Nanoscaled magnetism probed by synchrotron-radiation spectromicroscopy

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Synchrotron radiation (SR) provides high-brilliance, energy-tunable, and polarization-controllable X-ray that is powerful probe for materials science. X-ray magnetic circular dichroism (XMCD) spectroscopy using circularly polarized X-rays has been a popular experimental technique to investigate element-specific magnetic properties. By leveraging XMCD, variational techniques such as X-ray resonant magnetic reflectometry (XRMR) and X-ray-detected magnetic resonance (XFMR) are utilized to investigate layer-resolved magnetic properties or dynamics of spin presession. Magnetic contrast comes from XMCD is also utilized in X-ray magnetic imaging such as transmission X-ray microscopy (TXM) or scanning transmission X-ray microscopy (STXM). We performed XMCD-STXM experiment of several permanent magnet materials by using a STXM instrument at the Photon Factory, High-Energy Accelerator Research Organization (KEK)[1]. Magnetic domains of a commercial $SmCo_5$ magnet[2] and single-phase $SmCo_5$ magnet[3] are observed by STXM-XMCD. XMCD spectra in nanometer scale are obtained and element-specific spin and orbital magnetic moments are evaluated.

Synchrotron-radiation Mössbauer spectroscopy is another powerful technique to microscopic magnetism. By embedding a resonant isotope probe layer to atomic scale films, SR Mössbauer spectroscopy enables direct detection of local magnetic properties. Magnetic Friedel oscillation at the Fe(001) surface was observed by using *in situ* SR Mössbauer spectroscopy system developed at BL11XU of SPring-8[4].

New SR beamline for magnetic and spintronic materials BL-13U is now under construction at the latest SR facility of Japan, NanoTerasu in Sendai. BL-13U offers X-ray with wide energy range of 180–3000 eV and high-speed polarization switching[5]. This beamline will provide opportunities for state-of-the-art spectromicroscopy experiments on nanoscaled magnetism.

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Atomic-scale surface and interface magnetism based on ferromagnetic monatomic layer iron nitride

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A scanning tunneling microscopy (STM) is a unique tool to investigate surface structural and electronic properties of objects on the atomic scale. By utilizing tunneling current or voltage between the STM tip and sample, STM can also manipulate object's morphology and electronic states. Furthermore, STM allows to extract object's magnetic properties with atomic precision by coating the STM tip with magnetic materials (spin-polarized STM). We have conducted surface science researches and especially investigated atomic-scale spin related phenomena such as spin dynamics and stability of single atoms [1], multifunctional single molecule magnetic memory [2] by STM. In recent years, we extend our research to the fields of materials science and spintronics for practical purposes. Taking advantage of state-of-the-art STM techniques, we represent the first precise real-space determination of the wall width $Nd_2Fe_{14}B$ hard permanent magnets by spin-polarized STM [3]. In addition, we reveal for artificial lattices composed of ferromagnetic Fe and antiferromagnetic Mn thin films that atomic-scale surface and interface characterizations by spin-polarized STM can be effectively connected to macroscopic magnetic properties to achieve a comprehensive understanding of magnetic properties of the whole system [4]. In relation to the above our research activities, in this talk, we introduce a combined work of STM and synchrotron x-ray absorption spectroscopy/x-ray magnetic circular dichroism (XAS/XMCD) of the systems based on iron nitride, which has high potential as next-generation rare earth free permanent magnets. The results demonstrate the importance of a complementary microscopic and macroscopic approach to fully understand surface and interface magnetism toward developing novel magnetic thin film materials.

＜Sub-nanometer scale characterizations of point defects in monatomic layer iron nitride＞

