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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1987 B. E. The University of Tokyo
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Professional Employment
1992 Research Associate, Tohoku University
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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), Heidelberg University
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 Medal with Purple Ribbon (by His Majesty the Emperor of Japan)

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JAUNET-LAHARY, Titouan
KUMAR, Pushpander
SRAKAEW, Kritsana
TIRUMALASETTY PANDURANGA, Mahesh
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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.

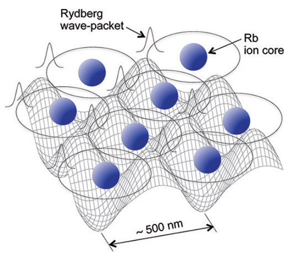

Figure 1. Metal-like quantum gas. A schematic of the many-body quantum simulator with ultracold Rydberg atoms, named "ultrafast quantum simulator," where electronic wave functions spatially overlap between neighboring atoms.[2]

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," *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).
- Y. Chew *et al.*, "Ultrafast Energy Exchange between Two Single Rydberg Atoms on a Nanosecond Timescale," *Nat. 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.[5–7] 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 degrees of freedom of atoms in our "ultrafast quantum simulator," generated by the repulsive force due to the strong interaction between Rydberg atoms in the neighboring lattice sites.[7] We have also proposed a new protocol including this repulsive force to introduce phonon motion into the quantum simulation.

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[8,10–13]

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 drastically reduce the noise effects. Moreover, this ultrafast two-qubit gate is faster than the fast two-qubit gate demonstrated recently by "Google Quantum AI" with superconducting qubits.[9] We are currently improving its key underlying technologies with optical tweezers and operating lasers.[10–13]

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

References

1) K. Tonomura *et. al.*, *Am. J. Phys.* **57**, 117 (1989).
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.*, *Phys. Rev. A* **110**, 053518 (2024).
11) T. P. Mahesh *et al.*, *Opt. Lett.* **50**, 403 (2025).
12) T. Denecker *et al.*, *Phys. Rev. A* **111**, 042614 (2025).
13) T. Tomita *et al.*, arXiv:2410.03241 (2024).

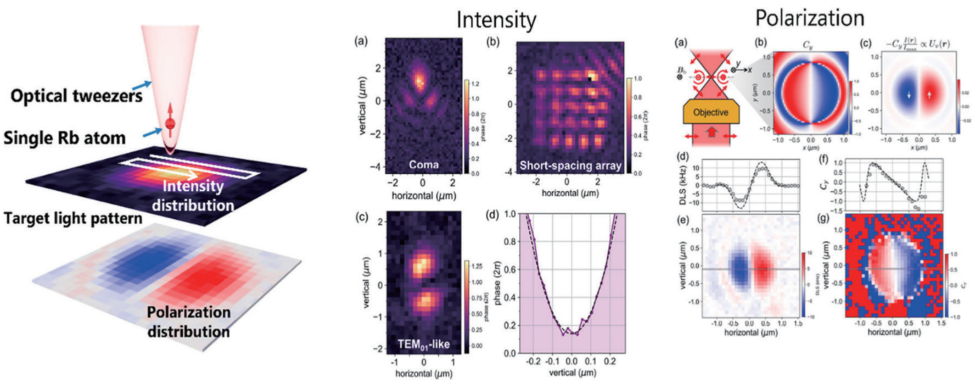

Figure 2. Atom Camera: Super-resolution scanning microscope of a light pattern with a single ultracold atom.[13] This would be useful for super-resolution diagnosis of the spatial profiles of the intensity and polarization of optical tweezers and gate operation lasers.

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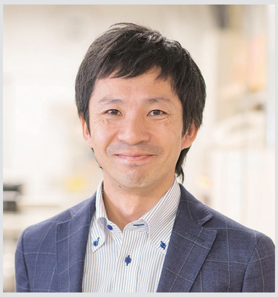
|| IMS International Internship Program

Electronic Property of Functional Low-Dimensional Material Systems

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



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2007 Associate Professor, Chiba University
2009 Visiting Associate Professor, Institute for Molecular Science
2013 Adjunct Lecturer, The Open University of Japan
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2014 Professor, Institute for Molecular Science
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2019 Visiting Professor, Kyoto University, Hiroshima University
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Keywords Photoelectron Spectroscopy, Molecular Assemble, Electronic State

Functional low-dimensional material systems (LMS) such as oriented molecular assembles on the surface have recently attracted considerable attention both for fundamental research and device applications because of peculiar properties not found in bulk materials or 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 would be conserved as in an isolated molecule even for solid phases due to the weak van der Waals interaction. To reveal characteristics of the LMS, 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 LMS 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).

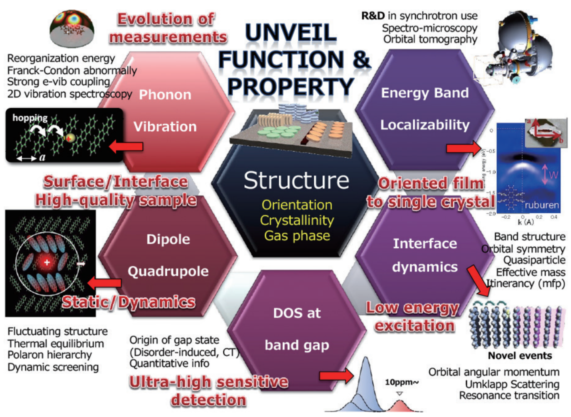

Figure 1. Overview of our agenda. A rich assortment of surface and interface structures of LMS to provide complicated spectral features of ultraviolet photoelectron spectroscopy.

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. Enantiospecific Mirror-Imaged Growth in Overlayers of Enantiopure Helicene on Au(111) without Commensurability[1)]

Two-dimensional crystallization of chiral molecules on achiral crystal surfaces typically exhibits mirror-imaged growth, defined by the substrate's mirror plane and the lattices of each enantiomer. While various commensurate molecular overlayers have shown such growth, the possibility of achieving substrate-defined enantiospecific structures in non-commensurate chiral molecular overlayers remains elusive. Here, enantiopure thiadiazole-[9]helicene on Au(111) is shown to form overlayers without commensurability, exhibiting substrate-defined mirror-imaged growth (Figure 2). This study experimentally demonstrates that the rotational orientation locking without two-dimensional interface potential minima can support mirror-imaged growth, suggesting that it can serve as benchmark for enantiospecific growth in a broader range of chiral molecular systems. The well-defined chiral molecular system will be useful for studying the mechanisms of chirality-induced spin selectivity at the interface in the near future.

