on the 4f electron state and its environment temperature. According to the calculated temperature dependence of  $\alpha$  in  $\text{Nd}_2\text{Fe}_{14}\text{B}$  magnets in a high temperature range, we can observe that the Gilbert damping does not develop in the low temperature range below about 450 K because the Fe precession hardly activates the 4f electron system without thermal assists. In the conference, assuming other rare-earth elements ( $R = \text{Nd}, \text{Dy}$  and so on), we will discuss the dominant factors and the material dependence of the temperature dependent Gilbert damping of the Fe sublattice magnetization in detail.

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## Magnetization Dynamics Study of High PMA thin films by Time-resolved MOKE Microscopy

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Ferromagnetic thin films with high perpendicular magnetic anisotropy (PMA) have been received a lot of attention for the spintronics application. We have been working the magnetization dynamics of various PMA materials. In this presentation, we will show two kinds of PMA materials.

Spin transfer torque magnetoresistive random access memories (STT-MRAMs), consisting of arrays of magnetic tunneling junctions (MTJs), need the ultra-thin PMA film in order to increase the capacity. To investigate the damping constant ( $\alpha$ ) in the ultra-thin PMA, we estimated  $\alpha$  of ultra-thin CoFeAl (CFA) and FeCo films by time-resolved magneto-optical Kerr effect and discuss the origin of large  $\alpha$  based on the microstructure observation and the first-principles calculation. For CFA a significant amount of Al diffusion observed near the interface of CFA and MgO layers which helps to increase in DOS near the  $E_F$  results high  $\alpha$ . For FeCo the detailed microstructure observation and first-principles calculation reveal that the significant lattice distortion causes the increase in  $\alpha$ .

In the permanent magnet, enhancement of coercivity in Nd-Fe-B magnet without using heavy rare-earth element is a very important topic. The possible way to achieve high coercivity is grain boundary diffusion process. However, no experimental method to evaluate exchange coupling between grains is established. In order to estimate the exchange coupling constant ( $J_{ex}$ ), at the grain boundary we chose Nd-Fe-B and NiFe bi-layer film with and without Mo spacer layer. With the help of macrospin modelling and magnetization dynamics we found 1 nm thick Mo layer is sufficient to decouple the strong exchange coupling between Nd-Fe-B and NiFe layer. This method will help to evaluate inter-granular exchange coupling of conventional Nd-Fe-B magnets.

## Constructing Macro Spin Model from Atomistic Spin Model

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Recently, micro magnetic model is widely accepted and utilized to simulate magnetic properties.

However, we cannot discuss the effect of thermal fluctuations by using this model. We develop a coarse-graining method to construct a macro spin model, which can treat thermal fluctuations [1], from the atomistic spin model derived from the first-principles calculations [2, 3, 4]. We report that some physical properties, such as magnetization along easy axis and domain wall width, evaluated from our macro spin model are consistent with those evaluated from the atomistic spin model. Usage and advantage of our model is also discussed.

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## First-principles study of chemical substitution effects in NdFe<sub>12</sub> and NdFe<sub>12</sub>N

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Experimental reports on NdFe<sub>12</sub>N and Sm(Fe,Co)<sub>12</sub> films have attracted renewed interest in RFe<sub>12</sub>-type compounds as a potential main phase of a new permanent magnet. One issue is search for a substitutional element which stabilizes the ThMn<sub>12</sub> structure with keeping good magnetic properties. In the present work, we theoretically study the effects of chemical substitution on the structural stability and magnetic properties of NdFe<sub>12</sub>(N) based on density functional theory. Main conclusions are the followings. (I) The calculation of NdFe<sub>11</sub>M (M=Ti, V, Cr, Mn, Co, Ni, Cu, Zn) [1] shows that M=Co has positive effect on stabilization, with keeping high magnetization. (II) Chromium substitution at Fe site raises the Curie temperature at low Cr concentrations [2]. (III) The formation energy of RFe<sub>12</sub> (R=rare earth) relative to R<sub>2</sub>Fe<sub>17</sub> and bcc-Fe has strong correlation with the size of R, and takes a minimum for R=Dy [3]. (IV) The magnetization is suppressed when N in NdFe<sub>12</sub>N is replaced with B and C [4], while the magnetic exchange coupling between the Nd and Fe(8j) sites is enhanced by the replacement [5].

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## Theoretical Study on Finite-Temperature Magnetic Properties in Sm(Fe<sub>1-x</sub>M<sub>x</sub>) systems

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Recently, Sm(Fe<sub>1-x</sub>M<sub>x</sub>)<sub>12</sub> (M=Co, V, Ti) has attracted much attention because these compounds are possible candidates for the main phase of novel strong permanent magnets. Unlike other 1-12 systems like NdFe<sub>12</sub> or PrFe<sub>12</sub>, Sm based 1-12 compounds exhibit uniaxial magnetocrystalline anisotropy without nitrogenation, by virtue of the prolate shape of the 4f electronic cloud of Sm ions. At this stage, it is useful to establish a microscopic model for these compounds to describe or to predict their intrinsic magnetic properties.

Thus in this contribution, we propose a microscopic, and effective model for Sm(Fe<sub>1-x</sub>M<sub>x</sub>)<sub>12</sub>, and study their finite temperature magnetic properties by using the model. We show the magnetization curves and  $K_1(T)$  and  $K_2(T)$  for Sm(Fe<sub>1-x</sub>M<sub>x</sub>)<sub>12</sub> (M=Co, V, Ti) for various temperatures. We find that the model describes quite well the temperature dependences of the anisotropy constants  $K_1(T)$  and  $K_2(T)$  of Sm(Fe<sub>1-x</sub>Co<sub>x</sub>)<sub>12</sub> compared with recent experimental data. We also find the FOMP-like behaviors in the magnetization curves along the hard axes of these compounds in lower temperatures than room temperature.



## Thermodynamic, Electronic and Dynamic Stability Study of Potential New Permanent Magnet from First-principles Calculations

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ThMn<sub>12</sub>-type compounds have been paid more attention in the permanent magnets research community in recent years due to the theoretical prediction that it might exhibit comparable/superior magnetization and magnetocrystalline anisotropy energy to the current commercially available strongest Nd<sub>2</sub>Fe<sub>14</sub>B permanent magnet. Due to the thermodynamic instability of the structure, it is very difficult for the experimentalist to synthesize the materials in bulk quantity. In this work, we investigated the effect of substitute/interstitial elements on the thermodynamic, electronic and dynamic stability of the NdFe<sub>12</sub> and SmFe<sub>12</sub> compounds using the density functional theory (DFT) for electronic structure and total energy calculations combining with the frozen phonon method to calculate phonon vibration spectrum.

The calculation results are summarized as following. (1) NdFe<sub>12</sub> and SmFe<sub>12</sub> have both thermodynamic and dynamic instabilities. The systems become thermodynamically stable with Ti and N addition. (2) The dynamic stability of both compounds is improved when pressure is introduced. (3) Co substitution also increases the dynamic stability in both NdFe<sub>11</sub>Co and SmFe<sub>11</sub>Co compounds. (4) The elastic and chemical contribution to the formation energy for both compounds are separated for investigating chemical and elastic effects, respectively. (5) The local distortion due to the substitutional elements is estimated for analyzing the size effect on the stability of the structure.

## Crystallographic and Micromagnetic Properties of Sm(Fe<sub>0.8</sub>Co<sub>0.2</sub>)<sub>12-x</sub>Ti<sub>x</sub> Micropowders

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Since the intrinsic magnetic properties of Sm(Fe<sub>0.8</sub>Co<sub>0.2</sub>)<sub>12</sub> film with the ThMn<sub>12</sub> (*I4/mmm*) structure were reported to be superior to those of Nd<sub>2</sub>Fe<sub>14</sub>B,<sup>1</sup> the stabilization of the Sm(Fe<sub>1-δ</sub>Co<sub>δ</sub>)<sub>12</sub> compound in bulk for the fabrication of high-performance permanent magnets has been a fascinating challenge. To this end, understanding the influence of the Fe-Co substitution on the stabilization of ThMn<sub>12</sub>-phase, structure, and magnetic properties of the Sm(Fe<sub>1-δ</sub>Co<sub>δ</sub>)<sub>12</sub> alloy is highly essential. In this study, we vary the composition of the Ti substituent of the Sm(Fe<sub>0.8</sub>Co<sub>0.2</sub>)<sub>12-x</sub>Ti<sub>x</sub> compound to identify the effects.

In particular, the Sm(Fe<sub>0.8</sub>Co<sub>0.2</sub>)<sub>12-x</sub>Ti<sub>x</sub> micropowders have been successfully prepared by the chemical synthesis of particulate Sm-Fe-Co-O and TiO<sub>2</sub> nanoparticles, followed by the calcination, the hydrogen reduction, and the reduction-diffusion process using calcium at 960 °C. The structural and micromagnetic analyses of the resulting micropowders have been carried out in details.

<sup>1</sup>Y. Hirayama, Y. K. Takahashi, S. Hirosawa, K. Hono, *Scripta Materialia*, **138**, 62–65, 2017.

