

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, high-resolution optical microscopy applied to nanomaterials, synchrotron-based spectroscopy of core-excited molecules and solid-state materials, vacuum-UV photochemistry, and the development of novel laser- and synchrotron-radiation sources.

Ultrafast Quantum Simulator and Computer

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Education

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Professional Employment

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2007 Visiting Professor, Tokyo Institute of Technology (–2008)
2009 Visiting Professor, The University of Tokyo (–2011)
2012 Visiting Professor (Humboldt Awardee), University of Heidelberg
2014 Visiting Professor, University of Strasbourg (–2016)

Awards

1998 Award by Research Foundation for Opto-Science and Technology
2007 JSPS Prize
2007 Japan Academy Medal
2008 Norman Hascoe Distinguished Lecturer, University of Connecticut, USA
2009 Fellow of the American Physical Society
2012 Humboldt Research Award (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.

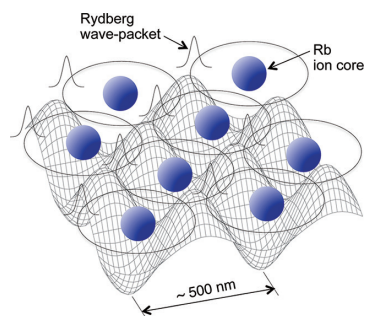


Figure 1. Metal-like quantum gas. A schematic of the many-body quantum simulator with ultracold Rydberg atoms, named “ultrafast quantum simulator,” where electronic wave functions spatially overlap between neighboring atoms.²⁾

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, S. Sugawa *et al.*, “Picosecond-Scale Ultrafast Many-Body Dynamics in an Ultracold Rydberg-Excited Atomic Mott Insulator,” *Phys. Rev. Lett.* **131**, 123201 (2023).

1. Development of an “Ultrafast Quantum Simulator” by Optical Control with Precisions on the Attosecond Temporal and Submicron Spatial Scales⁶⁾

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.

Very recently in 2023, quantum magnetism has successfully been simulated with this standard hardware assembled with ~30,000 Rb atoms.⁶⁾ Our novel scheme above completes the simulation in just several hundreds of picoseconds, accelerating the simulation speed by three orders of magnitude compared to any other quantum simulators of magnetism so far. This innovative acceleration solves the issue with external noise on the timescale of ~1 microsecond or slower in general, which has been one of the biggest concerns for quantum simulation. Moreover, we have succeeded in simulating the formation dynamics of “quantum entanglement,” which is difficult to measure in actual magnetic materials, on the fastest timescale of several hundred picoseconds, as schematically depicted in Figure 2.

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 Atoms⁷⁾

We have developed arbitrary two dimensional optical trap arrays for cold atoms with optical tweezers, which are necessary for quantum computing.

We have succeeded in loading a single atom into each trap of those arbitrary arrays, and reassembling those atoms with a movable optical tweezers. Such an array of cold atoms has been 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 have recently succeeded in executing a controlled Z gate,⁷⁾ 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, whose timescale is in general 1 microsecond or slower, and thus can be safely isolated from the noise. Moreover, the speed of this two-qubit gate is faster than that of the fast two-qubit gate demonstrated recently by “Google AI Quantum” with superconducting qubits.⁸⁾ 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, S. Sugawa *et al.*, *Phys. Rev. Lett.* **131**, 123201 (2023).
- 7) Y. Chew *et al.*, *Nat. Photonics* **16**, 724 (2022). (Cover-Page Highlight)
- 8) B. Foxen *et al.*, *Phys. Rev. Lett.* **125**, 120504 (2020).

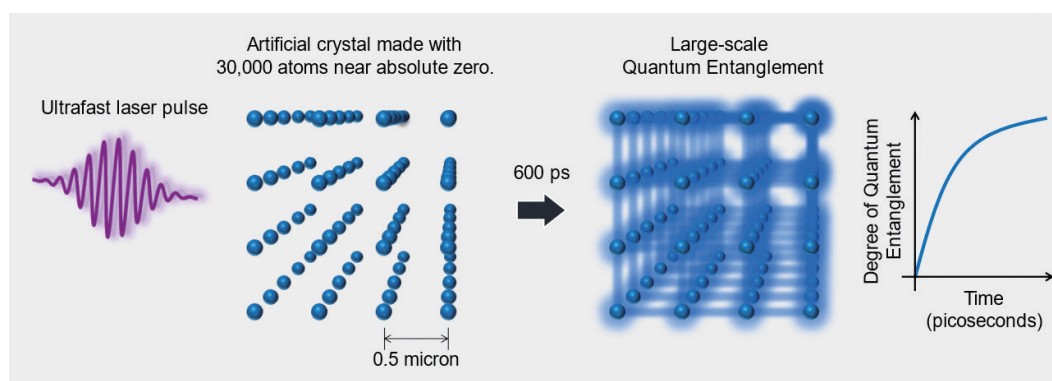


Figure 2. Conceptual diagram of ultrafast quantum simulation of magnetic material.⁶⁾ A large-scale array of 30,000 atoms, with a spacing of 0.5 micron, is controlled with a ~10 picosecond ultrafast laser pulse. After irradiating an ultrafast laser pulse, large-scale “quantum entanglement” is formed in only ~600 picoseconds. Image source: Prof. Seiji Sugawa (U. Tokyo).

Award

DE LÉSÉLEUC, Sylvain; The 12th Young Scientist Award of National Institutes of Natural Sciences (2023).

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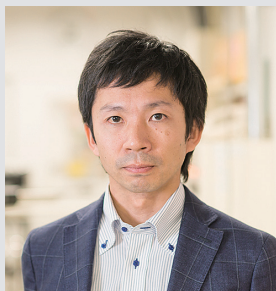
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Electronic Property of Functional Organic Materials

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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.

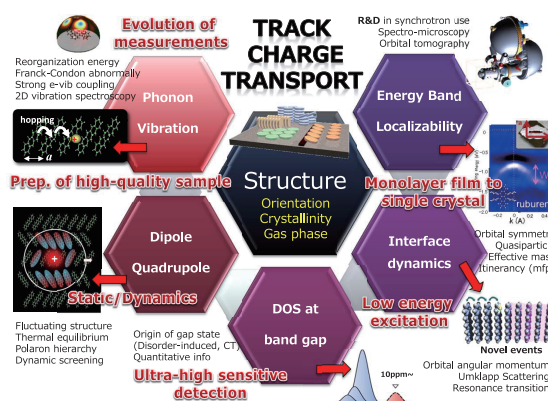


Figure 1. Overview of our agenda. A rich assortment of surface and interface structures of FOM to provide complicated spectral features of ultraviolet photoelectron spectroscopy.