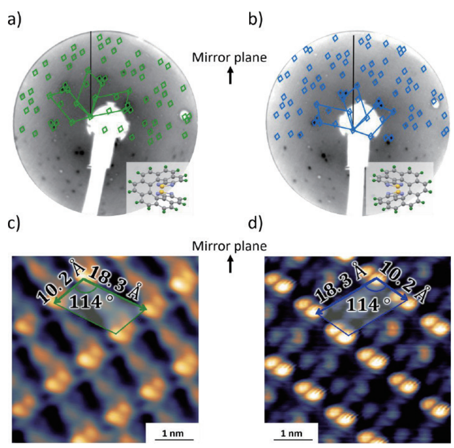

Figure 2. (a) and (b) are the distortion-corrected LEED images for (M)-TD[9]H and (P)-TD[9]H molecular layers on Au(111), observed at beam energies of 39.5 eV and 41.5 eV, respectively. (c) and (d) are STM images of (M)-TD[9]H and (P)-TD[9]H layers, respectively. The figure is after ref 1).

2. Fingerprinting Weak Electronic Interaction at a van der Waals Interface: Fano Signatures in Pentacene Monolayer on Graphite[2)]

The influence of van der Waals (vdW) interactions on the electronic structure at the interface between a pentacene monolayer and a graphite surface was investigated by using high-resolution angle-resolved photoelectron spectroscopy (ARPES) with synchrotron light sources. Upon cooling below 130 K, the pentacene molecules form a densely packed monolayer characterized by newly developed dispersive bands. These bands, observed using low-energy ARPES with photon energy varying from 7.2 to 8.5 eV, exhibit constant final state characteristics that overlap with nondispersive molecular orbital states (Figure 3). The results in variations for the photoemission intensity, both enhancement and suppressions the photoemission intensity, indicative of Fano resonance. Such resonance arises from the interaction between a discrete molecular state and a continuum state. The Fano profile analysis of spectral fine features reveals the wave-function connection by the broader spread in unoccupied states at the physisorbed interface. This discovery underscores the significant role of weak electronic coupling in shaping wave function connectivity, highlighted by the broader spread of unoccupied states. This spread serve as a spectral fingerprint for proving weak interactions at the vdW interface. The asymmetric parameter will provide quantifiable metrics for characterizing weak interactions, with further theoretical developments anticipated.

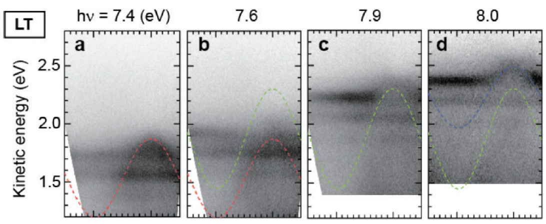

Figure 3. Photon energy dependence of the *E*–*k* map of pentacene on graphite (HOPG) recorded at LT (50 K for 8.0 eV and 14 K for 7.4, 7.6, and 7.9 eV) along with the fitting curves (POS: blue and red, NEG: green). The figure is after ref 2).

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

References

1) F. Nishino, K. Fukutani, J. Brandhoff, M. Gruenewald, E. Fuerch, M. Schaal, F. Otto, D. Stelter, R. Forker, Z. Zhang, T. Hirose, T. Fritz and S. Kera, *Appl. Phys. Express* **18**, 015502 (2025).

2) Y. Hasegawa, T. Yamaguchi, M. Meissner, T. Ueba, F. Bussoloti, S. Ideta, K. Tanaka, S. Yanagisawa and S. Kera, *Phys. Rev. B* **112**, 085301 (2025).

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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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 about 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 our 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. After two major upgrade projects, we now call our machine UVSOR-III. We have been developing novel light source technologies, such as free electron laser, coherent synchrotron radiation, structured light beams and laser Compton gamma-rays. We have been investigating beam physics which would be the basis of the future developments of the facility.

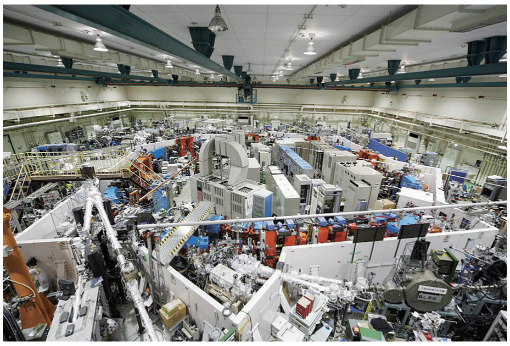


Figure 1. UVSOR-III Electron Storage Ring and Synchrotron Radiation Beamlines.

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

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 develop novel light sources and explore 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. We are now exploring their applications.

We have been exploring the possibility utilizing the temporal structures of undulator radiation, in collaboration with Saga Light Source, Toyama Univ. and Hiroshima 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 demonstrated a state-of-the-art technology to observe ultrafast properties of synchrotron radiation, in collaboration with Toyota Technological Institute.

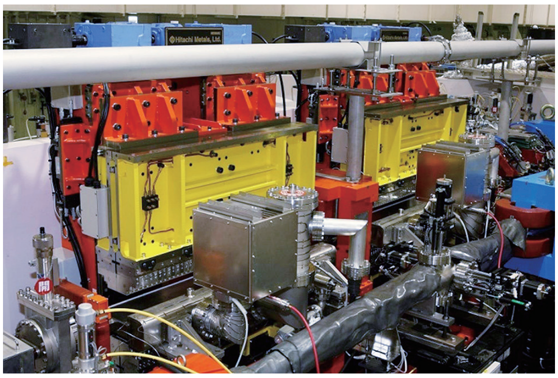


Figure 2. Twin Polarization-variable Undulators/Optical Klystron at UVSOR-III.

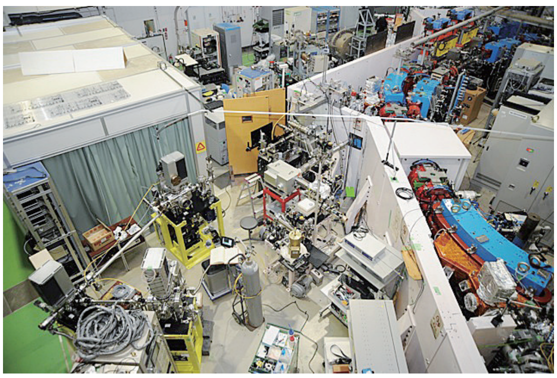


Figure 3. UVSOR BL1U experimental station for source development studies.

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. Some of the results have been published as bachelor thesis and master thesis in Hiroshima Univ.

We have developed a laser Compton scattering gamma-ray source at BL1U, which is capable of producing ultrashort, monochromatic and energy-tunable gamma-rays. They are now opened for public use.

We have been continuing experimental studies on the origin of the homochirality of biomolecules using intense circularly polarized undulator radiation at BL1U, in collaboration with Yokohama National Univ., Hiroshima Univ. and NIFS. This year, we have started a new project on exploring novel experimental techniques based on chiral undulator radiation, including optical vortex and other chiral structured light.