### Chemical synthesis and magnetic properties of $\text{Sm}(\text{Fe}_{1-x}\text{Co}_x)_{12-y}\text{Ti}_y$ particles

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Among R-Fe compounds (R: rare earth) as permanent magnetic materials,  $\text{RFe}_{12}$  compounds with the tetragonal  $\text{ThMn}_{12}$  structure are expected to exhibit superior magnetic properties due to the highest amount of Fe, although they need a stabilizer, i.e.,  $\text{R}(\text{Fe},\text{M})_{12}$  (M: Ti, V, Cr, Mn, Mo, W, Al, or Si) in order to maintain the  $\text{ThMn}_{12}$  structure in particle form. Ti can stabilize the  $\text{ThMn}_{12}$  structure with the smallest amount of substitution with Fe [1] for the smallest decrease in the magnetic properties. The  $\text{Sm}(\text{Fe}_{1-x}\text{Co}_x)_{11}\text{Ti}$  compounds have been produced as mixed phases by several methods [2, 3]. In this study, we focused on the synthesis of single-phase  $\text{Sm}(\text{Fe}_{1-x}\text{Co}_x)_{12-y}\text{Ti}_y$  ( $0 \leq x \leq 0.3$ ,  $0 \leq y \leq 1$ ) particles based on the chemical method and reduction-diffusion. Firstly, the mixture of Sm, Fe and Co acetates was heated in organic solvents and calcined after an addition of  $\text{TiO}_2$  particles ( $\sim 25\text{nm}$ ). Then, the reduction-diffusion was conducted to reduce the metal ions. Finally, to remove CaO and remained Ca, the products were rinsed with  $\text{N}_2$ -purged  $\text{mQH}_2\text{O}$ .

The resulting particles had the size of  $1.5 \pm 0.4 \mu\text{m}$  and contained  $\text{Sm}(\text{Fe}_{1-x}\text{Co}_x)_{11}\text{Ti}$  phase with 4 wt % of  $(\text{Fe}_{1-z}\text{Co}_z)_2\text{Ti}$  impurity phase (from Rievel analysis). They showed  $H_c = 0.3 \text{ T}$  and  $M_s > 110 \text{ emu/g}$  at an applied magnetic field of 50 T. The Sm : Fe : Co : Ti molar ratio of the resulting particles measured by ICP was 7.6 : 68.4 : 15.2 : 8.8.

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### Microstructure and magnetic properties of highly textured $\text{Sm}(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$ thin films with $\text{ThMn}_{12}$ structure

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The  $\text{ThMn}_{12}$ -type hard magnetic compounds composed of the  $\text{Sm}(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$  films with the excellent intrinsic hard magnetic properties of  $M_s \sim 1.78 \text{ T}$ ,  $H_A \sim 12 \text{ T}$  and  $T_C \sim 859\text{K}$  are reported [1], which are superior to those of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ . The purpose of the present investigation is to explore the possibilities for obtaining high coercivity in anisotropic  $\text{Sm}(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$  based alloys. We examined the grain boundary diffusion process, which is known as an effective methods to increase coercivity of fine grained magnets in the case of Nd-Fe-B magnets, on strongly textured polycrystalline thin films consisting of a large quantity of grain boundaries.

The out-of-plane XRD pattern of the  $\text{Sm}(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$  thin film shows strong (002) and (004) diffraction peaks. By tilting the sample, we clearly observe the superlattice peaks of (132) and (332), which indicates that this film has  $\text{ThMn}_{12}$  structure. The cross-sectional TEM image of a  $\text{Sm}(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$  thin film shows the columnar-shaped  $\text{Sm}(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$  grains on the NiTa/MgO/V underlayer using  $\text{SiO}_2$  substrate. The coercivity of 0.5 T before diffusion process can be further enhanced to 0.84 T by the diffusion of Cu-Ga into anisotropic polycrystalline  $\text{Sm}(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$  thin film.

[1] Y. Hirayama, Y.K. Takahashi, S. Hirosawa and K. Hono, Scr. Mater., 138, 62 (2017)

## SmFe<sub>12</sub>-based permanent magnets, intrinsic magnetic properties and toward development of bulk high-performance permanent magnet

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Intrinsic magnetic properties of the ThMn<sub>12</sub>-type phases in Sm(Fe<sub>1-x</sub>Co<sub>x</sub>)<sub>11</sub>Ti alloys with  $0 \leq x \leq 0.3$  and Sm<sub>1-y</sub>Zr<sub>y</sub>(Fe<sub>0.8</sub>Co<sub>0.2</sub>)<sub>11.5</sub>Ti<sub>0.5</sub> alloys with  $0 \leq y \leq 0.3$  are investigated for discussing the potential as permanent magnet material. Increase of Co substitution from  $x=0.1$  to  $0.3$  in Sm(Fe<sub>1-x</sub>Co<sub>x</sub>)<sub>11</sub>Ti increases the saturation magnetization of the ThMn<sub>12</sub>-type compound from 1.35 T to 1.52 T, while the largest magnetic anisotropy field of 10.9 T was achieved for  $x=0.2$ . The highest  $T_C$  of 830 K was found for the (Sm<sub>0.8</sub>Zr<sub>0.2</sub>)(Fe<sub>0.8</sub>Co<sub>0.2</sub>)<sub>11.5</sub>Ti<sub>0.5</sub> alloy, for which  $\mu_0 M_s$  and  $\mu_0 H_a$  are 1.53 T and 8.4 T at 300 K, respectively. The SmFe<sub>12</sub>-based alloys reach comparable intrinsic hard magnetic properties with those of Nd<sub>2</sub>Fe<sub>14</sub>B and even superior high temperature performance. The remaining challenge is to further develop those intrinsic properties into practically useful extrinsic ones by proper processing. We have therefore employed hydrogenation disproportionation desorption recombination (HDDR) process for the preparation of ultrafine-grained ( $461 \pm 186$  nm) powders. The effect of alloying elements such as Co, Cu, Ga and V on the HDDR process is investigated. Due to the high vapor pressure of Sm as well as the competing phases, recombination into the ThMn<sub>12</sub>-type phase can be realized only in a narrow processing parameters window. The resultant phase constitutions, chemical composition, microstructure and crystallographic relations at different process steps are discussed.

## Search for magnet compounds using Bayesian optimization

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Computer-aided materials-search is attracting much attention due to growing demands for high performance functional materials. Those based on informatics and massive data are especially considered to improve search efficiency significantly. In the realization, we need to obtain accurate data to describe magnetic properties of magnet compounds.

To provide a data set that represents a search space of practical interest, we have performed first-principles calculation of (R, Z) (Fe, Co, Ti)<sub>12</sub> with the ThMn<sub>12</sub> structure for 3630 systems (R=Y, Nd, Sm; Z=Zr, Dy). Then, we obtained theoretical values of magnetization, total energy, and Curie temperature within mean-field approximation.

In this paper, we show results of the Bayesian optimization using the data for (R, Z) (Fe, Co, Ti)<sub>12</sub> mentioned above, and discuss how it has advantage in search efficiency over the random search. We also propose an interpolation technique for calibrating calculated quantities with values from experiments or other theoretical calculations to provide more accurate data to the optimization scheme.

## Preparation of high performance Sm-Fe-N bulk magnets

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To obtain high performance Zn-bonded Sm-Fe-N bulk magnets, it is required to achieve following conditions; decreasing oxygen content in magnets, decreasing Zn content with well dispersibility, and increasing relative density. In this study, we adopted two processes; (i) preparing fine and low oxygen Zn powder, (ii) preparing low-oxygen Sm-Fe-N powder, applying the Arc-Plasma-Deposition (APD) for Zn deposition on the Sm-Fe-N powder and Spark plasma sintering (SPS) for increasing relative density of bulk magnets.

At first, we prepared fine and low oxygen Zn powder ( $d_{50}$  = 0.23  $\mu$ m, Oxy. content = 680 ppm) by the Hydrogen-Plasma-Metal-Reaction (HPMR) method. The Zn-bonded Sm-Fe-N magnets prepared using HPMR-Zn powder showed high coercivity of 2.66 MA/m (15 wt% Zn) and 2.41 MA/m (10 wt% Zn)<sup>[1]</sup>.

Second, low-oxygen Sm-Fe-N powder was prepared, and well dispersed and low-oxygen Zn was deposited on the Sm-Fe-N powders by the APD. The APDed Sm-Fe-N/Zn composite powders were sintered by Spark plasma sintering (SPS), and 3.3 wt% Zn-bonded Sm-Fe-N magnet showed high  $(BH)_{\max}$  of 153 kJ/m<sup>3</sup> with relatively high coercivity of 1.14 MA/m. In addition, we successfully obtained Zn-free low oxygen Sm-Fe-N magnet showing high  $(BH)_{\max}$  of 179 kJ/m<sup>3</sup> <sup>[2]</sup>.

This work was partially supported by MagHEM commissioned by NEDO and ESICMM commissioned by the MEXT.

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## I-36 (Invited)

### High-Performance Ferrite Magnets without Irreversible Low Temperature Demagnetization

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Ferrite magnets with magnetoplumbite structure have been used for various kinds of motors. Since these motors are operated under various conditions ferrite magnets without an irreversible demagnetization in wide temperature range are required. Generally, the intrinsic coercive force ( $H_cJ$ ) of ferrite magnets decreases with lowering temperature therefore the irreversible low temperature demagnetization has been the problem with ferrite magnets.

A mixture of raw materials for La-Ca-Co M-type ferrite with  $B_2O_3$  addition was attrition-milled with water and was calcined in air. The calcined granules were pulverized then ball-milled with water after  $SiO_2$  addition. The obtained slurry was wet-pressed into columnar-shape compacts under the magnetic field of 10 kOe. The compacts were sintered in air.

The magnetic properties of the sintered body measured with a direct current BH tracer showed a remarkable magnetic properties of  $Br$  = 4411 G and  $H_cJ$  = 6875 Oe. This  $H_cJ$  value is more than 25% higher than that of commercial high- $H_cJ$  type ferrite magnets (ex. FB12H; TDK co.). Furthermore, the  $H_cJ$  of the magnet is slightly increased with lowering temperature contrary to the conventional ferrite magnets. This result means overcoming an irreversible low temperature demagnetization which was a long-standing problem in ferrite magnets.

