RESEARCH ACTIVITIES
Photo-Molecular Science

We study the interaction of atoms and molecules with optical fields with its possible applications to active control of atomic and molecular functionality and reactivity. We also develop novel light sources to promote those studies. Two research facilities, the Center for Mesoscopic Sciences and the UVSOR Synchrotron Facility, closely collaborates with the Department.

The core topics of the Department include attosecond coherent control of gas- and condensed-phase atoms and molecules, high-resolution optical microscopy applied to nanomaterials, synchrotron-based spectroscopy of core-excited molecules and solid-state materials, vacuum-UV photochemistry, and the development of novel laser- and synchrotron-radiation sources.

Ultrafast Quantum Simulator and Computer

Department of Photo-Molecular Science
Division of Photo-Molecular Science II

OHMORI, Kenji
Professor
[ohmori@ims.ac.jp]

Education
1987 B. E. The University of Tokyo
1992 Ph.D. The University of Tokyo

Professional Employment
1992 Research Associate, Tohoku University
2001 Associate Professor, Tohoku University
2003 Professor, Institute for Molecular Science
Professor, The Graduate University for Advanced Studies
2004 Visiting Professor, Tohoku University (–2005)
2007 Visiting Professor, Tokyo Institute of Technology (–2008)
2009 Visiting Professor, The University of Tokyo (–2011)
2012 Visiting Professor (Humboldt Awardee), University of Heidelberg
2014 Visiting Professor, University of Strasbourg (–2016)

Awards
1998 Award by Research Foundation for Opto-Science and Technology
2007 JSPS Prize
2007 Japan Academy Medal
2008 Norman Hascoe Distinguished Lecturer, University of Connecticut, USA
2009 Fellow of the American Physical Society
2012 Humboldt Research Award
2017 Hiroshi Takuma Memorial Prize of Matsuo Foundation
2018 Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology of Japan

Member
Assistant Professor
SUGAWA, Seiji
DE LÉSÉLEUC, Sylvain
IMS Research Assistant Professor
TOMITA, Takafumi
Post-Doctoral Fellow
BHARTI, Vineet
KUNIMI, Masaya
Graduate Student
MIZOGUCHI, Michiteru
CHEW, Yee Lai
TIRUMALASETTY PANDURANGA, Mahesh
VILLELA, Rene
Secretary
KAWAMOTO, Minako

Keywords Quantum-Classical Boundary, Quantum Simulation, Quantum Computing

It is observed in a double-slit experiment by Tonomura and coworkers that single electrons recorded as dots on a detector screen build up to show an interference pattern, which is delocalized over the screen.[1)] This observation indicates that a delocalized wave function of an isolated electron interacts with the screen, which is composed of many nuclei and electrons interacting with each other, and becomes localized in space. This change, referred to as “collapse” in quantum theory, is often accepted as a discontinuous change, but a basic question arises: When and how the delocalized wave function becomes localized? Our objective is uncovering this mystery by observing the spatiotemporal evolution of a wave function delocalized over many particles interacting with each other. Having this objective in mind, we have developed coherent control with precisions on the picometer spatial and attosecond temporal scales. Now we apply this ultrafast and ultrahigh-precision coherent control to delocalized wave functions of macroscopic many-particle systems such as an array of ultracold rubidium (Rb) Rydberg atoms, as depicted schematically in Figure 1 and named “ultrafast quantum simulator,” envisaging the quantum-classical boundary connected smoothly.

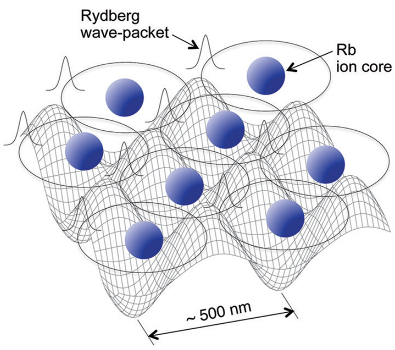


Figure 1. Metal-like quantum gas. A schematic of the many-body quantum simulator with ultracold Rydberg atoms, named “ultrafast quantum simulator,” where electronic wave functions spatially overlap between neighboring atoms.[2,7)]

Selected Publications

- H. Katsuki *et al.*, “Visualizing Picometric Quantum Ripples of Ultrafast Wave-Packet Interference,” *Science* **311**, 1589–1592 (2006).
- H. Katsuki *et al.*, “Actively Tailored Spatiotemporal Images of Quantum Interference on the Picometer and Femtosecond Scales,” *Phys. Rev. Lett.* **102**, 103602 (2009).
- K. Hosaka *et al.*, “Ultrafast Fourier Transform with a Femtosecond-Laser-Driven Molecule,” *Phys. Rev. Lett.* **104**, 180501 (2010).
- H. Goto *et al.*, “Strong-Laser-Induced Quantum Interference,” *Nat. Phys.* **7**, 383–385 (2011).
- H. Katsuki *et al.*, “All-Optical Control and Visualization of Ultrafast Two-Dimensional Atomic Motions in a Single Crystal of Bismuth,” *Nat. Commun.* **4**, 2801 (2013).
- N. Takei *et al.*, “Direct Observation of Ultrafast Many-Body Electron Dynamics in an Ultracold Rydberg Gas,” *Nat. Commun.* **7**, 13449 (2016).
- C. Liu *et al.*, “Attosecond Control of Restoration of Electronic Structure Symmetry,” *Phys. Rev. Lett.* **121**, 173201 (2018).
- M. Mizoguchi *et al.*, “Ultrafast Creation of Overlapping Rydberg Electrons in an Atomic BEC and Mott-Insulator Lattice,” *Phys. Rev. Lett.* **124**, 253201 (2020).

1. Development of an "Ultrafast Quantum Simulator" by Optical Control with Precisions on the Attosecond Temporal and Submicron Spatial Scales[3–10]

Quantum many-body problems are at the heart of a variety of physical functionalities including superconductivity and magnetism in solid materials. It is extremely hard, however, to solve such quantum many-body problems. In solving the Hubbard model with 1000 particles, for example, the diagonalization would take 10 to the power of 573 years even with the world's fastest supercomputers. In this project, we develop a novel quantum simulator that can simulate quantum many-body dynamics for more than 1000 particles within one nanosecond, combining our two unique experimental resources: "coherent control with attosecond precision"[8] and "a strongly-correlated ultracold Rydberg gas."[7,9,10]

We have completed a standard hardware of this ultrafast quantum simulator composed of an array of ultracold Rb atoms trapped in an optical lattice and excited to Rydberg levels with a coherent picosecond (ps) laser pulse, as schematically illustrated in Figure 2.[3,4,6,7,10] The broad bandwidth of the ps laser pulse has allowed us to excite the atoms in the neighboring lattice sites to Rydberg levels simultaneously for the first time. With this standard hardware, we have succeeded in creating an exotic electronic state with spatially overlapping wave-functions as shown schematically in Figures 1 and 2.[2,4,7,10] The degree of spatial overlap is actively tuned with ~50 nanometer precision. This exotic metal-like quantum gas under exquisite control opens up a completely new regime of many-body physics for simulating ultrafast many-body electron dynamics dominated by Coulomb interactions.[7,10]

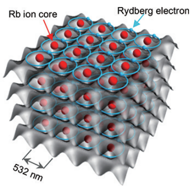


Figure 2. Schematic of the standard hardware of the ultrafast quantum simulator.[3,4,6,7,10]

We have also completed a readout interface of our ultrafast quantum simulator, which is the time domain Ramsey interferometry of ultracold Rydberg atoms with attosecond precision, whose contrast is almost 100% as shown in Figure 3.[5] The phase and visibility of this Ramsey interferogram are highly sensitive to the nature and strength of many-body interactions among the Rydberg atoms.