2. Accelerator Technology Developments

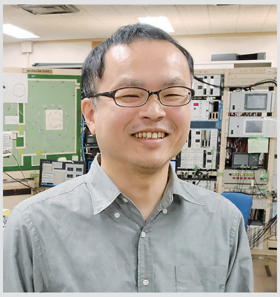
We carried out several upgrades on the UVSOR electron synchrotron since 2000. We designed a special beam optics intended to higher brightness. We developed necessary accelerator components, reconstructed the accelerator and commissioned it. We have commissioned six undulators successfully, three of which are variable polarization type and others in-vacuum type. 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 significantly lower emittance.

We are collaborating with Nagoya University, Hiroshima University and KEK Photon Factory to develop new accelerator technologies for the future plans of these facilities. 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.

Exploring a New Application of Synchrotron Radiation with Novel Light Source Technologies

UVSOR Synchrotron Facility
Division of Advanced Accelerator Research



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Keywords Synchrotron Radiation, Accelerator, Structured Light

UVSOR synchrotron at IMS is a compact low-energy synchrotron radiation facility which has been operating for more than 40 years. The light source performance is still in the word top level and continuous studies on the development of novel light source technologies have been conducted since the 1980s. From a viewpoint of exploring a new application of synchrotron radiation, UVSOR has advantages on the light source performance and the agility to immediately implement new concepts.

The waveform of electromagnetic radiation from an ultra-relativistic electron reflects the motion of the electron. This implies that, by controlling the electron motion in the magnetic field, one can control the properties of the radiation waveform in the nanometer or Angstrom scale. Our group has succeeded in generating the optical vortex beam which has helical phase plane and coherent double-pulse using insertion devices installed in the UVSOR synchrotron (Figure 1). The use of mutual coherence between the double-pulsed components enables time-domain interferometry experiments for controlling and monitoring the quantum state of matter using synchrotron radiation. Such an approach can be applied to the development of new spectroscopic and imaging methods using synchrotron radiation. We aim to develop novel measurement methods and their applications based on manipulating the motion of high-energy electrons in a synchrotron ring.

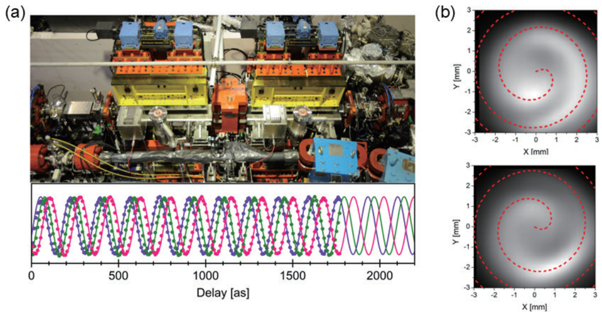

Figure 1. (a) Tandem-undulator system in the UVSOR-III synchrotron. Attosecond interference in photoexcitation of helium atoms is attached in the bottom panel. (b) Generation of optical vortex beam by synchrotron radiation.

Selected Publications

- T. Fuji, T. Kaneyasu, M. Fujimoto, Y. Okano, E. Salehi, M. Hosaka, Y. Takashima, A. Mano, Y. Hikosaka, S.-I. Wada and M. Katoh, "Spectral Phase Interferometry for Direct Electric-Field Reconstruction of Synchrotron Radiation," *Optica* **10**, 302 (2023).
- T. Kaneyasu, Y. Hikosaka, M. Fujimoto, H. Iwayama and M. Katoh, "Electron Wave Packet Interference in Atomic Inner-Shell Excitation," *Phys. Rev. Lett.* **126**, 113202 (2021).
- 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, T. Konomi, M. Katoh, H. Iwayama and E. Shigemasa, "Limitations in Photoionization of Helium by an Extreme Ultraviolet Optical Vortex," *Phys. Rev. A* **95**, 023413 (2017).
- T. Kaneyasu, Y. Takabayashi, Y. Iwasaki and S. Koda, "Beam Lifetime Study Based on Momentum Acceptance Restriction by Movable Beam Scraper," *Nucl. Instrum. Methods Phys. Res., Sect. A* **694**, 107 (2012).

1. Attosecond Interferometry Experiments Using Synchrotron Radiation

Attosecond interferometry experiments have been conducted using the light source development beamline BL1U in UVSOR-III synchrotron (Figure 2).[1)] In recent years, we have studied the interference phenomena between the photoelectron wave packets. The sequential interaction of an atom with a pair of coherent light pulses results in the production of a pair of photoelectron wave packets which interfere with each other during the propagation in a free space. The control and observation of photoelectron wave packet interference has been achieved so far by employing coherent pulse pairs generated by laser sources. In contrast, we have recently demonstrated that the phase coherent double pulses generated by a synchrotron light source can be utilized for the purpose of controlling the interference of photoelectron wave packets produced in the extreme ultraviolet wavelength range.[2)]

Figure 3 presents the time-domain photoelectron interferogram associated with the photoionization of 5p electron in xenon atom. This interferogram is composed of photoelectron spectra acquired at various phase shifter delays. During the measurement, the central wavelength of linearly polarized radiation was set to approximately 40 nm and the kinetic energy of the photoelectron was determined using a hemispherical electron energy analyzer. The interferogram exhibits periodic modulation with a period of approximately 140 as, which corresponds to the photon frequency. The clear modulation of this interferogram indicates that the photoelectron wave packet interference can be precisely controlled by varying the time delay, which was calibrated by the frequency-domain interferometry.[3)] Furthermore, the interferogram exhibits intensity modulation with a period of 3 fs. This effect can be attributed to the evolution of the spin–orbit wave packet produced in the Xe^+ ion, suggesting that the tandem undulator could be used to explore ultrafast quantum state dynamics with attosecond resolution.

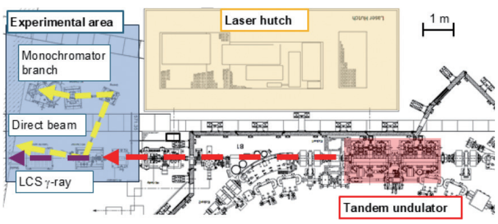

Figure 2. Layout of the light source development beamline BL1U.

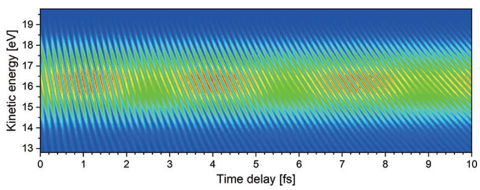

Figure 3. Time-domain photoelectron interferogram obtained for xenon 5p ionization.

