



## **Symposium on Frontier Researches of Functional Oxide Devices and Materials**

March 16, 2018-----Kobe, Japan

Organizers: Azusa N. Hattori (Osaka University)

Miho Kitamura (Photon Factory, KEK)

### **Scope**

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Metal oxides show various functionalities that could contribute to realize the nonvolatile memory devices with low power consumption, memristor, synaptic transistor and so on.

Behind recent advances and emerging trends in the field of multifunctional complex oxides and their utilization in electronic devices, both experimental and theoretical studies at surface/interface are essential.

In the upcoming Symposium on Frontier Researches of Functional Oxide Devices and Materials, the recent advances in the fundamental study and the application of metal oxides will be summarized by internationally recognized experts, who are exploiting functional oxides in their research in order to obtain novel materials with improved properties for electronics, magnetics, and other growing disciplines.

**Venue:** Andalucia, Ariston Hotel Kobe, Japan

**Fee:** ¥1,000 yen, Free for Student and EDTM member

### **Registration:**

mail to [foe-edtm@sanken.osaka.u-ac.jp](mailto:foe-edtm@sanken.osaka.u-ac.jp) with your Name and Affiliation

## Program

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		Chair	Miho Kitamura (KEK)
10:00~10:05	Hidekazu Tanaka (Osaka Univ.)		Opening remark
	Azusa N. Hattori (Osaka Univ.)		Introductory talk
10:05~10:50	Shik Shin (Univ. of Tokyo)		Keynote
	“Nondestructive operando nano-imaging of the resistivity switching in ReRAM by the development of ultrahigh resolution laser-photoelectron microscopy”		
10:50~11:20	Shinji Miwa (Osaka Univ.)		Invited
	“Electric-field-induced effect in Fe/MgO-based multilayer devices”		
11:20~11:50	Yusuke Wakabayashi (Osaka Univ.)		Invited
	“Surface structure analysis based on X-ray diffraction: solid-solid and solid-liquid interfaces”		
11:50~12:20	Naoya Shibata (Univ. of Tokyo)		Invited
	“Advanced atomic-resolution electron microscopy for real-space electromagnetic field imaging”		
12:20~13:50	Lunch		
		Chair	Azusa N. Hattori (Osaka Univ.)
13:50~14:35	Shriram Ramanathan (Purdue Univ.)		Keynote
	“Organismic Quantum Matter and Learning”		
14:35~15:05	Qiangfei Xia (Univ. of Massachusetts)		Invited
	“Hafnium Oxide and Silicon Oxide based Memristors”		
15:05~15:35	Michiko Yoshitake (NIMS)		Invited
	“Materials Curation <sup>TM</sup> : Examples for Metal Oxides”		
15:35~15:50	Break (conference photo)		
		Chair	Jobu Matsuno (RIKEN)
15:50~16:20	Masahito Mochizuki (Waseda Univ.)		Invited
	“Dynamical Phenomena and Device Functions of Magnetic Skyrmions”		
16:20~16:50	Ho Nyung Lee (Oak Ridge National Lab.)		Invited
	“Emergent Phenomena in 3d-5d Oxide Quantum Heterostructures”		
16:50~16:55	Hiroshi Kumigashira (KEK)		Closing remark

## Abstract

### ----- Nondestructive operando nano-imaging of the resistivity switching in ReRAM by the development of ultrahigh resolution laser-photoelectron microscopy

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Improvement of the reliability is one of the major challenges for ReRAM devices. The conductive filaments formation made by the redox cycle in the ReRAM plays the main role in the switching between high and low electronic resistivity states. However, it has been very difficult to observe the nondestructive measurements of the electronic states beneath the top electrodes under the device operation. Here, we succeeded to develop the ultra-high resolution photoelectron microscopy by using the CW VUV-laser (Laser-PEEM)<sup>1</sup> whose spatial resolution reached about 2.6 nm. The most prominent merit of the laser-PEEM has the ability to perform the nondestructive nano-imaging measurements of the electronic states under the operando condition, since the ultra-low energy photoelectron is very bulk-sensitive and it can penetrate the top electrode. We observed the several kinds of conductive filaments in ReRAM under the operando condition for the first time. The study of their formation mechanisms will contribute the reliability of the ReRAM devices.

Laser PEEM is also very powerful for the observation of magnetic domains. We will show the ferromagnetism of the interface of the LaTiO<sub>3</sub>/SrTiO<sub>3</sub> and several ferromagnetic and antiferromagnetic domain structures by using laser-PEEM.

1. T.Taniuchi, Y.Kotani, S.Shin, Rev. Sci. Instrum. **86**, 023701(2015).
2. T.Taniuchi, S. Shin, et al., Nature Commun. **7**, 11781(2016).