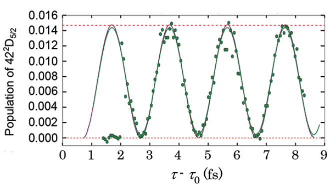


Figure 3. Time domain Ramsey interferometry of ultracold ^{87}Rb atoms with attosecond precision to be used as a readout interface of the ultrafast quantum simulator. Population of the $42^2D_{5/2}$ Rydberg state is plotted as a function of the delay τ between two laser pulses, where $\tau_0 \sim 50$ ps.[5]

2. Application of an "Ultrafast Quantum Simulator" to Quantum Computing[3,10]

We are developing a cold-atom based quantum annealer with the hardware of the ultrafast quantum simulator.[11] The cold-atom quantum annealer has advantages against the one with the superconducting qubits. Those advantages include scalability and efficiency. All to all connections among physical bits necessary for quantum annealing could also be easier with cold atoms than superconducting qubits.

So far we have developed arbitrary two dimensional optical trap arrays for cold atoms, which are necessary for quantum annealing,[11] in tight collaborations with Hamamatsu Photonics K.K.[3,10] Their examples are shown in Figure 4, the world's smallest arbitrary trap arrays whose nearest neighbor distance is only ~1 micron, which used to be typically ~4 micron in previous works.[12]

We have recently succeeded in loading a single atom into each trap of those arbitrary arrays, and reassembling those atoms with an optical tweezer. Accordingly we can prepare an array of atoms we desire, as exemplified in Figure 5.

These techniques mentioned above are also being applied to the development of gate-based quantum computing with cold atoms.

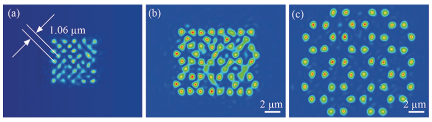


Figure 4. Examples of the world's smallest arbitrary arrays of optical traps. (a) Square lattice; (b) Kagome Lattice; (c) Hexagonal (Honeycomb) lattice.[10]

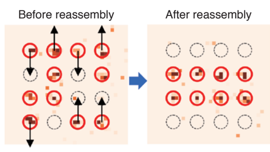


Figure 5. Assembly of an arbitrary array of single Rb atoms.[10]

3. Engineering Quantum Wave-Packet Dispersion with a Strong Nonresonant Femtosecond Laser Pulse[13,14]

A non-dispersing wave packet has been attracting much interest from various scientific and technological viewpoints. However, most quantum systems are accompanied by anharmonicity, so that retardation of quantum wave-packet dispersion is limited to very few examples only under specific conditions and targets. Here we demonstrate a conceptually new and universal method to retard or advance the dispersion of a quantum wave packet through "programmable time shift" induced by a strong non-resonant femtosecond laser pulse. A numerical simulation has verified that a train of such retardation pulses stops wave-packet dispersion.[13,14]

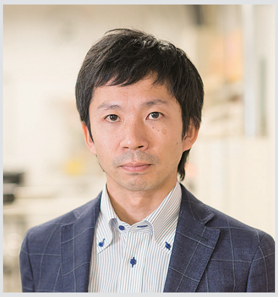
Our ultrafast quantum simulator and computer operates with atomic Rydberg levels,[3–10] whose level structure is anharmonic, so that its wave packet is dispersed and broadened quickly. The new control method for wave-packet dispersion developed here would serve as an enabling technology for our ultrafast quantum simulator and computer to enhance their functionalities.

References

1) K. Tonomura *et. al.*, *Am. J. Phys.* **57**, 117 (1989).
2) K. Ohmori, *Found. Phys.* **44**, 813–818 (2014).
3) Patent "Quantum Simulator and Quantum Simulation Method," H. Sakai (Hamamatsu Photonics K.K.), K. Ohmori (IMS) *et al.* (US 2020, JP 2021).
4) White Paper 2018 on Manufacturing Industries published by Ministry of Economy Trade and Industry, JAPAN.
5) C. Liu *et al.*, *Phys. Rev. Lett.* **121**, 173201 (2018).
6) Highlighted in "Quantum-Technology Innovation Strategy" by the Cabinet Office of Japan, January 2020.
7) M. Mizoguchi *et al.*, *Phys. Rev. Lett.* **124**, 253201 (2020).
8) H. Katsuki *et al.*, *Acc. Chem. Res.* **51**, 1174–1184 (2018).
9) N. Takei *et al.*, *Nat. Commun.* **7**, 13449 (2016).
10) S. Sugawa *et al.*, *Solid State Physics* **56**, 243 (2021). (Invited Paper/Cover-Page Highlight)
11) A. W. Glaetzle *et al.*, *Nat. Commun.* **8**, 15813 (2017).
12) D. Barredo *et al.*, *Science* **354**, 1021 (2016).
13) H. Katsuki *et al.*, *Phys. Rev. Res.* **3**, 043021 (2021).
14) Y. Ohtsuki *et al.*, *Phys. Rev. A* **104**, 033107 (2021).

Electronic Property of Functional Organic Materials

Department of Photo-Molecular Science
Division of Photo-Molecular Science III



KERA, Satoshi
Professor
[kera@ims.ac.jp]

Education
1996 B.E. Chiba University
1998 M.E. Chiba University
2001 Ph.D. Chiba University

Professional Employment
1998 JSPS Research Fellow
2001 Research Associate, Chiba University
2003 Research Associate, Institute for Molecular Science
2003 Postdoctoral Fellow, Wuerzburg University
2004 Assistant Professor, Chiba University
2007 Associate Professor, Chiba University
2009 Visiting Associate Professor, Institute for Molecular Science
2013 Adjunct Lecturer, The Open University of Japan
2013 Visiting Associate Professor, Soochow University
2014 Professor, Institute for Molecular Science
Professor, The Graduate University for Advanced Studies
Visiting Professor, Chiba University
2019 Visiting Professor, Kyoto University, Hiroshima University
2020 Visiting Professor, Tohoku University

Member
Assistant Professor
FUKUTANI, Keisuke
IMS Fellow
HASEGAWA, Yuri
Graduate Student
KANAZAWA, Shingo*
MIYATA, Takeshi†
Secretary
HAGIWARA, Hisayo

Keywords Photoelectron Spectroscopy, Molecular Assemble, Electronic State

Functional organic materials (FOM) have recently attracted considerable attention both for fundamental research and device applications because of peculiar properties not found in inorganics and small molecules. However, the mechanisms and the origin of various device characteristics are still under debate. Scientific discussions have been redundant because of long-standing beliefs that the electronic structure of FOM would be conserved as in an isolated molecule even for solid phases due to the weak van der Waals interaction. To reveal characteristics of FOM, it is essential to investigate precisely the electronic structure at various interfaces, including organic–organic and organic–inorganic (metal/semiconductor) contacts. Recently we realized that the weak electronic interaction manifests itself as small intensity modulations of fine structures in photoelectron spectra, depending on the adsorption and aggregation conditions on the surface. Thanks to recent instrumentation improvements, we can assess hidden fine features in the electronic states, *e.g.* electron–phonon coupling, quasi-particle states, very small densities of gap states, narrow band dispersion, and dynamic electronic polarization. To elucidate what really impacts on the electronic states of the FOM in their assembly as well as at the interface upon weak interaction, an evaluation of the wave-function spread of the electronic states is very important because the interface states are described as a delocalized molecular orbital state depending on the strength of weak electronic coupling (hybridization). Observing modifications of electron wave functions upon weak electronic coupling as well as strong electron–phonon coupling is a central issue on our agenda.

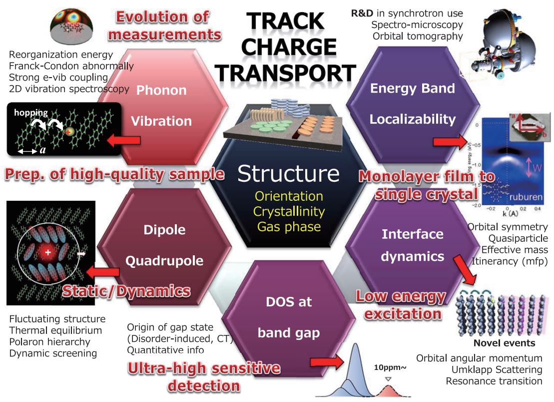


Figure 1. Overview of our agenda. A rich assortment of surface and interface structures of FOM to provide complicated spectral features of ultraviolet photoelectron spectroscopy.

