NATIONAL INSTITUTE FOR MATERIALS SCIENCE

Research Center for Magnetic and Spintronic Materials

2023 No.6

Magnetism: source of future innovations

Magnetism: source of future innovations

The explosive growth of the internet has significantly increased power consumption by data centers around the world. The spintronic data storage technologies they use enable them to read, write and store information. Many countries have been racing to develop high-performance spintronic devices to reduce global power consumption by data centers.

Magnetic materials are key components of electric vehicle motors.

Permanent magnets used in automobiles are critical to energy conservation and sustainability. Solving one of the challenges, temperature resistance, will greatly improve the performance of automobiles, including energy savings.

In addition, various magnetic phenomena driven by electron spin—the source of magnetism—may potentially be exploited to bring about significant technological innovations. These could include dramatically enhanced existing device performance in addition to substantially improved energy efficiency and reduced energy loss.

In pursuit of its mission to make society more sustainable, the Research Center for Magnetic and Spintronic Materials has been engaged in digital innovation research to develop functional magnetic and spintronic materials by leveraging unique magnetic phenomena.

Data:

Research Center for Magnetic and Spintronic Materials

Director: Seiji Mitani

Number of permanent researchers: 24

- Magnetic Functional Device Group
- Magnetic Recording Materials Group
- Spintronics Group
- Spin Physics Group
- Spin Caloritronics Group
- Nanostructure Analysis Group
- Spin Theory Group
- Green Magnetic Materials Group

Digital Transformation Initiative Center for Magnetic Materials (DXMag)

Cover Story _____

Thermoelectric permanent magnet

This magnetic slab was created by first alternately stacking and bonding a bismuth-antimony alloy ($Bi_{ss}Sb_{12}$) which exhibits large magneto-thermoelectric effects and neodymium permanent magnets. This multilayer junction was then cut at an angle to form a tilted stack. When a charge current passes through it, a temperature gradient in the direction perpendicular to the current flow is generated, cooling one side of the slab. This thermoelectric conversion phenomenon results from three



types of thermoelectric effects produced simultaneously, including the off-diagonal Peltier effect derived from the tilted multilayers. The neodymium permanent magnet component of the slab continuously applies a magnetic field to the $Bi_{88}Sb_{12}$ alloy component, increasing the alloy's thermoelectric conversion performance. Its continuous magnetic force may also be useful for other purposes. This is just one example of the use of many different potential combinations of materials to fabricate thermoelectric permanent magnets. Hybrid materials with more desirable thermoelectric properties may be created through design optimization (see p. 6).

<Press release on a related subject> https://www.nims.go.jp/eng/news/press/2023/11/202311300.html



Key Projects

Read on for an overview of the research projects being carried out at the Research Center for Magnetic and Spintronic Materials (CMSM).

Basic research on magnetic and spintronic materials that could be used to make society more sustainable



Green magnetic materials: creating revolutionary magnet-based, energy-efficient technologies

High-performance neodymium permanent magnets have been used to drive electric vehicle motors. Demand for them has been steadily growing amid increasing public interest in reducing dependence on fossil fuels. In this project, we will develop high-performance neodymium magnets free of heavy rare earth elements—a longstanding challenge. We have also begun developing recycling products containing neodymium magnets when they are ready to be disposed of. In addition, we will develop magnetic thermal management materials—a new class of energy conversion material capable of magnetic heat control. Finally, magnetic materials may play an important role in popularizing the use of hydrogen—a potential green energy source. Magnetic refrigeration materials that exhibit magnetocaloric effects (i.e., the ability to absorb and release heat when magnetic fields are applied) may be used to cool hydrogen to its liquefaction temperature. We have been researching and developing room temperature to cryogenic temperatures.

See Research 1 on p. 6 See Research 2 on **p. 7**



Dysprosium-free neodymium magnets. Their performance has been improved by con trolling their microstructures.



Greatly accelerating materials development through nanoscale analysis and theoretical calculations

free permanent magnet R&D. These achievements were made using technologies capable of analyzing materials' microstructures at the micro, nano and atomic scales. For example, crystalline grain boundaries and interfaces between thin films have been observed technologies (e.g., electron microscopes and three-dimensional atom probes), resulting in the development of materials design guidelines. In addition, comparing experimental results with theory-based computer simulations is a very useful approach to understanding the relationship between materials' microstructures and physical properties. In this project, the efforts of the groups carrying out materials R&D will be expedited through collaboration with other groups specialized in nanoscale analysis and theoretical calculations and by incorporating data-driven methods into their research



NOTE

Magnetoresistive devices

These devices are composed of a non-magnetic layer (made either of an insulator or a metal) sandwiched a fixed layer) while the other can reverse its magnetization (i.e., a free layer). This three-layer structure can change its electrical resistance by switching the relative magnetization directions of the two ferromagnetic layers between parallel and antiparallel. Magnetoresistive devices can be used in memory applications to record binary digital data, 0 or 1, by associating changes in electrical resistance, low or high, with the digits. The larger the change in electrical resistance (i.e., the higher the magnetoresistance ratio), the greater the performance of the device when used in sensors or memory

Digital innovation through cutting-edge magnet research

Electron spin is the source of magnetism. Efficient spin orientation control (or magnetization direction control) in magnetic materials is very important in developing more energy-efficient, higher-speed magnetic devices. There is high demand for developing larger-capacity hard disk drive (HDD) platters. Platters store binary digital data-0 or 1— in the form of magnetization directions within tiny ferromagnetic grains on their surfaces. In this project, we are aiming to increase the recording density of the iron-platinum (FePt)-based recording medium developed by NIMS and develop a new type of data recording system. If these more advanced recording media are successfully created, more sensitive and responsive read heads will also be needed to read data from them. A magnetoresistive device (see the boxed text above) is the core component in current read heads. This device has also been used to store data in magnetoresistive random access memory (MRAM)a next-generation memory candidate. Improving the performance of MRAM may promote its widespread use. NIMS has been developing higher-performance MRAM by optimizing the interfaces between its







*The non-magnetic layer of a tunnel magnetore-sistance (TMR) device is made of an insulator, while that of a giant magnetoresistance (GMR) device is



NIMS' R&D mission includes identifying potential applications for the magnetic and spintronic materials it develops in addition to improving the performance of existing magnetic and spintronic devices. In line with these missions, we have been investigating spinrelated physical phenomena and their mechanisms with the goal of creating energy-efficient, high-speed digital devices. These include sensors with novel working principles capable of systematically collecting large amounts of useful data and next-generation We have also been making our laboratories smarter to expedite materials search cycles. We have automated several systems, including a combinatorial film deposition system capable of forming films with gradually changing chemical compositions or thicknesses on the surfaces of substrates, and device fabrication and material property measurement systems. We have also actively incorporated data science into our analyses. Through these and other efforts, we are keeping our techniques and equipment up to date.



