RESEARCH ACTIVITIES
Photo-Molecular Science

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

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

Exploring Quantum-Classical Boundary

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Keywords Quantum-Classical Boundary, Quantum Simulation, Quantum Computing

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

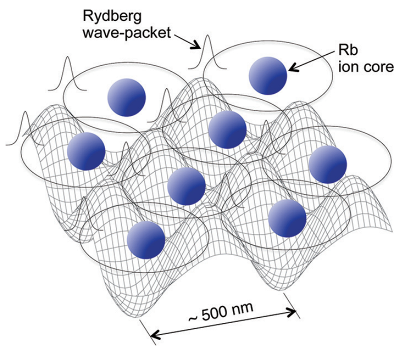

Figure 1. Metal-like quantum gas. A schematic of the many-body system of ultracold Rydberg atoms, where electronic wave functions spatially overlap between neighboring atoms.[2,7)]

Selected Publications

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

1. Development of an “Ultrafast Quantum Simulator” by Optical Control with Precisions on the Attosecond Temporal and Submicron Spatial Scales[3–9]

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

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

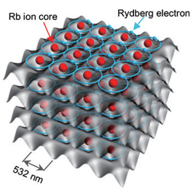

Figure 2. Schematic of the standard hardware of the ultrafast quantum simulator.[3,4,7]

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

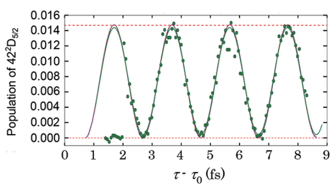

Figure 3. Time domain Ramsey interferometry of ultracold ^{87}Rb atoms with attosecond precision to be used as a readout interface of the ultrafast quantum simulator. Population of the $42^2D_{5/2}$ Rydberg state is plotted as a function of the delay τ between two laser pulses, where τ_0 ~ 50 ps. Adopted from Ref. 5).

2. Application of an “Ultrafast Quantum Simulator” to Quantum Computing[3]

We are developing a cold-atom based quantum annealer with the hardware of the ultrafast quantum simulator.[10] The cold-atom annealer has advantages against the one with the superconducting qubits. Those advantages include scalability and efficiency. All to all connections among physical bits necessary for quantum annealing could also be easier with cold atoms than superconducting qubits.

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

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

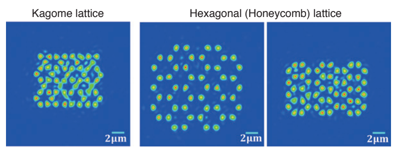

Figure 4. Examples of the world’s smallest arbitrary arrays of optical traps.[3]

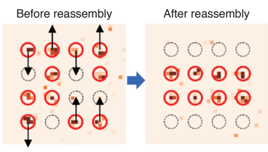

Figure 5. Assembly of an arbitrary array of single Rb atoms.

3. Engineering Quantum Wave-Packet Dispersion with a Strong Nonresonant Femtosecond Laser Pulse[12]

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

Our ultrafast quantum simulator operates with atomic Rydberg levels,[3–9] whose level structure is anharmonic, so that its wave packet is dispersed and broadened quickly. The new control method for wave-packet dispersion developed here would serve as an enabling technology for our ultrafast quantum simulator to enhance its functionality for quantum simulation and computing.

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3) US Patent (3rd Nov. 2020) “Quantum Simulator and Quantum Simulation Method,” H. Sakai (Hamamatsu Photonics K.K.), K. Ohmori (IMS) *et al.*, and one more patent publication.
4) White Paper 2018 on Manufacturing Industries published by Ministry of Economy Trade and Industry, JAPAN.
5) C. Liu *et al.*, *Phys. Rev. Lett.* **121**, 173201 (2018).
6) Highlighted in “Quantum-Technology Innovation Strategy” by the Cabinet Office of Japan, January 2020.
7) M. Mizoguchi *et al.*, *Phys. Rev. Lett.* **124**, 253201 (2020).
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11) D. Barredo *et al.*, *Science* **354**, 1021 (2016).
12) H. Katsuki *et al.*, *arXiv:*1910.08241 (2019).