Selected Publications

- Y. Nakayama, S. Kera and N. Ueno, *J. Mater. Chem. C* **8**, 9090–9132 (2020). [review]
- S. Kera, T. Hosokai and S. Duhm, *J. Phys. Soc. Jpn.* **87**, 061008 (7 pages) (2018). [review]
- J.-P. Yang, F. Bussolotti, S. Kera and N. Ueno, *J. Phys. D: Appl. Phys.* **50**, 423002 (45 pages) (2017). [review]
- S. Kera and N. Ueno, *J. Electron Spectrosc. Relat. Phenom.* **204**, 2–11 (2015). [review]

1. Experimental Observation of Anisotropic of Valence Band Dispersion in the Organic Semiconductor Crystal (DNTT)[1)]

Organic semiconductors based on the aromatic compounds having wide π-conjugation are attracting the attention of researchers because of their applications in various electronic devices. One of the central interests of these materials considering their physicochemical properties is their charge transport mechanism. In general, the conduction of charge carriers in most of the organic semiconductor solids is happening through the intermolecular hopping processes among the discrete molecular orbitals. However, some organic semiconductors with high charge-carrier mobility such as pentacene and rubrene exhibit continuous energy dispersion of valence bands, and the characteristics of the so-called "band transport" have been proposed.

The molecule focused on in this study is dinaphtho[2,3-b:2',3'-*f*]thieno[3,2-*b*]thiophene (DNTT). Although it has been proposed that the transport mechanism of DNTT is a band transport, the valence band dispersion has not yet been observed experimentally. In this study, we elucidate the valence band structure of DNTT single crystals using angle-resolved ultraviolet photoelectron spectroscopy (ARUPS) along three inequivalent crystallographic directions in the surface Brillouin zone. The valence band maximum is verified to be positioned at the Γ point, and the ionization energy of a DNTT single crystal is determined to be 5.02 eV at the VBM. The effective mass of hole is derived from the curvature of the experimental valence band at the Γ point in all three directions, where the lowest value of $2.6m_0$ is measured along the Γ–*S* direction.

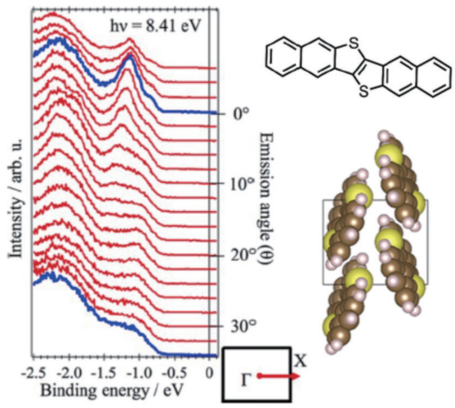


Figure 2. XeI-ARUPS spectra of the DNTT single crystal taken along Γ–*X* direction. The figure is after ref 1).

2. Accessing the Conduction Band Dispersion in $CH_3NH_3PbI_3$ Single Crystal[2)]

The conduction band dispersion in methylammonium lead iodide ($CH_3NH_3PbI_3$), which is a potential candidate for photovoltaic absorbers due to its high-light absorption coefficient, long carrier lifetime and diffusion length, low exciton binding energy, and easy fabrication, was studied both by angle-resolved two photon photoelectron spectroscopy (AR-2PPE) with low photon intensity (~0.0125 nJ/pulse) and angle-resolved low-energy inverse photoelectron spectroscopy (AR-LEIPS). Clear energy dispersions of the conduction band along Γ–*M* direction are observed by these independent methods under different temperatures for the first time, and the dispersion is found to be consistent with band calculation under a cubic phase. The effective mass of the electrons at Γ point is estimated to be $(0.20\pm0.05)m_0$ at 90 K. The observed energy position is significantly different between the AR-LEIPS and AR-2PPE, which is ascribed to the electronic-correlation effects depending on the difference of initial/final state probing processes. The present results also indicate that the surface structure of $CH_3NH_3PbI_3$ is given by a CH_3NH_3 + I plane, which provides the cubic dominated electronic property even at lower temperatures.

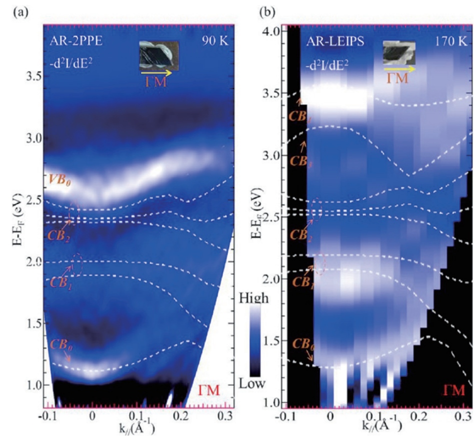


Figure 3. Second-derivative intensity maps of (a) AR-2PPE spectra and (b) AR-LEIPS spectra of $CH_3NH_3PbI_3$ single crystal. The figure is after ref 2).

3. Other Activities in UVSOR

We have conducted beamline R&D and user supports in collaboration with other universities. Experiments using photoelectron momentum microscope are developing at BL6U.[3)]

References

1) R. Takeuchi, S. Izawa, Y. Hasegawa, R. Tsuruta, T. Yamaguchi, M. Meissner, S. Ideta, K. Tanaka, S. Kera, M. Hiramoto and Y. Nakayama, *J. Phys. Chem. C* **125**, 2938–2943 (2021).
2) J. Yang, H. Sato, H. Orio, X. Liu, M. Fahlman, N. Ueno, H. Yoshida, T. Yamada and S. Kera, *J. Phys. Chem. Lett.* **12**, 3773–3778 (2021).
3) S. Makita, H. Matsuda, Y. Okano, T. Yano, E. Nakamura, Y. Hasegawa, S. Kera, S. Suga and F. Matsui, *e-J. Surf. Sci. Nanotechnol.* **19**, 42–47 (2021).

* carrying out graduate research on Cooperative Education Program of IMS with Chiba University
† carrying out graduate research on Cooperative Education Program of IMS with Chubu University

Light Source Developments by Using Relativistic Electron Beams

UVSOR Synchrotron Facility
Division of Advanced Accelerator Research

KATOH, Masahiro
Project Professor
[mkatoh@ims.ac.jp]

Education
1982 B.S. Tohoku University
1997 Ph.D. Tohoku University

Professional Employment
1986 Reseach Associate, National Laboratory for High Energy Physics
2000 Associate Professor, Institute for Molecular Science
2004 Professor, Institute for Molecular Science
Professor, The Graduate University for Advanced Studies
2019 Professor, Hiroshima University
Project Professor, Institute for Molecular Science

Member
Assistant Professor
FUJIMOTO, Masaki

Keywords Accelerator, Beam Physics, Synchrotron Radiation

UVSOR is a synchrotron light source providing low energy synchrotron light ranging from terahertz waves to the soft X-rays. Although it was constructed nearly 40 years ago, its performance is still in the world top level particularly among the low energy synchrotron light sources. This is the result of the continuous efforts on improving the machine. Our research group has been developing and introducing new accelerator technologies toward producing brighter synchrotron light with high stability, such as low emittance electron beam optics, novel insertion devices or state-of-the-art beam injection scheme. We have been developing novel light source technologies, such as free electron laser, coherent synchrotron radiation, optical vortices and laser Compton gamma-rays. We have been investigating beam physics which would be the basis of the future developments of the facility.