## Electric-field-induced effect in Fe/MgO-based multilayer devices

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An electric field at a surface is known to exhibit useful phenomena, such as the pinch-off phenomenon in field-effect transistors, inductions of Mott transitions, superconducting phases, ferromagnetic phases and magnetic anisotropy. All of these phenomena are attributable to one or both of two factors: electron accumulations or electric dipole inductions. In particular, electric-field control of magnetic properties at room temperature attracts much attention because of its great potential for enabling the construction of ultralow-power-consumption electric devices. Voltage-controlled magnetic anisotropy (VCMA) in Fe/MgO-based tunnel junctions [1] has shown that magnetization of nanomagnets can be controlled in fast periods (down to 0.1 ns) by electric fields, as indicated by a bistable precessional magnetization switching [2] and ferromagnetic resonance excitation [3]. The VCMA can be an ultimate technology for the operation of spintronics devices, such as a nonvolatile random access memory.

In this talk, we will show our recent progress on VCMA research. Firstly, we show VCMA-driven spin-torque utilizing interface state at Fe/MgO [4]. Secondly, we show a new mechanism to enhance VCMA in Fe/Pt/MgO tunnel junction proved by X-ray magnetic circular dichroism spectroscopy, which is an electric-quadrupole-induced VCMA [5,6].

1. T. Maruyama et al., Nat. Nanotechnol. **4**, 158 (2009)
2. Y. Shiota et al., Nat. Mater. **11**, 39 (2012)
3. T. Nozaki, SM et al., Nat. Phys. **8**, 491 (2012)
4. S. Miwa et al., Phys. Rev. X **7**, 031018 (2017).
5. S. Miwa et al., Nat. Commun. **8**, 15848 (2017)
6. T. Kawabe, SM et al., arXiv:1708.08549

## Surface structure analysis based on X-ray diffraction: solid-solid and solid-liquid interfaces

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Structural information is always required to fabricate specific materials; without having it, one cannot make designed structure. Especially, the structure of solid-solid or solid-liquid interfaces are challenging objects, because of limited way to measure whereas the functionality of them are huge. In order to study such interfaces, surface x-ray diffraction is potentially useful. It is a non-contact method, and the penetration depth of x-ray is large enough to measure the solid-liquid interface. The main disadvantage of this method is the difficulty in analysis. Here, the achievements which is provided by the recent development of the analyzing technique are presented. The first example is the epitaxial interfacial structure of perovskite oxides. Using the support of informatics, structure of  $\text{LaAlO}_3/\text{SrTiO}_3$  interface was clarified[1]. Second example is the structure of electric double layers formed at the gold/ionic liquid interface[2]. Surface x-ray diffraction is applicable on both cases. Our recent development on the analyzing technique will be presented.

1. M.Anada Y. Nakanishi-Ohno, M. Okada, T. Kimura and Y. Wakabayashi, *J. Appl. Cryst.*, **50**, 1611 (2017).
2. R. Yamamoto, H. Morisaki, O. Sakata, H. Shimotani, H.-T. Yuan, Y. Iwasa, T. Kimura, and Y. Wakabayashi, *Appl. Phys. Lett.* **101**, 053122 (2012).

## **Advanced atomic-resolution electron microscopy for real-space electromagnetic field imaging**

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Scanning transmission electron microscopy (STEM) boosted by aberration-correction technology has made possible to directly observe atomic structures in many materials and devices. In STEM, a very finely focused electron probe is scanned across the specimen and the transmitted and/or scattered electrons at each raster position are detected by the post-specimen detector(s) to form images. By elaborating special detector geometries, we not only image atomic structures of materials, but also can image local electromagnetic fields through differential phase contrast (DPC) imaging techniques. By developing high-speed segmented type detectors, DPC STEM imaging is now becoming very powerful to characterize materials and devices, such as pn junctions in semiconductor devices, electric fields of polar oxide interfaces and magnetic Skyrmions which cannot be observed by normal STEM imaging techniques using annular type detectors. In this talk, atomic-resolution DPC STEM and its prospects will be also discussed.

## **Organismic Quantum Matter and Learning**

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A hallmark of life is plasticity that enables reproduction, evolution and environmental adaptivity. We ask the question whether these remarkable features in nature can be realized in the physical world? We will first describe some examples of collective phenomena that ensures species survival in the animal kingdom emphasizing spatio-temporal dynamics. We will then contrast these observations with learning and decision making skills in non-neural organisms. Subsequently, we will describe our recent collaborative discoveries on adaptive oxide quantum materials that strikingly mimic organismic behavior and give examples of problems in machine intelligence, brain and behavioral sciences that may be emulated using such systems. Finally we will consider pathways for use of such synthetic matter to serve as a testbed to understand quintessential aspects of evolutionary dynamics that may be used to design animal-machine interfaces.

## **Hafnium Oxide and Silicon Oxide based Memristors**

Qiangfei Xia, Hao Jiang and Can Li

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We built a Ta/HfO<sub>2</sub> based device that has long retention time, superior endurance properties (120 billion cycles), and stable multiple level resistance switching behavior. We directly observed a Ta-rich O-deficient conductive channel that is responsible for the switching properties. Taking advantage of the analog switching behavior, we integrated the device with CMOS circuits, implementing large-scale crossbar arrays with demonstrated applications in high-precision signal and image processing.