RESEARCH 1

Creating magnetic thermal management materials



Uchida Distinguished Group Leader. Spin Caloritronics Group

Ken-ichi



Spin caloritronics is the science and technology of heat manipulation using electron spin—the source of magnetism. Ken-ichi Uchida has pulled into the lead in this field by shifting his focus from basic to applied research.

Change in research focus from physics to materials science

Magnetic, electrical, and thermal energies are mutually convertible. Although we use heat in our everyday lives, thermal energy is difficult to be controlled. Uchida has been pioneering spin caloritronics, a scientific field aiming to utilize and control heat in many different ways using electron spins. Uchida has been spearheading efforts to advance and popularize this field through various activities, including the hosting of the international spin caloritronics workshop in May 2023 in Tsukuba City where NIMS is based. He is now attempting to transition his focus from basic physics research to applied materials research.

"All previous spin caloritronics research projects have been basic in nature," Uchida said. "Our group's main research focus had likewise been the measurement and analysis of new physical phenomena. I believe we have developed the foundational lab techniques and expertise needed to apply spin caloritronics to the development of new materials. We have shifted our research focus accordingly."

The Uchida Magnetic Thermal Management Materials Project was launched in October 2022 under the Japan Science and Technology Agency (JST)'s ERATO Strategic Basic Research Program. This project aims to create magnetic thermal management materials-functional materials capable of magnetically converting, controlling, or transferring thermal energy.*

Thermoelectric permanent magnets: combining microscale physics and macroscale materials

Research on thermal energy conversion in the above project has already produced significant results. The creation of a thermoelectric permanent magnet-first reported in November 2023 (see the cover and the photo on p. 2)-may lead to the development of new technologies. This permanent-magnet-based material is capable of transverse thermoelectric conversion within it (i.e., energy conversion between charge and heat currents that flow orthogonally to each other) (figure).

"The figure of merit for spin-driven thermoelectric devices had previously been several orders of magnitude smaller than those of commercially available Seebeck-effect-based thermoelectric devices. This has resulted in a lack of interest in pursuing their practical use," Uchida said. "By comparison, the figure of merit for the thermoelectric permanent magnet we developed reaches a few tens of percent of that of the Seebeck-effect-based thermoelectric devices, finally convincing researchers of the feasibility of developing practical, spin-driven thermoelectric devices. Basic research in spin caloritronics was traditionally focused mainly on homogeneous thin films and single crystals for studying physical phenomena because their characteristics were well-known. However, this convention does not always align with our goal of creating practical technologies. Thermoelectric power generation

and electronic cooling requires the use of bulk materials capable of higher outputs. In addition, the energy conversion efficiency of thermoelectric devices can be improved by making them from composite materials and other materials with heterogeneous microstructures, thereby simultaneously inducing multiple physical phenomena leading to high efficiency. This thermoelectric permanent magnet is just one example of magnetic thermal management materials that could take many different forms." Uchida will continue engaging in pioneering research with the goal of making society more sustainable.

* This project focuses on three areas of research: thermal energy conversion, control, and transfer. Researchers in these areas have been working to create three types of magnetic thermal management materials, each representing one area: thermoelectric permanent magnets (the thermal energy conversion area), magnetic hybrid thermal switching materials capable of drastically changing their thermal conductivity in response to magnetization direction changes and/or magnetic field strength (the thermal energy control area), and phase-interface-controlled magnetic refrigeration materials capable of efficiently transferring heat to thermal media when magnetic fields are applied to them (the thermal energy transfer area).



Figure. Example of a hybrid material functioning as a thermoelectric permanent magnet

Existing Seebeck-effect-based longitudinal thermoelectric devices convert energy between charge and heat currents that flow in parallel to each other. These devices are composed of a complex cascaded structure of p-type and n-type semiconductors. By contrast, transverse thermo electric devices have much simpler structures, allowing them to be more resistant to energy loss, cheaper to manu facture, and more durable (see p. 2 for more details).

There is a growing need for sustainable and environmentally friendly magnetic materials to achieve carbon neutrality. We asked Hossein Sepehri-Amin, who has a strong track record in the development of bulk magnetic materials, about the current status of his research and development.

Magnetic Materials Contributing to Energy Issues

Magnetic materials play an important role in realizing net-zero CO2 emissions in our society due to their applications in green energy conversions; such as applications in electric vehicles, wind turbines, and etc. In addition, it is important the materials used for these applications remain "Green"; it means they do not depend on rare elements and offer high performance with low power consumption. To realize these, innovations in magnetic materials are required.

The Green Magnetic Materials Group, led by Hossein Sepehri-Amin, is mainly working on "Permanent Magnets," "Soft Magnets," and "Magnetocaloric Materials". Permanent magnets generate driving force in motors for electric vehicles and wind turbines, but the challenge is to reduce the amount of rare elements that are added to increase the coercivity of the magnets. Soft magnets are incorporated in power electronic devices that play a role in converting electrical DC and AC in electronic devices, and it is desirable to reduce energy loss by improving materials. Magnetocaloric materials are also called "magnetic refrigeration materials" and are responsible for the realization of eco-friendly cooling systems.

Integrating Multiple Research Methods at a High Level

RESEARCH 2

Leading to green innovation bulk magnetic materials



Hossein Sepehri-Amin

Group Leader, Green Magnetic Materials Group

The FIB-SEM, which combines a f cused ion beam (FIB) and a scal electron microscope (SEM), ca a three-dimensional image crostructure of m

The strength of the Green Magnetic Materials Group lies in its combinatorial research approach to design and develop desired properties in magnetic materials.

