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 collaborate with the Department.

The core topics of the Department include attosecond coherent control for the development of ultrafast quantum computers and simulators, chiro-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 Computer and Simulator

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Professional Employment
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2003 Professor, Institute for Molecular Science
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2004 Visiting Professor, Tohoku University (–2005)
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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 (Germany)
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
2021 National Medal with Purple Ribbon by His Majesty the Emperor of Japan

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Keywords Quantum Simulation, Quantum Computing, Attosecond

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 of 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.

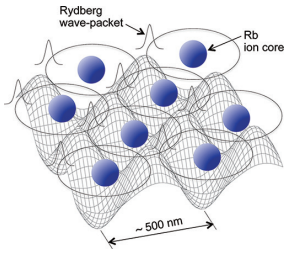

Figure 1. Metal-like quantum gas. A schematic of the many-body quantum simulator with ultracold Rydberg atoms, named "ultrafast quantum simulator," where electronic wave functions spatially overlap between neighboring atoms.[2)]

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," *Nature Physics* **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," *Nature Commun.* **4**, 2801 (2013).
- N. Takei *et al.*, "Direct Observation of Ultrafast Many-Body Electron Dynamics in an Ultracold Rydberg Gas," *Nature 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).
- Y. Chew *et al.*, "Ultrafast Energy Exchange between Two Single Rydberg Atoms on a Nanosecond Timescale," *Nature Photonics* **16**, 724 (2022).
- V. Bharti *et al.*, "Picosecond-Scale Ultrafast Many-Body Dynamics in an Ultracold Rydberg-Excited Atomic Mott Insulator," *Phys. Rev. Lett.* **131**, 123201 (2023).
- V. Bharti *et al.*, "Strong Spin-Motion Coupling in the Ultrafast Dynamics of Rydberg Atoms," *Phys. Rev. Lett.* **133**, 093405 (2024).

1. Development of an "Ultrafast Quantum Simulator" by Optical Control with Precisions on the Attosecond Temporal and Submicron Spatial Scales[2–7)]

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"[3)] and "a strongly correlated ultracold Rydberg gas."[4–6)]

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.[5,6)] 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. Recently in 2023, quantum magnetism has successfully been simulated with this standard hardware assembled with ~30,000 Rb atoms.[6)] Our novel scheme above has accelerated the simulation speed by three orders of magnitude compared to previous quantum simulators of magnetism. Moreover, we have succeeded in simulating the formation dynamics of "quantum entanglement," which is difficult to measure in actual magnetic materials, on the timescale of several hundred picoseconds.

Very recently in 2024 we have revealed the quantum entanglement between electronic and motional states in our "ultrafast quantum simulator," generated by the repulsive force due to the strong interaction between Rydberg atoms as seen in Figure 2.[7)] We have also proposed a new quantum simulation method including repulsive force between particles.

We continue upgrading this ultrafast quantum simulators, generously supported by the Q-LEAP program of the MEXT of Japan.

2. Development of an Ultrafast Quantum Computer with Cold Atoms[8,10,11)]

We develop a novel quantum computer with two dimensional arrays of ultracold Rb atoms trapped in optical tweezers. These atomic qubits are manipulated with an ultrafast laser for the first time, leading to a completely new quantum computer we refer to as an "ultrafast quantum computer." With this ultrafast quantum computer, we succeeded in executing a controlled Z gate,[8)] accelerating a two-qubit gate (a fundamental arithmetic element essential for quantum computing) of cold-atom quantum computers by two orders of magnitude. It is also two orders of magnitude faster than the noise from the external environment and operating lasers, and thus can be isolated from the noise. Moreover, this ultrafast two-qubit gate is faster than the fast two-qubit gate demonstrated recently by "Google AI Quantum" with superconducting qubits.[9)] We are currently improving key enabling technologies for optical tweezers and operating lasers.[10,11)]

We continue upgrading this ultrafast quantum computers, generously supported by the Moonshot program of the Cabinet Office of Japan.

References

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2) K. Ohmori, *Found. Phys.* **44**, 813–818 (2014).
3) H. Katsuki *et al.*, *Acc. Chem. Res.* **51**, 1174–1184 (2018).
4) N. Takei *et al.*, *Nat. Commun.* **7**, 13449 (2016).
5) M. Mizoguchi *et al.*, *Phys. Rev. Lett.* **124**, 253201 (2020).
6) V. Bharti *et al.*, *Phys. Rev. Lett.* **131**, 123201 (2023).
7) V. Bharti *et al.*, *Phys. Rev. Lett.* **133**, 093405 (2024).
8) Y. Chew *et al.*, *Nat. Photonics* **16**, 724 (2022). (Cover-Page Highlight)
9) B. Foxen *et al.*, *Phys. Rev. Lett.* **125**, 120504 (2020).
10) Y. Chew *et al.*, arXiv:2407.20699 (2024).
11) T. P. Mahesh *et al.*, arXiv:2408.02324 (2024).

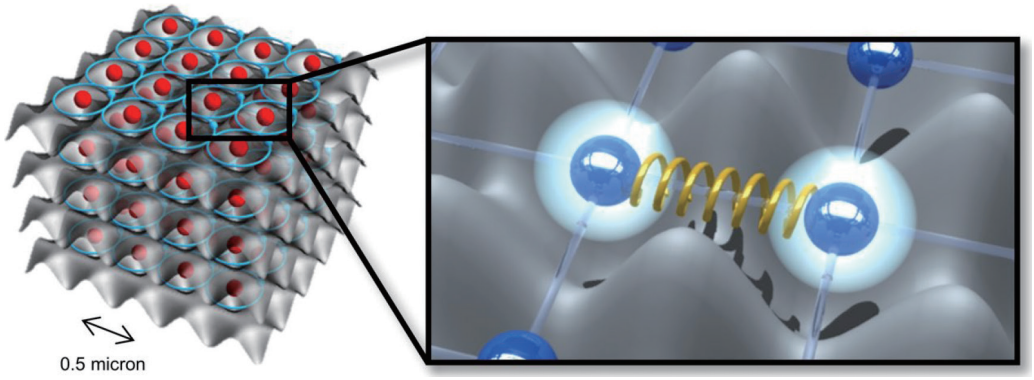

Figure 2. Conceptual diagram of the quantum entanglement between electronic and motional states in our ultrafast quantum simulation of magnetic material.[7)] Atoms in the optical lattice, trapped at a distance of 0.5 micron, are excited to the Rydberg state by the ultrafast excitation technique. Interaction between close Rydberg atoms results in the repulsive force, leading to the quantum entanglement between electronic and motional states of the atoms.

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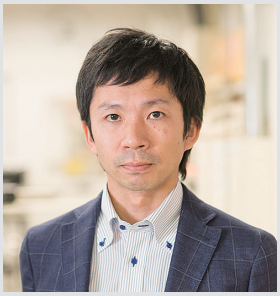
Electronic Property of Functional Organic Materials

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



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2013 Visiting Associate Professor, Soochow University
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Professor, The Graduate University for Advanced Studies
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2019 Visiting Professor, Kyoto University, Hiroshima University
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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.