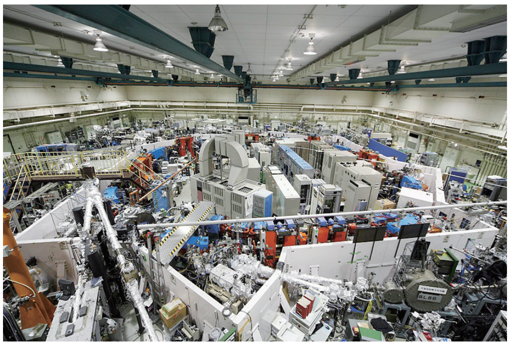


Figure 1. UVSOR-III Electron Storage Ring and Synchrotron Radiation Beamlines.

Selected Publications

- S. Bielawski, C. Evain, T. Hara, M. Hosaka, M. Katoh, S. Kimura, A. Mochihashi, M. Shimada, C. Szwaj, T. Takahashi and Y. Takashima, "Tunable Narrowband Terahertz Emission from Mastered Laser–Eelectron Beam Interaction," *Nat. Phys.* **4**, 390–393 (2008).
- M. Shimada, M. Katoh, M. Adachi, T. Tanikawa, S. Kimura, M. Hosaka, N. Yamamoto, Y. Takashima and T. Takahashi, "Transverse-Longitudinal Coupling Effect in Laser Bunch Slicing," *Phys. Rev. Lett.* **103**, 144802 (2009).
- M. Katoh, M. Fujimoto, H. Kawaguchi, K. Tsuchiya, K. Ohmi, T. Kaneyasu, Y. Taira, M. Hosaka, A. Mochihashi and Y. Takashima, "Angular Momentum of Twisted Radiation from an Electron in Spiral Motion," *Phys. Rev. Lett.* **118**, 094801 (2017).
- Y. Taira, T. Hayakawa and M. Katoh, "Gamma-Ray Vortices from Nonlinear Inverse Thomson Scattering of Circularly Polarized Light," *Sci. Rep.* **7**, 5018 (2017).
- S. Matsuba, K. Kawase, A. Miyamoto, S. Sasaki, M. Fujimoto, T. Konomi, N. Yamamoto, M. Hosaka and M. Katoh, "Generation of Vector Beam with Tandem Helical Undulators," *Appl. Phys. Lett.* **113**, 021106 (2018).
- Y. Hikosaka, T. Kaneyasu, M. Fujimoto, H. Iwayama and M. Katoh, "Coherent Control in the Extreme Ultraviolet and Attosecond Regime by Synchrotron Radiation," *Nat. Commun.* **10**, 4988 (2019).
- T. Kaneyasu, Y. Hikosaka, M. Fujimoto, H. Iwayama and M. Katoh, "Electron Wave Packet Interference in Atomic Inner-Shell Excitation," *Phys. Rev. Lett.* **126**, 1132202 (2021).

1. Light Source Technology Developments Based on Laser and Synchrotron

We have been developing light source technologies at the UVSOR-III electron storage ring using a dedicated experimental station BL1U, which was constructed under the support of Quantum Beam Technology Program of JST/MEXT aiming to develope novel light sources and exploring their applications. The BL1U is equipped with two undulators which constitute an optical klystron, a laser system which is synchronized with the accelerator beam and a dedicated beamline consisting of mirrors and a monochromator whose arrangement can be flexibly changed according to the types of the experiments.

In these years, we are focusing on generation of spatially structured light, such as optical vortex beams and optical vector beams from undulators, in collaboration with Hiroshima Univ. and Nagoya Univ. We have succeeded in producing such novel photon beams and are exploring their applications. More recently, we have started exploring the possibility utilizing the temporal structure of undulator radiation, in collaboration with Saga Light Source and Toyama Univ. So far, we have been demonstrated the coherent controls of atoms by using radiation from two undulators arranged in tandem. More recently, we have succeeded in observing ultrafast change of an electronic state of an atom, by using tandem undulator radiation.

We have been developing a laser Compton scattering gamma-ray source at BL1U, which is capable of producing monochromatic and energy-tunable gamma-rays. Now we are exploring their applications such as isotope imaging based on nuclear fluorescence resonance in collaboration with Kyoto Univ., AIST and QST, photon-induced positron annihilation lifetime spectroscopy in collaboration with Yamagata Univ. and AIST and an experimental verification on Delbruck scattering in collaboration with QST, AIST and Kyoto Univ. Theoretically we have proven that vortex photons carrying orbital angular momentum can be produced by non-linear Compton scattering of circularly polarized photons. We are planning its experimental demonstration at BL1U in collaboration with AIST.

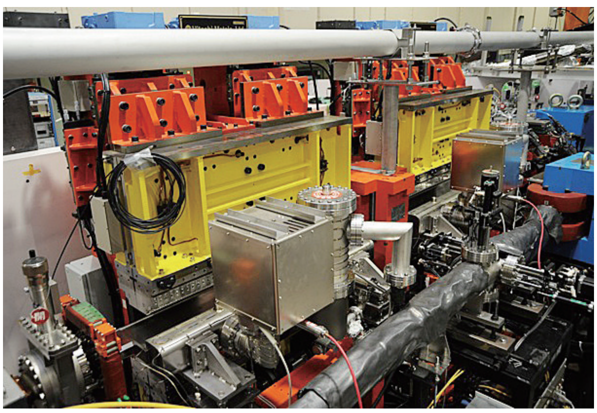


Figure 2. Twin Polarization-variable Undulators/Optical Klystron at UVSOR-III.

2. Accelerator Technology Developments for Electron Synchrotrons

We carried out several upgrade plans on UVSOR electron synchrotron since 2000. We designed a special beam optics intended to higher brightness. We developed necessary accelerator components, reconstructed the accelerator and commissioned it. We have constructed and commissioned six undulators successfully. Moreover, we have been continuously introducing new accelerator technologies such as the top-up operation in which the electron beam intensity is kept quasi-constant at a high beam current, 300mA, and the novel beam injection scheme with a pulsed sextupole magnet. As the result of all these efforts, now, the machine is one the brightest synchrotron light sources among the low energy machines below 1GeV in the world.

Currently, the storage ring is stably operated for many of the users, however, the requirements from the users for the stability is getting higher and higher. As a near-term upgrade plan, we are considering replacing some of the undulators to fit the changes of the users’ requirements on the wavelength. Also, we are seeking a possibility to reduce the emittance with the present magnet configuration. So far, we have found a few beam optics which would give lower emittance around 10 nm. Although they are not compatible with the operation of the narrow gap undulators, they may be used for special experiments which requires lower emittance. For a long-term plan, we continue the design study on a new light source facility. We have been investigating various accelerator systems such as a diffraction-limited synchrotron, an energy recovery linear accelerator and so on. Currently we are focusing on designing a synchrotron with the electron energy of 1 GeV and the circumference of around 70 m. We have designed a synchrotron which would give low emittance of around 6 nm under the achromatic condition.

We are collaborating with Nagoya Univ. and developing new technologies for the future plan. Accelerator magnets based on permanent magnets are being developed, which would contribute to the power consumption saving. New pulsed multipole magnet is also being developed to realize a novel beam injection scheme.

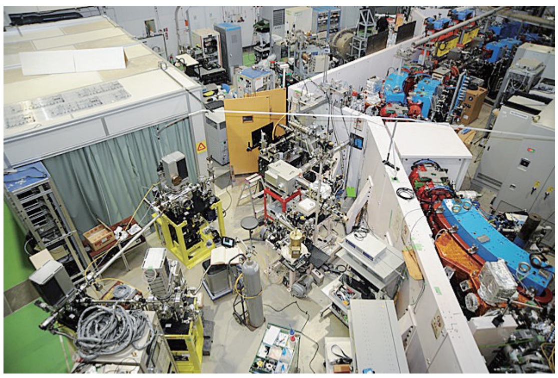
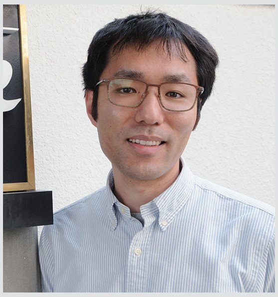


Figure 3. UVSOR BL1U experimental station for source development studies.