## Formation of $L1_0$ -ordered FeNi Films by Extracting Nitrogen from FeNiN Films

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Ferromagnetic materials possessing large uniaxial magnetic anisotropy energy ( $K_u$ ) and composed of earth abundant elements are required in order to realize the next-generation permanent magnets. We have focused on the  $L1_0$ -ordered FeNi alloy as a rare-earth free high  $K_u$  ferromagnetic material.<sup>1</sup> In this study, we tried to grow epitaxial FeNiN films by molecular beam epitaxy (MBE), and form  $L1_0$ -ordered FeNi films by extracting nitrogen from the FeNiN films<sup>2</sup> to investigate fundamental magnetic properties of  $L1_0$ -FeNi and the process of the formation of the  $L1_0$ -phase.

20 nm-thick FeNiN films were grown on SrTiO<sub>3</sub>(001), MgAl<sub>2</sub>O<sub>4</sub>(001), and MgO(001) substrates by MBE. Nitrogen atoms in the FeNiN films were extracted by furnace annealing at 300 °C for 4 h under hydrogen gas atmosphere. The samples were characterized by x-ray diffraction and a vibrating sample magnetometer.

Multi-domain epitaxial growth of  $a$ -axis-oriented FeNiN films on the (001) substrates was confirmed.  $a$ -axis-oriented  $L1_0$ -ordered FeNi films were successfully formed by the extraction of nitrogen atoms, and  $K_u$  was estimated to be approximately  $1.0 \times 10^6$  erg/cm<sup>3</sup> corresponding to the degree of order of 0.1. As a next step, we try to increase the degree of order for Fe-Ni and  $K_u$  by optimizing conditions of nitrogen extraction.

<sup>1</sup> K. Takanashi *et al.*, J. Phys. D: Appl. Phys. **50**, 483002 (2017).

<sup>2</sup> S. Goto *et al.*, Scientific Reports **7**, 13216 (2017).

## Crystal structure simulation by assimilating incomplete powder diffraction data: First application to magnetic material

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We are developing the structure prediction method to incorporate experimental data that is insufficient to determine crystal structure into theoretical structure search by data assimilation [1]. In this method, the penalty function  $D$  representing the "difference" between reference experimental data and theoretical value is defined, and we minimize the cost function  $F = E + D$  (where  $E$  is the potential energy). Since both  $E$  and  $D$  have the minimum value at the target structure and local minimum points of  $E$  and  $D$  are different,  $D$  can fill local minima of  $E$  and emphasize the target structure.

In the previous study [1], we defined the penalty function  $D$  by using the diffraction angles in the powder X-ray diffraction (XRD) pattern, but it is not useful when the crystal symmetry is very low. In this work, by adopting a penalty function explicitly including the XRD peak intensities, we are working on improving the success rate and robustness of structure search.

We apply this method to the magnetic material Nd<sub>2</sub>Fe<sub>17</sub>. The potential energy  $E$  is calculated using DFT. We confirm that this method can find the correct structure of Nd<sub>2</sub>Fe<sub>17</sub> within a moderate number of trials.

[1]. N. Tsujimoto, D. Adachi, R. Akashi, S. Todo, and S. Tsuneyuki, Phys. Rev. Mater. **2**, 053801 (2018)



## Large CPP-GMR effect in dual-spacer $\text{Co}_2\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si}/\text{AgMg}$ nanojunctions

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Current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) devices using the Co-based Heusler alloys have exhibited large magnetoresistance ratios, which have become promising candidates for reading heads of next-generation hard disk drives (HDDs). In this work, we fabricated CPP-GMR devices using half-metallic  $\text{Co}_2\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si}$  (CFMS) ferromagnetic electrodes and dual Ag-Mg spacers. [1] Magnetic properties and (001)-epitaxial growth of the CFMS films were investigated in the whole CPP-GMR stacks. A dramatically enhanced CPP-GMR effect was achieved in the dual-spacer devices. The CPP-GMR ratio of 73% was achieved at room temperature, which is much larger than 49% in single-spacer samples. Meanwhile, an enhancement of  $\Delta R A$  in the dual-spacer samples was observed to be  $27 \text{ m}\Omega\cdot\mu\text{m}^2$  compared to  $17 \text{ m}\Omega\cdot\mu\text{m}^2$  in single-spacer ones. The enhancement of CPP-GMR effect in dual-spacer devices could be attributed to the improved interfacial spin asymmetry according to the Valet-Fert model. This work indicates that the dual-spacer structures with Ag-Mg nonmagnetic layers are efficient to enhance CPP-GMR effect in half-metallic material based CPP-GMR systems.

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## Heusler Alloy-based CPP-GMR Junctions with an $\text{L1}_2\text{-Ag}_3\text{Mg}$ Ordered Alloy Spacer

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Current perpendicular-to-plane (CPP) giant magnetoresistance (GMR) effects are of interest for magnetic sensor applications, such as read head elements of hard disk drives[1]. In this study, Heusler alloy,  $\text{Co}_2\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si}$  (CFMS) based CPP-junctions layered with an  $\text{L1}_2$ -type  $\text{Ag}_3\text{Mg}$  ordered alloy spacer were investigated.

Single crystalline layered film samples were successfully fabricated including a CFMS |  $\text{Ag}_3\text{Mg}$  | CFMS structure with chemically ordered phases for each layer as shown in cross-sectional high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image. With an optimum condition, a maximum areal resistance change of  $25 \text{ m}\Omega\cdot\mu\text{m}^2$  and MR ratio of 63% were observed at room temperature[2]. CFMS layer thickness dependence of CPP-GMR was also investigated, in which the results suggest that the interfacial spin asymmetry enhanced for the  $\text{Ag}_3\text{Mg}$  based junctions compared with those using a conventional Ag spacer[3].

This work was partially supported by Grant-in-Aid for Scientific Research(S), Grant No. 25220910, from the JSPS, Advanced Storage Research Consortium (ASRC), and the ImPACT project. This work was a part of a cooperative program (Grant No. 17G0409) of the CRDAM-IMR, Tohoku University.

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## II-03

### Development of FePt-based granular film for HAMR media

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Hard disk drives (HDDs) is a key data storage device for data center due to the large capacity, non-volatility and low cost. As the digital information in the world rapidly increases, the development of HDD with high areal density is strongly required. According to Advanced Storage Research Consortium (ASRC), the current goal is to achieve 4 Tbit/in<sup>2</sup> in 2020.

Magnetic recording media is consisted of nano-size ferromagnetic grains dispersed in the non-magnetic matrix uniformly. To achieve 4 Tbit/in<sup>2</sup>, the size of the ferromagnetic grain should be 4 nm. However, currently used CoCrPt alloy does not have enough high anisotropy energy ( $K_u$ ) in order to maintain the magnetization one direction. FePt is a promising candidate for heat assisted magnetic recording (HAMR) media due to its high  $K_u$ . We demonstrated the uniform microstructure with the average grain size of 6 nm and high magnetic properties with the coercivity of 3.5 T in FePt-C granular film for the first in the world in 2008 [1]. In order to achieve 4 Tbit/in<sup>2</sup>, there are still issues, such as further refinement of the grain size, columnar grains and narrow distribution. To solve these problems, we did a lot of research work in the material science point of view. We will present an overview of our work on FePt media in this presentation. In addition to the media development, we try to control the magnetization of FePt media by exposing the circularly polarized light. We will show some results on the all optical switching in FePt media [2].

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## II-04

### Next generation magnetic recording technology --- FePt based heat assisted magnetic recording

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Have you ever considered that World could run out of computer hard drive space by 2020? For hard disk drives (HDD), most primary storage option would be to increase capacity, which meantime can also benefit to reduce the energy consumption with higher recording density devices. Nowadays,  $L1_0$ -ordered FePt nanogranular film based heat assisted magnetic recording (HAMR) is considered as the most promising candidate for the next generation storage technology which can further increase the recording areal density far beyond 2 Tb/in<sup>2</sup>.

Research team from Research Center for Magnetic and Spintronic Materials at NIMS as leading pioneer in this filed have contribute lots of effort in R&D of FePt based HAMR media. Close cooperation with companies from HDD community and University make the project targeting for the practical applications. In this presentation, I will briefly introduce our efforts on the microstructure optimization and fundamental mechanism study of FePt based HAMR media.



## Microwave-assisted switching behavior of CoCrPt based granular media

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Microwave-assisted magnetization switching (MAS) is a promising writing technology of next-generation hard disk drive. In MAS, the coercivity linearly decreases with the rf frequency until a critical frequency over which the MAS effect vanishes. This behavior is well described by the simulation based on macrospin model and also verified by the nanodot experiments. However, the previously reported MAS behaviors of granular media, which exhibit very broad rf frequency response, are apparently far from that mentioned above. Our recent micromagnetic simulation reveals that this very broad rf frequency response of granular media is owing to the effects of thermal agitation and low rf field amplitude. Therefore, we have examined the MAS behavior of CoCrPt-TiO<sub>2</sub> granular film under a large rf field of about 1 kOe. To realize the large rf field experiment, we elaborately designed the device structure to suppress the sample heating due to feeding a large rf power. Consequently, the large rf field MAS experiment successfully carried out, and we clearly confirmed the linear reduction of coercivity with the rf frequency and the presence of critical frequency in CoCrPt-TiO<sub>2</sub> granular film. On the other hand, the coercivity reduction rate with the rf frequency varies with the rf field amplitude, which has never been predicted by the theory and micromagnetic simulation. Moreover, the rf pulse exposure time dependent coercivity also exhibits the different rf frequency dependences.

## Modification of grain density in the FePt-C granular films

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*L10*-ordered FePt granular films are promising candidates for heat assisted magnetic recording with an areal density beyond 2 Tbit/in<sup>2</sup>. In particular, understanding of the grain growth behavior is of crucial importance to increase the grain density of FePt with a narrow size distribution, since the films grow through nucleation - nuclear growth/coalescence, the grain density decreases during the film deposition. Thus the grain density of the films is limited at the initial growth stage. Here we systematically investigated the microstructure of the FePt-C granular films as a function of films thickness with varying substrate temperature and volume fraction of C. We found that the grain density of FePt granular films is strongly influenced by the growth temperature and volume of C. The grain density of over 40 Tgrain/in<sup>2</sup> is realized in the FePt grown at 100 °C.