"We have not only fabricated bulk magnetic materials, but also elucidated the correlation between microstructure and magnetic properties by making full use of multi-scale analysis using electron microscopy, atom probe tomography (APT), and magnetic domain observations. In addition, the combination of multi-scale microstructure characterization and computational simulation is implemented to design the microstructure of magnetic materials and realize desired magnetic properties. In fact, this combinatorial approach in the field of permanent magnets has successfully led to the development of high performance Dy-free permanent magnets with strong resistance to magnetization reversal (see p. 4). Moreover, the development of permanent magnets with giant transverse thermoelectric conversions is another fold of our research interest in the Green Magnetic Materials group, which is being conducted in collaboration with the Spin Caloritronics group under the umbrella of the ERATO project led by Dr. Uchida (see left page.). However, designing high performance bulk magnetic materials without relying on scarce elements is a complicated task because multiple parameters need to be optimized simultaneously. This is where data science comes in. The Green Magnetic Materials Group has

been using data science to develop high-performance bulk magnets and also magnetic recording media responsible for information retention on a hard disk drive (HDD) under the umbrella of the "DXMag(see p.9 footnote)" project, which was launched in 2022.

"Machine learning assisted design of magnetic materials with desired microstructure is one fold of our research approach in the DXMag project. In addition, we are using "process informatics" to optimize the process required to realize the desired microstructure and magnetic properties in the materials. This approach will be a tool to realize high performance bulk magnetic materials that will contribute to a sustainable society. Another success of our research using data science is the so-called "media simulator" developed in collaboration with the Magnetic Recording Materials Group (see p. 6), which is based on deep learning of electron microscopy images as well as a machine learning approach to evaluate defects and magnetic anisotropy in FePt granular media. These approaches enable high-throughput characterization of FePt magnetic recording media, which is being actively applied in media development in collaboration with a HDD industry." said Sepehri-Amin.

With the fusion of competent technology and data-driven methods, research and development is being accelerated day by day.





The explosive growth of the digital information has led to a significant increase in power consumption in data centers around the world, which mainly use hard disk drives (HDDs) to store data. Yukiko Takahashi has been working on increasing HDD storage capacity and developing revolutionary magnetic recording technologies to make society more energy-efficient and sustainable.

"Magnetic fluctuation can be effectively pre-

vented by using magnetically anisotropic ma-

Increasing HDD capacity through microstructure optimization

The global datasphere (i.e., the complex system encompassing all types of data and their dynamic interactions with human groups and norms) is forecasted to grow to 175 zettabytes (1 ZB = 1 billion terabytes) by 2025,^{*1} causing data centers to consume about 10% of global power consumption. Reducing data center energy consumption will require HDDs with higher storage capacities.

Magnetic recording media (or HDD platter) is composed of nanosized ferromagnetic grains uniformly dispersed in a non-magnetic matrix, forming a granular microstructure. It stores digital information in their magnetic layers by switching the magnetization directions of a bit between upward and downward, representing the binary numbers 0 and 1. HDD capacity can be increased by reducing the size of the bit and increasing grain densities. However, this is difficult to achieve with cobalt-chromium-platinum alloy (CoCrPt) grains-the most common magnetic material used in currently available magnetic recording media-because reducing the CoCrPt grains makes them susceptible to magnetic fluctuation induced by ambient heat, disabling their ability to store information.

terials whose internal energy levels vary greatly depending on the direction in which they are magnetized." Takahashi said. "We focused on iron-platinum alloys (FePt) as a candidate magnetic recording material and began FePt R&D in around 2000. Because FePt exhibits strong magnetic anisotropy, rewriting data stored in it requires strong magnetic fields. To deal with this issue, it was adopted a data writing process called heat-assisted magnetic recording (HAMR). In this process, data storing FePt grains are heated to their Curie temperatures, causing them to lose magnetism. As soon as this state is achieved, their magnetization directions are reversed, changing the binary numbers associated with them from one value to another. To fabricate HAMR-compatible thin films, we evaluated various microstructures and their physical properties. As a result, we succeeded in 2008 for the first time ever in fabricating a thin film composed of FePt and carbon (C) with a microstructure highly compatible with HAMR." This success by Takahashi's group led HDD manufacturers around the world to prototype HAMR HDD products. Among them, Seagate Technology—a US data storage company—put a HAMR HDD with FePt recording media on

the market in 2020.

Data-driven search for next-generation magnetic materials

A roadmap published by the three major HDD manufacturers (Advanced Storage Research Consortium, ASRC) set a target recording density of 4 terabits per square inch (Tb/in²) to be achieved by 2028. Discovering revolutionary materials will be crucial to meeting this target. "In addition to FePt-C, we've examined many

different recording medium compositions, including FePt-AlF, and FePt-Cr₂O,," Takahashi said. "However, none have achieved a recording density of 4 Tb/in2. To make a breakthrough, we've incorporated a data-driven approach into our research."

Data-driven methods are used in materials science to efficiently and quickly search for materials with desirable physical properties. NIMS has constructed and operates DICE-one of the world's largest materials databases. To promote the development of innovative magnetic materials using DICE, the Digital Transformation Initiative Center for Magnetic Materials (DXMag)*2 was established in November 2022. Takahashi has been leading a research project at DXMag. "In this DXMag project, NIMS' data science



Figure 1. Conceptual diagram illustrating multi-level magnetic recording

The upper layer of a multi-level magnetic recording system has a Curie temperature higher than that of the lower layer. To rewrite data stored in the upper layer, the layer is irradiated with a low-intensity laser, causing its magnetization direction to reverse without affecting the lower layer. When recording data in the lower layer via magnetization reversal, the layer's temperature needs to be raised greatly by applying a high-intensity laser to it, causing the upper layer to undergo magnetization reversal as well. To correct this, the upper layer is once again irradiated with a low-intensity laser to restore its previous magnetization direction

experts and Green Magnetic Materials Group are leading efforts to construct a media simulator-a system capable of predicting the physical properties of magnetic materials and their fabrication processes (see p. 7)," Takahashi said. "At the same time, we-empirical researchers-are working to introduce Research Data Express (RDE)-an efficient experimental data aggregation system NIMS has developed capable of automatically collecting and standardizing the datasets generated by various lab instruments before transmitting them to DICE. In addition, we have adopted electronic lab notebooks and other tools to make data collection even more efficient." These efforts to develop a smart lab to expedite the search for promising magnetic materials are making steady progress.