Development and Utilization of Novel Quantum Beam Sources Using a High Energy Electron Beam

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research



TAIRA, Yoshitaka
Associate Professor
[yostaira@ims.ac.jp]

Education
2007 B.S. Nagoya University
2009 M.S. Nagoya University
2012 Ph.D. Nagoya University

Professional Employment
2011 JSPS Research Fellow
2012 Research Scientist, National Institute of Advanced Industrial Science and Technology (AIST)
2018 Senior Research Scientist, National Institute of Advanced Industrial Science and Technology (AIST)
2020 Associate Professor, Institute for Molecular Science
Associate Professor, The Graduate University for Advanced Studies

Awards
2010 Student Presentation Award, The 23rd Annual Meeting of the Japanese Society for Synchrotron Radiation Research
2010 Young Researcher Best Presentation Award, The 53rd Annual Meeting of the Japanese Society of Radiation Chemistry
2011 Nagoya University Outstanding Graduate Student Award
2012 Oral Presentation Award, The 9th Annual Meeting of Particle Accelerator Society of Japan
2012 Young Researcher Best Poster Award, 12th International Symposium on Radiation Physics
2013 Young Scientist Award of the Physical Society of Japan
2015 Young Researcher Best Presentation Award, Beam Physics Workshop 2015

Member
Assistant Professor
SUGITA, Kento
Post-Doctoral Fellow
SALEHI, Elham
Graduate Student
KOYAMA, Kazuki*
YAMAMOTO, Ryohei*

Keywords Electron Beams, Synchrotron Radiation, Gamma-Rays

Our group develop new electromagnetic wave sources using a high energy electron beam. In the UVSOR-III electron storage ring at the Institute for Molecular Science, a 750-MeV electron beam can be generated. Electromagnetic waves in a wide frequency range from ultraviolet waves to gamma-rays are emitted by interacting the electron beam with magnetic fileds and lasers.

Laser Thomson (Compton) scattering is a method to generate a high energy gamma-ray by the interaction between a high energy electron and a laser. We have developed ultra-short pulsed gamma-rays with the pulse width of sub-ps to ps range by using 90-degree laser Thomson scattering (Figure 1). We applied this ultra-short pulsed gamma-rays to gamma induced positron annihilation lifetime spectroscopy (GiPALS). A positron is an excellent probe of lattice defects in solids and of free volumes in polymers at the sub-nm to nm scale. GiPALS enables defect analysis of a thick material in a few cm because positrons are generated throughout a bulk material via pair production. Our group is conducting research on improving the properties of the material by using GiPALS.

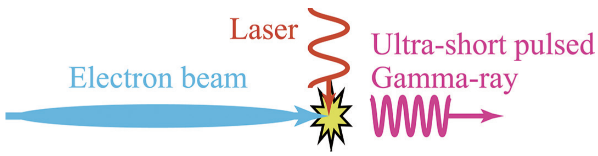


Figure 1. Schematic illustration of 90-degree laser Thomson scattering.

Selected Publications

- Y. Taira, M. Adachi, H. Zen, T. Tanikawa, N. Yamamoto, M. Hosaka, Y. Takashima, K. Soda and M. Katoh, "Generation of Energy-Tunable and Ultra-Short-Pulse Gamma Ray via Inverse Compton Scattering in an Electron Storage Ring," *Nucl. Instrum. Methods Phys. Res., Sect. A* **652**, 696 (2011).
- Y. Taira, H. Toyokawa, R. Kuroda, N. Yamamoto, M. Adachi, S. Tanaka and M. Katoh, "Photon-Induced Positron Annihilation Lifetime Spectroscopy Using Ultrashort Laser-Compton-Scattered Gamma-Ray Pulses," *Rev. Sci. Instrum.* **84**, 053305 (2013).
- Y. Taira, T. Hayakawa and M. Katoh, "Gamma-Ray Vortices from Nonlinear Inverse Thomson Scattering of Circularly Polarized Light," *Sci. Rep.* **7**, 5018 (2017).
- Y. Taira and M. Katoh, "Gamma-Ray Vortices Emitted from Nonlinear Inverse Thomson Scattering of a Two-Wavelength Laser Beam," *Phys. Rev. A* **98**, 052130 (2018).
- Y. Taira, M. Fujimoto, S. Ri, M. Hosaka and M. Katoh, "Measurement of the Phase Structure of Elliptically Polarized Undulator Radiation," *New J. Phys.* **22**, 093061 (2020).

1. Gamma-Induced Positron Annihilation Lifetime Spectroscopy (GiPALS)

Positron lifetime spectrum is calculated by measuring the time difference between a reference signal and a detector output for the annihilation gamma-rays, which is emitted when a positron annihilates with an electron inside material. A reference signal is the output of a photodiode located near the injection position of a laser. A BaF_2 scintillator and a photomultiplier tube is utilized to detect the annihilation gamma-rays. Two detectors are arranged at 180 degrees because two annihilation gamma-rays are generated at 180-degree direction. The annihilation gamma-rays are generated to whole solid angle. Therefore array detectors are effective to increase the count rate of the annihilation gamma-rays and to reduce the measurement time. We have developed the array detector using 10 detectors with a help of Equipment Development Center (Figure 2).

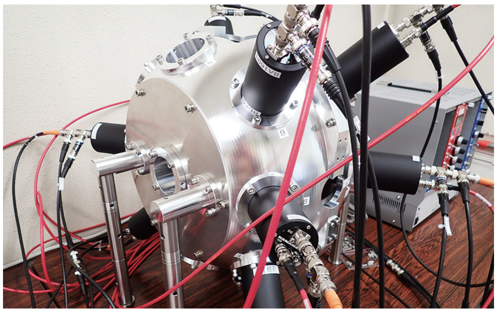


Figure 2. Positron lifetime measurement system using 10 detectors.

Users can currently utilize GiPALS at BL1U in UVSOR-III. A result of defect analysis for a GAGG scintillator was published in 2020.[1] We are also developing a new measurement method, gamma ray-induced age-momentum correlation (GiAMOC). On the other hand, we are planning to develop other measurement technique for the annihilation gamma-rays, such as a three-dimensional distribution imaging technique for defects, coincidence Doppler broadening, and spin polarized positrons generated from circularly polarized gamma-rays.

A new laser injection vacuum chamber was installed to the UVSOR-III electron storage ring in April 2021. As both the incoming and outgoing sides of the vacuum chamber are optical windows, the laser can be focused down to 10 micrometers. We have confirmed that the intensity of gamma-rays at the generation point has been improved by a factor of 40.

2. Short Wavelength Optical Vortices

An optical vortex is an electromagnetic wave with a helical phase structure. When an optical vortex beam is viewed in a plane transverse to the direction of propagation, an annular intensity profile is observed due to the phase singularity at the center axis. An important consequence of the optical vortex is that it carries orbital angular momentum (OAM) due to the helical phase structure.

While fundamental and applied research on optical vortices using visible wavelength lasers is widely studied, much less has been done in ultraviolet, X-rays, and gamma-rays energy ranges. We have proposed for the first time a method to generate a gamma-ray vortex using nonlinear inverse Thomson scattering of a high energy electron and an intense circularly polarized laser.[2] In our method, the circularly polarized laser is important because the helical phase structure arises from the transverse helical motion of the electron inside the circularly polarized laser field. When peak power of a laser achieves terawatt class, high harmonic gamma-rays are generated. Only gamma-rays more than the first harmonic carry OAM. High harmonic gamma-rays show the annular intensity distribution due to this characteristic.

There are few facilities in the world, where can carry out the experiment for the nonlinear inverse Thomson scattering using an intense circularly polarized laser in terawatt class. We carried out the experiment at Kansai Photon Science Institute in Japan, where a 150 MeV microtron and a petawatt laser are available. Although we were not able to achieve the measurement of an annular intensity distribution of high harmonic gamma-rays, we plan to continue the experiment this year.