2. Development of Atomic Fluorescence Polarimeter

Polarization represents one of the most significant characteristics of synchrotron radiation. The horizontally linear polarization of the radiation emitted from a bending magnet has been widely employed in various measurements for a long time. In addition, the rapid advancement of insertion devices in the 1990s has enabled arbitrary control of the polarization state of undulator radiation. However, the polarization state of the light changes according to the reflective properties of the beamline optics. Consequently, in order to ensure accurate measurements based on the polarization properties of synchrotron radiation, it is crucial to evaluate the polarization state of the light at the sample position.

To date, the synchrotron community has devoted considerable effort to the development of polarimeters for wavelengths shorter than those of vacuum ultraviolet (VUV) radiation. Simpler approach to measure the polarization of short wavelength light is to take advantage of the interaction of light with atoms and molecules in the gas phase. The use of a fluorescence polarimeter based on the conversion of VUV light into visible fluorescence light via atomic resonance allows the complete determination of the polarization state with a simple apparatus. In this study, the degree of linear polarization and the angle of inclination of the polarization axis of VUV light have been measured with a fluorescence polarimeter utilizing helium and neon atoms (Figure 4).[4)] This study demonstrates that a fluorescence polarimeter can be used in conjunction with a variety of atoms and molecules and thus extends the range of wavelengths to which this method can be applied.

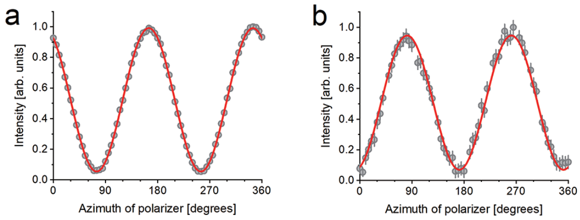

Figure 4. The fluorescence yield measured as a function of the polarizer angle for (a) helium and (b) neon atoms. The degree of linear polarization and angle of the polarization axis can be derived from the periodic modulation of the fluorescence yield.

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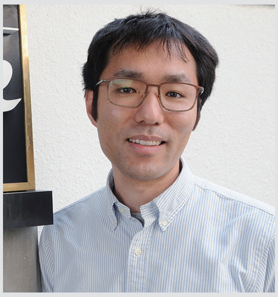
2) T. Kaneyasu, Y. Hikosaka, S. Wada, M. Fujimoto, H. Ota, H. Iwayama and M. Katoh, *Sci. Rep.* **13**, 6142 (2023).
3) Y. Hikosaka, T. Kaneyasu, S. Wada, H. Kohguchi, H. Ota, E. Nakamura, H. Iwayama, M. Fujimoto, M. Hosaka and M. Katoh, *Sci. Rep.* **13**, 10292 (2023).
4) T. Kaneyasu, H. Takeda, K. Hosaka and J. Adachi, *J. Electron Spectrosc. Relat. Phenom.* **279**, 147488 (2024).

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

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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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.

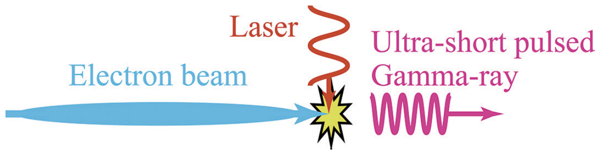


Figure 1. Schematic illustration of 90-degree inverse Thomson scattering.

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, 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).
- Y. Taira, Y. Yang, T. Shizuma and M. Omer, "Generation and Measurement of Gamma Rays with Axially Symmetric Polarization States via Compton Scattering," *Phys. Rev. Res.* **7**, 033130 (2025).

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. 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. A typical count rate is 20 cps.

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 using ^{22}Na, are being performed.

Meanwhile, we are also developing gamma-ray-induced spin-polarized positron annihilation spectroscopy using circularly polarized gamma rays. The spin-polarized positrons are generated from the circularly polarized gamma rays inside a sample. 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. 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 research and development.

2. Spatial Polarization Measurement of Gamma-Rays Generated Using Polarized Lasers

Inverse Thomson/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.

A novel Compton polarimeter was constructed to measure the linear polarization of MeV gamma rays. Gamma rays are irradiated onto an iron target, and the azimuth distribution of scattered gamma rays is measured by seven NaI detectors to determine the polarization axis of the gamma rays. The gamma rays expand over a diameter of 10 mm. By installing a collimator with a diameter of 1 mm on the gamma ray beam axis and irradiating only the gamma rays that pass through it onto the target, it is possible to measure the polarization axis at that position. Moreover, by scanning the collimator in two dimensions, it is possible to measure the spatial polarization distribution of gamma rays. Figure 2 shows the spatial polarization distribution of inverse Compton scattered gamma rays measured for the first time using the developed polarimeter. We were able to demonstrate that the polarization axis changes depending on the position of the beam cross section in both horizontal and vertical polarization. Gamma rays generated using a circularly polarized laser showed that the outer polarization changed to linear polarization and that the polarization axis was oriented in the azimuth direction. Gamma rays generated using radially and azimuthally polarized lasers were found to possess random polarization near the central axis and azimuth polarization states on the outer region.

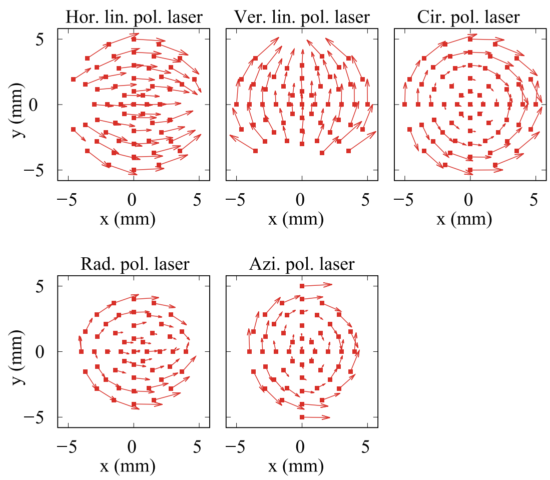
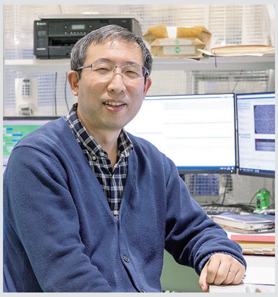


Figure 2. Measured spatial polarization distribution of gamma rays generated using five types of polarized lasers.

* CSC-IMS Scholarship Program

Symmetry Breaking in Crystal Microcosms Reflected in a Photoelectron Kaleidoscope

UVSOR Synchrotron Facility
Division of Advanced Solid State Physics



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Professor, The Graduate University for Advanced Studies

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Keywords Photoelectron Spectroscopy, Momentum Microscope, Electronic Spin Structure

The word "*kaleidoscope*" comes from the Greek "*kalos*," meaning beautiful, and "*eidos*," meaning shape or form. In a kaleidoscope, light repeatedly reflects off mirrors inside a tube, creating a beautiful pattern. The photoelectron angular distribution we study is similar to this kaleidoscope.