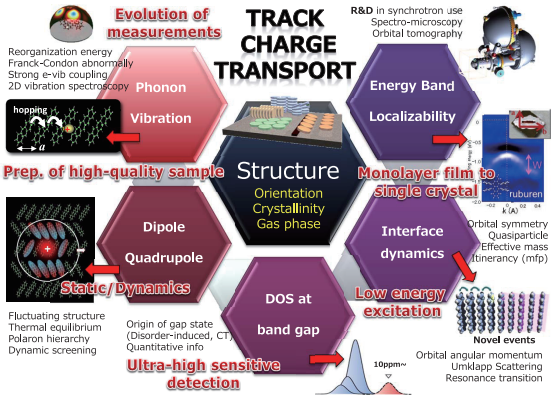

Figure 1. Overview of our agenda. A rich assortment of surface and interface structures of FOM to provide complicated spectral features of ultraviolet photoelectron spectroscopy.

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. Surface Characterization of 2D Covalent Organic Frameworks[1)]

Two-dimensional (2D) covalent organic frameworks (COFs) fabricated through on-surface synthesis were investigated as a honeycomb nanopore template for the growth of 3d-transition-metal nanoclusters (NCs) with a size of 2 nm on a metallic substrate. The evolution of these NCs and their electronic characteristics were studied employing scanning tunneling microscopy/spectroscopy (STM/STS), angle-resolved ultraviolet photoelectron spectroscopy (ARUPS), and X-ray photoelectron spectroscopy (XPS) under an ultrahigh-vacuum (UHV) condition at room temperature. The 2D COFs were synthesized on Cu(111) substrate utilizing 1,3,5-tris(4-bromophenyl) benzene (TBB) precursors, which engendered a honeycomb nanopore array of approximately 2 nm in size. In contrast to the behavior observed in the Co/Cu(111) system producing bilayer-stacking nanoclusters measuring 10–20 nm, STM imaging of Co/COFs/Cu(111) revealed the growth of Co NCs of approximately 1.5 nm within a single COF nanopore. This growth occurred without forming a monolayer film beneath the COFs, providing direct evidence that the 2D COFs on Cu(111) can effectively entrap Co atoms within the nanopore, giving rise to Co NCs (Figure 2). The spectroscopy measurements, STS, ARUPS, and XPS, confirmed the different local densities of states for Co NCs and COFs, corroborating the coexistence of Co NCs and COFs on the surface.

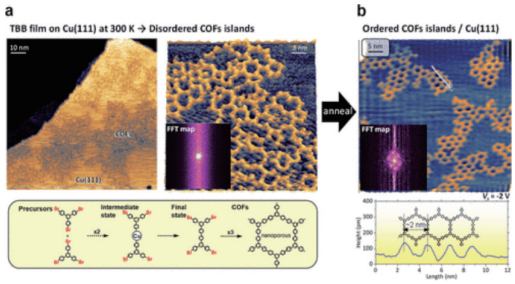

Figure 2. (a) UHV-STM topographic images obtained on the surface of TBB film grown on Cu(111) at 300 K with a schematic model of honeycomb lattice formation *via* the Ullman reaction. (b) STM topographic image of the ordered 2D-COFs islands on Cu(111) after the thermal annealing in UHV. The inset in the bottom panels denotes the FFT map. The figure is after ref 1).

2. Electronic Structure and Many-Body Interactions of Low-Mobility Carriers in Perylene Diimide Derivative[2)]

Despite the rapid progresses in the field of organic semiconductors, aided by the development of high-mobility organic materials, their high carrier mobilities are often unipolar, being sufficiently high only for either electrons or holes. Yet, the basic mechanisms underlying such significant mobility asymmetry largely remains elusive. We perform the ARUPS to reveal the occupied band structures and the many-body interactions for low-mobility hole carriers in a typical n-type semiconductor perylene diimide derivative (PTCDI-C8). The 50-nm thick PTCDI-C8 film consists of many needle-like crystallites, each of which are several hundreds of nanometres along its long axis by AFM, where the long axis of the crystallites was determined to be the crystallographic π-π stacking direction. The band dispersion is observed clearly even for random orientation of the molecular plane by thanks to large anisotropic nature of the BZ structure. The observed HOMO band by ARUPS exhibits strong renormalization to the calculated DFT results based on the single-particle electronic state (Figure 3). The analysis including many-body interactions elucidate that the significant mass enhancement can be understood in terms of strong charge–phonon coupling, leading to an important mechanism of polaron band transport of low intrinsic carrier mobility in organic semiconductor.

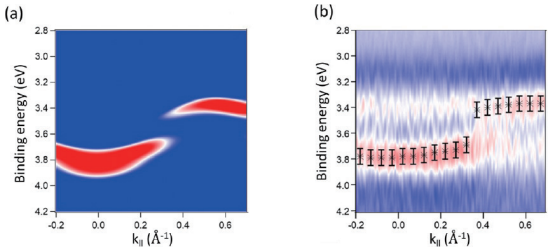

Figure 3. Second derivative plot of a simulated HOMO band dispersion with hole-intramolecular vibration coupling (a) and the observed HOMO by HeII ARUPS (b).

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)] The perspectives required for future light-source facility have been discussed with communities.[4)]

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3) F. Matsui, K. Hagiwara, E. Nakamura, T. Yano, H. Matsuda, Y. Okano, S. Kera, E. Hashimoto, S. Koh, K. Ueno, T. Kobayashi, E. Iwamoto, K. Sakamoto, S. Tanaka and S. Suga, *Rev. Sci. Instrum.* **97**, 083701 (10 pages) (2023).
4) S. Kera, F. Matsui, K. Tanaka, Y. Taira, T. Araki, T. Ohigashi, H. Iwayama, M. Fujimoto, H. Matsuda, E. Salehi and M. Katoh, *Electron. Struct.* **5**, 034001 (9 pages) (2023).

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Light Source Developments by Using Relativistic Electron Beams

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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.

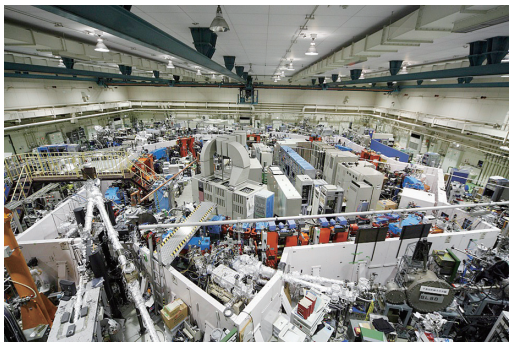


Figure 1. UVSOR-III Electron Storage Ring and Synchrotron Radiation Beamlines.