Energy-efficient multi-level magnetic recording system: targeting recording densities higher than 10 Tb/in²

In addition to developing new materials, Takahashi's group is pursuing other approaches to increasing HDD recording densities.

"One simple way of increasing HDD capacity is installing a larger number of platters," Takahashi said. "However, this isn't a good approach from a sustainability perspective because it increases the amount of energy needed to produce HDDs. Instead, our group has been developing a multilayered platter with individual layers capable of recording different information (i.e., multi-level magnetic recording). To achieve this, we have fabricated a three-dimensional (3D) recording medium (figure 1)."

Takahashi's group has been carrying out multi-level magnetic recording R&D since 2021 when their project was granted funding from the Japan Science and Technology Agency (JST)'s CREST Strategic Basic Research Program.

"We succeeded in fabricating a 3D recording medium consisting of a granular thin film core made of ruthenium (Ru) sandwiched between FePt thin films (figure 2)," Takahashi said. "Our magnetic property measurements indicated

Efficient data writing using new magnetization reversal mechanisms

In addition to the technologies mentioned above, Takahashi's group has been developing a new magnetization reversal control technology. Data centers already consume several percent of total global power consumption. It would be impossible to significantly reduce their power consumption using currently available technologies. Photonics-electronics convergence technology may be a game changer in making data centers greener. This technology aims to replace some electronic communications and data processing with more energy-efficient, faster optical communications and data processing. Incorporating this technology into HDDs will require magnetic manipulation using light. Takahashi's group has been investigating the possibility of inducing magnetization reversal using a special type of laser beam called circularly polarized light (figure 4). Currently, reversing the direction of magnetization just once in a magnetic recording material requires the application of a laser beam hundreds of times. Her group is working to reduce the number of required applications by modifying the material. "Achieving this requires us to measure the magnetization dynamics (i.e., magnetization

reversal behavior) of ferromagnetic grains. We therefore developed an original measurement instrument." Takahashi said. "This instrument. equipped with a superconducting magnet, is able to heat FePt-C thin films to their Curie temperature of 700 kelvin (K)—a capability unseen elsewhere in the world. The instrument is also able to measure extremely rapid changes in energy levels that occur within a thin film during magnetization reversal. We have been using the



that this medium would be compatible with multi-level magnetic recording (figure 3). We're getting close to making three-level magnetic recording a reality. We will continue to improve our 3D medium with the goal of achieving a recording density of 10 Tb/in²."

instrument to study microstructures that exhibit desirable responses when circularly polarized light is applied to them."

Takahashi and her colleagues have been engaged in different types of research projects to put these energy-efficient HAMR HDD technologies into practical use, thereby making data centers greener.

*1. Source: IDC Global DataSphere, Nov. 2018

*2. The Digital Transformation Initiative Center for Magnetic Materials (DXMag. Director: Tadakatsu Ohkubo) was established within the NIMS Research Center for Magnetic and Spintronic Materials (CMSM) to promote magnetic materials R&D under the framework of the MEXT-funded DxMT projects.



Figure 2. Transmission electron microscope (TEM) im age of a three-dimensional recording medium

This medium is composed of a middle ruthenium (Ru) granular thin film sandwitched between the upper and lower FePt thin films. Both the Ru and FePt films have been grown epitaxially



Figure 3. Magnetic properties of the three-dimensional recording medium

(Left) Magnetization curves illustrating the relationship between an external magnetic field applied to the FePt medium and how strongly the medium was magnetized. The upper and lower FePt films exhibited magnetization reversal at magnetic field of H_{c1} and H_{c2} , respectively. (Right) Graph showing how the magnetization of the FePt films was influenced by their temperatures. The Curie temperatures (T_c) of the upper (blue) and lower (red) FePt films differed by approximately 100 K. These results indicate that the upper and lower FePt films have different magnetic properties and their magnetization directions can be controlled separately using different laser intensities.



Figure 4. Conceptual diagram illustrating how magnetization reversal can be induced using circularly polarized light

The magnetization directions of individual ferromagnetic grains on the surface of a platter can be switched by reversing the rotational direction (either right- or left-hand) of the circularly polarized light applied to the grains

RESEARCH 4

Promoting digital innovation through magnetic thin film research



Yuya Sakuraba

Group Leader, Magnetic Functional Device Group

film deposition system consisting of two multi-target combinational sputtering chambers and a chamber equipped with a substrate transfer robot. This system is capable of forming various types of films for multilayer devices, including layers with compositional gradients. In addition, automated laser processing machines and automatic electrical/magnetic resistance measurement systems are available to expedite the search for materials with desirable characteristics and ensure device optimization.

Developing advanced sensors is vital to increasing the storage capacity of hard disk drives (HDDs) and systematically collecting large amounts of useful data. Yuya Sakuraba has been carrying out materials research focused mainly on thin film sensor materials.

Creating next-generation HDD read heads using half-metals

The Magnetic Functional Device Group led by Sakuraba has long been researching and developing CPP-GMR (current-perpendicular-to-the-plane giant magnetoresistance) devices for use in HDD read/write heads (see p. 5 for details). The electrical resistances of CPP-GMR devices are lower than those of the magnetic tunnel junction (MTJ) devices currently used in commercially available HDD read heads. This feature enables CPP-GMR devices to be miniaturized without compromising their read speeds, making them promising candidates for use in next-generation HDD read heads. However, their low sensitivity is an issue.

To address this, Sakuraba's group has been searching for materials suitable for use in CPP-GMR devices and controlling their microstructures with a main focus on half-metals^{*} with unique magnetic properties (see the example on p. 15).

"These devices are composed of thin films only several nanometers in thickness," Sakuraba said. "Our group has been trying to control thin film microstructures at the atomic scale and understand their physical properties in close collaboration with the Nanostructure Analysis Group and the Spin Theory Group at the Research Center for Magnetic and Spintronic Materials."

World's first flexible heat flux sensor

In addition to the CPP-GMR device R&D described above, Sakuraba is using his expertise in thin film materials to research and develop heat flux sensors. Heat fluxes are continuously generated by the human body, its organs and other biological systems. He believes that useful information can be derived from heat flux data.