Iron nitride compounds in the iron rich phases such as $Fe_{16}N_2$ or Fe_4N, have been intensively studied as promising candidates for the rare-earth free permanent magnet due to their large magnetic anisotropy and room-temperature ferromagnetism. However, the impact of morphological changes of iron nitride from bulk to thin films on electronic and magnetic properties has been rarely identified. Furthermore, as the film thickness decreases down to atomic layers, spatial modulations of electronic and magnetic properties caused by microscopic structural changes become non-negligible. We have intended to fabricate a monatomic layer of iron nitride with the Fe_4N stoichiometry (Fe_2N) grown on Cu(001) and investigated its structural and electronic properties on the atomic scale by STM. We here experimentally discovered orbital selective tunneling effect by STM; the information on the orbital characters (e.g. s, p, d orbitals) of the surface electronic states can be separately extracted with atomic precision by the strict control of the STM tip-sample distance [5]. The results reveal the dominant role of microscopic electronic structures derived from the strong Fe-N bonding on the magnetism of Fe_2N. In addition, we find from sub-nanometer scale STM spectroscopy across a point defect on Fe_2N that the point defect modulates surface electronic states up to third nearest neighbor Fe atoms. In combination with the results of XAS/XMCD measurements and theoretical calculations, it is suggested that the point defect, which induces atomic-scale spatial modulation of electronic states of Fe_2N, acts as "functional core" and enhances the spin magnetic moment of surrounding Fe atoms.

＜Fabrication of high-quality FeCo ordered alloy thin films assisted by nitrogen surfactant epitaxy＞

An $L1_0$-type FeCo ordered alloy attracts much attention as a rare-earth free magnetic material due to its large magnetic anisotropy, large magnetic moment, and high Curie temperature. However, the $L1_0$ structure is a non-equilibrium state of FeCo phase and hence even its fabrication method has not been established yet. The main problem could be caused by the atomic-scale disorder at the Fe/Co interface during growth processes. Thus, unambiguous microscopic characterizations of structural, electronic and magnetic properties of $L1_0$-FeCo are required toward realizing high-quality $L1_0$-FeCo thin films. To improve the quality of FeCo ordered alloy thin films, we intend to incorporate the nitrogen surfactant effect of monatomic layer iron nitride (Fe_2N) into the alternate atomic-layer Fe and Co deposition. In this method, the nitrogen surfactant effect of Fe_2N with high lateral lattice stability can effectively suppress the interdiffusion at the Fe/Co interface during the deposition and annealing processes, which results in atomically flat surface and interface. We have revealed the validity of the nitrogen surfactant epitaxy on the fabrication of high-quality $L1_0$-type alloy thin films for FeNi [2, 3]. In this work, we grow 1 monolayer (ML) Co on Fe_2N/Cu(001), which is the initial stage for the fabrication of FeCo ordered alloy thin films by means of nitrogen surfactant effects. The correlation between the structural changes at the Fe/Co interface and magnetic properties upon sample heat treatment up to 370, 570, 620, and 670 K is investigated by STM and XAS/XMCD. First, the XMCD signal of bare Fe_2N is element specifically extracted. We confirm that bare Fe_2N shows greater XMCD signal in the grazing geometry than that in the normal geometry, revealing its strong in-plane magnetic anisotropy as previously reported [7]. Adding 1 ML Co activates the nitrogen surfactant effect, leading to structural transformation from Co/Fe_2N/Cu(001) to CoN/Fe/Cu(001). Accordingly, the enhancement of the out-of-plane magnetization of the Fe layer is observed. The perpendicular magnetic anisotropy is further enhanced by annealing up to 570 K. Atomically-resolved STM observations reveal that, while ordering of the Fe/Co interface is improved up to the annealing temperature of 570 K, point defects appear at 620 K and the interdiffusion of Fe and Co atoms takes place at 670 K. Combining XAS/XMCD and STM observations, we find that the microscopic structural changes on annealing, which cannot be accessed by conventional macroscopic techniques such as low energy electron diffraction, dominantly determine the macroscopic magnetic properties of the whole system. The results demonstrate that the nitrogen surfactant epitaxy efficiently suppresses the interdiffusion at the Fe/Co interface and keep atomically flat surface/interface, and possibly reflect the intrinsic out-of-plane magnetization of $L1_0$ FeCo.