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. 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).
- T. Fuji, T. Kaneyasu, M. Fujimoto, Y. Okano, E. Salehi, M. Hosaka, Y. Takashima, A. Mano, Y. Hikosaka, S. Wada and M. Katoh, "Spectral Phase Interferometry for Direct Electric-Field Reconstruction of Synchrotron Radiation," *Optica* **10(2)**, 302–302 (2023).

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 (Figure 2), 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 (Figure 3).

In collaboration with Hiroshima Univ. and Nagoya Univ., we have succeeded in producing spatially structured synchrotron radiation such as vortex beam and vector beam, and we are exploring their applications.

We have been exploring the possibility utilizing the temporal structure of undulator radiation, in collaboration with Saga Light Source and Toyama Univ. We have succeeded in the coherent controls of atoms and in observing ultrafast change of an electronic state of an atom by using radiation from two undulators arranged in tandem. We have started developing state-of-the-art technology to observe ultrafast properties of synchrotron radiation, in collaboration with Toyota Technological Institute.

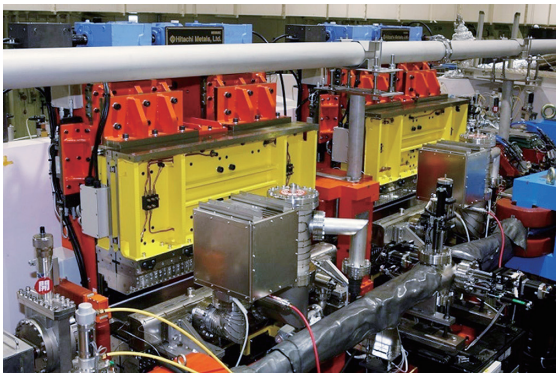


Figure 2. Twin Polarization-variable Undulators/Optical Klystron at UVSOR-III.

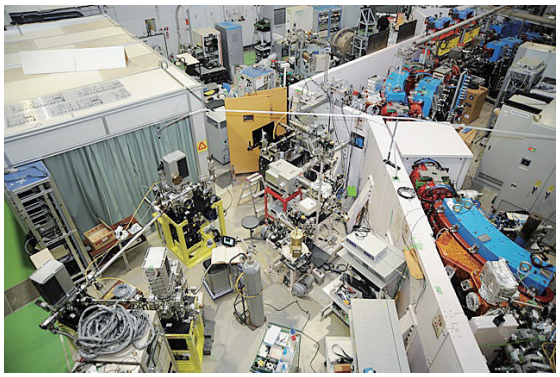


Figure 3. UVSOR BL1U experimental station for source development studies.

In these years, we are interested in the quantum nature of synchrotron radiation photons. We have established a technique to store only one electron in the synchrotron. We are working on experimental studies on photon emission from a single electron.

We have been developing a laser Compton scattering gamma-ray source at BL1U, which is capable of producing monochromatic and energy-tunable gamma-rays.

We continue experimental studies on the origin of the homochirality of biomolecules using intense circularly polarized undulator radiation at BL1U, in collaboration with Yokohama National Univ. and Hiroshima Univ. Recently we have started a new project on this subject, which includes specialist of plasma physics from NIFS.

2. Accelerator Technology Developments for Electron Synchrotrons

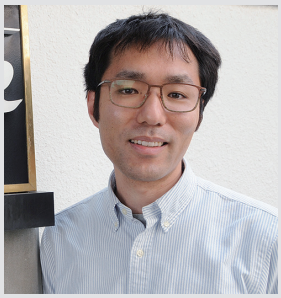
We carried out several upgrade plans on UVSOR electron synchrotron since 2000. We designed a special beam optics intended to higher brightness. We developed necessary accelerator components, reconstructed the accelerator and commissioned it. We have constructed and commissioned six undulators successfully. 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, however, the requirements from the users for the higher brightness is getting stronger, because new light sources and upgrade plans are being realized all over the world. We had seeked 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. However, they are not compatible with the operation of the narrow gap undulators. Then, we started a design study on a new light source facility. Currently we are focusing on designing a synchrotron with the electron energy of 1 GeV and the circumference of around 70 m. In parallel, we are designing a magnetic lattice which has same beam energy and circumference as the present machine but would give lower emittance.

We are collaborating with Nagoya Univ., Hiroshima Univ. and KEK Photon Factory and are developing new accelerator 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.

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

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research



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2018 Senior Research Scientist, National Institute of Advanced Industrial Science and Technology (AIST)
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Associate Professor, The Graduate University for Advanced Studies

Awards
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
2021 Outstanding Presentation Award, 64th Annual Meeting of the Japanese Society of Radiation Chemistry
2023 Young Scientist Award of the Japanese Positron Science Society

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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.

Inverse 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 inverse Thomson scattering (Figure 1). This ultra-short pulsed gamma-rays were applied to gamma-ray-induced positron annihilation pectroscopy (GiPAS). A positron is an excellent probe of atomic scale defects in solids and of free volumes in polymers at the sub-nm to nm scale. GiPAS enables defect analysis of a thick material in a few cm because positrons are generated throughout a bulk material via pair production.

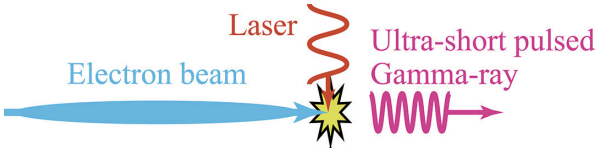

Figure 1. Schematic illustration of 90-degree inverse Thomson scattering.

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, T. Hayakawa and M. Katoh, "Gamma-Ray Vortices from Nonlinear Inverse Thomson Scattering of Circularly Polarized Light," *Sci. Rep.* **7**, 5018 (2017).
- 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).
- Y. Taira, R. Yamamoto, K. Sugita, Y. Okano, T. Hirade, S. Namizaki, T. Ogawa and Y. Adachi, "Development of Gamma-Ray-Induced Positron Age-Momentum Correlation Measurement," *Rev. Sci. Instrum.* **93**, 113304 (2022).
- Y. Taira *et al.*, "Measurement of the Spatial Polarization Distribution of Circularly Polarized Gamma Rays Produced by Inverse Compton Scattering," *Phys. Rev. A* **107**, 063503 (2023).

1. Gamma Ray-Induced Positron Annihilation Spectroscopy (GiPAS)

In GiPAS, defect analysis is performed by measuring the energy spectrum and emission time distribution (positron lifetime spectrum) of annihilation gamma rays, which are generated when a positron annihilates with an electron inside material. Gamma-ray-induced positron annihilation lifetime spectroscopy (GiPALS) is a technique that measures the time difference distribution between a reference signal and a detector output of annihilation gamma rays. The reference signal is the output of a photodiode placed near the collision point between the electron beam and the laser, which detects the laser just before it generates gamma rays. 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.