"For example, sensors can be used to collect heat flux data from various locations in a building. This information could then be used to improve the building's energy efficiency by enabling the strength and direction of air conditioning to be promptly adjusted," Sakuraba said. "Another example would be detecting mechanical failures in machines by monitoring heat flux patterns within them."

Although heat flux sensors are already available commercially, they all operate using the Seebeck effect. Because these sensors convert energy between charge and heat currents that flow in parallel to each other, they need to have complex structures, increasing their thermal resistance and thereby causing a difficulty of versatile usage.

In light of this, Sakuraba focused on the anomalous Nernst effect, a type of thermoelectric conversion exhibited by magnetic materials. Because this effect causes energy conversion between charge and heat currents that flow diagonally to each other within magnetic

*A half-metal is a type of ferromagnetic material that acts as a conductor to electrons of one spin orientation, but as an insulator to those of the opposite orientation (i.e., having a spin polarization of 100%). By contrast, most other ferromagnetic materials have a relatively low spin polarization of approximately 50%.

materials, anomalous-Nernst-effect-based heat flux sensors can have simple thin film layered structures, reducing their thermal resistance. After a process of trial and error, Sakuraba fabricated a flexible heat flux sensor composed of magnetic thin films on the surface of a plastic film (photo on p.5) and demonstrated for the first time in 2018 that this sensor was able to function using the anomalous Nernst effect.

"The thin film heat flux sensor I developed has advantages over Seebeck-effect-based heat flux sensors in that it is flexible and cheaper to produce," Sakuraba said. "These advantages would be significant if the device is adopted for use as an IoT sensor. Its current sensitivity is approximately 1 microvolt at a heat flux of 1 watt per square meter (i.e., $\approx 1 \,\mu\text{V/W/m^2}$). This sensitivity needs to be improved before it can be put into practical use. To achieve this goal, our group will continue to work closely with other groups specialized in analysis and theoretical research."

Sakuraba's group succeeded in prototyping a bendable, flexible neat flux sensor (photo on p.5) and a large-area, higher-sen sitivity heat flux sensor (photo below).

Various magnetic interactions and unique physical phenomena are known to occur in magnetic thin film materials. Shinya Kasai has been researching the fundamental mechanisms behind these interactions and phenomena with the goal of putting them into practical use. He is also building systems that can be used to efficiently search for magnetic thin film materials with desirable characteristics.

Efforts to improve magnetization switching efficiency

£

Magnetoresistive random-access memory (MRAM) is an outstanding example of a spintronic technology. Improving the performance of MRAM will require a deeper understanding of electron spin-the source of magnetism—and precise control of its behavior.

MRAM stores data using tunnel magnetoresistance (TMR) devices composed of a stack of many thin films in layers only nanometers thick. The core of a TMR device is a three-layer structure composed of two ferromagnetic layers separated by an insulation layer (see p. 5 for details).

"We've focused our MRAM research on making its data writing more energy-efficient and improving its data reading efficiency," Kasai said. "My Spin Physics Group colleagues and I have been seeking to identify ways of increasing magnetization switching efficiency by leveraging various physical phenomena and magnetic interactions."

This may be achieved by efficiently generating spin current-a flow of electron spinsand controlling its direction.

A multilayered device is composed of a stack of different materials, including magnetic thin films. Searching for promising magnetic thin film materials and evaluating their performance in multilayered devices require fabrication and optimization of these devices. These processes can be significantly expedited by fully automated film deposition systems (for details, see NIMS NOW vol. 23, no. 1, p. 3) and a cluster of microfabrication machines capable of manufacturing three-dimensional, multilayered, microfabricated devices with only minimal damage (photo). In addition, fully auto mated magnetic property measurement systems are being developed.



"We've been working to achieve energy-efficient magnetization switching by improving the charge-spin current conversion efficiency of multi-layered systems composed of ferromagnetic and various other materials," Kasai said. "In addition, we've recently begun studying ways of controlling spin within ferrimagnetic and antiferromagnetic materials-potential alternatives to ferromagnetic materials."

Magnetic skyrmion: a promising information carrier for artificial neural networks

Basic research is indispensable in developing next-generation electronic devices with novel working principles. Kasai's research is currently focused on magnetic skyrmionstopologically stable, nanoscale, vortex-like spin configurations that form in ferromagnetic materials-due to their potential to serve as information carriers (figure). Magnetic skyrmion-based devices may be used to write binary digital data-0 or 1-by associating the formation and annihilation of magnetic skyrmions with the digits. Kasai's group has been investigating ways of manipulating

magnetic skyrmions using electric currents and voltages and searching for new materials that may be used to make data writing more energy-efficient in these devices.

"In physical reservoir computing—a type of artificial neural network-the highly non-linear appearance and disappearance of magnetic skyrmions may be used to perform time-series data processing," Kasai said. "We have succeeded in validating the mechanisms behind the formation and annihilation of magnetic skyrmions. We will work on developing functional magnetic skyrmion-based devices and improving their physical properties."



Figure. Schematic diagram of a magnetic skyrmior A skyrmion has a topologically stable, radial spin configuration. The outer spins point upward while the inner spin point downward

RESEARCH 5

Developing new computing mechanisms through a deeper understanding of spin properties



Shinya Kasai

Group Leader, Spin Physics Group



Aiming to fabricate revolutionary MTJs after setting a world record



Sukegawa Group Leader, Spintronics Group

Non-volatile magnetoresistive random access memory (MRAM) is a promising next-generation memory technology that uses magnetic tunnel junctions (MTJs) to retain information. The performance of MTJs is measured using their tunnel magnetoresistance (TMR) ratios. Hiroaki Sukegawa's group achieved the world's largest TMR ratio and has since been working to further improve the quality of MTJs in an effort to develop higher-capacity, stable MRAM.

Setting a world record using an automated film deposition system capable of performing elaborate processes

MTJs (also called TMR devices) that constitute MRAM cells store binary digital data-0 or 1in the form of low or high electrical resistance states. These states can be reversed by applying a magnetic field or other means (see p. 5 for details). The amount of change in electrical resistance can be measured using the TMR ratio. MRAM with a larger TMR ratio is more conducive to faster operation, higher energy efficiency and larger storage capacity. For this reason, memory device researchers around the world have been racing to develop MRAM with a larger TMR ratio. Tohoku University set a TMR ratio record of 604% in 2008 that remained unbroken until recently.