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 Covalent Organic Frameworks¹⁾

Covalent organic frameworks (COFs), as a large class of porous organic materials, have attracted intense research in the past few decades due to the great potential for applications. Substituting carbon with silicon in organic molecules and materials has been an attractive way to modify their electronic structure and properties. Silicon-doped graphene-based materials are known to exhibit exotic properties, yet conjugated organic materials with atomically precise Si substitution have remained difficult to prepare. We present the on-surface synthesis of one- and two-dimensional covalent organic frameworks whose backbones contain 1,4-disilabenzene (C_4Si_2) linkers. Silicon atoms were first deposited on a Au(111) surface, forming a $AuSi_x$ film on annealing. The subsequent deposition and annealing of a bromo-substituted polyaromatic hydrocarbon precursor on this surface led to the formation of the C_4Si_2 -bridged networks, which were characterized by a combination of high-resolution scanning tunnelling microscopy and X-ray photoelectron spectroscopy taken at BL2B of UVSOR supported by density functional theory (DFT) calculations. Each Si in a hexagonal C_4Si_2 ring was found to be covalently linked to one terminal Br atom. For the linear structure obtained with the pyrene-based precursor, the C_4Si_2 rings were converted into C_4Si pentagonal siloles by further annealing.

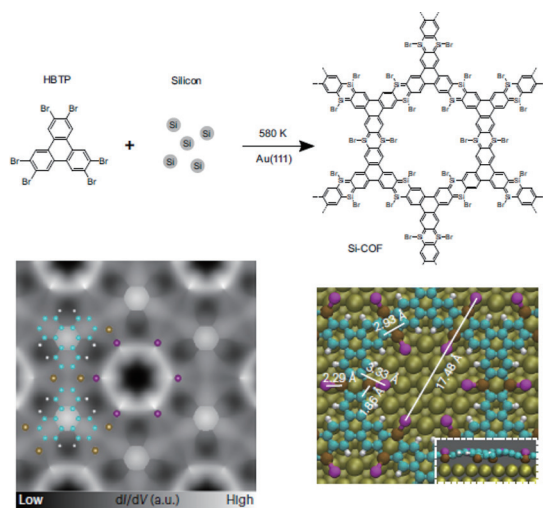


Figure 2. Top: Scheme of on-surface reaction for the aryl-Si coupling reaction on Au(111). Bottom: High-resolution constant height dI/dV map of the COF taken with a CO tip and the top and side views of DFT simulation. The figure is after ref 1).

2. Photoemission Tomography of Br-Doped Perylene Monolayer²⁾

Energy level alignment at the organic-metal interface is an

important issue in the field of organic electronics to understand the efficiency of charge injection and fundamental conduction mechanisms. The spectroscopic data of organic cations seem slightly insufficient, comparing to that of organic anions using strong molecular acceptor and alkali metal doping. Halogens are fundamental elements and have been widely used as hole dopants for organic electronics since the very early stages of research. Perylene doped with bromine exhibit a dramatic increase in conductivity. In this study, we observed the geometric and electronic structures of a monolayer of perylene molecules on the surfaces of Au(110) and Au(111). The effect of bromine doping was studied using C-K NEXAFS, angle-resolved photoemission spectroscopy (ARPES), and X-ray photoelectron spectroscopy (XPS). C-K NEXAFS can be used to observe vacant states, particularly a hole state, which is essential for p-type organic semiconductors. Moreover, the inclined molecular orientation is analyzed by measuring the polarization dependence. ARPES and 2D momentum map using a photoelectron momentum microscope at BL6U of UVSOR is a powerful technique for discussing the changes in the molecular orbital character upon the doping.

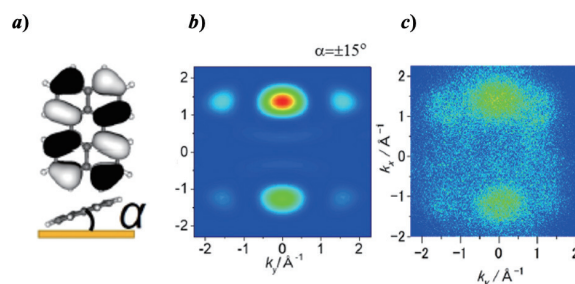


Figure 3. (a) HOMO distribution of a perylene molecule. (b) Simulated photoemission momentum maps of HOMO. (c) Experimental momentum map at binding energy of 1.5 eV. The figure is after ref 2).

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.^{2,3)} The perspectives required for future light-source facility have been discussed with communities.⁴⁾

References

- 1) K. Sun, O. J. Silveira, Y. Ma, Y. Hasegawa, M. Matsumoto, S. Kera, O. Krejčí, A. S. Foster and S. Kawai, *Nat. Chem.* **15**, 136–143 (2023).
- 2) O. Endo, F. Matsui, S. Kera, W.-J. Chun, M. Nakamura, K. Amemiya and H. Ozaki, *J. Phys. Chem. C* **126**, 15971–15979 (2022).
- 3) Y. Hasegawa, F. Matsui and S. Kera, *e-J. Surf. Sci. Nanotechnol.* **20**, 174–179 (2022).
- 4) H. Ota *et al.*, *J. Phys: Conf. Ser.* **2380**, 012003 (5 pages) (2022).

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Light Source Developments by Using Relativistic Electron Beams

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Professional Employment

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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.

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–Electron 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).
- S. Matsuba, K. Kawase, A. Miyamoto, S. Sasaki, M. Fujimoto, T. Konomi, N. Yamamoto, M. Hosaka and M. Katoh, "Generation of Vector Beam with Tandem Helical Undulators," *Appl. Phys. Lett.* **113**, 021106 (2018).
- Y. Hikosaka, T. Kaneyasu, M. Fujimoto, H. Iwayama and M. Katoh, "Coherent Control in the Extreme Ultraviolet and Attosecond Regime by Synchrotron Radiation," *Nat. Commun.* **10**, 4988 (2019).
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- 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).