A digital oscilloscope is used to store the waveforms of the photodiode and the BaF_2 detector, and calculate the time difference distribution. One digital oscilloscope for four BaF_2 detectors is used as a pair of detection systems. 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. A detection system with eight detectors and two digital oscilloscopes was constructed (Figure 2). Time resolution is 140 ps in full width at half maximum, which is high despite the use of a 52-mm thick BaF_2 scintillator. The count rate is 20 cps.

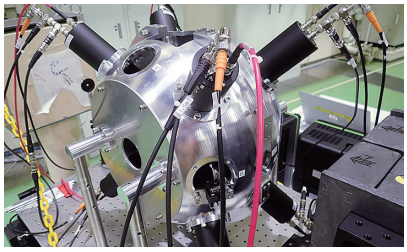

Figure 2. Gamma-ray-induced positron annihilation lifetime spectroscopy system using eight detectors and two digital oscilloscope.

Currently, user applications of GiPALS are underway at BL1U of UVSOR, and users from universities, research institutes, and private companies are using the system. Measurements of samples under special environments such as stress loading, high temperature, gas atmosphere, laser irradiation, hydrogenation, etc., which are difficult to measure with conventional methods, are being performed.

Meanwhile, we are also developing gamma-ray-induced spin-polarized positron annihilation spectroscopy using circularly polarized gamma rays. If the electron spins of a sample are ordered in a particular direction and the positrons are also spin-polarized, the Doppler broadening spectra of annihilation gamma rays and the positron lifetime will change. The spin-polarized positrons are generated from the circularly polarized gamma rays inside a sample. From this change, it is possible to obtain information about the electron spins around defects in magnetic materials. To demonstrate the principle of circularly polarized gamma-ray-induced spin-polarized positron annihilation spectroscopy, a pure iron sample is mounted between permanent magnets and the positron lifetime and Doppler broadening are measured. We have not been able to measure the difference in positron lifetime due to the helicity inversion of circularly polarized gamma rays, but we will continue our research and development.

2. Measurement of Gamma-Rays Generated by Using Polarized Lasers

Inverse Compton scattering of a polarized laser by energetic electrons is an excellent method to generate polarized gamma rays. The development and use of linearly and circularly polarized gamma rays have been conducted. The polarization state of linearly and circularly polarized lasers is homogeneous across their cross sections. However, it is possible to produce lasers with spatially variant polarization states. An example is the axially symmetric polarization state, referred to as an axially symmetric polarized laser or a cylindrical vector beam. Although the polarization characteristics of gamma rays produced by linearly or circularly polarized lasers have been theoretically clarified, that of gamma rays generated by axially symmetric polarized lasers have not. If gamma rays with novel polarization characteristics can be generated, it is possible to develop new ways to use gamma rays.

The spatial distribution of the gamma rays, which reflects the polarization characteristics, was measured with a two-dimensional CdTe imaging sensor. Gamma rays were generated through 90-degree collisional inverse Compton scattering between an electron beam and an axially symmetric polarized laser. The results showed that the spatial distribution of gamma rays generated from axially symmetric polarized lasers was changed compared to that of linearly or circularly polarized gamma rays. Comparing the linearly polarized gamma rays with those generated by the axially symmetric polarized laser, a node that appears at an outer scattering angle along the polarization axis of the linearly polarized gamma ray was absent in the gamma ray generated by the axially symmetric polarized laser. Compared to the circularly polarized gamma rays, the gamma rays generated by the axially symmetric polarized laser showed a spatial distribution that was slightly expanded in a specific direction rather than concentric, while the circularly polarized gamma rays showed a concentric spatial distribution. This was thought to be due to the relatively intense polarization component of the axially symmetric polarized laser. These results suggested that axially symmetric polarized lasers generate gamma rays with different polarization states. In the near future, a polarimeter of gamma rays will be constructed to investigate the spatial polarization distribution of gamma rays.

Award

TAIRA, Yoshitaka; Young Scientist Award of the Japanese Positron Science Society (2023).

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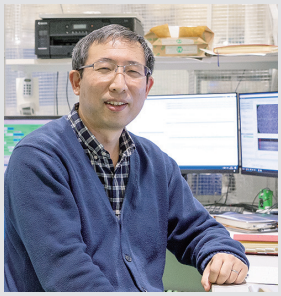
Novel Spin and Chiral Materials Science by Photoelectron Cinemato-Microspectroscopy

UVSOR Synchrotron Facility
Division of Advanced Solid State Physics



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Education
1995 B.S. The University of Tokyo
1997 M.S. The University of Tokyo
2000 Ph.D. The University of Tokyo

Professional Employment
2000 Assistant Professor, Nara Institute of Science and Technology
2011 Guest Professor, Physik Insitut, Universität Zürich, Switzerland (–2012)
2013 Associate Professor, Nara Institute of Science and Technology
2018 Senior Researcher, Institute for Molecular Science
2021 Professor, Institute for Molecular Science
Professor, The Graduate University for Advanced Studies

Awards
2007 NAIST Award (NAIST foundation)
2008 The Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology, Awards for Science and technology (Research Category)
2009 Young Scientist Award of the Physical Society of Japan
2021 The NAGAI Foundation for Science & Technology Encouragement Award

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Keywords Photoelectron Spectroscopy, Momentum Microscope, Electronic Spin Structure

Imagine cherry blossom petals fluttering down. Under a uniform gravitational field, the slight asymmetry of the petals creates anisotropy in the airflow, causing the petals to rotate. Capturing the movement of this dynamic 3D-structure as a *cinema* is important for understanding the physics behind this.

When electrons in a material are excited by photons, they are emitted into the vacuum as photoelectrons. Interestingly, the angular distribution of these photoelectrons shows a truly beautiful holographic pattern derived from the motion of valence electrons and the arrangement of atoms in the material. Photoelectron momentum microscope (PMM) is an analyzer that can instantaneously image the behavior of electrons of material and device surfaces. PMM combines imaging-type photoelectron spectroscopy and microscopy techniques to visualize the electronic state (band dispersion, composition, and spin polarization) in reciprocal lattice space of a selected μm-sized area. We have constructed the world's first dual beamline (soft x-ray; SX and vacuum ultraviolet; VUV) PMM system at the IMS UVSOR synchrotron facility!

What we are currently very interested in is chiral phase transition phenomena, for example. The layered material TaS_2 exhibits an attractive behavior in which it undergoes a phase transition to a chiral structure at low temperatures and its electrical conductivity changes by two orders of magnitude. We succeeded in capturing a video of the hysteresis change in the valence band dispersion during this phase transition. The results serve as a benchmark experiment for *in situ* observation of material responses to various external fields.

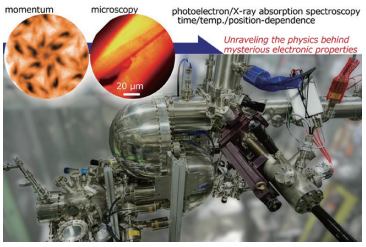

Figure 1. Photoelectron momentum microscope station at UVSOR synchrotron facility. Multimodal measurements hold the key to unlocking the mysteries of the electronic properties of materials.