Electronic Property of Functional Organic Materials

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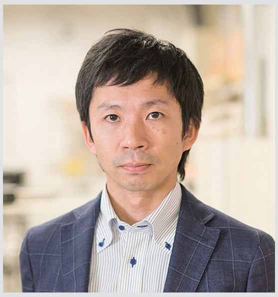

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Keywords Photoelectron Spectroscopy, Molecular Assemble, Electronic State

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.

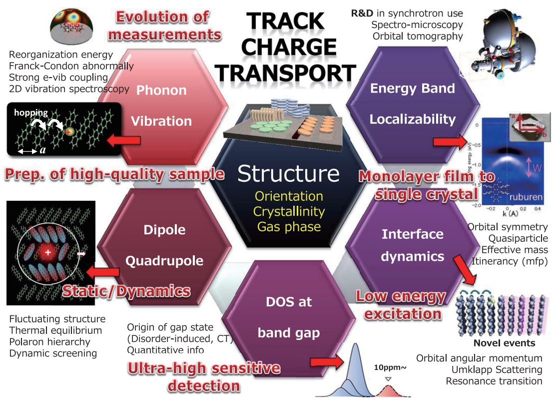

Figure 1. Overview of our agenda. A rich assortment of surface and interface structures of FOM to provide complicated spectral features of ultraviolet photoelectron spectroscopy.

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. The Role of Initial and Final States in Molecular Spectroscopies: A Case Study of DBP[1,2)]

Interpreting experimental spectra of organic semiconductor films is challenging, and understanding the relationship between experimental data obtained by different spectroscopic techniques, UPS, IPES, 2PPE and optical absorption requires a careful consideration of the initial and final states for each process. We present a coherent framework that is capable of treating on equal footing most spectroscopies. We develop a simple model for the expected energy level positions by the spectroscopies and relate them to the energies of molecular states. Molecular charging energies in photoionization processes, as well as adsorption energies and the screening of molecular charges due to environmental polarization, are taken into account as the main causes for shifts of the measured spectroscopic features. We explain the relationship between these quantities, as well as with the transport gap, the optical gap and the exciton binding energy.[1)]

Our considerations serve as a model for weakly interacting systems where wave function hybridizations between adjacent molecules are negligible as demonstrated for DBP films on graphite.[2)]

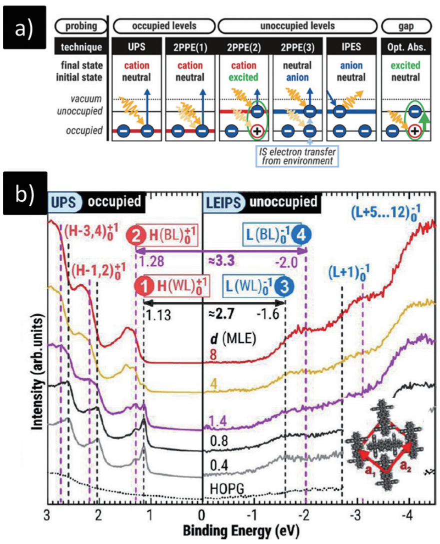

Figure 2. a) Scheme of the typical spectroscopic processes in UPS, 2PPE, IPES, and optical absorption in relation to the initial and final states. b) Thickness dependences of XeI-UPS and IPES (LEIPS) of DBP films on graphite. The energy positions of bilayer (BL) are shifted from wetting layer (WL) of monolayer. The figure is after ref 1,2).

2. Impacts of Electrostatic Interactions on the Energy Levels of Organic Semiconductor Blends[3)]

Halogenation of conjugated molecules represents a powerful approach to tune the electronic structure of molecular thin films through inductive effects and long-range intermolecular electrostatic interactions. We provide a comprehensive experimental and theoretical analysis of the prototypical blend formed by pentacene (PEN) and perfluoropentacene (PFP) to relate structure with electronic properties. We find a mixed-stack structural motif in standing and lying orientations depending on the substrate nature. In the standing orientation, the ionization potential lies in between the values of the pure components, in line with the established picture of averaged molecular quadrupole moments. For the lying orientation, however, we experimentally observe an ionization potential lower than both pristine values, which seems at odds with this simple rationale. Electrostatic simulations based on the knowledge of the atomistic structure of the films capture the complex experimental scenario for both orientations. In particular, the ultralow ionization potential of films formed by lying molecules is identified as a signature of the monolayer structure, where quadrupolar interactions are responsible for a difference of *ca.* 0.4 eV in the highest occupied molecular orbital energy as compared to thicker films with the same molecular orientation.