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Automated interpretation of magnetic domain structure using feature extended Landau free energy model

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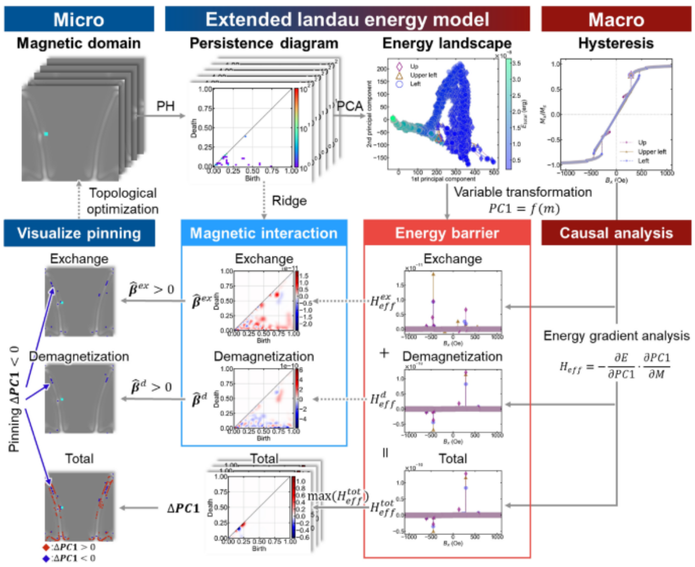
Interpretating magnetic domain structure data is of paramount significance as it offers critical insights into the functionalities of a wide array of magnetic materials. This understanding is a cornerstone for designing next-generation electronic devices, capable of achieving both high-speed performance and low power consumption. However, the inherent complexity of interactions in nanoscale magnetic materials often poses daunting challenges. Understanding the underlying mechanisms or pinpointing specific locations through human observation is strenuous, often relegating device design to an iterative, trial-and-error process.

To circumvent these challenges, we have formulated an innovative "Extended Landau Free Energy Model". This model harmoniously blends the principles of topology and data science, thereby automating the interpretation of image data [1-6]. Through its application, we discerned that the anti-magnetic field effect predominantly steers the process of information recording in nanomagnets. The model has also enabled us to successfully visualize the spatial distribution of energy barriers that inhibit efficient information recording.

Building upon these findings, our model has further facilitated the proposal of a novel device structure, characterized by markedly reduced energy consumption. Endowed with high explanatory prowess and deeply rooted in the principles of physics, this machine learning model holds substantial promise. It is anticipated to be instrumental in elucidating the mechanisms of diverse materials where they currently remain undefined.



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Interfacial Imaging on Magnetic Junctions by Electron Microscopy

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In spintronics, a large magnetoresistance (MR) ratio is a key parameter to achieve better performance in magnetic random access memory (MRAM) and a magnetic bipolar operation.[1)] Theoretically coherent tunnelling has been predicted to exceed 1,000% at room temperature.[2),3)] However, experimentally recent demonstration of 631% tunnelling MR (TMR) ratio using a CoFe/MgO/CoFe(001) magnetic tunnel junction (MTJ) is the largest reported to date.[4)] To investigate the origin of this departure, we categorised MTJs into four types; (a) polycrystalline, (b) epitaxial, (c) lattice softened and (d) lattice matched (without dislocations) MTJs.[5)] We then characterised the differences in their interfaces using cross-sectional transmission electron microscopy (TEM) and non-destructive imaging by scanning electron microscopy (SEM).

We correlated their interfacial crystalline structures with their local magnetic properties, namely spin-polarised electron transport. The reduction in a TMR ratio was found to be caused by the density of interfacial dislocations and disorder.. In our study, the minimum dislocation density has been achieved at a $Co_{0.2}Fe_{0.6}B_{0.2}$/MgO interface. This has been achieved by the post-annealing of an amorphous junction rather than growing a junction epitaxially. On the other hand, a half-metallic ferromagnet has shown larger TMR ratios at low temperature but the presence of disordered phases formed at MgO interfaces induces large spin fluctuation with increasing temperature (see Fig. 1). For the improvement of the MR ratios, it is critical to eliminate such interfacial dislocations and disordering.

One approach is to use a ferromagnetic layer with soft lattice nature. We found a Co-Mn alloy can reduce the interfacial dislocation significantly against the MgO barrier, which is also found to hold soft lattice nature.[6)] Our findings can be fed back to the growth and fabrication processes of MTJs for their optimisation and improvement.