On the other hand, optical vortices in the ultraviolet wavelength range can be generated using a helical undulator. Similar with a nonlinear inverse Thomson scattering, an electron obeys a helical trajectory inside an undulator. Therefore, high harmonic radiation emitted from a helical undulator forms the helical phase structure.

Generation of an optical vortex from a helical undulator has been demonstrated at UVSOR-III. Recently, we newly revealed that undulator radiation with the phase structure can be generated from an elliptically polarized undulator.[2] We derived the analytic expressions for the emitted electric fields were fully derived and the radiation's phase structure was found to change according to polarization. Average phase gradients of the undulator's radiation were measured using a double slit interferometer. The measured phase gradients of the first through third harmonics were compared with the calculated results.

References

1) K. Fujimori, M. Kitaura, Y. Taira, M. Fujimoto, H. Zen, S. Watanabe, K. Kamada, Y. Okano, M. Katoh, M. Hosaka, J. Yamazaki, T. Hirade, Y. Kobayashi and A. Ohnishi, *Appl. Phys. Express* **13**, 085505 (4 pages) (2020).
2) Y. Taira, M. Fujimoto, S. Ri, M. Hosaka and M. Katoh, *New J. Phys.* **22**, 093061 (2020).

Award

TAIRA, Yoshitaka; The Outstanding Presentation Award of the 64th Annual Meeting of the Japanese Society of Radiation Chemistry (2021).

* carrying out graduate research on Cooperative Education Program of IMS with Nagoya University

Angle-Resolved Photoemission Study on Strongly Correlated Electron Materials

UVSOR Synchrotron Facility
Division of Advanced Solid State Physics

TANAKA, Kiyohisa
Associate Professor
[k-tanaka@ims.ac.jp]

Education

2000 B.S. The University of Tokyo
2005 Ph.D. The University of Tokyo

Professional Employment

2005 Postdoctoral Fellow, Stanford University and Lawrence Berkeley National Laboratory
2008 Assistant Professor, Osaka University
2013 Associate Professor, Osaka University
2014 Associate Professor, Institute for Molecular Science
Associate Professor, The Graduate University for Advanced Studies

Member

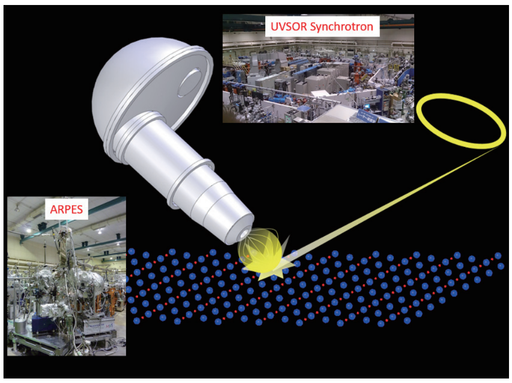
Assistant Professor
IDETA, Shin-ichiro*

Graduate Student
HOSOYA Tomoki†
FURUTA Kanji†
MATSUNAGA, Kazuya†
YAMAMOTO, Ryo†

Keywords Strongly Correlated Electron System, Synchrotron Light, Photoemission

Strongly correlated electron materials has attracted more attentions in the last few decades because of their unusual and fascinating properties such as high-T_c superconductivity, giant magnetoresistance, heavy fermion and so on. Those unique properties can offer a route toward the next-generation devices. We investigate the mechanism of the physical properties as well as the electronic structure of those materials by using angle-resolved photoemission spectroscopy (ARPES). ARPES is a powerful experimental technique, directly measuring the energy (E) and momentum (k) relation, namely the band structure of solids. In the last quarter of a century, the energy resolution and angular resolution of ARPES have improved almost three order of magnitude better, which makes us possible to address the fine structure of the electronic structure near the Fermi level: Superconducting gap, kink structure and so on. The main target materials of our group is high-T_c superconductors, such as cuprates and iron pnictides and use UVSOR-III as a strong light source.

Our group is also developing high-efficiency spin-resolved ARPES system. Spintronics is a rapidly emerging field of science and technology that will most likely have a significant impact on the future of all aspects of electronics as we continue to move into the 21st century. Understanding magnetism of surfaces, interfaces, and nanostructures is greatly important for realizing the spintronics which aims to control and use the function of spin as well as the charge of electrons. Spin-resolved ARPES is one of the most powerful experimental techniques to investigate the magnetic properties of such materials.



Selected Publications

- K. Tanaka, W. S. Lee, D. H. Lu, A. Fujimori, T. Fujii, Risdiana, I. Terasaki, D. J. Scalapino, T. P. Devereaux, Z. Hussain and Z.-X. Shen, "Distinct Fermi-Momentum-Dependent Energy Gaps in Deeply Underdoped Bi2212," *Science* **314**, 1910–1913 (2006).
- W. S. Lee, I. M. Vishik, K. Tanaka, D. H. Lu, T. Sasagawa, N. Nagaosa, T. P. Devereaux, Z. Hussain and Z.-X. Shen, "Abrupt Onset of a Second Energy Gap at the Superconducting Transition of Underdoped Bi2212," *Nature* **450**, 81–84 (2007).
- K. Tanaka, N. Hieu, G. Vincini, T. Masui, S. Miyasaka, S. Tajima and T. Sasagawa, "Quantitative Comparison between Electronic Raman Scattering and Angle-Resolved Photoemission Spectra in $Bi_2Sr_2CaCu_2O_{8+\delta}$ Superconductors: Doping Dependence of Nodal and Antinodal Superconducting Gaps," *J. Phys. Soc. Jpn.* **88**, 044710 (2019).
- S. Ideta, N. Murai, M. Nakajima, R. Kajimto and K. Tanaka, "Experimental Investigation of the Suppressed Superconducting Gap and Double-Resonance Mode in $Ba_{1-x}K_xFe_2As_2$," *Phys. Rev. B* **100**, 235135 (7 pages) (2019).

1. Development of Spin-Resolved ARPES with Image-Spin Detection

Spintronics is a rapidly emerging field of science and technology that will most likely have a significant impact on the future of all aspects of electronics as we continue to move into the 21st century. Understanding magnetism of surfaces, interfaces, and nanostructures is greatly important for realizing the spintronics which aims to control and use the function of spin as well as the charge of electrons. Spin- and angle-resolved photoemission spectroscopy (spin-resolved ARPES) is one of the most powerful experimental techniques to investigate the magnetic properties of such materials, where one can know the "complete" information of the electronic states of materials; energy, momentum, and spin direction. Recent development of high energy and angle resolved photoelectron analyzer as well as the contemporary light sources such as third generation synchrotron radiation make it possible for the photoemission spectroscopy to investigate not only band structures but many body interactions of electrons in solids. However, appending the spin resolution to photoemission spectroscopy is quite difficult because of an extremely low efficiency (10^{-4}) of Mott-type spin detections. Recently, very-low-energy-electron-diffraction (VLEED-type) spin detector with 100 times higher efficiency than that of conventional Mott-type has been developed and spin-resolved ARPES has been started to be realized. So far, most of the spin-resolved ARPES systems in the world are using the single-channel detector and efficiency is still a problem.

Beamline BL5U at UVSOR has been totally reconstructed by our group, and opened for users as high photon flux and high energy resolution ARPES beamline since 2017. As a new function for this beamline, we have started high-efficient spin-resolved ARPES project with multi-channel detection (we call "image-spin" detection). The goal of this project is to realize the 100 times better efficiency and the 10 times better momentum resolution than the current spin-resolved ARPES system in the world, which can be a breakthrough in this field.

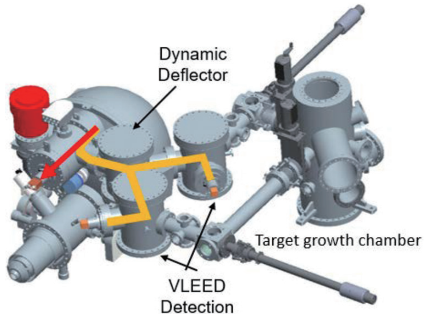


Figure 1. Current setup of image-spin ARPES.