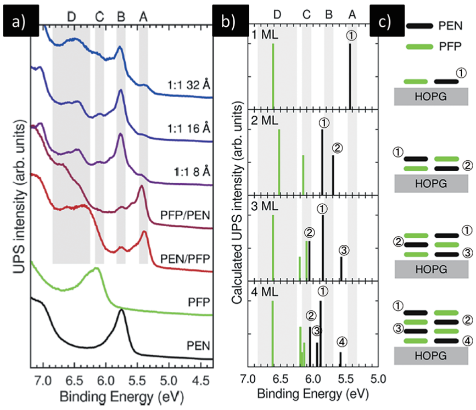

Figure 3. (a) UPS of blends PEN/PFP films of lying orientation on graphite. The spectra of PEN and PFP monolayers are shown as reference. b) Calculated energy levels of PEN:PFP layers; black and green bars correspond to the IPs of PEN and PFP, respectively. The calculated intensity (bar height) depends on the depth z of the ionized layer from the surface in Figure 3c). The figure is after ref 3).

3. Other Activities in UVSOR

We have conducted beamline R&D and user supports in collaboration with other universities. Experiments using photoelectron momentum microscope is started at BL6U.[4)]

References

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4) F. Matsui *et al.*, *Jpn. J. Appl. Phys.* **59**, 067001 (2020).

* IMS-IIPA Program
† IMS with Osaka University (Cadet Program)

Light Source Developments by Using Relativistic Electron Beams

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Keywords Accelerator, Beam Physics, Astrobiology

UVSOR is a synchrotron light source providing low energy synchrotron light ranging from terahertz wave to soft X-rays. Although it was constructed about 30 years ago, its performance is still in the world top level among low energy synchrotron light sources. This is the result of the continuous effort on improving the machine. Our research group has been developing and introducing new accelerator technologies toward producing bright and stable synchrotron light, such as low emittance electron beam optics, novel insertion devices or state-of-the-art beam injection technique. We have been developing novel light sources, such as free electron laser, coherent synchrotron radiation, optical vortices and laser Compton gamma-rays. We have been investigating beam physics which would be the basis of the future developments of the facility.

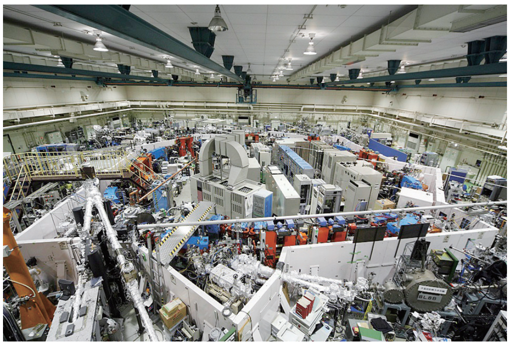

Figure 1. UVSOR-III Electron Storage Ring and Synchrotron Radiation Beamlines.

Selected Publications

- S. Bielawski, C. Evain, T. Hara, M. Hosaka, M. Katoh, S. Kimura, A. Mochihashi, M. Shimada, C. Szwaj, T. Takahashi and Y. Takashima, "Tunable Narrowband Terahertz Emission from Mastered Laser–Eelectron Beam Interaction," *Nat. Phys.* **4**, 390–393 (2008).
- M. Shimada, M. Katoh, M. Adachi, T. Tanikawa, S. Kimura, M. Hosaka, N. Yamamoto, Y. Takashima and T. Takahashi, "Transverse-Longitudinal Coupling Effect in Laser Bunch Slicing," *Phys. Rev. Lett.* **103**, 144802 (2009).
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- 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, "Controlling the Orbital Alignment in Atoms Using Cross-Circularly Polarized Extreme Ultraviolet Wave Packets," *Phys. Rev. Lett.* **123**, 233401 (2019).