Selected Publications

- K. Hagiwara, F. Matsui *et al.*, "Development of Dual-Beamline Photoelectron Momentum Microscopy for Valence Orbital Analysis," *J. Synchrotron Radiat.* **31**, 540 (2024).
- F. Matsui *et al.*, "Soft X-Ray Photoelectron Momentum Microscope for Multimodal Valence Band Stereography," *Rev. Sci. Instrum.* **94**, 083701 (2023).
- F. Matsui *et al.*, "Domain-Resolved Photoelectron Microscopy and μm-Scale Momentum-Resolved Photoelectron Spectroscopy of Graphite Armchair Edge Facet," *J. Phys. Soc. Jpn.* **91**, 094703 (2022).
- F. Matsui and S. Suga, "Coupling of k_z-dispersing π Band with Surface Localized States in Graphite," *Phys. Rev. B* **105**, 23526 (2022).
- F. Matsui and H. Matsuda, "Projection-type Electron Spectroscopy Collimator Analyzer for Charged Particles and X-Ray Detections," *Rev. Sci. Instrum.* **92**, 073301 (2021).
- F. Matsui *et al.*, "Photoelectron Momentum Microscope at BL6U of UVSOR-III synchrotron," *Jpn. J. Appl. Phys.* **59**, 067001 (2020).

1. Visualization of Phase Transition and Phase Separation

The most distinctive advantage of PMM[1] is its snapshot-style of measuring photoelectron constant energy intensity distribution pattern in real and reciprocal spaces. Recently, we succeeded in the development of on-the-fly scans for temperature-dependent Fermi surface and valence band measurements during phase transitions, taking advantage of the rapid acquisition (seconds to minutes) of photoelectron patterns.

1T-TaS_2, which exhibits various phase transitions in structural and electronic properties, has fascinated researchers for more than half a century. However, despite many angle-resolved photoelectron spectroscopy (ARPES) studies on 1T-TaS_2, the basic understanding of charge density wave (CDW) formation is far from a common consensus. As for the mechanism of the metal–insulator transition, the Mott insulator model, band insulator model, and one-dimensional metal model have been so far proposed.

In order to solve the mystery of CDW formation in 1T-TaS_2, we conducted the following photoemission measurements.[2] (i) Band-dispersion mapping from selected μm-scale chiral CDW domains, (ii) characterization of 3D CDW nesting vectors by complete Brillouin-zone-dispersion measurements and (iii) on-the-fly observation of valence band and core-level photoelectron spectra during temperature changes.

Figure 2 shows a series of 1T-TaS_2 valence band cross sections scanned in 2D real space in the low-temperature CDW phase. Here, a domain boundary between two different twin domains is recognized. Typical areas of domains are tens to hundreds of μm^2. We exploited the microscopic capabilities of PMM to analyze the electronic structure of selected single domains. By measuring the valence band pattern during temperature change in real time, the hysteresis behavior of the phase transition was visualized in detail.

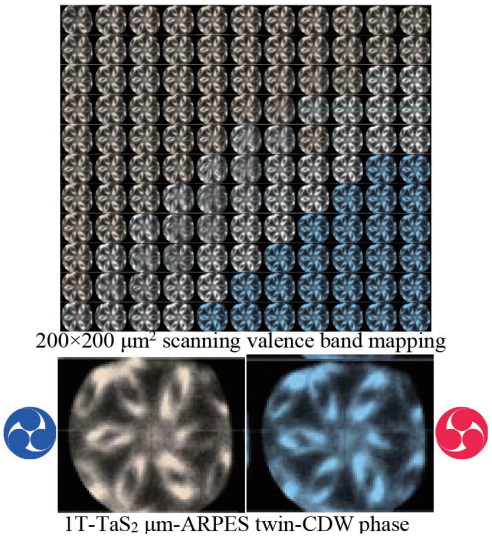

Figure 2. Valence band photoelectron patterns of 1T-TaS_2 surface in the low-temperature CDW phase. The field of view for the ARPES snapshot measurements was a few μm in diameter.

2. Analyzing Polarization of Atomic Orbitals and Electron Spins

The combination of PMM with synchrotron SX and VUV undulators paves the way for comprehensive characterization of atomic orbitals in the Fermi surface and valence band on the μm scale.[3]

Figure 3 shows the 2D momentum (k_x, k_y) distribution of photoelectron intensity of the Au(111) at the Fermi level.[3] Figures 3(a) and (b) show the 2D patterns measured using p-polarized SX light from BL6U, while s-polarized VUV light from BL7U is used in the case of Figures 3(c) and (d). In both conditions, the photoelectron pattern in the entire Brillouin Zone is covered. Shockley surface state centered at the Γ point as small circular contours (Figure 3(a)) is clearly observed with p-polarized excitation with splitting due to spin–orbit coupling (Figure 3(b)), but these features exhibit very weak intensity for normal incidence light (Figure 3(c)). The Shockley surface state mainly comprises 6s and $6p_z$ orbitals. In the normal-incidence geometry, the transition-matrix element from the initial s and p_z orbitals becomes 0 at the photoemission direction orthogonal to the excited electric vector (Figure 3(d)). The relationship between the orbital angular momentum and the effects of the transition matrix elements can be directly investigated using this normal incidence geometry.

Finally, our main goal is to employ these techniques with spin polarization sensitivity. We have just started obtaining spin-polarized valence band dispersion data for typical materials having spin polarized surface states.

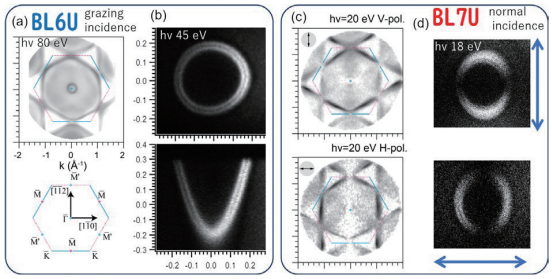

Figure 3. Fermi surface of Au(111) surface measured using grazing incident SX (a,b) and normal incident VUV (c,d) beam. (b,d) The Shockley surface state at the center is zoomed and analyzed. Parabolic band dispersion with splitting due to spin–orbit coupling, *i.e.* Rashba effect, is well resolved.

References

1) F. Matsui, S. Makita, H. Matsuda, T. Yano, E. Nakamura, K. Tanaka, S. Suga and S. Kera, *Jpn. J. Appl. Phys.* **59**, 067001 (2020).
2) F. Matsui, K. Hagiwara, E. Nakamura, T. Yano, H. Matsuda, Y. Okano, S. Kera, E. Hashimoto, S. Koh, K. Ueno, T. Kobayashi, E. Iwamoto, K. Sakamoto, S. Tanaka and S. Suga, *Rev. Sci. Instrum.* **94**, 083701 (2023).
3) K. Hagiwara, E. Nakamura, S. Makita, S. Suga, S. Tanaka, S. Kera and F. Matsui, *J. Synchrotron Radiat.* **31**, 540 (2024).