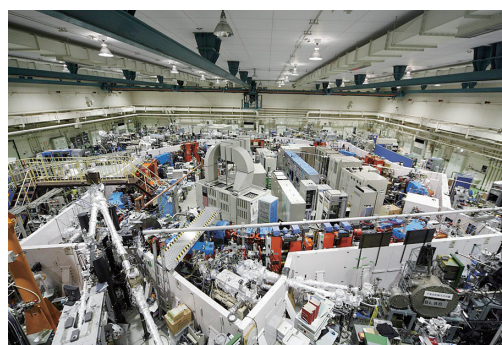


Figure 1. UVSOR-III Electron Storage Ring and Synchrotron Radiation Beamlines.

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 develop novel light sources and exploring their applications. The BL1U is equipped with two undulators which constitute an optical klystron, a laser system which is synchronized with the accelerator beam and a dedicated beamline consisting of mirrors and a monochromator whose arrangement can be flexibly changed according to the types of the experiments.

In 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. In these years, we are focusing on 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.

We have been developing a laser Compton scattering gamma-ray source at BL1U, which is capable of producing monochromatic and energy-tunable gamma-rays. Currently we are interested in controlling the wave properties of gamma-ray photons. Theoretically we have shown that vortex photons carrying orbital angular momentum can be produced by nonlinear Compton scattering of circularly polarized photons. We are challenging its experimental demonstration.

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.

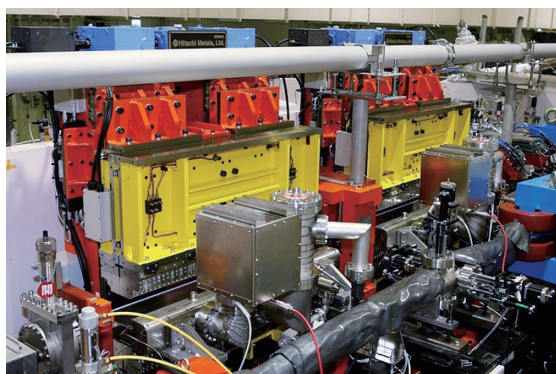


Figure 2. Twin Polarization-variable Undulators/Optical Klystron at UVSOR-III.

2. Accelerator Technology Developments for Electron Synchrotrons

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

Currently, the storage ring is stably operated for many of the users, however, the requirements from the users for the stability is getting higher and higher. As a near-term upgrade plan, we are considering replacing some of the undulators to fit the changes of the users' requirements on the wavelength. Also, we are seeking a possibility to reduce the emittance with the present magnet configuration. So far, we have found a few beam optics which would give lower emittance around 10 nm. Although they are not compatible with the operation of the narrow gap undulators, they may be used for special experiments which requires lower emittance. For a long-term plan, we continue the design study on a new light source facility. We have been investigating various accelerator systems such as a diffraction-limited synchrotron, an energy recovery linear accelerator and so on. Currently we are focusing on designing a synchrotron with the electron energy of 1 GeV and the circumference of around 70 m. We have designed a synchrotron which would give low emittance of around 5 nm under the achromatic condition.

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

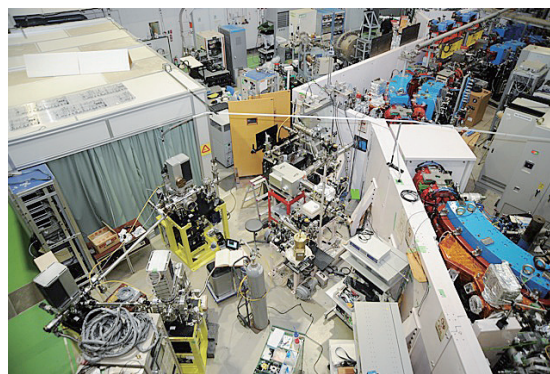
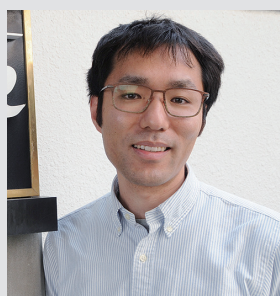


Figure 3. UVSOR BL1U experimental station for source development studies.

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

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Awards

2010 Young Researcher Best Presentation Award, The 53rd Annual Meeting of the Japanese Society of Radiation Chemistry
2011 Nagoya University Outstanding Graduate Student Award
2012 Oral Presentation Award, The 9th Annual Meeting of Particle Accelerator Society of Japan
2012 Young Researcher Best Poster Award, 12th International Symposium on Radiation Physics
2013 Young Scientist Award of the Physical Society of Japan
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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 fields 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 spectroscopy (GiPAS). A posi-

tron 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. Our group is conducting research on improving the properties of the material by using GiPAS.

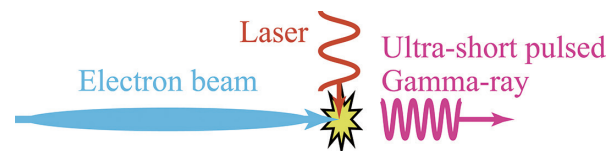


Figure 1. Schematic illustration of 90-degree inverse Thomson scattering.

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 gamma-ray-induced positron annihilation lifetime spectroscopy (GiPALS), positron lifetime spectrum is calculated by measuring the time difference between a reference signal and a detector output for the annihilation gamma rays, which is emitted when a positron annihilates with an electron inside material. A reference signal is the output of a photodiode located near the injection position of a laser. A BaF₂ 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 BaF₂ detector, and calculate the time difference distribution. One digital oscilloscope for four BaF₂ 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 BaF₂ scintillator. The count rate is 20 cps.

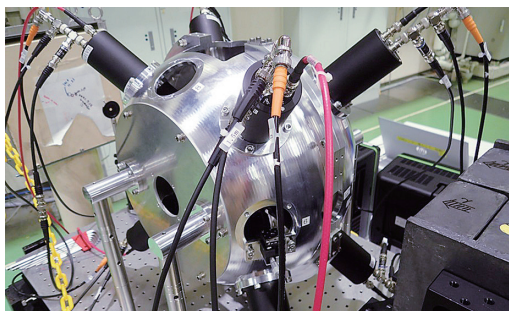


Figure 2. Positron annihilation lifetime measurement system using eight detectors and two digital oscilloscope.