1. Light Source Technology Developments Based on Laser and Synchrotron

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

In these years, we are focusing on generation of spatially structured light, such as optical vortex beams and optical vector beams from undulators, in collaboration with Hiroshima Univ. and Nagoya Univ. We have succeeded in producing such novel photon beams and are exploring their applications. More recently, we have started exploring the possibility utilizing the temporal structure of undulator radiation, in collaboration with Saga Light Source and Toyama Univ. So far, we have been demonstrated the coherent controls of atoms by using radiation from two undulators arranged in tandem. It has been widely believed that such experiments were possible only with precisely controlled laser beam. Our results may open a new research field on the application of synchrotron radiation.

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

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

Currently, the storage ring is stably operated for many of the users, however, the requirements from the users for the stability is getting higher and higher. We are improving cooling water system and developing various feedback systems. 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. For a long-term plan, we continue the design studies on the new low emittance optics of the present synchrotron and on new accelerator systems such as a linear accelerator based free electron laser or a diffraction limited storage ring light source.

We are collaborating with Nagoya University Synchrotron Radiation Research Center (NUSR) for the accelerator technology developments. Accelerator magnets based on permanent magnets are being developed, which would contribute to the power consumption saving in the future plan. Various high brightness electron sources are being developed and tested. New beam diagnostic technologies toward beam stabilization are being developed. Several PhD students from the University are involved in these studies.

We are also collaborating with Accelerator Research Laboratory at KEK for the compact Energy Recovery Linac (cERL) project, which is a novel electron accelerator toward a diffraction-limited synchrotron light source and a free electron laser.

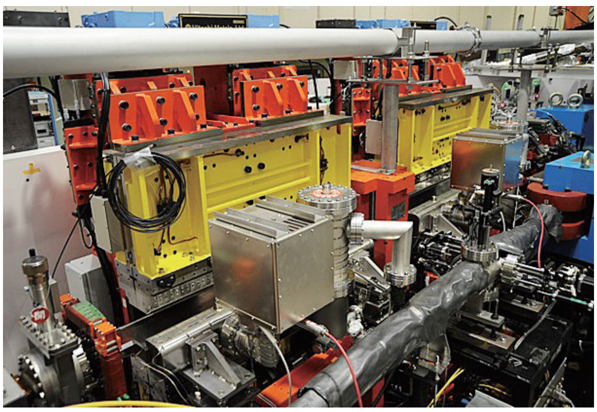


Figure 2. Twin Polarization-variable Undulators/Optical Klystron at UVSOR-III.

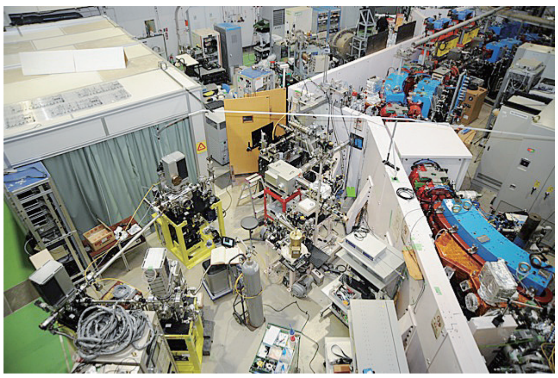
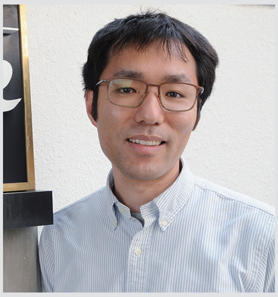


Figure 3. UVSOR BL1U experimental station for source development studies.

* carrying out graduate research on Cooperative Education Program of IMS with Nagoya 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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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 fileds and lasers.

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

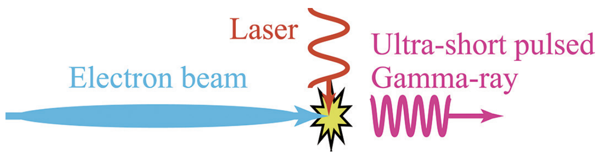

Figure 1. Schematic illustration of 90-degree laser Thomson scattering.