In 2020, we set up the spin detection system and finished the adjustment of the electron lens parameters of the spin detection part (Figure 1). Finally, we successfully obtained spin-resolved signal of Au(111) surface as shown in Figure 2. According to the rough estimation, the efficiency is 100 times better and the momentum resolution is several times better than the current spin-resolved ARPES system in the world.

2. Strong Relationship between ARPES Superconducting Spectral Weight and T_c[1,2]

Our resent ARPES study on high-T_c cuprate superconductors $Bi_2Sr_2CaCu_2O_{6+\delta}$ (Bi2212) and $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi2223) indicated strong relationship between the superconducting spectral weight and the critical temperature T_c. To see the detail, we are performing temperature and carrier concentration dependent measurements on several kinds of high-T_c cuprate superconductors.

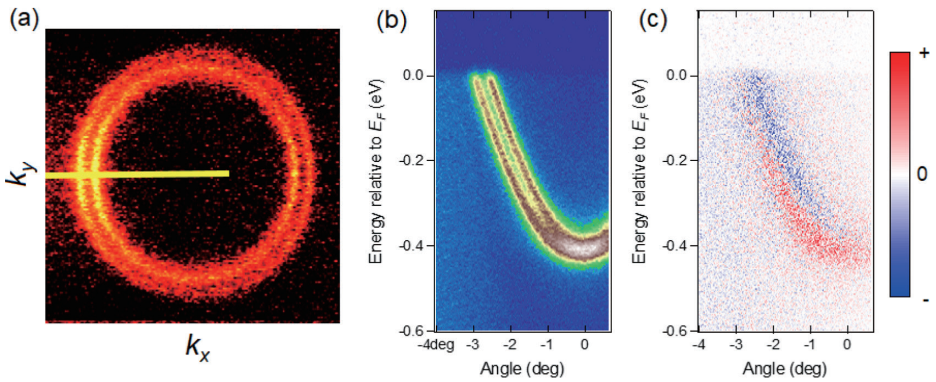


Figure 2. (a) Fermi surface of Rashba spin splitting in Au(111) surface states and (b) image plot of normal ARPES at yellow bar in (a). (c) Spin-resolved ARPES showing the spin polarization (blue–red scale).

References
1) G. Vincini *et al.*, *Supercond. Sci. Technol.* **32**, 113001 (2019).
2) K. Tanaka *et al.*, *J. Phys. Soc. Jpn.* **88**, 044710 (2019).

* Present Address; Hiroshima University
† carrying out graduate research on Cooperative Education Program of IMS with Nagoya University

Photoemission Imaging in *k* and *r* Spaces

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research

MATSUI, Fumihiko
Senior Researcher

Complete measurements of photoelectron, including three- dimensional spin degrees of freedom are challenging techniques for pioneering electron spin physics and spintronics. We are developing a new system that combines a photoelectron momentum microscope (PMM), two-dimensional spin filter, and a synchrotron light source. Our goal is to achieve reliable electronic structure analysis by complete photoelectron measurement of band dispersion and core levels, and link it to developments and applications of materials and devices.

1. Photoelectron Momentum Microscope

We have built a new PMM station for 3D momentum-resolved photoelectron spectroscopy with a microscopic field of view at the soft X-ray beamline BL6U of UVSOR. The details of the specification evaluation result are described elsewhere.[1)] In brief, the energy, spatial, and momentum resolutions of the analyzer were estimated to be 20 meV, 50 nm, and 0.012 Å^{-1}, respectively. Samples can be cooled down to 8 K and heated up to 400 K.

A gold checkerboard pattern on a Si wafer imaged by a microscopy mode is shown in Figure 1. Elemental specific information is obtained by spectro-microscopy as shown in Figure 1(b) and (c). Figure 2 shows an example of 3D valence band dispersion imaging by a momentum mode.

With a photon energy range up to 800 eV covered by the BL6U, core-level excitation of a variety of important elements including C, N, O and transition metals is possible. Specific atomic sites and electronic states can be selectively characterized by the resonant Auger process. Resonant momentum-resolved photoelectron spectroscopy is a method unique to this station that opens the door to elemental- and orbital-selective valence band dispersion analysis.

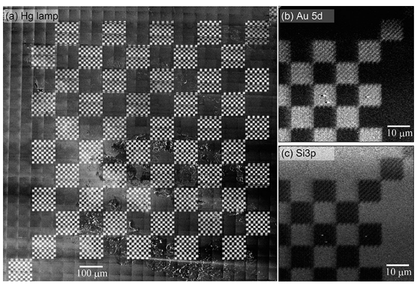


Figure 1. Real-space valence photoelectron images of a gold checkerboard pattern observed using (a) Hg lamp and (b)–(c) synchrotron radiation ($h\nu$ = 60 eV) as excitation.[2)]

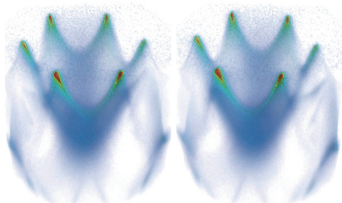


Figure 2. (a) Stereograph of valence band dispersion of graphite crystal surface. A parabolic π band with six Dirac points is imaged.

2. Original Analyzers towards Spin Imaging

Furthermore, we are aiming at highly efficient and comprehensive measurement of spin distribution as the final goal. *Omnidirectional photoelectron acceptance lens* (OPAL)[3)] together with *Projection-type electron spectroscopy collimator analyzer* (PESCATORA)[4)] enables photoelectron holography measurement of the full hemisphere. Moreover, we invented *Right angle deflection imaging analyzer* (RADIAN)[5)] for spin vector analysis with ***k***/***r***-space resolution. We are expanding the MM system based on our original inventions.

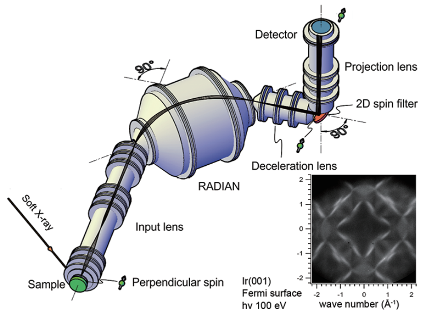


Figure 3. Schematic diagram of 3D-spin distribution projection analysis system.[5)] Inset is the Fermi surface of the Ir(001) thin film for a 2D spin filter target measured by UVSOR-PMM.

References

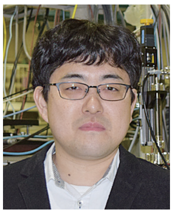
1) F. Matsui *et al.*, *Jpn. J. Appl. Phys.* **59**, 067001 (2020).
2) S. Makita, F. Matsui, *et al.*, *e-J. Surf. Sci. Nanotechnol.* **19**, 42 (2021).
3) H. Matsuda and F. Matsui, *Jpn. J. Appl. Phys.* **59**, 046503 (2020).
4) F. Matsui and H. Matsuda, *Rev. Sci. Inst.* **92**, 073301 (2021).
5) H. Matsuda and F. Matsui, *J. Electron Spectrosc. Relat. Phenom.* **245**, 147001 (2020).

Award

MATSUI, Fumihiko; NAGAI Foundation for Science & Technology Encouragement Award (2021).

Soft X-Ray Absorption Spectroscopy for Observing Chemical Processes in Solution

Department of Photo-Molecular Science
Division of Photo-Molecular Science III



NAGASAKA, Masanari
Assistant Professor

Soft X-ray absorption spectroscopy (XAS) observes local structures of liquids with different light elements. We have developed liquid cells and devices with precise absorbance control and observed several chemical processes in solution by using *operando* XAS.[1] In this year, we have developed an ultrathin liquid cell for XAS of liquids in the low-energy region below 200 eV.