Photoelectron spectroscopy is a technique that uses photon to eject electrons from a solid. By analyzing the ejected electrons' energy and motion, we can determine the properties of the material. As the photoelectrons travel within a solid crystal, they form a complex angular distribution, much like the patterns in a kaleidoscope. This complex pattern holds astonishing information. The diffraction pattern of core-level photoelectrons reveals the local atomic arrangment of the specific element sites that make up the crystal, and the angular distribution of valence photoelectrons tells us about the fundamental electronic properties of the material.

We have developed a new analytical instrument, the photoelectron momentum microscope (PMM). 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. Specifically, the "broken symmetry" in the kaleidoscopic patterns created by photoelectrons contains crucial information that determines the characteristics of a material.

Our research focuses on a range of intriguing phenomena, including twinning crystal growth, phase transitions, magnetism, and superconductivity. We are particularly fascinated by the interplay between domain boundaries and electronic properties, where the delicate balance between order formation and fluctuations creates a rich and complex environment. We are pioneering a new technique to unravel the mysteries of electronic properties within individual microcrystalline regions (Figure 1).

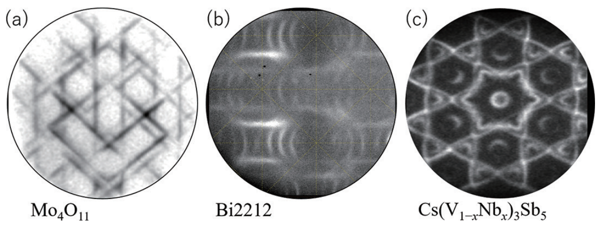

Figure 1. Photoelectron momentum distributions of Fermi surfaces from (a) a quasi-2D crystal, (b) a high-temperature superconducting copper oxide and (c) a Kagome-lattice superconductor.

Selected Publications

- T. Kobayashi, F. Matsui *et al.*, "Temperature-Dependent Electronic Structure of a Quasi-Two-Dimensional Conductor η-Mo_4O_{11}," *Sci. Rep.* **15**, 9034 (2025).
- K. Hagiwara, F. Matsui *et al.*, "Development of Dual-Beamline PMM for Valence Orbital Analysis," *J. Synchrotron Radiat.* **31**, 540 (2024).
- F. Matsui *et al.*, "Soft X-Ray PMM for Multimodal Valence Band Stereography," *Rev. Sci. Instrum.* **94**, 083701 (2023).
- T. Kato, F. Matsui *et al.*, "Fermiology and Origin of T_c Enhancement in a Kagome Superconductor $Cs(V_{1-x}Nb_x)_3Sb_5$," *Phys. Rev. Lett.* **129**, 206402 (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).

1. Photoelectron Feels the Broken Symmetry at the Surface

PMM can also map conduction band dispersion. The conduction band structure is imprinted on the energy-loss background of secondary electrons (SEs) that accompany the primary photoemission process. The interaction of these secondary electrons with the conduction band states reduces their emission intensity into the vacuum, creating a negative contrast pattern of the conduction band.[1,2] This is analogous to the negative photoelectron diffraction patterns seen in the angular distribution of energy-loss electrons accompanying core-level photoemission.[3] Consequently, a standard photoelectron spectroscopy setup, typically used for occupied states, can also visualize the unoccupied conduction band.

Figure 2(a) shows the overall valence band dispersion of graphite, where darker regions indicate a stronger photoelectron signal. We tuned the photon energy to 68 eV to match the bulk L symmetry point. By selectively detecting photoelectrons from a terrace with a single termination type, we confirmed a clear surface symmetry breaking (Figure 2(b)).[4]

Figure 2(c) displays the momentum-resolved kinetic energy distribution of SEs. The unoccupied band dispersion appears as a negative contrast due to the absorption of photoelectrons by the conduction band states. Figure 2(d) presents a series of angular patterns at various kinetic energies. A key finding is that while most patterns exhibit six-fold symmetry, those at kinetic energies around 20 eV show a three-fold symmetry. This suggests that as photoelectrons escape from the solid into the vacuum, they are influenced by the three-fold symmetric structure of the top surface of graphite.

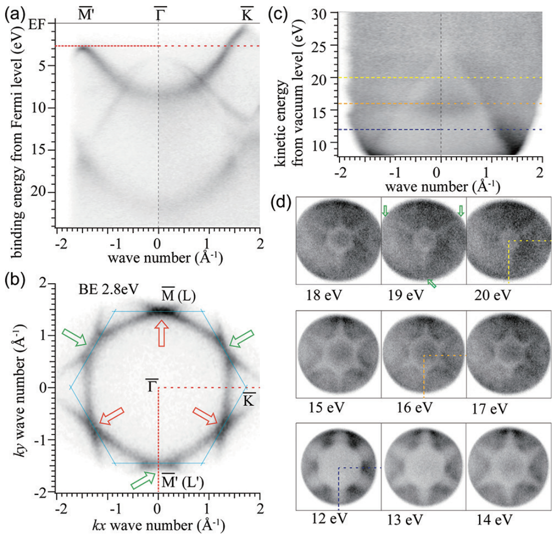

Figure 2. (a) The overall dispersion of the valence band of graphite. Dark areas correspond to larger photoelectron signals. (b) Momentum distribution indicated as a red line in (a). (c) The momentum resolved kinetic energy distribution of SE electrons. The conduction band signature appears as a lighter grey contrast. (d) A series of SE angle patterns at several kinetic energies. Note that at kinetic energies of 19 and 20 eV, the patterns appear as three-fold symmetric.

2. Spins, in Which Direction Are You Oriented?

That's the fundamental question at the heart of spin physics. Spin, a crucial quantum number, enriches the properties of materials, magnetism and superconductivity for instance. To truly understand those phenomena, we need to clarify not only their electronic structures but also the behavior of electron spins.

Spin-polarized photoelectron momentum microscopy (SP-PMM) is a powerful technique for detecting spin polarization in 2D real and reciprocal spaces. Its main advantage is efficiency, providing instant, spin-resolved snapshots of valence band structures. Our SP-PMM at the UVSOR synchrotron facility uses both grazing-incidence soft X-rays and normal-incidence vacuum ultraviolet light. A unique feature of our instrument is a spin rotator placed just before the spin detector, which allows for precise spin manipulation.

The Au(111) surface state is a classic example of spin–orbit coupling. The loss of bulk symmetry at the surface splits the band dispersion into two branches due to the Rashba effect (Figures 3(a), (b)). The in-plane spin components of these split bands are known to point in opposite directions, and observing this state is a common milestone in spin-polarized ARPES.

However, a review of the literature on the Au(111) Rashba state revealed a significant contradiction: The spin orientation of the outer bands is ambiguously assigned as either counterclockwise (ccw) or clockwise (cw). This confusion has been made worse by numerous secondary publications.