## II-07

### Development of spin-torque-oscillator for microwave-assisted-magnetic-recording

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The maturity of the perpendicular recording method has stagnated the areal density growth of hard disc drives (HDD) in the last several years. Microwave assisted magnetic recording (MAMR) has been proposed as an alternative recording technology to increase the areal density beyond 2 Tbit/in<sup>2</sup> [1-3]. A spin-torque-oscillator that produces a microwave frequency of 20-30 GHz induced by a current density below  $1.0 \times 10^8$  A/cm<sup>2</sup> is needed for MAMR writer, which has not been realized yet. In this work, all-in-plane STO is introduced for MAMR that consist of a spin-injection-layer (SIL) and a field-generating-layer (FGL) with an in-plane easy axis separated with a metallic spacer. The working principle of all-in-plane STO is introduced first and merits for MAMR application is discussed. It was found that magnetization switching of SIL to opposite direction of applied external magnetic field is essential for oscillation of FGL with a large oscillation cone angle and frequency of 20-30 GHz. We designed SIL and FGL to reduce the critical current density,  $J_c$ , required for the magnetization switching of SIL. It was found that the material with low spin polarization and small saturation magnetization for SIL is beneficial for an easy magnetization switching of SIL. A large spin polarized materials in FGL is needed to reduce  $J_c$  for the magnetization switching of SIL. We will present our progress in the experimental development of all-in-plane STO.

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## II-08

### Evaluation of Structural Order in Magnetic Films by Anomalous X-ray Diffraction using Synchrotron X-rays

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Half-metallic ferromagnets including Heusler alloys, have attracted great interest for long years because of their potential to enhance various kind of spin dependent phenomena such as tunneling magnetoresistance, giant magnetoresistance, and spin transfer torque.<sup>1)</sup> In these systems, formations of super-structures, e.g. the  $L2_1$  ordered structure in Heusler alloy, are intrinsically required to develop half-metallicity which has perfectly spin-polarized conduction electrons. Therefore, evaluating their degree of order is a key part for realizing a completely half-metallic thin-film.

Anomalous X-ray diffraction (AXRD) using synchrotron X-rays is a promising tool to estimate degree of order in these materials quantitatively. Here, we report the current status of AXRD at the beamline BL13XU in the third-generation synchrotron facility, SPring-8, including an *in-situ* instrument for annealing process. An *In-situ* measurement is crucial to eliminate extrinsic effects such as oxidation.

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## Inducing out-of-plane precession of magnetization for microwave assisted magnetic recording using an oscillating spin polarizer in spin torque oscillator

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Microwave assisted magnetic recording (MAMR) is one of the promising technologies for maintaining the continuous increase of the recording density of the hard disk drives (HDDs)<sup>1</sup>. One key component of MAMR is the spin torque oscillator (STO)<sup>2</sup>, which is responsible for the generation of ac magnetic field to assist in writing information. In this study, we investigated the spin dynamics of a current-perpendicular-to-plane giant magnetoresistance type device, and demonstrate the out-of-plane precession of a 7-nm-thick Fe<sub>67</sub>Co<sub>33</sub> layer induced by the spin torque from the other magnetic layer also under oscillation. The power spectra from experiment were well interpreted by simulation. The STO device showed characteristics that have potential for practical application in HDDs.

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### Reduction of write current with ultra-thin Spin-Hall-effect electrode in Voltage-Control Spintronics Memory (VoCSM)

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VoCSM is a magnetic random access memory (MRAM), employing the voltage-controlled-magnetic-anisotropy (VCMA) effect as a bit selecting principle and the Spin-Hall-effect (SHE) as a writing principle. Considering its advantages such as high density, low energy-consumption, and read disturb mitigation, ultra-high efficient writing, VoCSM is one of dominant candidates for future nonvolatile main memory, which can be used in high-speed computing techniques such as deep learning and neural network. In this study, we controlled the amount of over-etching the SHE electrode by using a highly-selective etching method. As a result, thickness of SHE electrode reached to the highest efficient thickness [1]. Write current has reduced to 37  $\mu$ A, which is far smaller than that of same sized memory cells such as spin transfer torque MRAMs.

This work was partly supported by the ImPACT Program of the Council for Science, Technology and Innovation (Cabinet Office, Government of Japan).

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### Thermally Induced Write Errors in Voltage-Driven Magnetization Switching

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Magnetization switching driven by subnanosecond pulsed voltages has opened a way of realizing ultralow-energy consumption nonvolatile magnetic memories. However, a substantial improvement of write error rate (WER) is demanded for practical applications. Although current materials engineering has brought a WER of the order of  $10^{-5}$ , a deeper understanding of voltage-driven magnetization dynamics is necessary for further improving the WER. In this work, we investigate in detail the WER in perpendicularly magnetized magnetic tunnel junctions by varying the pulse width and the external magnetic field. We show that the thermal fluctuation gives rise to magnetization transition between the precession orbits and that this becomes an additional source of write errors. We also show that the WER increase can be avoided by choosing a proper write voltage pulse width. These findings would be of great importance for promoting the developments of various voltage-driven magnetic devices.

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### III-03

#### Pulse shape control of write error rate of a voltage-torque MRAM

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Voltage control of interfacial magnetic anisotropy has attracted much attention as a key phenomenon for low power writing in voltage-torque magnetic random access memories (VT-MRAMs). The write error rate (WER) due to thermal agitation is an inevitable issue for practical application. Much effort has been devoted to improving materials to increase thermal stability and therefore reduce the WER. Here we propose a novel approach to reduce the WER by shaping the voltage pulse. Based on the macrospin simulations we found that introduction of a moderate fall time to the voltage pulse can reduce the WER.

The mechanism of the reduction of the WER is as follows. Without fall time the damping torque during the precession tilts the magnetization to the direction of the applied magnetic field. Therefore, the magnetization at the end of the pulse deviates from the equilibrium direction, which causes the WER. However, the torque due to the anisotropy field during the fall time pulls magnetization away from direction of the applied magnetic field. Tuning the fall time to the optimal value the magnetization at the end of the pulse can reach close to the equilibrium direction, and therefore the WER can be reduced.

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### III-04

#### Materials engineering for voltage-controlled magnetic anisotropy effect

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Voltage-controlled magnetic anisotropy (VCMA) effect is drawing much attention as a promising approach for low power spin manipulation in future spintronic devices, such as voltage-torque magnetoresistive random access memory (MRAM). VCMA effect is observed at the interface between ultrathin ferromagnetic metal and dielectric layers. Physical origin of the purely electronic VCMA effect are explained by the modification of electronic structure at the interface through charge accumulation/depletion, or electric-field induced magnetic dipole. Most important technical issue in the voltage-torque MRAM is to achieve highly-efficient VCMA effect to show the scalability. In this presentation, we will discuss physical origin of the VCMA effect and materials design to enhance the VCMA effect, especially employing the large spin orbit interaction in 5d transition materials.

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### Voltage controlled perpendicular magnetic anisotropy in quantum wells

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Voltage controlled magnetic anisotropy (VCMA) is a promising method to manipulate the magnetization for future spintronics applications. Recently, not only large but also non-linear VCMA effect has been observed in Fe/MgO based magnetic tunnel junctions (MTJs)<sup>1,2</sup>. In order to further enhance the VCMA effect, and figure out the possible origin of the phenomenon, we investigated the VCMA effect in Fe/MgAl<sub>2</sub>O<sub>4</sub> based MTJs, where large perpendicular magnetic anisotropy was also observed as in Fe/MgO.<sup>3,4</sup> Due to the perfect lattice matching, we succeeded in fabricating the MTJs with quantum well effect, where the density of states was modified. As a result, the peak value of VCMA ratio was enhanced to ~400 fJ/Vm, and unusual modification of VCMA is realized based on the quantum well effect.

This work was supported by the ImPACT Program of the Council for Science, Technology and Innovation (Cabinet Office, Government of Japan).

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### Perpendicular magnetic anisotropy in W-doped epitaxial Fe/MgO heterostructures

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Interface perpendicular magnetic anisotropy (PMA) is one of the most important factors in the research of perpendicular magnetic tunnel junctions. Recently, in order to reduce the power consumption of magnetic memory devices, voltage control magnetic anisotropy (VCMA) is being intensively investigated. Single-crystalline Fe/MgO heterostructure is known to have large PMA energy<sup>1</sup> and VCMA coefficient<sup>2</sup>, and moreover they are increased by doping heavy metal elements such as Ir and W to the interface of polycrystalline Fe/MgO<sup>3</sup>. According to the first principles calculation, large PMA energy can be expected to be obtained when the lattice constant of W is close to that of Fe<sup>4</sup>. In this study, we investigated the effect of W doping to the single-crystalline Fe/MgO interface. Results of PMA and VCMA will be presented and discussed with the orbital magnetic moment of Fe. This work was partly supported by the ImPACT Program of the Council for Science, Technology and Innovation.

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### III-07

#### Analysis of lattice distortion of $L1_0$ -MnGa ultrathin films grown on CoGa

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It is expected to utilize  $L1_0$ -MnGa thin films to magnetic tunnel junctions for magnetoresistive random access memory (MRAM) with high recording density because those show small magnetization and high perpendicular magnetic anisotropy (PMA) [1]. Recently, it was reported that the 1-to-3-nm-thick MnGa films with a lattice distortion can be grown at room temperature on a  $B2$  ordered paramagnetic CoGa buffer layer [2]. However, the correlation between the lattice distortion and magnetic properties is not yet clear. Here we report the quantitative evaluation of the lattice distortion and its correlation with PMA properties. The in-plane ( $a$ ) and out-of-plane ( $c$ ) lattice constants of the MnGa layer were determined by a reflection high-energy electron diffraction and X-ray diffraction, respectively. Magnetic properties were measured by a polar magneto-optical Kerr effect. Large decrease in the  $c/a$  values with decreasing the thickness was clearly observed. The thickness dependence of  $H_c$  was similar to that of the  $c/a$  value, indicating the deterioration of PMA due to the lattice distortion. The work was partially supported by the ImPACT program.