User applications are currently underway at BL1U in UVSOR-III, including measurements of bulk materials and in situ measurements of defect formation under stress loading.

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.

Inverse Compton scattering of a polarized laser by energetic electrons is an excellent method to generate polarized gamma rays. A 100% polarized laser can generate 100% polarized gamma rays, but polarization varies depending on the scattering angle of the gamma rays. We have experimentally measured the spatial polarization distribution of circularly polarized gamma rays using magnetic Compton scattering that can measure the circular polarization of MeV gamma rays. Measurements of the asymmetry of gamma-ray transmission relative to the magnetized iron at each scattering angle clearly show that gamma rays are circularly polarized near the central axis, and they change from circular to linear polarization as the scattering angle increases.¹⁾

2. Gamma-Ray Vortices

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

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

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

As there is a laser with a pulse energy of 50 mJ and a pulse width of 130 fs (FWHM) in UVSOR-III, the laser strength parameter is 0.4 if the beam size can be focused to 3 μm (rms). Nonlinear inverse Thomson scattering experiments were performed in 2022, but no higher harmonic gamma-ray generation was observed. Concentric fringes were observed in the laser focusing pattern, suggesting that the laser energy was dispersed and the laser strength parameter was reduced. Further improvements will be made and re-experiment is planned in the future.

Reference

- 1) Y. Taira *et al.*, *Phys. Rev. A* **107**, 063503 (2023).

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

Application of X-Ray Microscopy

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Keywords

X-Ray Microscopy, Computer Tomography, Soft X-Ray Absorption Spectroscopy

A synchrotron-based scanning transmission X-ray microscope (STXM) is a technique to perform 2-dimensional (2D) X-ray absorption spectroscopy with high spatial resolution. The schematic image is shown in Figure 1. A monochromatic X-ray is focused by an X-ray focusing lens, a Fresnel zone plate, on a sample as a diameter around 30 nm through an order select aperture and the transmitted X-ray is detected. By scanning the sample 2-dimensionally, an X-ray absorption image is obtained. Then, by noticing the near edge X-ray absorption fine structure (NEXAFS) of the specific element, 2D chemical state of the sample can be obtained. Since characteristics of UVSOR is suitable for using extreme ultra-violet and soft X-ray region, the STXM in UVSOR, BL12, is suitable to analyze soft materials and organic materials. The X-ray range from 55 to 770 eV is a unique feature of BL12 and enables to approach lithium K-edge (55 eV~) with spatial resolution at 72 nm.¹⁾ The advantages of STXM, such as high transmittance of X-ray and relatively wide working distance, gain flexibility of the sample and its environment. Therefore, we have been developing special observation/analytical techniques mainly by designing sample cells for STXM.²⁾ Espe-

cially, nowadays, an in-situ/operando analytical technique is attracting more attentions of researchers because that is an important technique to understand intrinsic state of the samples. Recently, heating and cooling of the sample, humidity control system and electrochemistry, 2D orientation of molecules, 3D chemical state mapping, a sample transfer system without exposing to air and microscopic analysis of chemical state of lithium have been developed to explore a new field of science.³⁾ These techniques are difficult to perform by using the other microscopic techniques, such as an electron microscope.

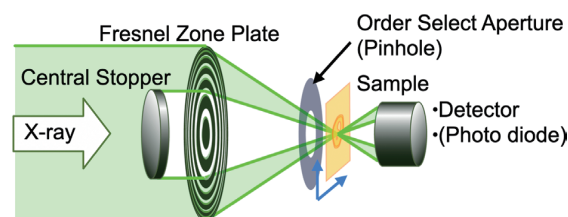


Figure 1. Schematic optical system of STXM.

Selected Publications

- T. Ohigashi and N. Kosugi, "Developments in Sample Environment for a Scanning Transmission X-Ray Microscope at UVSOR-III Synchrotron," *J. Electron Spectrosc. Relat. Phenom.* **266**, 147356 (2023).
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1. 3-Dimensional near Edge X-Ray Absorption Fine Structure of an Isolated Cell Nucleus

Computer tomography (CT) is an arithmetic method to reconstruct a 3D structure from serial tilted 2D X-ray absorption images without any destructive process. Reconstructed images of CT with full-rotation (or 180° rotation) data acquisition have quantitative values of X-ray line absorption coefficient. Therefore, by changing the X-ray energies around the absorption edge, 3D nano-NEXAFS can be performed. CT is typically performed by using full-field imaging X-ray microscopy because of shorter acquisition time of 2D X-ray transmission images. On the other hand, in regard to the radiation dose, STXM-CT is one order less than by the full-field imaging CT. This advantage is preferable to analyze 3D nano-NEXAFS of organic and bio samples with complicated structures, such as a cell nucleus. To establish STXM-CT, we have designed a rotating sample cell.^{4,5)}

An isolated cell nucleus of a HeLa S3 cell was chemically fixed with glutaraldehyde. After the critical point drying, the cell nucleus was glued on a tip of a tungsten needle (TP-001, Micro Support co., ltd.) by a crystal bond. 50 energy stacks around O K-edge (530 ~ 538 eV) dataset, $f(x,y,\theta,E)$, were obtained with tilting the sample 3.6° each, in total 180° rotation. In the energy stack, X-ray absorption images were acquired by scanning 8×8 μm² area of the sample with 160 nm pitch. Then, the dwell time was 1 ms per pixel. In total, the whole measurement process took ~12 hours. As pre-reconstruction process, the X-ray absorption images in all the energy stacks were grouped according to the X-ray energy. The 2D cross sectional images were reconstructed from each group and the 3D image was obtained by stacking those images. Finally, a 3D NEXAFS mapping dataset, $F(x,y,z,E)$, was obtained by sorting the reconstructed 3D images by the X-ray energy. For example, a 3D volume projection image is shown in Figure 2(a). A reconstructed cross sectional image in the plane shown in Figure 2(a) by a red dashed line and its local XAS spectra are shown in Figure 2(b) and 2(c), respectively. O K-edge spectra were extracted from structures of cell nucleolus (red and green areas) and cytoskeleton (yellow and blue areas) in Figure 2(b). Figure 2(d) shows RGB color distribution of these chemical components by performing SVD

fitting to the 2D NEXAFS by using aXis2000 software. The colors of the plots and of the RGB map are coincident with those of the area in Figure 2(b) except for the yellow plot. In Figure 2(d), the distribution of the green color is not only at the cell nucleolus but also slightly at the cytoskeleton. In the case of the measurement of biological samples, the measurement under cryo condition is necessary to keep samples from radiation damage.