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Angle-Resolved Photoemission Study on Strongly Correlated Electron Materials

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2000 B.S. The University of Tokyo
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Keywords Strongly Correlated Electron System, Synchrotron Light, Photoemission

Strongly correlated electron materials have 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 strucature 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 (Figure 1).

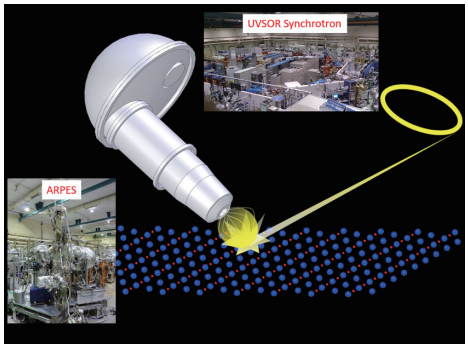

Figure 1.

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. Evidence of Strong Violation from Fermi Arc Picture in Heavily Underdoped Bi2212

Understanding the complex nature of cuprate superconductors, especially the origin of their high superconducting transition temperature (T_c), is one of the greatest challenges in condensed matter physics. An early experimental observation in this regard is the famous Uemura plot. It shows that the T_c in the high-T_c cuprate superconductors is correlated with the muon spin relaxation rate σ_0 that is proportional to the superfluid density divided by the effective mass ($\sigma_0 \propto n_s/m^*$). This relation holds not only for all high-T_c cuprate superconductors but also for many other superconductors. While there have been various attempts at a theoretical explanation of the Uemura relation, a connection to other experimentally probed properties of cuprates is still lacking.

ARPES is a powerful experimental technique, directly measuring 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, especially the superconducting (SC) gap. Indeed, the first direct experimental evidence of *d*-wave symmetry of the SC gap has been reported by ARPES, where the SC gap follows the d-wave order parameter $\Delta(k) = \Delta_0(\cos k_x a - \cos k_y a)/2$, here a is the lattice constant. In ARPES field, T_c of cuprates has been believed to be explained by so-called "Fermi arc" picture. According to this picture, Fermi arc is a region where the SC gap closes above T_c in the momentum space and the "effective SC gap" Δ_{SC} can be defined by the gap magnitude at the edge of the Fermi arc as shown in Figure 2 (a). Since Δ_{SC} and T_c show linear correlation among different doping and even among different families of cuprates as shown in Figure 2 (d), this picture is well accepted as a phenomenological model to explain T_c in cuprates.

Recently, we have studied heavily underdoped Bi2212, whose T_c is only 30 K but still high-T_c, and found strong violation from the Fermi arc picture. In this doping, there is almost no temperature dependence of the SC gap across T_c as shown in Figure 2 (b)–(c). Therefore, Δ_{SC} should be extremely small (~1 meV) and should not be enough to explain 30 K of T_c. To understand the origin of the superconductivity in this doping, we have performed new way of ARPES analysis, where information of the spectral weight difference across T_c can be estimated. Interestingly, this new analysis shows that the doping dependence of the spectral weight along the Fermi surface quantitatively scales with the superfluid density (and T_c) for a wide range of carrier concentrations (not shown). Our new results represent the first evidence showing the close relationship between the superfluid density and T_c from ARPES. Our results also show that the whole Fermi surface contributes to superconductivity and the superfluid density can be more important than the SC gap. This is completely new idea to understand high-T_c superconductivity from ARPES.

Since it is well known that ARPES intensity strongly depends on the matrix element, which can be changed by measurement geometry, polarization, photon energy and so on, our spectral weight analysis should be affected by the measurement conditions. To make sure that our observation is universal, we have to perform many ARPES studies by changing measurement geometry, polarization and photon energy. To perform measurements with different measurement geometry, a low temperature 6-axis manipulator is necessary, since measurement geometry should be able to be changed to study the same sample surface with different geometries. We are using our home-made 6-axis manipulator which is one of the lowest temperature 6-axis manipulators in the world to complete this study.

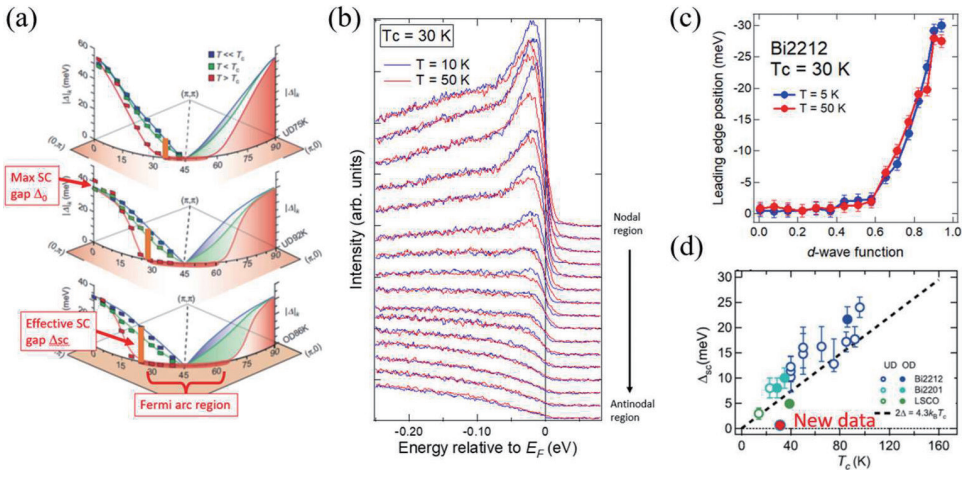

Figure 2. (a) Temperature and doping dependence of SC gap in cuprates and Fermi arc picture. (b) Temperature dependence of ARPES spectra along Fermi surface of heavily underdoped Bi2212. (c) Temperature dependence of SC gap obtained by the leading-edge position. (d) Comparison of the effective SC gap in different families of cuprates including current study.

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Soft X-Ray Spectro-Microscopy and Scattering for Life Science—beyond Organelle Mapping

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Division of Advanced Photochemistry

ARAKI, Tohru
Senior Researcher

Beside running the user programs using the BL4U at the UVSOR, which is a STXM (Scanning Transmission X-ray Microscopy) beamline. STXM is one of the x-ray based spectro-microscopy techniques providing us a label-free chemical mapping for a wide variety of sciences like energy materials, environmental and earth science, and many in the industrial science as well. I want to push this technique applying for a life science, especially with soft x-rays, which has been studied by mainly hard x-ray regimes such as protein crystallography and small angle scattering at the synchrotron radiation facilities so far.