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### III-08

#### Tunneling anisotropic magnetoresistance (TAMR) in spin-dependent Fe quantum well

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A lot of the size (thickness) and the cost of conventional magnetic tunnel junctions (MTJs) is spent on auxiliary layers. Alternatively, TAMR effect relies on a single magnetic layer, where the local magnetization determines the tunneling resistance. The relativistic spin-orbit coupling (SOC) occurring in a variety of magnetic heterostructures leads to TAMR, and sizable TAMR has been observed in systems with strong SOC effect, like GaMnAs [1], Pt/Co/Pt [2], and IrMn [3].

In this work, we investigated the TAMR in a quantum-well (QW) structure of Cr /Fe ( $t_{Fe}$ = 5–8 monolayers (MLs)) /MgAl<sub>2</sub>O<sub>4</sub>. The QW states are formed due to the mismatch of  $\Delta_1$  bands of Fe and Cr [4]. The tunneling conductance ( $G$ ) showed clear resonant tunneling, and a large TAMR up to 5% was found. This sizable TAMR indicates that the QW states strongly modulate the interfacial SOC. This work was partly supported by the ImPACT Program of Council for Science, Technology and Innovation, Japan, and JSPS KAKENHI Grant No. 16H06332.

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### Spin-orbit torque switching in in-plane nanomagnets characterized by planar Hall effect

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Spin-orbit torque (SOT) induced magnetization switching has been expected to be a new writing method of magnetoresistive random access memories (MRAMs) [1-3], as SOT-MRAMs, especially those with in-plane easy axis schemes, can allow for sub-ns and field-free switching depending on the easy axis direction [4]. It is of high importance to systematically and statistically investigate the dependence of switching properties with the design of nanomagnet towards low-power and reliable operations. Here we develop a procedure to statistically measure the SOT switching of in-plane nanomagnet array, where the planar Hall effect (PHE) is utilized to detect the magnetization direction. Using the procedure, we systematically evaluate the SOT switching properties of in-plane nanomagnets as a function of their size, aspect ratio, and easy axis angle at various current pulse widths. Based on the obtained results along with macrospin and micromagnetic simulations, we discuss the favorable design for SOT-MRAM applications. A portion of this work was supported by ImPACT Program of CSTI and JST-OPERA.

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### Spin-orbit torque switching in perpendicular-magnetized Co/Pt multilayers

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Spin-orbit torque (SOT) devices has been gaining attention in the field of spintronics owing to their fast operation capabilities and design flexibility, as well as non-volatility [1-3]. Being utilized in high-reliability applications, SOT devices require high thermal stability and large SOT generated by unit current in nanoscale dimensions. In this work, to explore those materials meeting these requirements, we study SOT-induced magnetization switching in Hall-bar devices using Co/Pt multilayer structures with a Pt buffer layer together with a measurement of effective fields. The switching efficiency, defined as a ratio of the areal anisotropy energy density to switching current density, increases with increasing the number of Co/Pt stacking. This trend is in accordance with the stacking number dependence of effective fields per unit current density. Moreover, we can achieve high thermal stability factor  $\Delta$  ( $=E/kBT$ , where  $E$  is energy barrier against thermal fluctuation,  $k_B$  the Boltzmann constant, and  $T$  the absolute temperature) of more than 100 in nanoscale devices down to 60 nm in wire width [4]. Our results indicate that Co/Pt multilayers are promising for nanoscale SOT-MRAM devices possessing high thermal stability and small switching current.

This work was partly supported by ImPACT Program of CSTI, R&D Project for ICT Key Technology of MEXT, and JST-OPERA.

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### III-11

#### Equiatomic quaternary CoFeMnSi Heusler alloy for spintronic applications

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Highly spin-polarized current sources are an essential requirement for spintronic devices, which are expected to provide non-volatile, reconfigurable logic functions and ultralow power consumption. The equiatomic quaternary CoFeMnSi Heusler alloy is interesting in view of its theoretical spin gapless electronic structure, high Curie temperature and experimentally observed high spin polarization [1,2]. Recently, we succeeded in growing single-crystalline CoFeMnSi films that have full *B2* and partial *L21* ordering, which were still expected to show a half-metallicity even though the pseudo gap in the majority spin band could be lost due to the non-ideal Y-type structure [3]. Magnetic tunnel junctions with CoFeMnSi as electrode was also studied and a relatively high TMR ratio of about 101 and 521% was obtained at 300 and 10 K, respectively [4]. The Gilbert damping constant studies was done considering the low total density of states for spin gapless semiconducting materials [5].

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### III-12

#### Development of Mn-based nano-layer films toward spintronic applications

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MnGa alloys thin films with *L1<sub>0</sub>* and *D0<sub>22</sub>* ordered structure are attractive materials applicable to spin-transfer-torque based devices, such as magnetoresistive random access memory (MRAM) with high memory capacity and high frequency devices with a range of THz.[1] Recently, we have developed a room temperature growth process for an highly-ordered *L1<sub>0</sub>* and *D0<sub>22</sub>*-MnGa nano-layer films using *B2*-CoGa buffer layer.[2-7] This achievement was an important step for the development of practical device applications. This is because the nano-layer is needed to manipulate magnetization with low energy consumption by electrical stimulation. In this presentation, we introduce our recent achievements of the development of Mn based nano-layer for practical devices.

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### Fabrication and Magnetoresistance of Fully Epitaxial Magnetic Tunnel Junctions with a Rock-Salt Type ZnO/MgO bilayer tunnel barrier

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Fully epitaxial magnetic tunnel junction (MTJ) with a semiconducting tunnel barrier is one of the fundamental building blocks of a vertical-type spin field-effect transistor. In this study, we report on fabrication of fully epitaxial Fe/ZnO/MgO/Fe structures and conducted a detailed investigation on its structural properties, where the ZnO has a metastable rock-salt (RS) crystal structure.

Films were prepared by molecular beam epitaxy. The structure of the MTJ is Au(10 nm)/Co(10 nm)/Fe(5 nm) top electrode/ZnO(1.2 nm)/MgO (1 nm)/Fe(30 nm) bottom electrode on MgO(001) substrates.

Reflection high-energy electron diffraction (RHEED) images of the ZnO layer grown at 100°C, 170°C and 230°C showed streaky patterns similar to the MgO(001), indicating the formation of the single-crystalline RS-ZnO(001) layer. We also confirmed by the RHEED observation that the Fe top electrodes grown on the RS-ZnO tunnel barrier are single-crystalline, resulting in a fully epitaxial Fe(001)/RS-ZnO(001)/MgO(001)/Fe(001) structure.

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### Tunnel magnetoresistance of equiatomic quaternary CoFeCrAl Heusler alloy films

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Equiatomic quaternary CoFeCrAl Heusler alloy has been predicted to be a ferrimagnetic spin-gapless semiconductor [1]. In this study, we have investigated the tunnel magnetoresistance (TMR) in magnetic tunnel junctions (MTJs) using the epitaxially grown CoFeCrAl film. Samples were prepared by magnetron sputtering. The stacking structure of the MTJs was MgO(001) sub./ Cr (40)/ CoFeCrAl (30)/ Mg (0.4)/ MgO (2)/ CoFe (5)/ IrMn (10)/ Ta (3)/ Ru (5) (thickness is in nm). The XRD measurements indicated that the CoFeCrAl films have the B2 ordered phase. The maximum TMR ratio at RT and 10 K for the MTJ annealed at 400°C was 75% and 153%, respectively. This low TMR ratio might be due to the slight increase of the electronic states in the half-metal gap as indicated by our first principle calculations for the fully B2 ordered CoFeCrAl.

This work was partially supported by CREST (No. JPMJCR17J5), JSPS Core-to-Core Program, and KAKENHI (No. 17F17063).

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### III-15

#### High Performance CoFeB-MgO Magnetic Tunnel Junctions for Nonvolatile VLSI

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Since first demonstrations of the TMR ratios of over 10% at room temperature by Miyazaki group [1] and Moodera group [2], magnetic tunnel junctions (MTJs) have been actively developed for industrial applications. Nonvolatile VLSIs using MTJs are attracting attention for realization of low-power-consumption IoT edge-devices toward an arrival of the IoT era [3,4]. A perpendicular-anisotropy CoFeB/MgO based MTJ (p-MTJ) with a synthetic ferrimagnetic reference layer is an essential building block in the nonvolatile VLSIs [5-7]. Here, we review development in the p-MTJs. In particular, we describe about the material design knowledge of double CoFeB/MgO p-MTJs with thermal tolerance for annealing at temperature of 400°C, which is standard requirement for the integration with CMOS in back-end-of line process [8].

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### III-16

#### Perpendicular Magnetic Anisotropy in Epitaxial Fe<sub>80</sub>Al<sub>20</sub>/MgAl<sub>2</sub>O<sub>4</sub> Heterostructures

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Magnetic tunnel junctions (MTJs) with large perpendicular magnetic anisotropy (PMA) at ferromagnet (FM)/oxide barrier interfaces play a key role in state of the art magnetic random access memory (MRAM). With demand of increasing the MRAM capacity, MTJs need to be constantly downscaled which leads to increased thermal instability of FM layers. Therefore, many attempts were examined to compensate the thermal instability by further strengthening of the PMA at FM/oxide barrier interfaces. Recently, relatively large PMA at Co<sub>2</sub>FeAl/MgO [1] and Co<sub>2</sub>FeAl/MgAl<sub>2</sub>O<sub>4</sub> heterostructures were reported [2]. For these cases, the Al is found to diffuse from Co<sub>2</sub>FeAl into MgO or MgAl<sub>2</sub>O<sub>4</sub>, resulting in promotion of the Fe-O orbital hybridization to enhance the interfacial PMA [3]. However, in bcc Fe-Co alloys, Fe-rich composition tends to show larger PMA than Co-rich ones due to stronger hybridization of interfacial Fe-orbitals with O-orbitals [4]. Considering this, we replaced Co in Co<sub>2</sub>FeAl with Fe, i.e. Fe<sub>3</sub>Al. Here recent results of sputter-deposited Fe<sub>80</sub>Al<sub>20</sub> ultrathin films in combination with an MgAl<sub>2</sub>O<sub>4</sub> barrier, including microstructure analysis will be presented.