The MTJ developed by Sukegawa's group achieved a TMR ratio of 631% in March 2023, breaking the 15-year-old world record.

"The decisive factor influencing MTJ performance is the quality of the interfaces," Sukegawa said. "An MTJ consists of two magnetic layers separated by an insulation layer. We used a cobalt-iron alloy and magnesium oxide (MgO) to fabricate the magnetic and insulation layers, respectively, and made both layers from single crystals. We employed an automated film deposition system developed by NIMS (photo right) which enabled us to create mag-

netic layers using a sputtering deposition process and the insulation layer using an electron beam evaporation process. By selecting film deposition processes most suitable for specific film forming materials, we were able to create high-quality, smooth interfaces."

In addition, Sukegawa's group inserted a metallic magnesium layer only 0.6 nanometers in thickness between the MgO insulation layer and the lower magnetic layer to absorb excess oxygen from the MgO/magnetic layer interface, thereby improving the MTJ's structural stability. The group also took other unique structural stabilization measures, such as spraying a trace amount of oxygen gas on the upper surface of the MgO layer immediately before the upper magnetic layer was stacked on it.

Understanding unexplained phenomena: key to a breakthrough

The record TMR ratio of 631% resulted from optimizing the MTJ's interfacial structures by forming all of its layers in single crystals under ideal film deposition conditions. However, to create commercially viable MRAM products, some modifications will be necessary: polycrystalline MTJ layers can be produced more cost efficiently than monocrystalline ones and it is desirable for these layers to be fabricated using existing industrial sputtering processes. Sukegawa's group has been investigating ways of making these modifications.

"In addition to the issues we identified, we made a significant discovery," Sukegawa said. "We plotted the TMR ratio as a function of the MgO insulation layer thickness and found that the ratio oscillated widely between about 500% and 600%. Although this phenomenon had previously been known to occur, its cause had been assumed to be interfacial defects; removing them would therefore make the phenomenon disappear. However, improved interfacial quality actually increased the magnitude of the oscillation. I believe that understanding this and other unexplained phenomena-in addition to pursuing larger TMR ratios-will lead to the development of revolutionary MRAM devices."



Automated film deposition system developed by Sukegawa's group consisting of four peripheral sputtering chambers-including one with an electron beam evaporation source-and a central chamber equipped with a substrate transfer robot. The preprogrammed robot automatically transfers a substrate to a specific sputtering chamber that enables target atoms/molecules to be deposited and grown on the substrate under optimum conditions "This system is able to consistently perform high-precision tasks that are difficult to achieve manually. For example, it can supply a small amount of oxygen gas to a sputtering chamber by opening a gas valve only momentarily," Sukegawa said.

Numerical calculation plays a crucial role in validating the physical properties of newly developed materials and showing guidelines for materials development. The Spin Theory Group led by Yoshio Miura has applied theoretical condensed matter physics for development of magnetic and spintronic materials, and supported experimental efforts to improve material performance and discover new materials.

Identifying the cause of magnetic sensor performance deterioration

First principles calculations can predict the various physical properties of materials (e.g., their structural, dynamical, electrical, optical, and magnetic properties) based solely on quantum mechanics and condensed matter physics independent of experiments.

"I recently dealt with issues concerning a magnetoresistive device for magnetic memory and sensors (see p. 5 for details)," Miura said. "The performance of this device was very sensitive to temperature and its magnetoresistance ratio-the device's performance indicator-significantly decreased at room temperatures. The experimentalists who developed this device and consulted me about this issue to identify its cause. I therefore carried out first principles calculations to find an origin of the reduction of the magnetoresistance at finite temperature."

A magnetoresistive device has a complex multilayered structure of magnetic thin films, including a three-layer core functional structure (i.e., the magnetic tunnel junction (MTJ)) composed of a non-magnetic layer sandwiched between two ferromagnetic layers. Miura's calculations focused on the MTJ interface.

The first principles calculations can basically treat physical properties at the absolute zero temperature of -273.15°C. To make predictions possible at ambient temperatures (i.e., finite temperatures) at which the device works actually, Miura incorporated other theories such as the statistical physics into first principles calculations.

"At finite temperatures, spin-dependent transport properties of MTJ are significantly influenced by thermal fluctuation of magnetization," Miura said. "Accordingly, I improved my spin-dependent transport calculations by including thermal fluctuations of interface magnetization in MTJs using statistical thermodynamics. The calculation results showed that the interface in fact caused reduction of magnetoresistance and that reduction due to increased temperatures can be prevented by modifying interfacial atomic structures (figure). We reported these findings to the experimental researchers and they have been making modifications to their materials in magnetoresistive devices accordingly. I know these modifications are not easy to realize in real experiments, but it's important for us to guide them in a more promising direction."

Contributing to data-driven magnetic materials development

"In a project in which we are collaborating with NIMS researchers specialized in data-driven materials development, we took the first step by performing first principles calculations," Miura said. "Our collaborators are now using the calculation results to construct machine learning models capable of high throughput material developments." In another research project, our group is trying to develop iridium (Ir) free antiferromagnetic



Data-driven materials development has been widely adopted by researchers. The members of Miura's group have begun participating in various data-driven research projects.

materials-a scarce chemical element-due to a rapid rising in Ir prices. Ir is a key element in the antiferromagnetic materials used in currently available magnetoresistive devices to fix the magnetization direction. Miura is trying to identify Ir-free antiferromagnetic material alternatives that could be used to maintain or improve the current performance of magnetoresistive devices. He has been trying to apply the theoretical condensed matter physics to give new directions for developments of magnetic materials.



Figure. By performing first principles calculations, Miura estimated the exchange stiffness of spin moments at inter faces of the magnetic tunnel junction (MTJ) between the cobalt-manganese-silicon (Co₂MnSi) alloy ferromagnetic layer and the magnesium oxide (MgO) insulation layer at ambien emperatures. The interfacial exchange stiffness constant plotted on the v-axis of the graph represents the stiffness of the spin moment at the interface. The higher the stiffness, the more endurance the spin to thermal fluctuation

Calculation results found that spin moments in the Co-terminated Co_2MnSi interface is easy to fluctuate at finite temperatures (the red bar for Co^1 at far left in the graph). By contrast, the Co₂MnSi interface with an iron (Fe) layer inserted underneath it maintained a larger exchange stiffness at interfaces, making the durability to thermal fluctuations of interface magnetization(the red bar for Fe¹ at far right).