One of the most effective soft X-ray microscopes, the so-called soft X-ray tomography (SXT), can offer unparalleled 3D organelle mapping of entire biological cells larger than 10 micrometres. The SXT uses a fixed photon energy, just before the O K-edge absorption at 520 eV, to observe the hydrated sample using the "water window" energy region. This allows us to see the absorption contrast of the biological sample, which comes from the carbon and nitrogen content inside them. The distinctive LAC (linear absorption coefficient) at 520 eV of each organelle enables the delineation of these structures. I intend to extend this analysis by employing the full potential of the SXAS (soft X-ray absorption spectroscopy) technique, which allows for the utilisation of a range of photon energies. The utilisation of SXAS-based spectro-microscopy and spectroscopy-scattering enables the differentiation of analogous organelles and oxidation states of metal/ion within cellular structures. In order to establish this methodology, two key steps must be taken: Firstly, a basic spectral interpretation of organelles must be conducted, and secondly, the sample preparation and specimen environment must be optimised. It is of particular importance to refrain from any modifications of the samples and to maintain the native states of the cells, including the loss of metals or ions, throughout both the sample preparation and the data collection process, in order to prevent radiation damage.

In the current year, I conducted a collaborative research project on *Ramazzottius varieornatus*, a tardigrade that is renowned for its anhydrobiotic capabilities, enabling it to survive in harsh, arid environments. My colleague prepared the thin section samples embedded in resin for a comparative morphometric analysis of their microscopic anatomy using both scanning electron microscopy (SEM) and STXM. Figure 1 (a) illustrates a representative optical microscope image. In order to enhance contrast, a staining agent must be applied to the sample prior to conducting a SEM analysis. However, it offers a higher spatial resolution than other techniques. In comparison, STXM is a label-free method that provides a lower spatial resolution, yet still achieves a reasonable resolution of tens of nanometres. Figure 1 (b) depicts a STXM transmission raw image collected at the protein characteristic photon energy. Figure 1 (c) presents a RGB composite map, comprising the three STXM images acquired at distinct photon energies. The images provide insight into the structural and compositional characteristics of different organelles and biological molecules.

These results prove the STXM's capabilities. However, there is still work to be done to fully understand the images and extract all the information they contain. One possible approach is the cryogenically vitreous ice-covered sample prepared by the plunge-freeze method. This is currently regarded as the gold standard for such sample preparation in the fields of Cryo-EM and Cryo-SXT.

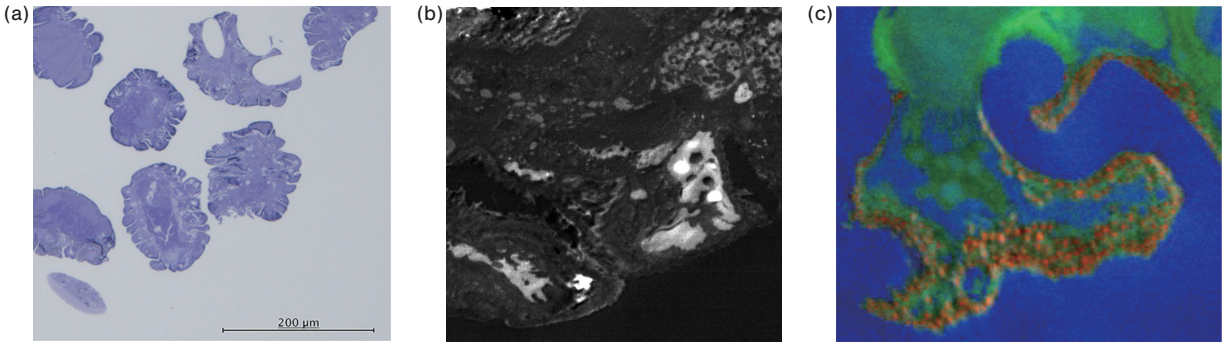
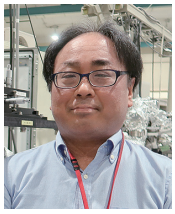


Figure 1. (a) The optical microscope images of the stained tardigrade thin section tissue samples. (b) The STXM transmission image of the tardigrade thin section tissue sample. (30 × 30 μm). (c) The RGB composite map of the tardigrade samples obtained by the STXM images at the three different photon energies. (16 × 14 μm).

Development of Resonant Soft X-Ray Scattering Spectroscopy for Soft Matter

UVSOR Synchrotron Facility
Division of Advanced Photochemistry



IWAYAMA, Hiroshi
Senior Researcher

Soft matter, such as liquid-crystal and polymer, is essential in various technological applications. Interesting soft matter properties come from their mesoscopic structures and dynamics. Small-angle X-ray and Neutron Scattering (SAXS/SANS) are powerful tools for revealing mesoscopic structures. However, since SAXS is sensitive to modifications of electron densities, their contrast difference between similar chemical species is weak.

Resonant soft X-ray scattering (RSoXS) is an emerging characterization technique for mesoscopic structural analysis. Soft matter molecules mainly consist of light elements such as carbon, nitrogen, and oxygen, whose X-rays resonant energies are in the soft X-ray region. Resonant scattering is strongly sensitive to its element, chemical species and the molecular alignment relative to polarization vector. In particular, molecular alignment sensitivity enables us to investigate twisted structure of soft matter in mesoscopic scale.

This year, we developed a new RSoXS spectrometer. The feature of the new spectrometer is that a soft X-ray detector can be rotated along a scattering angle in vacuum. Compared to X-rays used for SAXS measurements, wavelengths of soft X-rays are much longer. Since a scattering vector, q, is proportional to $\sin(\theta)/\lambda$, where 2θ and λ are scattering angle and a wavelength of soft X-ray, a scattering angle becomes wider compared to SAXS. For examples, to analyze the structure from 5 nm to 500 nm, we need to measure the scattering angle 2θ from 0.5 to 60 degrees. To achieve this wide scattering angle measurement, we developed a new RSoXS spectrometer, where a soft X-ray detector can be rotated along the 2θ in the vacuum chamber, as shown in Figure 1.

For the performance evaluation, we investigated twisted bend (TB) liquid-crystal molecules with the new RSoXS spectrometer. The TB liquid-crystal phase is the newest nematic phase, only identified in 2011. There are many outstanding mysteries about the nature of its nanoscale organization and behavior. Our collaborators elucidate how the number of monomer units in a linear TB oligomer influences the structure of its nanoscale helix, an important TB phase structure–property relationship.[1] Figures 2 show a schematic view of TB liquid-crystal molecules and a scattering image at a photon energy of 285 eV ($\lambda \sim 4.3$ nm), which is corresponding to carbon K-edge resonance. We find a sharp diffraction ring corresponding to the half-pitch periodicity of the helical filaments ($d \sim 128$ nm ($q \sim 0.005$ Å^{-1})). We also confirmed that the detector can be rotated along 2θ and we can measure scattering angles from 0 to 60 degrees. We started to analyze molecular orientation orders of liquid-crystal molecules and crosslinked structures of polymers with the spectrometer.