1. Development of the Ultrathin Liquid Cell for XAS in the Low-Energy Region

XAS below 200 eV is important for chemical research since it includes K-edges of Li and B and L-edges of Si, P, S, and Cl. Recently, we have established soft X-ray transmission argon gas window that is effective from 60 to 240 eV.[2] From soft X-ray transmission calculations, soft X-rays below 200 eV can transmit argon gas with the optical length of 2.6 mm. As shown in Figure 1, we have developed the ultrathin liquid cell that realize the 2.6 mm optical length of argon gas. XAS spectra of 2 M LiCl aqueous solution at Li K-edge and Cl L-edge were successfully obtained by using this liquid cell.

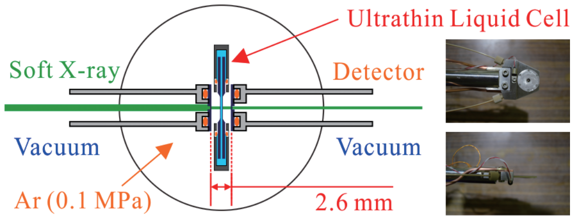


Figure 1. The schematic and photographs of the ultrathin liquid cell for XAS measurements of liquids in the low-energy region.

References

1) M. Nagasaka and N. Kosugi, *Chem. Lett.* **50**, 956–964 (2021).
2) M. Nagasaka, *J. Synchrotron Radiat.* **27**, 959–962 (2020).

Distribution of Biological Molecules in a Cell Nucleus Analyzed by 3-Dimensional Spectro-Microscopy

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research

OHIGASHI, Takuji
Assistant Professor

Scanning transmission X-ray microscopy (STXM) is a promising tool to analyze 2-dimensional chemical state of a sample with high spatial resolution around 30 nm. We have been developing computer tomography (CT) for STXM to perform 3-dimensional spectro-microscopy.[1,2] An isolated cell nucleus of a HeLa S3 cell was used as a sample. 50 datasets of 2-dimensional X-ray absorption spectra (2D XAS) of the sample around O K-edge was acquired with rotating the sample 3.6° each (180° rotation in total). 3D XAS of the cell nucleus is reconstructed from 50 datasets of 2D XAS. Distributions of DNA (red) and protein (green) are obtained by fitting reference spectra to the 3D XAS by single value decomposition algorithm (Figure 1). In cross sectional images, nucleoli and network structure of protein around them can be distinguished clearly. A goal of this research is to elucidate chemical and morphological change of biological molecules through a process of apoptosis.

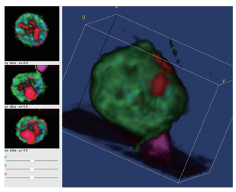


Figure 1. 3-dimensional distributions of DNA (red) and protein (green) in a HeLa S3 cell nucleus. Left panels are cross sectional images and a right panel is volume rendering image.

References

1) T. Ohigashi, A. Ito, K. Shinohara, S. Toné, Y. Inagaki, H. Yuzawa and N. Kosugi, *Microsc. Microanal.* **24**, 400–401 (2018).
2) T. Ohigashi, Y. Inagaki, A. Ito, K. Shinohara and N. Kosugi, *J. Phys.: Conf. Ser.* **849**, 012044 (2017).

Mesoscopic Structures of Liquid-Crystal Molecules Probed by Resonant Soft X-Ray Scattering

UVSOR Synchrotron Facility
Division of Advanced Photochemistry

IWAYAMA, Hiroshi
Assistant Professor

We study soft matters such as liquid-crystal materials with soft x-ray, whose energy region covers K-edge energies of carbon, nitrogen and oxygen. Soft matters exhibit their intriguing properties due to mesoscopic physical structures by self-organizations. To understand properties of soft matters, we need to investigate their structure in the mesoscopic scale.

1. Resonant Soft X-Ray Scattering Method

Resonant soft x-ray scattering measurements can probe mesoscopic structures and periodic spatial variations of the orientation of molecules with both elemental and chemical environment sensitivity and have orders of magnitude scattering intensity enhancement over conventional small angle (non-resonant) x-ray scattering, which is sensitive only to the electron density modulations.

In this year, we performed RSoXS experiments at UVSOR BL3U for the first time. Our sample is S-MHPOBC, which is a chiral smectic liquid crystal molecule and shows ferro- and antiferroelectric phases depending on its temperature.

We successfully obtained diffraction images at the wavelength of 4.34 nm (285 eV), which correspond to C 1s-to-π* core excitations. From the analysis of images, we found a resonant enhancement of diffractions corresponding to a period length of 6.4 nm. Considering single molecular length of 3 nm, this shows the sample is antiferroelectric under the measurement conditions. We investigate intermediate phases between ferro- and antiferroelectric one by changing sample temperatures.

Visiting Professors

Visiting Professor

FUKUI, Ken-ichi *(from Osaka University)*

Ionic Liquid/Organic Semiconductor Film Interfaces for Efficient Carrier Transport

Local analyses of electrolyte/organic semiconductor electrode interfaces at controlled electrode potentials are of fundamental importance to understanding the origin and properties of the electric double layer (EDL) at the interfaces, which is necessary for their application to EDL-organic field effect transistors (OFETs). Ionic liquids (ILs) gated EDL-OFETs can be operated with ultralow threshold voltage (~0.1 V), and high electric field of the EDL restrict the hole carrier at the organic molecular facing the interface, thus a few molecular layer film works as the efficient device. By adopting newly developed electrochemical ATR-FUV (EC-ATR-FUV) system for the analyses of electronic states of the device consisting of a two-layer-thick C9-DNBDT-NW film and an IL (EMIM-FSA), we have succeeded to obtaining the hole carrier density as low as 1/500 hole per a C9-DNBDT-NW molecule. The spectrum analyses also provided the information of interaction between the organic thin film and the IL. Further analyses of the system using UVSOR are in progress.

Visiting Professor

SHIMADA, Kenya *(from Hiroshima University)*

High-Resolution Angle-Resolved Photoemission Study of Topological Materials

By means of high-resolution angle-resolved photoemission spectroscopy (ARPES), we clarify electronic structures of solids (band structures, Fermi surface, spin polarization) to understand their physical properties from the microscopic point of view. This year, we have examined the antiferromagnetic topological insulator families $(Bi_2Te_3)_nMnBi_2Te_4$ (n = 1,2). We have observed the electronic band structures specific to the surface termination, which are fully consistent with detailed density functional theory (DFT) calculations. It indicates the surface termination is important for the surface states on $(Bi_2Te_3)_nMnBi_2Te_4$. We have also examined the spin texture of a photocatalyst BiOI. There are two Bi-I sectors connected via the centrosymmetric point, and we found that the topmost iodine layer had a helical spin texture with a spin polarization up to ~80%. Based on the detailed theoretical considerations, the high spin polarization on each sector is protected by the non-symmorphic lattice symmetry (P4/nmm) together with the strong spin–orbit interaction. We have confirmed that the spin-momentum-layer locking effect in BiOI. To improve the spatial resolution of high-resolution ARPES on the HiSOR beamline, we have introduced a highly precise XYZ translator with the absolute accuracy of <1μm and developed the spatial mapping mode.

Visiting Associate Professor

KATSUKI, Hiroyuki *(from Nara Institute of Science and Technology)*

Coherent Control in Condensed Systems

Coherent control is a technique to manipulate quantum states of a target system utilizing the interference of wavefunctions. Highly designed ultrashort laser pulses, both temporally- and spatially-modulated, are used to manipulate the amplitudes and phases of the target wavefunctions. Current my research is focused on the coherent control in various condensed phase systems including strongly coupled systems composed of cavity photons and molecular excited states. Especially, vibrational polaritons which are composed of the molecular vibrational states and mid-infrared cavity photons are of great interest due to the possibility to manipulate the dynamics of chemical reactions. This is possible since the ground state potential surface is locally modulated by the formation of polaritons. Now we are preparing a femtosecond pump–probe and other nonlinear spectroscopic setups to track the ultrafast dynamics of such strongly coupled vibrational polariton systems.