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## Reactive deposition technology for a uniform spinel $\text{MgAl}_2\text{O}_4$ barrier toward high quality magnetic tunnel junctions

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Epitaxial magnetic tunnel junctions (MTJs) with a spinel  $\text{MgAl}_2\text{O}_4$  (MAO) tunnel barrier showed high tunnel magnetoresistance (TMR) ratios over 200% at 300 K by a post oxidation method [1] and a direct rf sputtering of an  $\text{MgAl}_2\text{O}_4$  sintered target [2]. In this work, we explored an oxygen reactive sputtering as an alternative method of MAO tunnel barrier preparation. Fe/MAO using rf sputtering of  $\text{Mg}_{19}\text{Al}_{81}$  with Ar- $\text{O}_2$  gas mixture /Fe(001) MTJ stacks on an MgO(001) substrate showed TMR ratios around 150% at 300 K in a wide range of resistance  $\times$  area ( $RA$ ) =  $10^2 \sim 10^6 \Omega \mu\text{m}^2$ , as well as flat and lattice matched interfaces with upper and lower Fe electrodes. This result suggests that the reactive sputtering is a promising process for high quality MAO-based MTJs.

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## Fabrication of an $\text{MgAl}_2\text{O}_4$ -based Fully-Epitaxial Magnetic Tunnel Junction on an 8" Si Wafer

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Full-epitaxial magnetic tunnel junctions (MTJs) with (001) oriented  $\text{Mg}_2\text{AlO}_4$  spinel barrier have been successfully developed on single crystal 8-in silicon wafers. MTJs exhibited high MR ratio exceeding 230% at room temperature. A crucial key was the use of the NiAl seed layer which has some roles such as suppression of Si-diffusion and promotion of (001) orientation for the subsequently deposited layers. TEM analysis revealed that each layer was highly (001) oriented with atomically flat interfaces thanks to the NiAl seed with the remarkably high orientation and flatness. The development would open up a novel approach to yield higher performance for spintronics devices by using 3-dimensional integration process [1].

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### III-19

#### Magneto-static coupling in magnetic tunnel junctions

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Magneto-static coupling has been studied in closely-packed magnetic structures such magnetic random access memory. In the structures, the magneto-static coupling between cells should be suppressed because the coupling causes degradation of stored information. In contrast the coupling is essential in new computing architectures such as magnetic logic, in which information is transferred sequentially through the cells by magnetization switching driven by the coupling. In this study we investigated magneto-static coupling strength in closely located magnetic tunnel junctions (MTJs), which is useful to read out the information transfer with a large voltage signal output. We prepared multilayers of IrMn/CoFe/Ru/CoFeB/MgO/CoFeB/MgO/Ta on Si/SiO<sub>2</sub> substrates. The CoFeB (2.2 nm) sandwiched by MgO layers was a free layer. The films were patterned into pairs of two elliptical cells of 150 x 50 nm (left, small) and 450 nm x 150 nm (right, large), locating along the long axes. The gap between the cells (*g*) were varied with 50 nm and 600 nm. The resistance and magnetoresistance (MR) of the two-cell MTJs were well consistent with the parallel connection of small and large single MTJ cells, indicating no effect of short circuiting. When *g* > 200 nm the two-cell MTJs showed MR loops with two-step sharp switching, indicating small magneto-static coupling. When *g* < 100 nm, all the samples showed single-step switching, indicating that magnetization switching in the large cells propagated to the smalls cell through strong magneto-static coupling.

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### III-20

#### X-ray absorption spectroscopy for voltage-controlled magnetic anisotropy effect

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Voltage-controlled magnetic anisotropy (VCMA) in Fe|MgO-based solid state devices has shown that magnetization can be controlled in fast periods (down to 0.1 ns) by electric fields. Because the VCMA mechanism can be purely electronic, this is an ultimate technology for the operation of spintronics devices, such as nonvolatile random access memory, where high-speed operation with high writing endurance is indispensable. In this poster, we show our recent studies on VCMA effect using X-ray absorption spectroscopy and first principles study to see its microscopic origin. Specifically, we introduce orbital magnetic moment and electric quadrupole mechanisms for the VCMA effect.

This work was supported by the ImPACT Program of the Council for Science, Technology and Innovation (Cabinet Office, Government of Japan).



## Manipulation of antiferromagnetic spin using tunable parasitic magnetization in doped Cr<sub>2</sub>O<sub>3</sub> film

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Antiferromagnets are key component for future spintronic devices, as its superior properties has been anticipated. However, the manipulation, especially 180° control (unidirectional anisotropy control), of antiferromagnetic spin has been rarely demonstrated. Magnetoelectric Cr<sub>2</sub>O<sub>3</sub> is one of few material which can achieve nonvolatile 180° manipulation of antiferromagnetic spin in electrical mean and thus suitable for future memory and storage applications. In this study we developed Cr<sub>2</sub>O<sub>3</sub> film exhibiting a tunable parasitic magnetization, with maintaining its ↑↓↑↓-type antiferromagnetic order. We demonstrated the nonvolatile 180° control of antiferromagnetic single domain by magnetic field, similar to that demonstrated by electric field. The tunable parasitic magnetization enables easy manipulation and detection of antiferromagnetic spin and provides a platform for further understanding antiferromagnets and research opportunities in innovative spintronics device applications.

## Magnetoelectric control of antiferromagnetic domain state

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In an antiferromagnet, magnetic moments are fully compensated and thus, the stray field and the spin-polarization are not generated. Because of these limitation, the application of the antiferromagnet is limited; for example, the spin-valve film in which the antiferromagnet pins the ferromagnetic magnetization by an exchange bias. Recent progress of spintronics shed a light to the antiferromagnet from the new point of view based on the compensated spins; the low power operation by spin current, the THz precession, and the spin/domain manipulation by voltage etc. Among various antiferromagnets, Cr<sub>2</sub>O<sub>3</sub> exhibits the magnetoelectric effect which enable us to control the antiferromagnetic spin/domain state by an electric field. Based on this mechanism, up to date, we have reported the magneto-electric switching of perpendicular exchange bias[1], the energy condition of this switching[2] and the dynamical switching below 10 ns[3]. These phenomena are dominated by the antiferromagnetic domain state which we focus on in this presentation. We also address the direct/indirect observations of the antiferromagnetic domain state by using the soft X-ray magnetic circular dichroism[4] and the dynamical domain-wall propagation

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### III-23

#### Preparation and characterization of a novel heterostructure of graphene/Co<sub>2</sub>Fe(Ge<sub>0.5</sub>Ga<sub>0.5</sub>) for high-performance spintronic device application

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A novel heterostructure consisting of single-layer graphene (SLG) synthesized by high-vacuum chemical vapor deposition (CVD) on a half-metallic Co<sub>2</sub>Fe(GeGa) (CFGG) Heusler alloy was demonstrated in this work. In the SLG/CFGG heterostructure, comparing with other SLG/ferromagnet systems, the chemical bonding between SLG and CFGG was found to be rather weak through an x-ray absorption spectra study of SLG near the C *K*-edge, which indicates a quasi-freestanding nature of the SLG synthesized on CFGG. Depth resolved magnetic circular dichroism (XMCD) measurement suggests there is no degradation on the magnetic property of CFGG at the interface of SLG/CFGG heterostructure, which further confirms the physical nature of the interface between SLG and CFGG. First-principle calculation results show the Dirac cone of SLG and half-metallic band structure of CFGG at the interface are both preserved in the SLG/CFGG heterostructure. Those striking features make the SLG/CFGG heterostructure an extremely promising building block for developing high performance spintronic devices such as magnetic field sensor, magnetic random access memory and spin-transistor.

### III-24

#### Magnetic tunnel junctions with semiconductor Cu(In,Ga)Se<sub>2</sub> spacers

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Magnetic tunnel junction (MTJ) is a key device to realize the spintronic applications such as read heads of ultrahigh density hard disk drives and non-volatile magnetic random access memories (MRAMs). To develop these applications, the high magnetoresistance (MR) ratio should be achieved with relatively low resistance area product (*RA*) smaller than 1 Ωμm<sup>2</sup>. However, reduction of *RA* is a difficult task for current MTJs, because of the large bandgap of oxide insulating spacers. To overcome the above issues, we examined Cu(In,Ga)Se<sub>2</sub> (CIGS) semiconductor spacers instead of oxide insulators. MTJs with CIGS spacer layers show high MR ratio of 40 % at room temperature at low *RA* range smaller than 1 Ωμm<sup>2</sup>. Further enhancement of the MR ratio can be realized by replacing the spacer layer material from CIGS to CuGaSe<sub>2</sub>.

This work was supported by the ImPACT Program of the Council for Science, Technology and Innovation (Cabinet Office, Government of Japan).