Selected Publications

- Y. Taira, M. Adachi, H. Zen, T. Tanikawa, N. Yamamoto, M. Hosaka, Y. Takashima, K. Soda and M. Katoh, "Generation of Energy-Tunable and Ultra-Short-Pulse Gamma Ray via Inverse Compton Scattering in an Electron Storage Ring," *Nucl. Instrum. Methods Phys. Res., Sect. A* **652**, 696 (2011).
- Y. Taira, H. Toyokawa, R. Kuroda, N. Yamamoto, M. Adachi, S. Tanaka and M. Katoh, "Photon-Induced Positron Annihilation Lifetime Spectroscopy Using Ultrashort Laser-Compton-Scattered Gamma-Ray Pulses," *Rev. Sci. Instrum.* **84**, 053305 (2013).
- Y. Taira, T. Hayakawa and M. Katoh, "Gamma-Ray Vortices from Nonlinear Inverse Thomson Scattering of Circularly Polarized Light," *Sci. Rep.* **7**, 5018 (2017).
- Y. Taira and M. Katoh, "Gamma-Ray Vortices Emitted from Nonlinear Inverse Thomson Scattering of a Two-Wavelength Laser Beam," *Phys. Rev. A* **98**, 052130 (2018).
- Y. Taira and M. Katoh, "Generation of Optical Vortices by Nonlinear Inverse Thomson Scattering at Arbitrary Angle Interactions," *Astrophys. J.* **860**, 45 (2018).

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

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

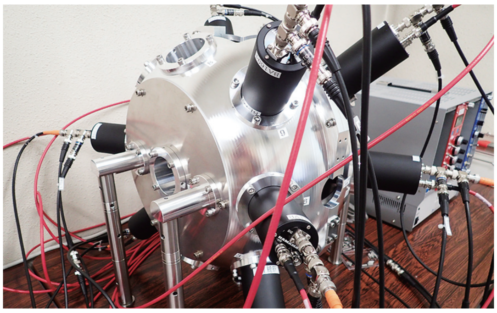

Figure 2. Positron lifetime measurement system using 10 detectors.

Users can currently utilize GiPALS at BL1U in UVSOR-III. A result of defect analysis for a GAGG scintillator was published in 2020.[1)]

We plan to change the laser injection position to increase the ultra-short pulsed gamma-ray intensity in the next fiscal year. In the current laser injection position, the opposite side of the laser injection window is the vacuum duct. Thus, the laser hits the vacuum duct and generates gas. This gas induces the background gamma-rays, which is generated by the interaction with an electron beam. As a result, the laser cannot be tightly focused and therefore the intensity of the gamma-ray is weak. The laser can be focused down to few tens of micrometers at the new laser injection position. We estimate that the intensity of the gamma-rays will be increased more than 20 times.

On the other hand, we are planning to develop other measurement technique for the annihilation gamma-rays, such as a three-dimensional distribution imaging technique for defects, spin polarized positrons generated from circularly polarized gamma-rays, and age-momentum correlation (AMOC).

2. Short Wavelength Optical Vortices

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

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

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

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

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

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Angle-Resolved Photoemission Study on Strongly Correlated Electron Materials

UVSOR Synchrotron Facility
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Professional Employment
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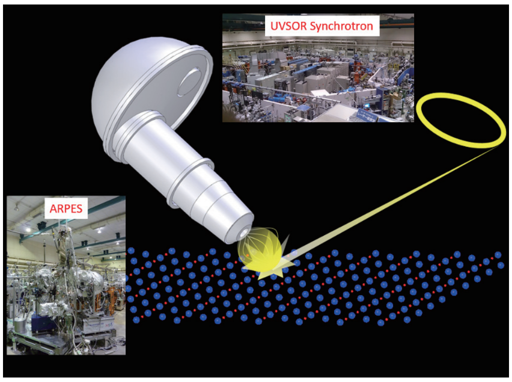
Keywords Strongly Correlated Electron System, Synchrotron Light, Photoemission