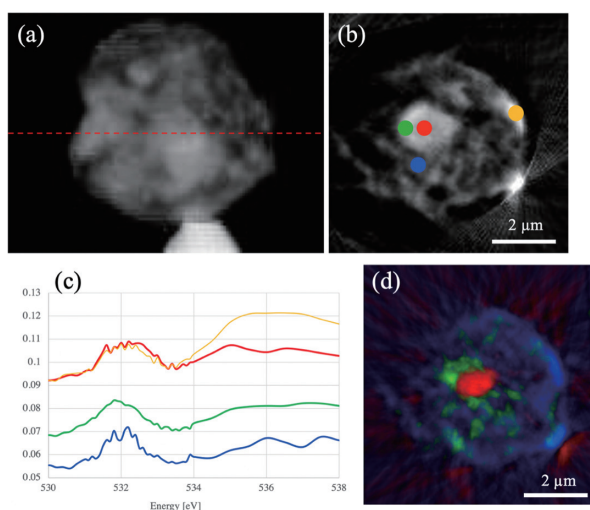


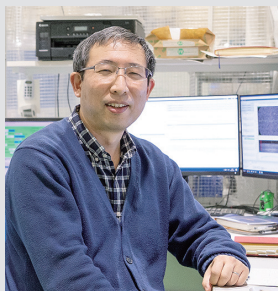
Figure 2.³⁾ (a) A 3D volume projection image of an isolated cell nucleus of HeLa S3 cell, (b) a reconstructed cross sectional image of the cell nucleus at a red dashed line, (c) X-ray absorption spectra extracted from 2(b) and (d) a RGB color distribution of the spectra. Colors of the plots and the RGB color distribution are coincident with those of areas in (b).

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Novel Spin and Chiral Materials Science by Advanced Photoemission Methodologies

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Keywords

Photoelectron Spectroscopy, Momentum Microscope, Electronic Spin Structure

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. Analyzing “art” based on physical laws lead to discoveries that connect the world of atoms with practical technology and applications, and this is what makes us so excited.

We have constructed an advanced photoelectron momentum microscope (PMM) experimental station at the UVSOR Synchrotron Facility of IMS. The PMM is a novel concept analyzer for imaging photoelectron holograms and Fermi surface patterns from the selected μm -sized area. The combination of domain-resolved photoelectron microscopy and μm -scale momentum-resolved photoelectron spectroscopy techniques is essential for the investigation of fragile radiation sensitive materials and complicated phase-separated systems.

Electron spins, which we pay particular attention to, are the source of various physical properties and functions such as

magnetism, superconductivity, and topology. We are developing a unique 3D spin vector imaging system and element-selective resonant photoelectron diffraction/spectroscopy technique for the complete photoelectron analysis. We aim to pioneer cutting-edge spin materials science through comprehensive and detailed characterization of electrons.

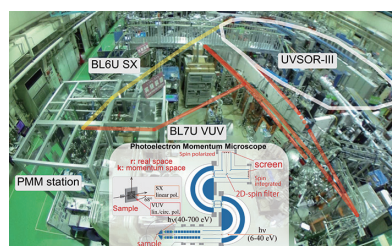


Figure 1. Photoelectron momentum microscope station at UVSOR synchrotron facility. Soft X-rays from BL6U for k_z dispersion and core-level excitations and vacuum ultraviolet light from BL7U at normal incidence for atomic orbital analysis make this station unique.

Selected Publications

- 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).
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- F. Matsui, S. Makita, H. Matsuda, E. Nakamura, Y. Okano, T. Yano, S. Kera and S. Suga, “Valence Band Dispersion Embedded in Resonant Auger Electrons,” *J. Phys. Soc. Jpn.* **90**, 124710 (2021).
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- F. Matsui, S. Makita, H. Matsuda, T. Yano, E. Nakamura, K. Tanaka, S. Suga and S. Kera, “Photoelectron Momentum Microscope at BL6U of UVSOR-III synchrotron,” *Jpn. J. Appl. Phys.* **59**, 067001 (2020).

1. Chiral Charge Density Wave Revisited

1T-TaS₂ has fascinated researchers for half a century as a system that undergoes a phase transition with three structural changes from high to low temperatures. In the lowest temperature phase, a chiral charge density wave (CDW) structure ($\sqrt{13}\times\sqrt{13}$ -R±13.90° (Figure 2(a) and (b)) is observed by electron diffraction and scanning probe microscopy. Regarding the electronic structure, the first paper on two-dimensional ARPES, which observed the average of both chiral structures,¹⁾ had a strong impact, and it greatly influenced subsequent papers. Only recently has the nesting vector, which is the basis of charge density waves, begun to be accurately discussed in measurements using chiral single domains. In a single layer, unpaired electron spins are isolated in a 13-Ta-atom units, commonly known as the Star of David (six pointed star). Isolated spins are stabilized by interlayer interactions. Recently, STM revealed coexistence of Mott-insulator and band-insulator domains on the cleaved surface,²⁾ which has attracted attention as a great opportunity to elucidate the CDW mechanism (Figure 2(c) and (d)). Here, spin-resolved micro-ARPES using PMM will be a decisive tool to clarify the physics behind this complex CDW phenomena.