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### Theoretical study for tunnel magnetoresistance and magnetic anisotropy in magnetic tunnel junctions with semiconductor $\text{CuIn}_{1-x}\text{Ga}_x\text{Se}_2$ spacers

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Both high magnetoresistance (MR) ratios and low resistance-area products ( $RA$ ) are required for magnetic tunnel junctions (MTJs) to realize read heads of ultrahigh density hard disk drives and non-volatile magnetic random access memories (MRAMs). Recently, Kasai *et al.* successfully fabricated new MTJs with semiconductor  $\text{CuIn}_{1-x}\text{Ga}_x\text{Se}_2$  (CIGS) spacers and found that these MTJs exhibit high MR ratios and low  $RA$  values.<sup>1</sup> In this work, we theoretically investigated magnetotransport properties of the CIGS-based MTJs by means of the first-principles calculations. We revealed that spin-dependent tunneling transport of  $\Delta_1$  wave functions can provide high MR ratios in these MTJs,<sup>2</sup> which is the same mechanism for the giant MR ratios in the MgO-based MTJs. To examine the applicability of these MTJs to spin-transfer-torque and voltage-torque MRAMs, we further studied magnetic anisotropy at the interfaces. It was found that Fe/CuInSe<sub>2</sub> interface can have a giant perpendicular magnetic anisotropy,<sup>3</sup> which is highly beneficial for both the two types of MRAMs.

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### A first-principles study on perpendicular magnetic anisotropy in W-inserted Fe/MgO and Fe/MgAl<sub>2</sub>O<sub>4</sub> interfaces

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Required properties for future spintronic materials are the high spin polarization with low resistance, large perpendicular magnetic anisotropy with small magnetic damping constant  $\alpha$  and a small saturation magnetization with large Curie temperature. Recently, voltage-induced magnetization reversal was proposed in the writing process because of the small writing energy as compared with the current-induced magnetization reversal[1]. In the voltage-induced magnetization switching, the large perpendicular magnetic anisotropy (PMA) and the large voltage dependence of the PMA of heterojunction between ferromagnetic layer and Oxide layer are crucial for application. Here, we will show that the insertion of W layer between Fe/MgO and Fe/MgAl<sub>2</sub>O<sub>4</sub> interfaces can enhance the PMA around 3 mJ/m<sup>2</sup>[2]. We will discuss the origin of the large PMA of these interfaces on the basis of the second order perturbation of the spin-orbit interaction.

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### III-27

#### Huge voltage-controlled magnetic anisotropy effect of monoatomic 5d-transition-metal layer inserted into MgO/Fe interface

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Voltage-controlled magnetic anisotropy (VCMA) effect is a key issue for realizing fast magnetization switching with ultra-low energy consumption in magnetic tunnel junctions. Further enhancement of the VCMA effect is required for the application of the voltage-induced magnetization switching to magnetoresistive random access memories. In this work, we theoretically investigated the effect of monoatomic 5d-transition-metal layer inserted into Fe/MgO interface on the VCMA.

We performed a first-principles calculation by using the Vienna ab initio simulation package. We evaluated the magnetic anisotropy energy (MAE) and the VCMA for Cu/Fe/X/MgO films (X = Hf, Ta, W, Re, Os, Ir, Pt and Au), and discussed the origin of the VCMA on the basis of the second-order perturbation theory with respect to spin-orbit coupling. We found that the VCMA is -173 and 298 fJ/Vm for the Fe/Os/MgO and Fe/Ir/MgO films, respectively, which are more than 10 times larger than that for the Fe/MgO film without 5d-transition-metal layer. We will also discuss the origin of the VCMA enhancement observed in the inserted Os and Ir layers.

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### III-28

#### Anomalous field-like torque on Py layer via Edelstein effect at W/Pt interface

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Magnetization switching induced by spin-orbit (SO) torque is central issue in spintronics for more efficient devices such as storage or logic device. A threshold current density for the switching is proportional to inverse of a spin-torque efficiency. So several materials have been intensively studied so far. Recently there are several challenges for novel SO materials like oxidized metals (CuO<sub>x</sub>[1], WO<sub>x</sub>[2]) or artificial SO lattice (Pt/FM/Ta[3], Pt/W/FM[4]), where FM is ferromagnet. Especially, for making novel artificial lattice structures, we could design new materials based on conventional SO materials whose properties are well known. In this study, we investigated Py/W/Pt trilayer system by means of spin-torque ferromagnetic resonance (ST-FMR) measurement. We found that Py(5nm)/W(2nm)/Pt(1nm) has anomalous behavior because the detected signal consists of almost Lorentzian spectrum. This implies that comparable field-like (FL) field to Oersted field emerges and the signs are opposite each other. We concluded that this FL field is originated from Edelstein effect at W/Pt interface, diffusive spin transport in W layer, and spin scattering at Py/W interface.

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### Observation of strong spin orbit interaction in Cu by doping nitrogen impurities

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It is well known that Cu is a material with very weak spin orbit interaction (SOI). However, it has been reported that SOI in Cu is enhanced by oxidation or impurity doping [1, 2]. In this research, we have demonstrated the enhancement of SOI in Cu(N) in comparison to the non-doped Cu. This approach can provide further comprehensive study for the enhancement of SOI by impurities doping in weak SOI materials. In addition, this is industrially more profitable method as enhancement of SOI due to the cheapness of Cu and N.

Cu(N) thin films were deposited on SiO<sub>2</sub> substrate by radio frequency magnetron sputtering while N<sub>2</sub> gas partial pressure was varied between 0.03 and 0.1 Pa during the sputtering.

We confirmed the enhancement of SOI due to nitrogen doping by two measurement methods. From the weak-antilocalization (WAL) measurement method, it is revealed that spin relaxation rate in Cu(N) is stronger than pure Cu. Spin relaxation length for highly nitrogen doped Cu(N) films is 7.1 nm, which is 20 times shorter than that for pure Cu films. In addition, spin-torque generation efficiency investigated by the spin transfer torque ferromagnetic resonance (STT-FMR) method shows the finite spin torque efficiency of 0.34 in Cu(N)/Co bilayer. It is much higher than that in pure Cu/Co bilayer (~0.04).

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### Spin Hall effect from hybridized 3d-4p orbitals

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We show that a paramagnetic B2 CoGa compound possesses large enough spin Hall angle to allow robust current-induced spin-orbit torques switching of perpendicularly-magnetized ferrimagnetic MnGa films in CoGa/MnGa/Oxide heterostructures[1]. The spin Hall efficiency estimated via spin Hall magnetoresistance and harmonic Hall measurements is  $+0.05 \pm 0.01$ , which is surprisingly large for a system that does not contain any heavy metal element. First-principles calculations corroborate our experimental observations and suggest that the hybridized Co 3d – Ga 4p orbitals is responsible for the intrinsic spin Hall effect. Our results suggest that efficient spin current generation can be realized in intermetallic by alloying a transition metal with a p-orbital element and by Fermi level tuning.

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## Electronic Tunneling through Epitaxial $\text{Co}_x\text{Fe}_{3-x}\text{O}_{4+\delta}$ (001) films with Perpendicular Magnetic Anisotropy

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Recently, it has been reported that magnetic insulator  $\text{CoFe}_2\text{O}_4$  (001) films grown epitaxially on  $\text{MgO}$  (001) substrates have large perpendicular magnetic anisotropy [1, 2]. If  $\text{CoFe}_2\text{O}_4$  films with perpendicular magnetic anisotropy are successfully grown on nonmagnetic metal or semiconductor layers, they can be applied for spintronics devices, e.g. spin filtering devices [3, 4], with perpendicular magnetization, which have advantage for high magnetic recording density and fast magnetic switching. In this presentation, we report successful growth of  $\text{Co}_x\text{Fe}_{3-x}\text{O}_{4+\delta}$  (001) films with perpendicular magnetic anisotropy on nonmagnetic metal  $\text{TiN}$  layers and electron tunneling characteristics through the  $\text{Co}_x\text{Fe}_{3-x}\text{O}_{4+\delta}$  films.

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## Spin-current induced synchronization between spin-Hall nano-oscillators

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Application of spintronics technology to high-frequency devices and artificial neural networks, such as microwave generator, phased array radar, and spoken-digit recognition, is becoming an exciting topic [1]. A key element in these devices is a spin-torque oscillator (STO), which can emit a high-power microwave signal on the order of micro Watts without using a resonator. In particular, an excitation of frequency and/or phase synchronization between STOs is an important subject because it leads to an enhancement of the emission power from the devices and enables us to control the phase of the microwave signal.

In this work, a theoretical proposal on new mechanism of the coupling between STOs is reported. We consider an array of spin-Hall nano-oscillators connected by a common nonmagnetic electrode. We notice that the oscillations of the magnetizations in the oscillators are spontaneously coupled due to spin-current injection through the common electrode. Deriving the coupling torque formula from the spin transport theory and solving the Landau-Lifshitz-Gilbert equation numerically, it is revealed that an antiphase synchronization is excited between the oscillators.

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## Radio-frequency amplification induced by Joule heating induced magnetization dynamics

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Magnetic tunnel junction (MTJ) is promising device for microwave application in spintronics. Currently, microwave amplification has been investigated, however, the obtained amplification gain has been less than 1 [1-2]. In this study, we observed the microwave amplification gain larger than 1 due to the strong heat-driven spin-torque [3]. The MTJ film stack, bottom layer[CoFeB(2.0)|MgO barrier(1.1)|FeB(1.7)|MgO cap(1.0)|top layer, was fabricated on Si|SiO<sub>2</sub> substrate (unit nm) with the junction size of 110 nm. The microwave amplification gain is measured by  $S_{11}$  parameter of dc-biased magnetic tunnel junctions. The observed gain is 1.2 with a single MTJ. This research was supported by Bilateral Programs (MEXT), MIC, ImPACT program of the Council for Science, Technology, and Innovation (Cabinet Office, Government of Japan), and JSPS KAKENHI (Grant No. JP16H03850).

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## IV-03

### Large emission power in vortex spin torque oscillator

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Spin torque oscillator (STO) is a device transforming dc current to radio frequency (rf) power through spin transfer phenomenon and magneto-resistance (MR) effect. Because the rf power of STO is roughly proportional to the square of the MR ratio<sup>1)</sup>, the improvement of MR ratio is the key to enhance the rf power. In this work, we optimized ferromagnet/MgO interface for the improvement of MR ratio and rf power of STO.