The use of state-of-the-art analytical instruments—including electron microscopes and three-dimensional atom probe (3DAP)—can provide detailed insights into the microstructures. The Nanostructure Analysis Group works with researchers engaged in materials development by performing multiscale microstructure analyses using these instruments. We asked Taisuke Sasaki, the group leader, about his group's activities.

probe (3DAP) capable of imaging 3D elemental distribution within material specimens

Multiscale, multifaceted analyses of metallic materials

nic level

The properties of materials are closely related to the microstructures that develop during their fabrication processes and their constituent elements. Because the microstructures that affect properties exist on a wide range of length scales from microscale to atomic, multiscale microstructural analyses are vital. The Nanostructure Analysis Group led by Sasaki studies the relationships between the microstructures and properties of various metallic materials, including magnetic materials, through the complementary use of various analytical instruments.

Sasaki's group extensively uses transmission electron microscope (TEM), focused ion beam scanning electron microscope (FIB-SEM) and three-dimensional atom probe (3DAP).

TEM provides the information on the structure and elemental distribution with nano- to atomic-scale resolution using an electron beam transmitted through the sample and characteristic. SEM, although inferior in resolution to TEM reveals the microstructures in 3D SEM images can be obtained by serial sectioning, which repeats microfabrication of the sample surface using FIB and SEM observation. FIB-SEM can be also used for site-specific specimen preparation for TEM and 3DAP analysis. 3DAP, on the other hand, is a state-of-theart nanoanalytical instrument that can map out the three-dimensional elemental distribution with atomic resolution to draw "nanoscale three-dimensional elemental maps.

detail at the micro- to nanoscale. In addition,

"While 3DAP is a powerful tool for three-dimensional analysis of elemental distribution at the nanoscale, it is not suitable for analysis of crystal structures, and elemental distribution at the microscale," Sasaki said. "For a full understanding of the microstructure, we use TEM and/or SEM, complementing each other's weaknesses."

Contributing to research on next-generation HDD read head

The Nanostructure Analysis Group is responsible for microstructure analysis and assists in proposing guidelines for designing high-performance materials being developed in the Research Center for Magnetic and Spintronic Materials. Their contributions are greatly appreciated by the researchers involved in materials development.

Nanostructure Analysis Group

One of the examples is the nanostructure analysis of CPP-GMR (current-perpendicular-to-the-plane giant magnetoresistance) devices, which could be applicable as read heads in hard disk drives (HDD), etc. Very thin three-layer structure consisting of ferromagnetic and non-magnetic metal layers is the core component of these devices.

"This collaboration began when Yuya Sakuraba (Magnetic Functional Device Group; see p. 8) visited me with his CPP-GMR devices," Sasaki said. "He was looking for clues on how to further increase the magnetoresistance ratio. Then, I analyzed the element distribution and interface structure between the ferromagnetic and non-magnetic metal layers in the device on an atomic scale using a combination of scanning transmission electron microscopy (STEM) and energy dispersive X-ray spectroscopy (EDS) (Figure 1)."

The microstructure information was then

shared with Yoshio Miura (Spin Theory Group; see p. 11)—an expert in first-principles calculations. By comparing the microstructure data with the results of first-principle calculations, Sakuraba was able to gain greater insight into the ideal microstructures for increasing the magnetoresistance ratio of his devices.

Based on these studies, the Magnetic Functional Device Group succeeded in developing a CPP-GMR device with the world's highest performance at low temperatures. The three groups continue to collaborate with the goal of achieving another world record.

Proactively developing high-performance magnets with less scarce elements

The activities of Sasaki's group are not limited to assisting with materials development through microstructure analysis; they also leverage the knowledge gained from microstructure analysis to carry out materials development themselves. Among these projects is the development of neodymium magnets, which they started about a decade ago. Neodymium magnets—the strongest permanent magnets currently available—are composed mainly of neodymium (Nd), iron (Fe) and boron (B). Demand for neodymium magnets has been rapidly growing in recent years due to their use as traction motors in electric vehicles (EVs).

The magnetic properties of neodymium magnets decrease with increasing temperature, and coercivity is used as an indicator of their



Grain boundary



Figure 2. (Left) SEM image of a Dygrain boundary diffusion treated neodymium magnet. (Right) STEM-EDS image of the vicinity of the grain boundary. The colored areas indicate the presence of elements. While the iron (Fe) concentrations at the grain boundary are low, neodymium (Nd) and praseodymium (Pr) concentrations are high. It can also be seen that Dy is enriched only on the grain surface.

heat resistance. Because EV motors operate at temperatures above 150°C, the neodymium magnets used in these applications are doped with dysprosium (Dy)—a heavy rare earth element—to improve their heat resistance.

However, due to the scarcity of Dy, reducing the amount of Dy used is an urgent issue. NIMS has been addressing this issue for many years by investigating the relationship between microstructures and coercivity through in-depth multi-scale microstructural analyses. Through these efforts, NIMS has made a number of discoveries that have led to the development of magnet design guidelines.

"Some time ago, the high concentrations of Nd enrichment at the grain boundaries were thought to be the origin of the coercivity development. However, in 2012, Hossein Sepehri-Amin (see p. 14) and his colleagues found that significant amounts of Fe were present at the grain boundaries and suggested that reducing the Fe content could improve the coercivity of neodymium. To verify these findings, we thoroughly analyzed their microstructure and magnetic properties at the grain boundaries of commercial neodymium magnets and those with higher coercivity by doping with gallium (Ga) using SEM, TEM and 3DAP. We found that there were smaller amounts of Fe at the grain boundaries in the high-coercivity Ga-doped magnets than in the low-coercivity commercial magnets," said Sasaki.

Sasaki's group then started to improve the coercivity of the neodymium magnets while reducing Dy content. Instead of alloying Dy, they adopted the grain boundary diffusion process of eutectic alloys in which Dy, praseodymium (Pr), and copper (Cu) are diffused along the grain boundaries as established by Sepehri- Amin and his colleagues in 2010.