To resolve this issue, we performed a case study on the Au(111) surface state using our SP-PMM. By carefully investigating our multichannel detection system, we were able to unambiguously determine the precise spin orientation of the Au(111) surface state (Figure 3(c)).[5]

Our PMM at the UVSOR synchrotron facility uses both grazing-incidence soft X-rays and normal-incidence vacuum ultraviolet (VUV) light with variable polarization. This normal-incidence setup is particularly useful for directly investigating the relationship between orbital angular momentum and transition matrix elements (Figure 3(d)).[6]

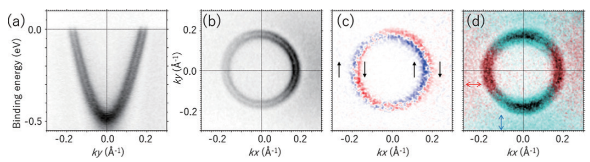

Figure 3. (a) The dispersion of the Au(111) surface state. (b) A cross-section of the surface state at the Fermi level. (c) The determined spin orientation of the surface state. (d) Intensity distributions excited by horizontal (magenta) and vertical (cyan) VUV polarization, respectively.

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3) F. Matsui *et al.*, *J. Phys. Soc. Jpn.* **81**, 013601 (2012).
4) F. Matsui *et al.*, *Phys. Rev. B* **105**, 235126 (2022).
5) F. Matsui *et al.*, *J. Phys. Soc. Jpn.* **94**, 114707 (2025).
6) K. Hagiwara *et al.*, *J. Synchrotron Radiat.* **31**, 540 (2024).

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

Angle-Resolved Photoemission Study on Strongly Correlated Electron Materials

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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).

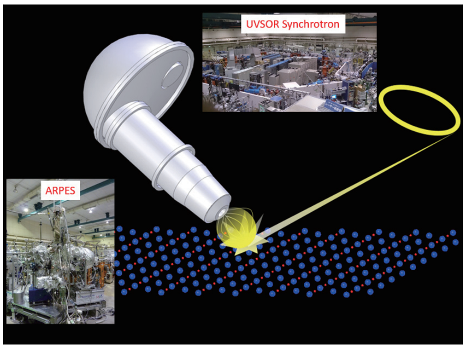

Figure 1.

Selected Publications

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

Spintronics is an emerging field that aims to utilize the spin as well as the charge of electrons, and its progress is expected to strongly shape the future of electronics. To realize spin-based devices, it is crucial to understand the magnetism of surfaces, interfaces, and nanostructures at a fundamental level. Spin- and angle-resolved photoemission spectroscopy (spin-resolved ARPES) is one of the most powerful experimental methods for this purpose, because it can provide the full information of the electronic states—energy, momentum, and spin orientation. However, conventional Mott-type spin detectors suffer from an extremely low efficiency of about 10^{-4}, which has been a serious obstacle for decades. The development of very-low-energy-electron-diffraction (VLEED) detectors, with roughly 100 times higher efficiency, has made spin-resolved ARPES feasible in practice, yet most existing systems still use single-channel detection, where efficiency and angular resolution remain limited.

To overcome these limitations, our group reconstructed the BL5U beamline at UVSOR in 2017, creating a high-photon-flux and high-energy-resolution ARPES station. Building on this foundation, we initiated a long-term project to establish a next-generation spin-resolved ARPES system with multi-channel detection, which we call "image-spin" detection. The aim of this project is not only to improve detection efficiency and momentum resolution by factors of 100 and 10, respectively, but also to develop a platform that enables user-friendly and systematic spin-resolved measurements at synchrotron light sources. By pushing the limits of both efficiency and resolution, such a system can provide unprecedented opportunities for studying spin-dependent band structures, many-body interactions, and exotic electronic phases that cannot be accessed with conventional methods.

In 2024, we made substantial progress with the introduction of a spin manipulator combined with an ultra-bright electron gun. In our previous report, we demonstrated that this system could already achieve 100 times higher efficiency and 10 times better momentum resolution than conventional single-channel systems, but these improvements were restricted to one axis (the in-plane x-axis of the sample) and to a photon energy range of only 40–80 eV. Over the past year, careful optimization of the electron lens parameters allowed us to extend spin detection to two in-plane axes (x and y directions), as shown in Figures 2 and 3. At the same time, the usable photon energy range was expanded to 21–120 eV. This not only enables the study of a wider variety of materials, but also allows systematic comparisons across different excitation energies, which is particularly important for disentangling bulk and surface contributions in complex systems.

An additional improvement in 2024 was the refinement of the spin target handling. The magnetization procedure, which had previously required frequent manual intervention, was motorized to allow precise and reproducible control during experiments. This change enhanced the stability of the spin-resolved signals and improved the reliability of the data.

Our plan for FY2025 is to complete the lens calibration for the out-of-plane (z-axis) direction, so that spin polarization can be measured along all three spatial axes. Achieving this goal will make it possible to obtain the full spin information of electronic states in a truly three-dimensional manner, establishing a comprehensive framework for future spin-resolved studies at UVSOR.

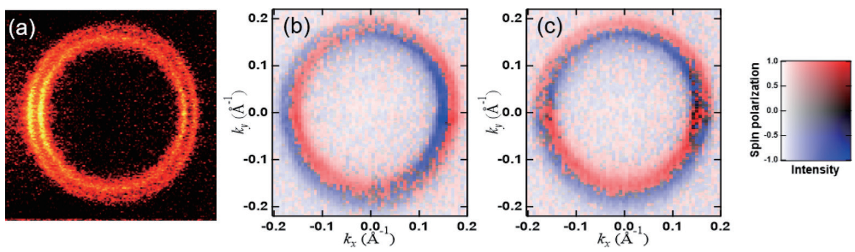

Figure 2. (a) Fermi surface image of Au(111) obtained using ordinary ARPES. Spin-resolved Fermi surface image of Au(111) with the spin detection axis aligned along the k_y (b) and k_x (c) directions.

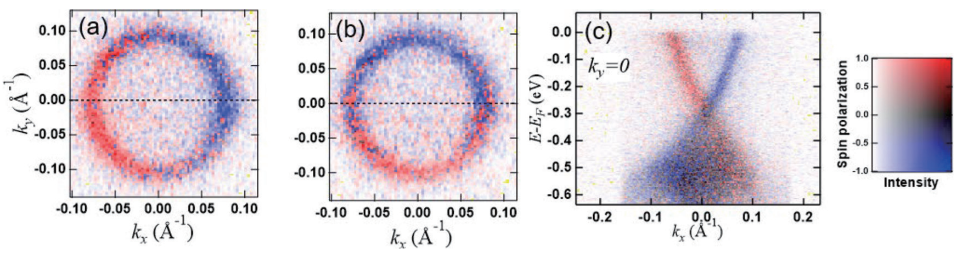

Figure 3. Spin-resolved Fermi surface images of the topological insulator Bi_2Se_3 with spin the detection axis aligned along the k_y (a) and k_x (b) directions. (c) Spin ARPES image along k_y = 0 in (a).