Figures 2(e) and 2(f) shows the constant energy contour and band dispersion of 1T-TaS₂, respectively, measured by UVSOR-PMM³⁾ at 30 K.⁴⁾ The ellipsoidal electron pockets around the M points are modified and exhibit the so-called “windmill” rotational symmetry modulation around the Γ point due to the CDW formation. The microscopic field-of-view enabled selective observation of one of the two types of twinned CDW domains. Although we have expected the for-

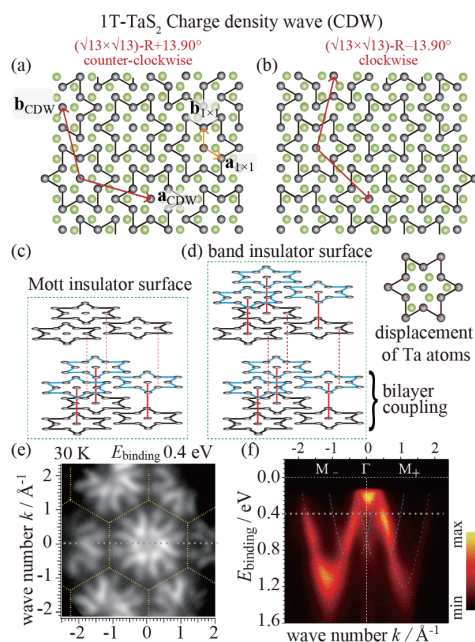


Figure 2. (a) and (b) Lateral atomic structure models of 1T-TaS₂ in the CDW phase. (c) and (d) Structure models of different surface terminations. (e) Constant energy contour at the binding energy of 0.4 eV. (f) Valence band dispersion along the $M\Gamma M$ axis.

mation of a sharp gap on the band dispersion due to CDW, noticeable intensity remained even around the region of $E_{\text{binding}} = 1$ eV and $|k| = 1 \text{ \AA}^{-1}$ (Figure 2(f)). This result may be due to the observed mixture of multiple electronic states in the Mott insulator and band insulator at different surface terminations, which was pointed out in STM.²⁾ Further detailed PMM works will be described elsewhere.

2. Original Analyzer for 3D-Atomic Structure Imaging and 3D-Spin Vector Analysis

Photoelectron holography is an element specific 3D atomic imaging technique. Local atomic arrangements of dopant atoms can be characterized. Compositional crossover of multiple-site Ag doping in Bi₂Se₃ from substitution to intercalation was revealed (Figure 3).⁵⁾

We are aiming at highly efficient and comprehensive measurement of atomic structure and spin distribution. Omnidirectional photoelectron acceptance lens (OPAL)⁶⁾ together with Projection-type electron spectroscopy collimator analyzer (PESCATORA)⁷⁾ enables photoelectron holography measurement of the full hemisphere. Moreover, we invented Right angle deflection imaging analyzer (RADIAN)⁸⁾ for spin vector analysis with k/r -space resolution. We are expanding the MM system based on our original inventions.

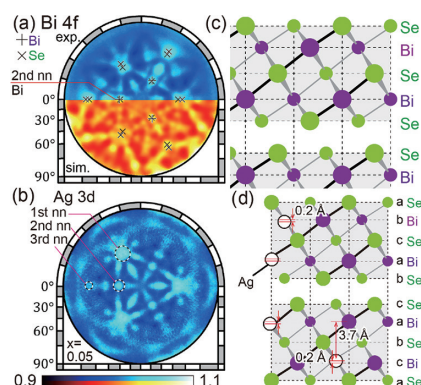


Figure 3. (a) Bi4f and (b) Ag3d photoelectron holograms of Ag-doped Bi₂Se₃. Atomic structure models of (c) pristine and (d) Ag-doped Bi₂Se₃ deduced by holography analysis.

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- 4) F. Matsui *et al.*, *Rev. Sci. Instrum.* **94**, 083701 (2023). [Editor's pick]
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Angle-Resolved Photoemission Study on Strongly Correlated Electron Materials

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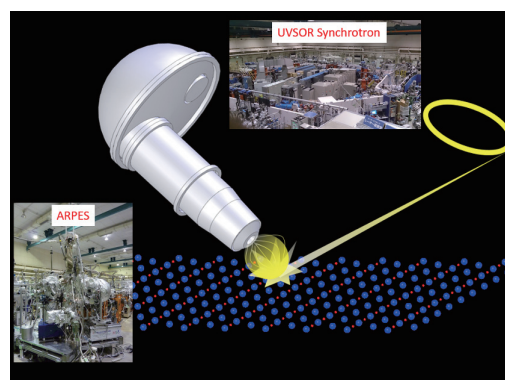
ISHIHARA, Mayumi
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Keywords Strongly Correlated Electron System, Synchrotron Light, Photoemission

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 structure near the Fermi level: Superconducting gap, kink structure and so on. The main target materials of our group is high- T_c superconductors, such as cuprates and iron pnictides and use UVSOR-III as a strong light source.

Our group is also developing high-efficiency spin-resolved ARPES system. Spintronics is a rapidly emerging field of science and technology that will most likely have a significant

impact on the future of all aspects of electronics as we continue to move into the 21st century. Understanding magnetism of surfaces, interfaces, and nanostructures is greatly important for realizing the spintronics which aims to control and use the function of spin as well as the charge of electrons. Spin-resolved ARPES is one of the most powerful experimental techniques to investigate the magnetic properties of such materials.



Selected Publications

- K. Tanaka, W. S. Lee, D. H. Lu, A. Fujimori, T. Fujii, Risdiana, I. Terasaki, D. J. Scalapino, T. P. Devereaux, Z. Hussain and Z.-X. Shen, "Distinct Fermi-Momentum-Dependent Energy Gaps in Deeply Underdoped Bi2212," *Science* **314**, 1910–1913 (2006).
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- 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 $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ Superconductors: Doping Dependence of Nodal and Antinodal Superconducting Gaps," *J. Phys. Soc. Jpn.* **88**, 044710 (2019).
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1. Development of Spin-Resolved ARPES with Image-Spin Detection

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

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

In 2020, we set up the spin detection apparatus shown in Figure 1(a) and finished tuning the electron lens parameters of the spin detection section. With this apparatus, we succeeded in obtaining a spin-resolved signal on the Au(111) surface as shown in Figure 2(c). According to rough estimates, the

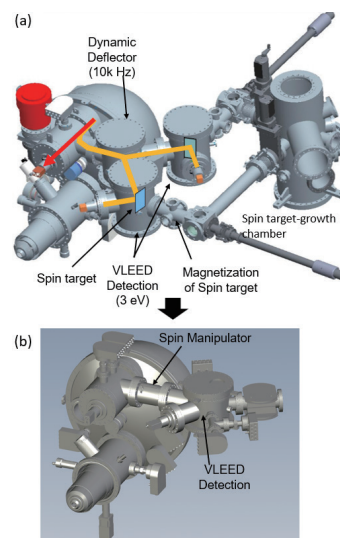


Figure 1. Previous (a) and current (b) setup of image-spin ARPES.

efficiency of spin-resolved ARPES was 100 times higher than that of the single-channel detection systems currently used in the world. However, the spin-resolved ARPES bands were broad compared to the normal ARPES ones shown in Figure 2(b), meaning that the momentum resolution was not so good. With this apparatus, the instrument required two VLEED detection chambers and a high-quality spin target of the same quality to detect spin information in the x , y , and z directions of the sample. In addition, the spin targets had to be magnetized frequently during the measurement.