"We succeeded in achieving excellent coercivity with only 10% the amount added compared to Dy-alloyed neodymium magnets. Our SEM and TEM analyses revealed that the grain boundary Fe contents were indeed reduced and Dy was found only locally on the surfaces of individual grains (Figure 2)," said Sasaki.

Other ongoing projects in the Nanostructure Analysis Group include the advancement of analytical techniques, such as three-dimensional imaging of hydrogen distribution at nanoscale using 3DAP, and the automation of the specimen preparation for TEM and 3DAP analysis. Algorithms are being developed to enable the instruments, e.g. FIB-SEM, to fully automate the "artisanal" process of preparing specimens from a target area— processes that have relied on the skill and experience of experts (see NIMS NOW vol. 23, no. 1, p. 15).

"The strength of NIMS is that when researchers involved in materials development want to look at the microstructures, we're here to do the microstructure analysis in a timely manner and work with them to find clues to improve the properties," Sasaki said. "In addition, we can also accelerate materials development by engaging in materials development ourselves." He will continue to contribute to the Center's materials research while continuing to develop the group's research.

Z. Wang et al., Acta Mater., 248, 118774 (2023).

Figure 1. Interface structure and elemental distribution of a CPP-GMR device using half-metallic Heusler alloy as ferromagnetic layers.

his device is composed of ferromagnetic layers made of Co,FeGa, Ge, Ag-a half-metallic Heusler alloyand a non-magnetic layer made of silver (Ag). The device was first analyzed at low magnification using a scanning transmission electron microscope (STEM evaluate the thickness of its ferromagnetic and non-magnetic layers and the roughness of the interfaces, followed by an analysis of the interface structure and element distribution at the interfaces at the atomic scale using a STEM and energy dispersive X-ray spectroscopy (EDS). These analyses revealed that the ferro magnetic layer terminates with a layer of Co atoms and that a layer of alternating Ga and Ag atoms is formed between the Co and Ag layers. These results provided clues to further improve the magnetoresistance ratio. B. Büker et al., Phys. Rev. B, 103, L140405 (2021).

NIMS NEWS

Report on NIMS Award Symposium 2023

On 6th and 7th November 2023,

NIMS held the academic symposium entitled "NIMS Award Symposium 2023".

NIMS has been awarding "NIMS Award" to researchers or groups that had made outstanding achievements in materials research field since 2007, and for 2023, the 16th. The candidates for NIMS Award were nominated by world-leading scientists and fairly reviewed by the NIMS Award Committee which consists of neutral experts.

NIMS Award 2023 went to Prof. Dierk



NIMS Award Commemorative Lecture by Prof. Dierk Raabe

Raabe (Director of Max-Planck-Institut für Eisenforschung GmbH (Germany)) and he gave a commemorative lecture at the symposium.

Invited lectures were given by Prof. Sudarsanam Suresh Babu of the University of Tennessee, Knoxville (USA), Prof. Simon P. Ringer of The University of Sydney (Australia), Prof. Sung-Joon Kim of Pohang University of Science and Technology (Korea) and Prof. Tadashi Furuhara of Tohoku University, as introduction to leading research in the structural materials research field.

In addition, there were presentations by prominent Japanese invited speakers and NIMS researchers, and 85 posters were presented.



Prof. Raabe, NIMS Award winner, received commemorative medal from Dr. Kazuhiro Hono, NIMS president.

During the core time of the poster session, participants engaged in a vigorous exchange of opinions on the latest research results, and provided the participants a fruitful good opportunity for the academic exchange that contributed to the creation of new global network.





Invited talk by Prof. Sudarsanam Suresh Babu



Invited talk by Prof. Sung-Joon Kim



Invited talk by Prof. Tadashi Furuhara

Research Center for Magnetic and Spintronic Materials

MSNOW INTERNATIONAL

2023 No. 6

NIMS researchers were selected as "Highly Cited Researchers 2023"

Four researchers of NIMS and three related researchers were selected as "Highly Cited Researchers in 2023" by Clarivate Analytics Inc.

Highly Cited Researchers have demonstrated significant and broad influence reflected in their publication of multiple highly cited papers. These highly cited papers rank in the top 1% by citations for a field or fields.



Katsuhiko Ariga Jonathan P. Hill Takashi Taniguch Kenji Watanabe



My name is Cédric Bourgès, a French researcher working on functional ceramics materials for energy-saving applications. Following my Ph.D. graduation, I got the chance to have my first postdoctoral experience in Japan as JSPS Fellow in Tohoku University, Sendai which made me discover this beautiful country. I experienced a culture rich and attractive, from the private and professional point of view, pushing me to extend my stay. I successfully integrated NIMS in 2019 in the group of Dr. Takao Mori and discovered a more international environment stuffed with the highest level of facilities which created an inextinguishable flame of motivation to give the best of me as a scientist. Thanks to the numerous opportunities offered by NIMS, I have applied and been accepted as ICYS fellow in 2022 which gave me a chance to prove my capacity as an independent researcher with the best support that the institute can provide! Since my arrival in Japan, I always enjoyed my daily life, discovering wonderful places between the mountains and the sea, and feeling comfortable and safe due to the kind and respectful people. Living in Japan, the place where my son is born, is a dream come true...



BOURGÈS Cédric (France) ICYS research fellow

Enjoy family time around the windmill of Kasumigaura Park in Tsuchiura.



NIMS NOW International 2023. Vol.21 No.6 National Institute for Materials Science

http://www.nims.go.jp/eng/publicity/nimsnow/

©2024 All righit reserved by the National Institute for Materials Science photo by Naohiro Tsukada (STASH) (cover, p.2), Michihito Ishikawa (p.6-15) editorial design by Barbazio Inc.

on the cover: Thermoelectric permanent magnet (see p.2)

To subscribe, contact: Dr. Yasufumi Nakamichi, Publisher Public Relations Office, NIMS 1-2-1 Sengen, Tsukuba, Ibaraki, 305-0047 JAPAN Phone: +81-29-859-2026, Fax: +81-29-859-2017, Email: inquiry@nims.go.jp



ISSN 2436-3510