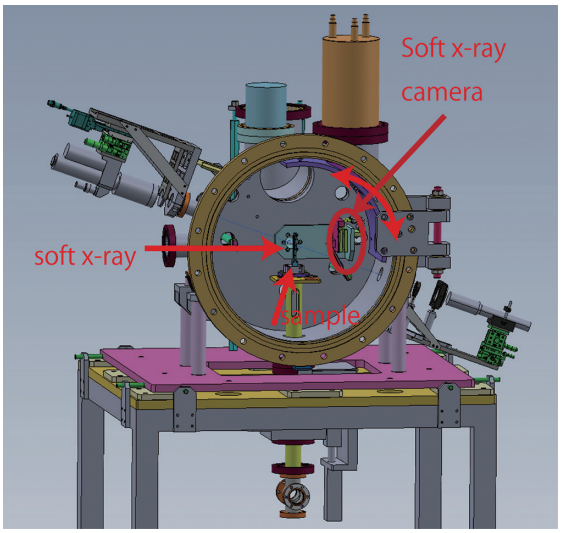


Figure 1. Schematic draw of a new RSoXS spectrometer.

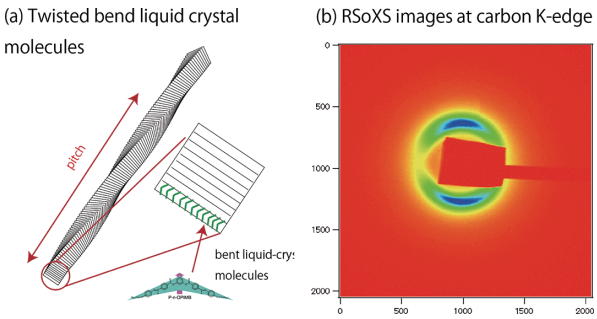


Figure 2. (a) Schematic view of twisted bend liquid crystal molecules. (b) Scattering image at a photon energy of 285 eV.

Reference

1) Y. Takanishi, F. Araoka and H. Iwayama, *RSC Adv.* **12**, 29346 (2022).

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

Department of Photo-Molecular Science
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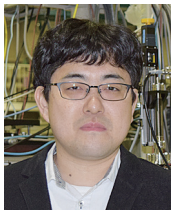

NAGASAKA, Masanari
Assistant Professor

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.[1,2] In this year, we investigated cononsolvency mechanism of polymer solutions[3] and isolated water molecules in aqueous acetonitrile solutions.[4]

1. Mechanism of Polymer Cononsolvency Explored via Oxygen K-Edge XAS

The cononsolvency of poly(*N*-isopropylacrylamide) (PNIPAM), dissolving in pure methanol and water but being insoluble in aqueous methanol solutions, was investigate by O K-edge XAS.[3] The cononsolvency emerges from the aggregation of PNIPAM with methanol clusters, leading to the collapse of the hydrophobic hydration of PNIPAM.

2. Probing Isolated Water Molecules in Aqueous Acetonitrile Solutions

O K-edge XAS of aqueous acetonitrile solutions exhibited a sharp peak around 537 eV, which was like that of water vapor and was not observed in liquid water. The inner-shell calculations revealed that the sharp peak profiles were derived not from water clusters but from isolated water molecules surrounded by acetonitrile molecules.[4] It means that the isolated water molecules in aqueous acetonitrile solutions can be analyzed using O K-edge XAS, which separates the contributions of small water clusters.

References

1) M. Nagasaka and N. Kosugi, *Chem. Lett.* **50**, 956–964 (2021).
2) M. Nagasaka, H. Yuzawa and N. Kosugi, *Anal. Sci.* **36**, 95–105 (2020).
3) M. Nagasaka, F. Kumaki, Y. Yao, J. Adachi and K. Mochizuki, *Phys. Chem. Chem. Phys.* **26**, 13634–13638 (2024).
4) M. Nagasaka, *J. Phys. Chem. Lett.* **15**, 5165–5170 (2024).

Visiting Professors

Visiting Professor
FUKUHARA, Takeshi *(from RIKEN)*

Quantum Gas Microscope for Rb-85 Atoms with a Tunable Atom–Atom Interaction

Ultracold quantum gases in optical lattices provide a clean and controllable platform for studying quantum many-body systems; especially they enable us to emulate a variety of fundamental models in solid-state physics. An "artificial" quantum spin system described by the spin-1/2 Heisenberg model can be realized by using ultracold bosonic atoms in optical lattices. In this realization, the anisotropy of the spin–spin interaction can be controlled by changing the interatomic interaction via a Feshbach resonance. Although, for quantum gas microscope experiments, rubidium-87 atoms have been utilized, in this study we performed experiments using rubidium-85 atoms which have a Feshbach resonance at a magnetic field of $B \sim 155$ Gauss. We succeeded in observing rubidium-85 atoms in a triangular lattice at the single-atom level. We also confirmed the Feshbach resonance through atom loss spectroscopy. This system is expected to be used for a quantum simulation of frustrated magnets that follow the spin-1/2 triangular-lattice antiferromagnetic Heisenberg model.

Visiting Professor
MATSUSHITA, Tomohiro *(from Nara Institute of Science and Technology)*

Development of Analysis Methods for Photoelectron Momentum Microscope

The photoelectron momentum microscope introduced at UVSOR is a highly powerful tool for observing the composition and electronic structure of samples using photoelectron spectroscopy. This pioneering analytical apparatus enables the observation of Fermi surfaces and band structures in various systems, including surface atomic sites, thin films, interfaces, surface adsorbates, and polycrystalline materials. Currently, we are developing two methods for analyzing the data. First, the observed photoelectron intensities from valence band depend on the transition probabilities of photoelectrons, resulting in modulation of these intensities. To calculate this intensity modulation, we are developing an analysis tool based on the first-principles calculation code OpenMX. Second, we are utilizing information technology to process data and extract hidden information. Specifically, we are applying principal component analysis to the data obtained from this instrument to visualize the behavior of phase transitions.

Visiting Associate Professor
NAKAYAMA, Yasuo *(from Tokyo University of Science)*

Epitaxially-Grown Single-Crystalline Organic Molecular Semiconductors

Single-crystalline organic semiconductor materials exhibiting "band transport" realize considerably high charge carrier mobility of over 10 $cm^2V^{-1}s^{-1}$ and have potential applications as flexible and efficient electronic devices. Whereas current organic semiconductor electronics are mostly composed of polycrystalline or amorphous molecular solids, our group has been working on single-crystalline organic semiconductor solids and their heterojunctions formed by "molecular beam epitaxy" techniques to pursue potential applications as flexible and efficient electronic devices. Recently, we discovered as a collaborative work with an IMS group that an n-type molecule C_{60} forms well-ordered epitaxial heterojunctions on the single-crystal surface of a high-mobility p-type molecule dinaphthothienothiophene (DNTT). This work was selected as one of the "Spotlight2023" articles. In addition, our group is also engaged in exploration into the fundamental properties of molecular single-crystal materials themselves, and have published collaborative works with the UVSOR facility on vibrational properties of the single-crystals of DNTT and pentacene.