We prepared a circular shaped vortex-STO having the structure of substrate/buffer/PtMn(15)/CoFe(2.5)/Ru(0.86)/CoFeB(1.6)/CoFe(0.8)/MgO(1.0)/CoFe(0.5)/FeB(3)/CoFe(0.5)/MgO(0.9)/Ta/Ru with 500 nm  $\phi$ . The unit in the parenthesis is nm. Here, CoFe(0.5)/FeB(3)/CoFe(0.5) is free layer. The MR ratio of the magnetic tunnel junction (MTJ) was 190 %. The power spectrum density of the vortex-STO measured under the perpendicular field of 3.5 kOe and the dc bias voltage of 465 mV. The integrated power was 10.1  $\mu\text{W}^2$ , which is the highest rf power from STOs reported to date and comparable to some of the commercially available crystal oscillators.

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## IV-04

### Tunnel magnetoresistance properties of magnetic tunnel junctions sputtered with the off-axis cathode

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The high tunnel magnetoresistance (TMR) ratio in magnetic tunnel junctions (MTJs) is required for numerous spintronic applications. In the MTJs with oxide barriers, the reduction of the defects originates from O<sup>-</sup> ion bombardment should be important to improve TMR properties. Off-axis sputtering technique using the cathode orthogonal to substrate surface could reduce the damage on the oxide barrier due to the bombardments, whereas there were few investigations using the off-axis sputtering, to our knowledge. In this report, the CoFeB/MgO/CoFeB-based MTJs, the conventional type of MTJs, prepared using off-axis sputtering were investigated to examine whether this technique is useful for exploration of oxide barrier materials.

Sputtered MTJs composed of Si/ SiO<sub>2</sub>/ Ta(5)/ Ru(10)/ Ta(5)/ CoFeB(10)/ MgO(2)/ CoFeB(4)/ Ta(3)/ Ru(5) showed the high TMR ratio of 330% by annealing at 450°C compared to our MTJs previously prepared with conventional cathodes, which implied effectiveness of off-axis sputtering to explore the various oxide barrier materials. Reduction of oxygen vacancies in an oxide barrier sputtered with an off-axis cathode could be important for further improvement of the barrier quality and the TMR ratio.

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## Tuning of magnetic anisotropy in cobalt ferrite thin films by epitaxial strain

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One of the most important properties for magnetic materials is magnetic anisotropy (MA). Particularly, magnetic thin films possessing large perpendicular magnetic anisotropy (PMA) are useful for functional spintronics devices and high-density magnetic recording technology. As a means of controlling MA, a method utilizing magnetoelastic effects is effective. In fact, cobalt-ferrite (CFO) thin films grown on a MgO

(001) substrate with 0.6% of in-plane tensile stress exhibits an induced large PMA  $K_u$  of 14.7 Merg/cm<sup>3</sup> [1], whereas CFO films grown on Mg<sub>2</sub>AlO<sub>4</sub> (001) substrate suffering 3.6% in-plane compressive stress, exhibits as large as negative  $K_u$  of -60 Merg/cm<sup>3</sup> [2]. In this study, we explored appropriate compounds for a buffer layer between CFO thin films and MgO (001) substrates to introduce a larger in-plane tensile strain, thereby realizing the larger PMA. We found that spinel-type compound of Mg<sub>2</sub>SnO<sub>4</sub>(001) can efficiently introduce large strain into CFO thin films. We carefully investigated the relationship between the lattice distortion and the induced  $K_u$ . According to the results of the reciprocal space map measurements and the magnetic torque measurements, we found that the  $K_u$  and distortion are in a linear relationship, meaning that phenomenological magnetoelastic model is valid despite the large lattice distortion of several percent. Our results indicate that the larger  $K_u$  can be realized by introducing further distortion.

[1] T. Niizeki, et al., Appl. Phys. Lett., 103 162407 (2013).

[2] T. Tainosho, et al., in preparation.

## Microscopic Origin of Interfacial Perpendicular Magnetic Anisotropy Studied by X-ray Magnetic Circular Dichroism

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Perpendicular magnetic anisotropy (PMA) is the important subjects to develop the spintronics devices. Microscopic origin has to be clarified explicitly by detecting orbital magnetic moments ( $L$ ) using x-ray magnetic circular dichroism (XMCD). Relationship between  $L$  and PMA energy  $K$  is described by

$$K = \xi/4(L_z - L_x) + \xi^2/\Delta E_{\text{ex}}(L_x^{\text{ud}} - L_z^{\text{ud}}),$$

where  $\xi$  is spin-orbit coupling constant,  $\Delta E_{\text{ex}}$  is exchange splitting,  $L_i^{\text{ud}}$  is the orbital angular momentum matrix elements between up and down spin states in  $i$  direction [1]. The first term directly corresponds to the orbital moment anisotropy. Second term related to the spin-flipped transition. In this presentation, several cases where the second term becomes negligible or dominant are discussed.

- (i) **Fe/MgO interface:** Angle-dependent XMCD detects the orbital moment anisotropy in Fe, which is originated from the chemical bonding between 3d<sub>z<sup>2</sup></sub> and O 2p<sub>z</sub> to tune the electron occupancy in Fe 3d states. Without using heavy-metal atoms,  $K$  can be described mainly by the first term [2].
- (ii) **Co/Pd interface:** While Co 3d states are explained by the anisotropic orbital moments,  $L$  of Pd 4d states are isotropic, resulting in the contribution of the second term [3].

Interfacial spin-orbit couplings are discussed from the view point of  $L$  detected by XMCD.

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## IV-07

### Potential spintronic devices based on anti-perovskite transition metal nitrides

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Transition metal nitride magnetic thin films with antiperovskite structures,  $\gamma'$ -Fe<sub>4</sub>N and  $\varepsilon$ -Mn<sub>4</sub>N, exhibit unique spin transport properties as well as galvanomagnetic effects. Thus, they could potentially find application in future spintronic devices, in addition to contributing to the advancement of fundamental physics. Fe<sub>4</sub>N is not a half-metal itself; however, the polarization of its spin-resolved conductivity reaches almost 100% with a negative sign. As evidence of such properties, an inverse tunneling magnetoresistance ratio of  $-75\%$  and inverse current induced magnetization switching phenomena for magnetic tunnel junctions with the Fe<sub>4</sub>N electrodes were observed at room temperature. Furthermore, the anisotropic magnetoresistance of the Fe<sub>4</sub>N and Mn<sub>4</sub>N reveals a specific temperature dependence, which was explained by the theoretical models that invoke the  $s$ - $d$  scattering of electrons. The recent experimental work on Fe<sub>4</sub>N found an inverse current perpendicular to the plane giant magnetoresistance, efficient spin-current generation, and anisotropic thermoelectric conversion phenomena. These findings demonstrate physical insights that N atoms provide dramatic change in the spin transport properties of host Fe in antiperovskite unit cells, which is resulted from the hybridization effects between  $2p$ -orbitals of N and  $3d$ -orbitals of Fe.

## IV-08

### Room Temperature Measurement of Bio-Magnetic Field using Tunnel Magneto-Resistance Sensor

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Medical diagnosis and basic research have been actively conducted in recent years by measuring the biomagnetic field coming from the human body. Currently, SQUID is used to measure this biomagnetic field, however, facility investment and running cost of liquid helium are problems. In this research, we aim to measure this biomagnetic field using tunnel magneto-resistance (TMR) sensor. The TMR sensor is a magnetic field sensor using magnetic tunnel junction (MTJ), which has characteristics of room temperature operation and low cost, it can be expected to widely spread biomagnetic field measurement.

The MTJ multilayer film was deposited by DC/RF sputtering on thermally oxidized Si wafer. In order to reduce noise, the MTJ multilayer film was micro-fabricated in array using photolithography process. In order to measure a weak biomagnetic field signal, a dedicated amplification and filter circuit was developed and measurement was performed in the shield room to avoid external magnetic field noise.

Measurement of magnetocardiography (MCG), which is a measurement of the magnetic field coming from the heart of the human body, was performed using TMR sensor. We succeeded in observing MCG R peak (with heartbeat) in real time and QRS peaks with good S/N ratio in averaging<sup>1)</sup>. In addition, we tried to measure the magnetoencephalography (MEG) which is the magnetic field signal of the brain. We performed experiments to alpha waves, which are known to have relatively large signals, and succeeded in measuring signals considered to be from the brain by averaging<sup>1)</sup>.

1) K. Fujiwara, *et al.*, Appl. Phys. Express **11**, 023001 (2018)



## Controlling the all-in-all-out magnetic domains in pyrochlore iridate thin films and heterostructures

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Topological materials are expected to possess high current-spin conversion efficiency based on the spin-momentum locked band structures. Antiferromagnetic insulating pyrochlore iridates ( $R_2\text{Ir}_2\text{O}_7$ ,  $R$ : rare-earth) have been predicted to exhibit a topologically nontrivial metallic state at the boundary of magnetic domains because of the possible topological band structures inherent in this class of compounds.

Here, we report on the growth of pyrochlore iridate thin films and heterostructures, aiming at controlling the magnetic domains and boundaries towards controllable creation and annihilation of the domain wall conduction. As a result, we successfully established fabrication of the pyrochlore iridate thin films utilizing solid-state epitaxy [1]. Additionally, two antiferromagnetic magnetic domains are controlled and detected by magnetoresistance. Finally, we observed magnetic domain wall conduction at the  $\text{Eu}_2\text{Ir}_2\text{O}_7/\text{Tb}_2\text{Ir}_2\text{O}_7$  heterostructure with independently controlling the magnetic domains of each layer [2].

[1] T. C. Fujita *et al.*, Sci. Rep. **5**, 9711 (2015). [2] T. C. Fujita *et al.*, Phys. Rev. B **93**, 064419 (2016).

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