We then developed an all-silicon based memristor with p-Si/SiO<sub>2</sub>/n-Si geometry. The device exhibits repeatable resistance switching with high ON/OFF conductance ratio ( $10^4$ ) and competitive retention at 300 oC. Furthermore, we observed self-rectifying switching behavior with a rectifying ratio up to  $10^5$ . We physically characterized the formation/rupture of a silicon rich conductive filament and attributed the self-rectifying behavior to the formation of a p-i-n diode within the junction. We further built 3-dimensinal stacked memristor arrays up to 5 layers of crossbars with high yield, and demonstrated effective blocking of sneak path currents in the arrays owing to the self-rectifying behavior.

1. H. Jiang et al., Sci. Rep. 6, 28525 (2016).
2. C. Li et al., Nat. Commun. 8, 15666 (2017).
3. C. Li et al., Nat. Electron. 1 (2018). Published online Dec. 4, 2017.



## Materials Curation™ : Examples for Metal Oxides

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Abstract (100-200 words)

We proposed a technique of searching materials from perspective views, Materials Curation™, that utilizes various relations between physical quantities interdisciplinary [1-3]. The composition dependence of work function values of metal carbides and nitrides has been predicted via this technique [4]. In this talk, how the relations are derived and utilized is briefly explained through examples in metal oxides.

Formation enthalpy of metal oxides is taken as an example, which is one of the basic quantities. Formation enthalpy connects to various material properties, such as energy band gap, redox potential in electrochemistry, adsorption energy of oxygen on metals and interface bonding.

The logic of the connections is presented and the connections are demonstrated through experimentally obtained values and experimental results.

1. M. Yoshitake, *KinouZairyou*, **33**, 48 (2013).
2. M. Yoshitake, presented in JSAP annual meetings, 27pB6-1 (2013.3), 19a-F4-1 (2014.3), 17p-A23-1 (2014.9), 11p-D13-2 (2015.3), 21p\_S322\_1 (2016.3).
3. M. Yoshitake, *International Journal of Science and Research*, **4**, 571 (2015).
4. M. Yoshitake, *J. Vac. Sci. Technol. A* **32**, 061403 (2014).

## Dynamical Phenomena and Device Functions of Magnetic Skyrmions

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Skyrmions, swirling topological spin textures in magnets, are currently attracting great research interest because they have numerous advantageous properties for application to high-performance magnetic memory devices. They exhibit several peculiar dynamical phenomena owing to their (i) finite topological charges, (ii) vortex-like nature, and (iii) particle-like nature. The peculiarities show up especially in their current-driven dynamics. In this talk, following two issues will be discussed.

(1) Magnon-current-driven dynamics [1]: Unidirectional rotation of skyrmions occurs in the presence of temperature gradient, whereas it is forbidden in thermal equilibrium. We theoretically argue that this rotation is driven by the topological Hall effect of diffusive magnon currents due to the emergent magnetic field from the skyrmion spin textures.

(2) Electric-current-driven dynamics [2,3]: Translational motion of skyrmions can be driven by spin-polarized electric currents in metallic chiral-lattice magnets via the spin transfer torque effect. The required current density turned out to be five or six magnitudes smaller than that required for driving ferromagnetic domain walls, which opens a route to spintronics applications. We argue that the skyrmion motion is affected neither by impurity pinning nor by damping effects. This surprisingly high mobility of skyrmions originates from their particle-like nature and finite topological charges.

[1] M. Mochizuki *et al.*, *Nature Materials* **13**, 241 (2014).

[2] J. Iwasaki, M. Mochizuki, N. Nagaosa, *Nature Commun.* **4**, 2391 (2013).

[3] J. Iwasaki, M. Mochizuki, N. Nagaosa, *Nature Nanotech.* **8**, 742 (2013).

## **Emergent Phenomena in 3d-5d Oxide Quantum Heterostructures**

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The controlled synthesis of heterostructures by epitaxial growth to create interfaces between a wide variety of oxide-based quantum materials provides tremendous opportunities for producing materials with remarkable physical properties and functionalities. The strong interfacial orbital hybridization and spin-orbit entangled magnetic interface point to correlated oxide superlattices as ideal test bed for interface-induced Dzyaloshinskii-Moriya (DM) interactions. Recent studies in the field of 3d-5d oxide heterostructures have revealed interesting physical properties at interfaces between iridates and manganites. In this talk, we will present our observations on (1) charge transfer induced magnetism and anomalous Hall effect (AHE) in non-inversion symmetry broken  $(\text{SrMnO}_3)_m(\text{SrIrO}_3)_n$  superlattices and (2) the topological Hall effect (THE) in inversion symmetry broken  $(\text{LaMnO}_3)_m(\text{SrIrO}_3)_n$  heterostructures. The critical role of the DMI and its interfacial control will be discussed together with the great potential of 5d TMOs for developing novel quantum materials and spintronic devices.

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