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

UVSOR Synchrotron Facility
Division of Advanced Photochemistry

ARAKI, Tohru
Senior Researcher

As a beamline scientist, I oversee the user science programs, in both academia and indsutry that utilize the BL4U-STXM (Scanning Transmission X-ray Microscopy) beamline at the UVSOR research facility. STXM is a form of X-ray absorption-based spectromicroscopy that provides label-free chemical mapping.

STXM is a specialized imaging technique that falls under the category of X-ray absorption-based spectromicroscopy. By measuring how different materials absorb X-rays at specific energies, STXM enables label-free chemical mapping, allowing researchers to visualize the distribution of various chemical components within biological and material samples without the need for dyes or stains. It has a wide range of applications in various fields, including energy materials, environmental and earth sciences, and industrial polymer studies. My current focus is on "beyond organelle mapping," which requires the advanced spectroscopy to identify the biomacromolecules. Shinohara *et al.* conducted a related study at the BL4U (*cells* 2019). The team presented the quantitative mapping of DNA, RNA, histones, and general proteins in mammalian cells, nuclei, and a chromosomes. This was achieved through the spectral fitting of the reference spectra. To accommodate a broader range of biological samples, including cells and tissues, it is essential to achieve higher chemical sensitivity and enhanced accuracy. "To establish this methodology, two key steps must be taken. First, a basic spectral interpretation of organelles must be conducted. Second, the sample preparation and specimen environment must be optimized. It is imperative to refrain from altering the samples and to preserve 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." This is my statement from last year. However, due to technical difficulties and the need for a direct comparison between STXM and SEM, I have been working on the biological sample embedded in resin. The sample was ultra-thin sliced using a diamond knife to create a sample 100 nm thick for the STXM measurement. The bottom figures show the STXM results of *Ramazzottius varieornatus*, a tardigrade that is renowned for its anhydrobiotic capabilities, enabling it to survive in harsh, arid environments. The data was collected at the Carbon K-edge absorption for both active (hydrated) and inactive tardigrade samples. The experiment was conducted with the support of ExCELLS. My collaborators prepared the samples and conducted the SEM experiment. The spectra displayed in the Figure 1 correspond to the five significant components that were analyzed using principal component analysis and cluster analysis techniques of the active tardigrade. The following presentation offers a visual representation of the two component maps of C4 and C1 spectra for both active and inactive tardigrades. The distribution of the two components differs between two samples. To further elucidate these results, additional STXM data analysis and the scheduled immunoelectron microscopy experiment will be conducted.

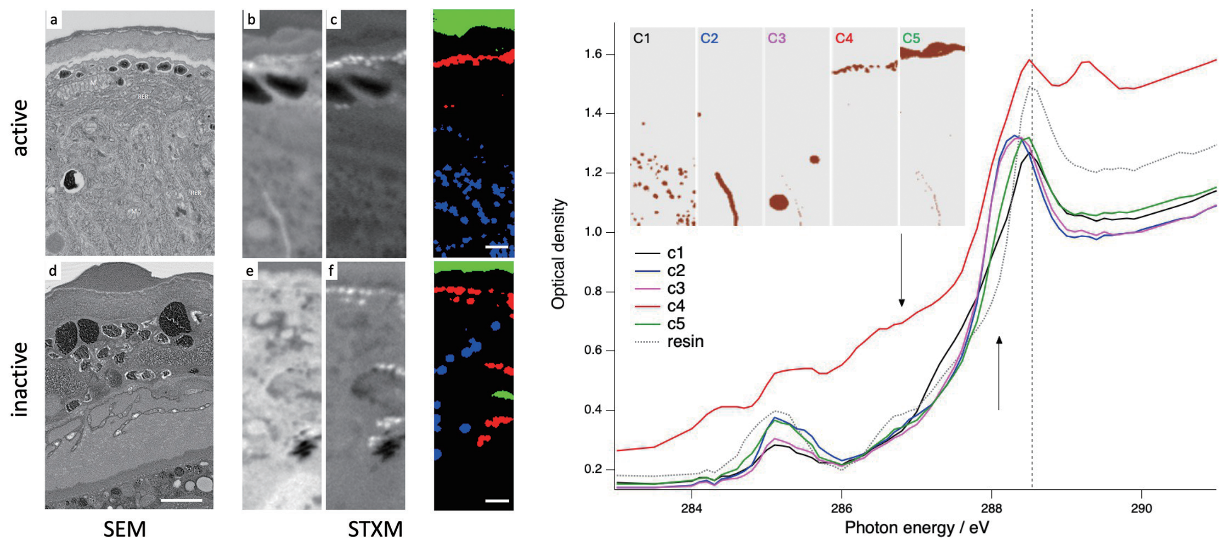
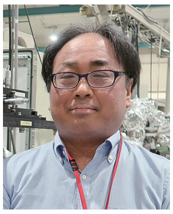


Figure 1. [left] SEM and STXM images of active (top) and inactive (bottom) tardigrade samples. (Scale bar 1 μm) RGB composite component map (red and blue: Two significant components, green: Resin). [right] 5 components map and the corresponding spectra of active tardigrade sample. (Two arrows indicate the photon energies used for the STXM images (b, e: 288.1 eV, c, f: 286.8 eV). The 5 significant components map is shown in C1–C5 images.)

Development of Resonant Soft X-Ray Scattering Spectroscopy for Photoresists

UVSOR Synchrotron Facility
Division of Advanced Photochemistry



IWAYAMA, Hiroshi
Senior Researcher

For further miniaturization of semiconductors, the era of EUV (13.5 nm) exposure has arrived, down from the 197 nm of the conventional ArF laser. This reduction in wavelength to less than one-tenth requires further thinning of the photosensitive materials (photoresists), leading to active development efforts of photoresist industry. Since the depth of focus decreases in proportion to the wavelength, EUV exposure requires a thickness of only a few tens of nanometers, compared to conventional photosensitive materials of around a few hundred nanometers. To preserve the 10-nm pattern, a thin base layer between the silicon substrate and photoresist is essential. The EUV exposure technology utilises a two-layer polymer film that is several tens of nanometers thick.

Resonant soft X-ray reflectivity is a technique that can determine the film thickness of different chemical species by utilizing the difference in X-ray resonance energy. We conducted experiments on a bilayer polymer film of PMMA (40 nm thick) and PVPh (70 nm thick). Figure 1 shows a schematic diagram of the sample.