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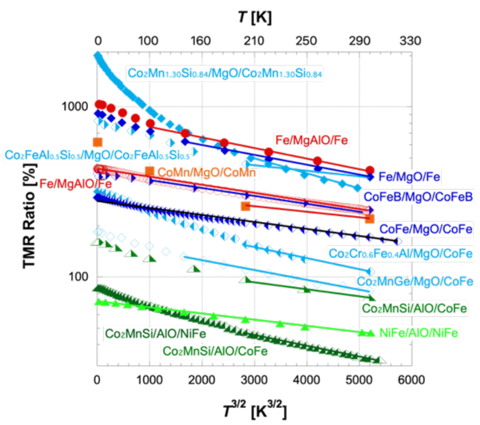

Fig.1 Temperature-dependence of TMR ratios of four types of MTJs.[5)]

Nanostructure characterization of magnetic materials by SEM/TEM/APT

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For the development of high-performance magnetic materials, microstructure control is effective in improving microstructure-sensitive properties such as coercivity, magnetization, magnetoresistance (MR) ratio, *etc*. Because the microstructures that influence properties could exist over a wide range of micro to atomic scales, their direct visualization requires the complementary use of different microscopy techniques. This talk will present examples of microstructural analysis of neodymium magnets, current-perpendicular-to-plane giant magnetoresistive (CPP-GMR) devices, and nanocrystalline soft magnets, using scanning electron microscope (SEM), transmission electron microscope (TEM), and atom probe tomography (APT), and discuss how an in-depth understanding of the microstructure is effective in proposing guiding principles for the enhancement of the important properties in applications.

(1) Nd-Fe-B magnets - Coercivity enhancement by grain boundary diffusion (GBD) process[1)]

GBD process is a way to improve the coercivity with minimal use of heavy rare-earth (HRE) elements. The coercivity enhancement is attributed to the HRE-rich shell formed on the surface of $Nd_2Fe_{14}B$ grains, Fig. 1. However, the coercivity enhancement has been limited to ~2 T when the HRE-free sintered magnets are treated with the Dy-GBD process. We clarified the formation mechanism of the HRE-rich shell through SEM and TEM analyses and learned that further coercivity enhancement is expected by increasing the HRE content in the HRE-rich shell. Then, we applied the two-step GBD process, originally proposed in the study of hot-deformed magnets, to demonstrate a high coercivity of 2.8 T in a fine-grained sintered magnet.

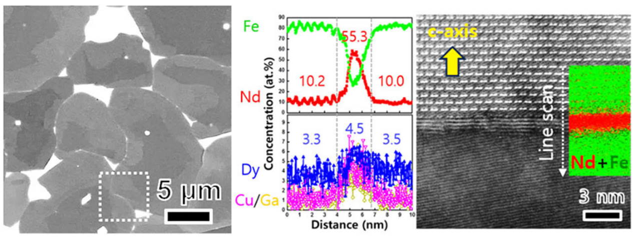

Fig. 1: SEM image showing HRE-rich shell formation on $Nd_2Fe_{14}B$ grain surface and HAADF-STEM and EDS elemental map of a sintered magnet treated with Dy-GBD.

(2) Influence of sample shape on APT analysis of soft magnetic nanocrystalline materials[2)]

APT can map out the elemental distribution in nanoscale in 3D. However, the laser irradiation conditions significantly affect the mass resolution because the laser-assisted field evaporation occurs. Recently, we found that the laser conditions, the material's thermal conductivity, and the tip geometry, such as tip radius and taper angle, affect the quantitatively. I will show an analysis result of soft magnetic nanocrystalline material and the technique to improve the quantitatively.

(3) CPP-GMR devices - Effect of atomic-scale interfacial structure on the banding matching and MR properties

Electronic band matching at the interface between ferromagnetic and nonmagnetic metals affects the spin-dependent transport properties such as the giant magnetoresistance (GMR) effect. Jung *et al.* found the insertion of very thin 0.21-nm-thick NiAl layers at the $Co_2FeGa_{0.5}Ge_{0.5}$(CFGG)/Ag interfaces leads to an increase in MR ratio[4)]. We analyzed the CFGG/Ag interfaces with Ni insertion using STEM/EDS analysis and revealed the interface structure in atomic scale, *i.e.*, the Co atoms in a second termination layer from the Ag interface are replaced with Ni monolayer, Fig. 2[5)]. The proposed structure is implemented into first-principles calculations of ballistic transmittance and revealed that substituting the Co termination layer with Ni improved electronic band matching of majority spin electrons.