To overcome these problems, we have introduced a new “spin manipulator” that can change the spin direction of the passing electrons in any direction. The new setup shown in Figure 1(b) requires only one VLEED detection chamber and the users do not need to magnetize the spin target during the measurement. The installation of the spin manipulator and optimization of the spin target deposition conditions have greatly improved the spin-resolved images, and in 2022, we were able to obtain spin-resolved images with momentum resolution comparable to that of normal ARPES, as shown in Figure 2(d). We are currently optimizing the lens parameters of the spin manipulator to obtain spin information in the remaining two axial directions.

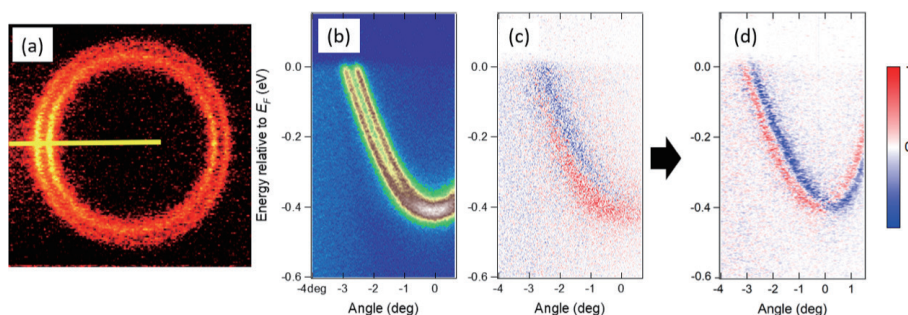


Figure 2. a) Fermi surface of Rashba spin splitting in Au(111) surface states and (b) image plot of normal ARPES along the yellow bar in (a). Spin-resolved ARPES data showing the spin polarization (blue–red scale) with the previous experimental setup (c) and the current setup (d).

Soft X-Ray Spectro-Microscopy and Spectro-Scattering for Life Science Research

UVSOR Synchrotron Facility
Division of Advanced Photochemistry



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Scanning Transmission X-ray Microscopy: STXM is a mainstream of Soft X-ray Spectromicroscopy techniques due to that versatility since it is a photon-in & photon-out technique, which allow us to have some freedom of the sample specimen environments, like under magnetic field or wet condition. STXM utilizes the NEXAFS: Near Edge X-ray Absorption Fine Structure as the image contrast mechanism. When we study the soft materials ex. polymers, which consist of Carbon, Nitrogen, and Oxygen as a main element, through the NEXAFS spectral features we can obtain the chemical bonding/functional group information about the samples. Combined with the focused soft x-ray beam, about several tens of nanometer, we can achieve the chemical component mapping of the samples. If we look at the phase-separated polymer blend, we can speciate each domain with such high spatial resolution. Figure 1 shows an example of the chemical component map; blend of PS: Polystyrene, PMMA: Polymethylmethacrylate, and PVC: Polyvinylchloride mixed with nano-clays. The thin-section sample was prepared by a ultramicrotome (~100 nm thick).

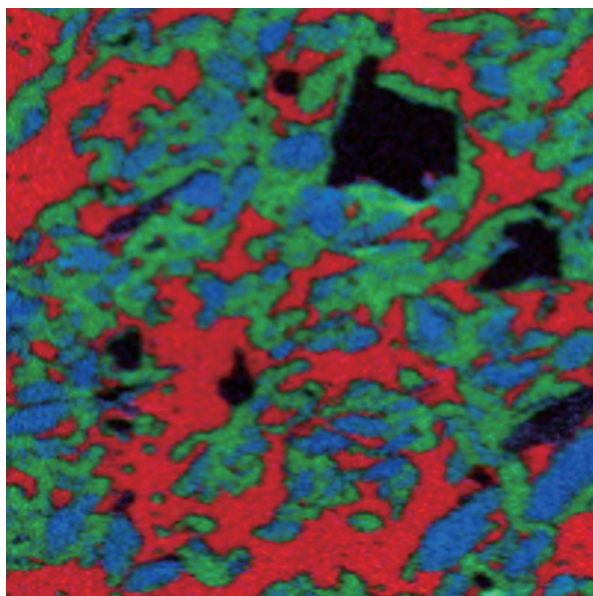


Figure 1. STXM chemical component map of the Polymer-Nano Clay Blend. R:PS, G: PMMA, B: PVC. The image size is 20 μm by 20 μm .

Before joined the IMS, at the Diamond Light Source in the UK, I had worked with my collaborators to study many

environmental science cases to understand the interaction between minerals and organics including bacteria with using STXM in the Soft and Tender X-ray regimes.^{2,3)}

Since January 2023 I have been responsible to operate the BL4U STXM beamline at the UVSOR to maintain the user program including the industrial research, especially for the soft materials research like the radiation sensitive rubbery materials, which is difficult to study by the electron microscopy.

Another important mission is to plan the future UVSOR project,¹⁾ which will be under the Research Center for Autonomous Functions by Tailor-made Photon Science. Toward the new approach using the combination of the low energy range photon from infrared to soft/tender x-rays, currently I am working with many scientists in the Life Science field to start the feasibility/trial studies using STXM and other soft x-ray techniques such as RSoXS: Resonant Soft X-ray Scattering, counterpart of STXM in reciprocal space. Figure 2 illustrates my concept to study the soft materials including biological samples using the three types of x-ray technique based on the NEXAFS spectroscopy.