The complex permittivity in the X-ray region is written as (1-δ(hν))-i(β(hν)), where (1-δ) and β correspond to the refractive index and absorptivity, respectively. In the X-ray region, δ and β are much smaller than 1. Furthermore, δ and β are related by the Kramers-Kronig (KK) relation. Thus, β(hν) is measured *via* the XAFS absorption spectrum. Figure 2 shows the refractive index δ calculated from each absorption spectrum using the KK transformation. Just as the pre-edge structure of the absorption spectrum differs depending on the molecular species, the refractive index δ also has a complex structure at the absorption edge that depends on the chemical species. Near the core absorption edge, selecting the resonance energy allows scattering contrast between chemically distinct species, even if their electron densities are nearly identical.

Figure 3 shows reflectivity measurements at both non-resonant and resonant energies. At non-resonant energy, a single period is observed, indicating a film thickness of 110 nm. The bilayer film appears as a single film. At resonant energy, a complex vibrational structure appears, reflecting the properties of a bilayer film. Details are still under analysis; according to these results, a method for analyzing multilayer polymer films using resonant soft X-ray reflection is being developed.

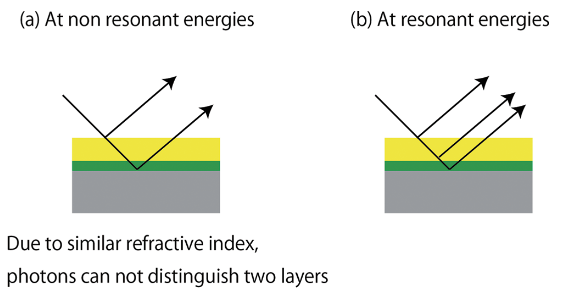


Figure 1. Schematic of (a) nonresonant and (b) resonant scattering of two polymer layers.

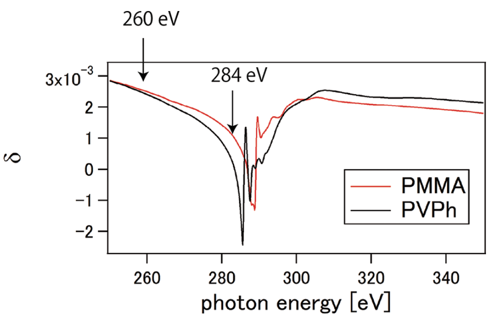


Figure 2. X-ray energy dependence of the refractive index δ of PMMA and PVPh polymers.

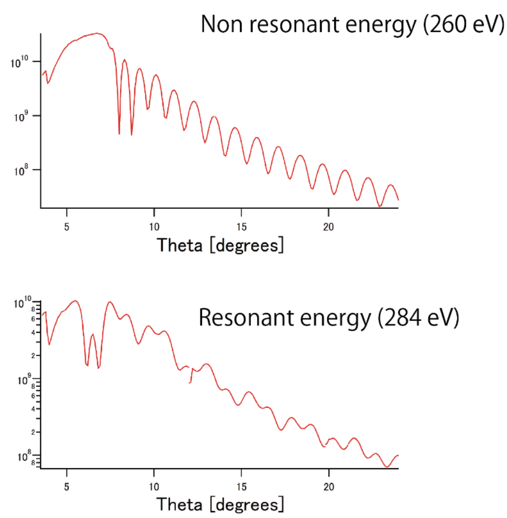
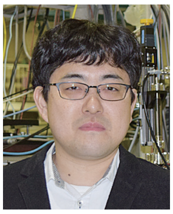


Figure 3. Reflectance spectra of (PMMA, PVPh) two-layer polymer film at off-resonance and resonant energies.

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

Chemical processes in solutions were investigated using *operando* soft X-ray absorption spectroscopy (XAS) with different light elements.[1,2] In this year, the metal-ligand delocalization of metal porphyrin complexes was investigated from the ligand side,[3] and inner-shell calculations of large molecular systems were developed for polymers in solutions.[4]

1. Metal-Ligand Delocalization of Metal Porphyrin Complexes in Solutions

Metal-ligand delocalization of metal porphyrin complexes such as iron protoporphyrin IX was proved from the ligand side using N K-edge XAS.[3] The C=N π* peaks of porphyrins are useful for discussing the central metal dependence of metal-ligand delocalization and the hydration structures of metal porphyrins in solutions.

2. Inner-Shell Calculations of Polymers in Solutions

Inner-shell spectra of poly(*N*-isopropylacrylamide) (PNIPAM) in solutions were calculated by extracting the 5-mer PNIPAM chains with terminated H atoms, including the second coordination shells of solvent molecules, from the snapshots of the molecular dynamics simulations.[4] The C=O π* peaks of PNIPAM at the O K-edge reflected the structural changes of the polymer chains and the coordination of the C=O groups with solvent methanol and water molecules.

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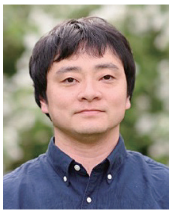
Visiting Professors

Visiting Professor

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

Development of Analysis Methods for Photoelectron Momentum Microscope

Following last year, we have continued the development of data analysis tools for the photoelectron momentum microscope introduced at UVSOR. This instrument is a highly powerful tool for observing the electronic structure of samples, including spin, by means of photoelectron spectroscopy. Using the first-principles calculation code OpenMX, we have enabled the calculation of photoelectron transition probabilities, so that the observed photoelectron intensities from the valence band can be analyzed. We have begun comparing the experimental results of twisted graphene obtained using the momentum microscope with theoretical calculations. In addition, we applied principal component analysis to the 1T-TaS_2 data obtained with this instrument, attempting to visualize the behavior of the phase transition. In this way, we are developing a framework that integrates first-principles calculations with information-theoretical approaches.



Visiting Associate Professor

SHIBUTA, Masahiro *(from Osaka Metropolitan University)*

Vibration-Resolved Unoccupied Molecular Orbitals by Two-Photon Photoemission Spectroscopy

Carrier-vibration couplings in organic thin films are important to understand the carrier mobilities in organic devices. So far, hole-vibration couplings have been studied, resolving fine structures of occupied orbitals using photoelectron spectroscopy. However, it was difficult to analyze electron–vibration couplings because we must analyze the unoccupied orbitals. Two-photon photoemission (2PPE) spectroscopy is a powerful method to observe unoccupied states with high resolution (20 meV), where a first photon injects an electron from a substrate into an adsorbed molecule, and the excited electron is extracted by a second photon. In fact, we have successfully resolved fine structures in the lowest unoccupied molecular orbital (LUMO)-derived peaks for small polyaromatic molecular films (*e.g.*, naphthalene, anthracene, phenanthrene) due to electron–vibration couplings. These results are opening the door to understand the electron mobility in the organic films. Furthermore, 2PPE can track the time-dependent behavior of the fine structures, which will unveil the energy modification and/or molecular motion of the excited state in the ultrafast regime.