As such, microstructure analysis reveals the effects of micro- to even atom-level microstructure on properties, allowing the design of higher-performance magnetic materials.

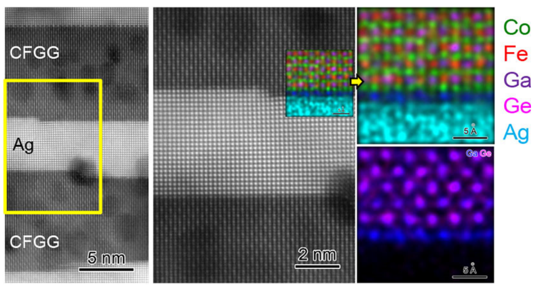

Fig. 2: HAADF-STEM image of a CFGG/Ag/CFGG pseudo-spin valve and atomic-resolution EDS elemental map.

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Voltage-control of magnetization dynamics by using topological insulators

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The voltage control of magnetic properties is both fundamentally and technologically important for next generation magnetic devices such as magnetoresistive random-access memories (MRAMs) and spin-wave-based logic devices. Voltage control of magnetic anisotropy (VCMA) in magnets promises energy-efficient reversal of magnetization in MRAMs by means of the so-called voltage torque. The voltage control of magnetic damping is also desirable to increase the performance of spin-wave-based logic gates and magnon-based transistors.

Herein, we theoretically study the voltage control of magnetic anisotropy by using a contact of three-dimensional topological insulators (TIs). We formulate a uniaxial magnetic anisotropy[1)] and effective damping constant[2)] at the ferromagnet/TI interface as a function of an applied voltage[3)]. We proposed the field-effect-transistor (FET)-like devices which consists of TI and magnetic-topological-insulator (MTI) as shown in Fig. 1. We also demonstrate a reversal of magnetization by using the TI-based voltage-control of magnetic properties. This device realizes magnetization switching via spin-orbit torque (SOT) and VCMA which originate from 2D-Dirac electronic structure. We theoretically investigate influences of electronic circuit delay, noise, and temperature on write-error-rate (WER) in voltage-controlled magnetization switching operation of an MTI-based device by means of the micromagnetic simulation[4)]. We reveal that the device operation is extremely robust against circuit delay and signal-to-noise ratio. We demonstrate that the WER on the order of approximately 10^{-4} or below is achieved around room temperature due to steep change in VCMA as shown in Fig. 2. Also, we show that the larger SOT improves thermal stability factor. This study provides a new perspective for developing voltage-driven spintronic devices with ultra-low power consumption.

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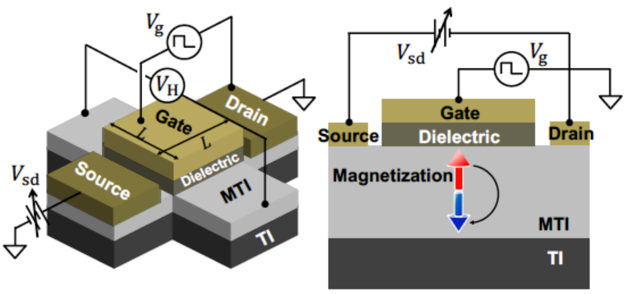

Fig. 1 Schematic illustration of an MTI-based device with a FET-like structure consisting of magnetic-TI (MTI) and TI film.

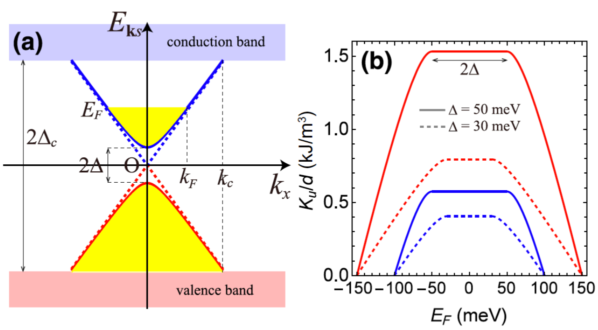

Fig. 2 (a)Massless (dashed line) and massive (solid line) surface state dispersions, (b) Scaled magnetic anisotropy energy K_u/d as a function of Fermi level E_F for different values of bulk gap and surface gap.