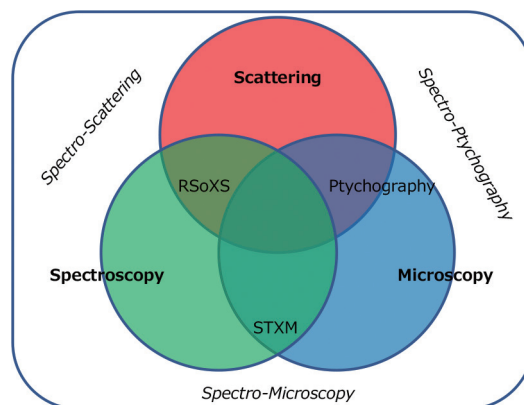


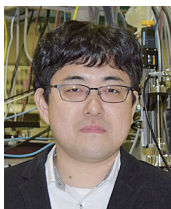
Figure 2. Concept of the NEXAFS based submicron chemical speciation methods.

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

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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 have developed time-resolved XAS of photochemical reactions by synchronizing soft X-ray probe pulses with the laser pump pulses.³⁾

1. Time-Resolved XAS of Photochemical Reactions in Solutions

The time-resolved XAS system has been developed at the soft X-ray beamline BL-13A of the Photon Factory, KEK.³⁾ The laser pump pulses (515 nm, 290 fs) were almost coaxially introduced to the liquid cell with the soft X-ray probe pulses. A trigger clock system and a frequency synchronization mod-

ule were used for the synchronization of the laser pulses with soft X-ray pulses.

By using this measurement system, we have measured the time-resolved N K-edge XAS spectra of iron phenanthroline $[\text{Fe}(\text{phen})_3]^{2+}$ aqueous solutions during the photoexcitation process with the time resolution of 45 ps. The C=N π^* peaks of the ligands in the photoexcited (high spin) state of $[\text{Fe}(\text{phen})_3]^{2+}$ are shifted to the lower photon energy compared to those in the ground (low spin) state. The temporal evolution of the peak intensity difference as a function of the delay time of the soft X-ray pulses with the laser pulses has determined that the time constant of the relaxation process from the high spin state to the low spin state is 550 ± 12 ps.

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Mesoscopic Structural Analysis of Polymer Materials

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Polymer composites, made by mixing multiple materials, have become increasingly important in developing polymer materials with higher functionality. Structural analysis is required for each constituent material (chemical species). However, with conventional small-angle X-ray and neutron scattering methods (SAXS/SANS), it is not easy to analyze the structure of each constituent material because the overall structural information is obtained simultaneously.

1. Resonant Soft X-Ray Scattering for Polymer Materials

Recently, we have developed resonant soft X-ray scattering (RSoXS) method. RSoXS has element, molecule and

molecular orientation selectivity, making it possible to observe specific mesoscopic structures that cannot be observed with conventional SAXS. In particular, soft X-rays have the advantage of being able to selectively observe light elements such as carbon, nitrogen, and oxygen. Last year, we succeeded in analyzing the twisted structure of self-assembled liquid-crystal helical nanofilaments without electron density modulation.¹⁾

This year, we started researching polymers. In order to understand the performance of epoxy resins, it is necessary to analyze the crosslinked structure, which is usually composed of light elements and cannot be observed by conventional SAXS. Therefore, by using our resonant soft X-ray scattering method, we focused on the elements peculiar to crosslinked molecules and started to analyze the crosslinked structures of polymers by utilizing the resonance scattering of these elements.

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Visiting Professors



Visiting Professor

MASE, Kazuhiko (from *High Energy Accelerator Research Organization*)

Development of New Nonevaporable Getter (NEG) with a Low Activation Temperature

Nonevaporable getter (NEG) is a functional material that evacuates residual gases at room temperature by forming an active surface when heated in ultra-high vacuum (UHV). Recently we have developed a new NEG thin film that was prepared by the following simple procedure, sublimation of high-purity Ti under UHV in the range of 10^{-7} to 10^{-8} Pa, followed by N₂ introduction (partially nitrated high-purity Ti). We confirmed that partially nitrated high-purity Ti deposited on inner surfaces of a vacuum vessel pumps H₂, H₂O, O₂, and CO gases even after 30 cycles of pumping, baking at 185 °C for 6 hours, cooling down to room temperature, introduction of high-purity N₂, and exposure to air. We applied surface-partially nitrated high-purity Ti deposition to the inner surface of the vacuum ducts in the upstream section of BL-12C in the Photon Factory 2.5 GeV ring and baked them at 250 °C. Pressure in the section reached 2.2×10^{-8} Pa without ion pumps after isolation from a turbomolecular pump with a gate valve. Partially nitrated high-purity Ti deposition is also applicable to UVSOR beamlines.



Visiting Professor

FUKUHARA, Takeshi (from *RIKEN*)

Quantum Gas Microscopy of a Frustrated XY Model in Triangular Optical Lattices

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. We have prepared Bose-Einstein condensates (BECs) in triangular optical lattices for the study of frustration physics, and implemented quantum gas microscope, which makes it possible to observe such quantum gases at the single atom level. By regarding the phase of BECs as a spin and by implementing antiferromagnetic spin–spin couplings via a lattice shaking technique, we have realized frustrated XY spin model. The frustration leads to two-fold ground states corresponding to two chiral modes. We have investigated the relaxation dynamics from the ferromagnetic phase to the spiral phases with the chiral modes by dynamically changing the spin–spin coupling. The domain formation of the chiral modes has been observed through high-spatial-resolution time-of-flight measurements.



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 $10 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and have potential applications as flexible and efficient electronic devices. However, current organic semiconductor electronics are mostly built on heterojunctions composed of polycrystalline or amorphous molecular solids, in contrast to the inorganic semiconductor cases where epitaxial growth of single-crystalline is one of the most essential technologies for modern electronic applications. Our group has been working on single-crystalline heterojunctions of organic molecular semiconductors by “molecular beam epitaxy” techniques. Recently, we have published that on the single-crystal surface on phthalocyanine-copper (CuPc), which is one of the most well-studied organic semiconductors, perfluorinated CuPc exhibits epitaxial growth. As this is an analogical finding to our previous collaborative achievement with IMS for epitaxial perfluorinated pentacene on the single-crystal pentacene, further studies on its electronic properties will be of our next targets.