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

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



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

1. Quantitative Comparison between ARPES and ERS on Multilayer Cuprates Superconductor[1)]

It has been well known that one of the most efficient ways to increase the critical temperature (T_c) of high-T_c cuprate superconductors (HTSCs) is to increase the number of neighboring CuO_2 planes (n). T_c of the optimally doped region ($T_{c:max}$) generally increases from single layer (n = 1), double layer (n = 2), to triple layer (n = 3) and then decreases for n > 4. In order to explain the n dependence of T_c, several mechanisms have been proposed. However, it has been unclear which parameter governs the n dependence of $T_{c:max}$ because of the lack of detailed knowledge about the electronic structure of the multilayer cuprates. In this study, we have performed angle-resolved photoemission spectroscopy (ARPES) and electronic Raman scattering (ERS) to clarify the electronic structure of optimally doped triple-layer $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi2223) which has the highest $T_{c:max}$ (~110 K) among Bi-based HTSCs.

Since the superconducting gap in the cuprates has a d-wave symmetry, we need k-selective experimental probes. Although ARPES and ERS are the most powerful k-selective probes, the gap sizes estimated from these two techniques are not always identical. To clarify the origin of the discrepancy, we have examined a direct comparison of ARPES and ERS through the Kubo formula analysis. In a previous study on the double layer Bi2212, we proved that this method is valid and advantageous.[2)]

In ERS study on optimally doped Bi2223, we found that B_{1g} spectra, which is sensitive to the antinodal region in the k-space, showed double peaks as shown in Figure 1a (blue curve). This is the first observation of multiple peaks in B_{1g} spectra in HTSCs. On the other hand, ARPES study on Bi2223 reveals two Fermi surfaces (FSs), which can be attributed the FS closer to the Γ point to that of the outer CuO_2 plane (OP) and the other to that of the inner CuO_2 plane (IP), assuming that the doping level is higher in the OP than in the IP. From those observations, we think that the double peak in ERS is originated from two different bands which form different FSs observed by ARPES.

To confirm our interpretation of the double peak, we calculated the Raman spectra from the ARPES, using the Kubo formula. For triple layer compounds, by separating the IP and OP bands of ARPES, we can calculate their separate contribution to the Raman spectra, and verify if the two B_{1g} Raman peaks truly originate from the two separate bands. The ARPES intensities for the IP and OP bands were separated by a Gaussian fit of the energy distribution curves using three Gaussian peaks, one for the IP and OP band each and one for the high energy incoherent intensity that originates from the strong correlations effects in the antinodal part of the k-space.

The calculated B_{1g} and B_{2g} Raman spectra for the optimally doped Bi2223 are compared with the experimental ones in Figure 1. One can find that the calculated spectra from the ARPES data successfully reproduce the experimental Raman spectra. The striking result is that, in the B_{1g} configuration, the IP and OP bands exhibit peaks at different energies that are close to the experimental B_{1g} peak energies. Here the OP peak position is slightly underestimated, which may be due to the fact that a small portion of the antinodal part of the momentum space is missing in our input ARRPES data. By summing the separate contribution of IP and OP, we obtain a thick orange line. A rather good correspondence of the calculated and experimental IP and OP peaks provides strong proof that the double B_{1g} Raman peak truly originates from the two separate bands of Bi2223 and, therefore, that it is a signature of the double superconducting gap of this material.

This results clarify systematic doping dependence of superconducting gaps of IP and OP in ERS, which reveals that the both the pair-breaking energy and the gap ratio are larger in triple layer cuprates than in single and double layer cuprates (not shown).

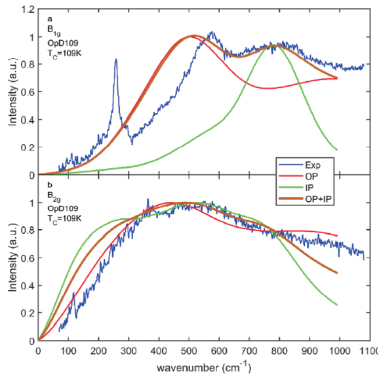

Figure 1. Comparison of the Raman spectra calculated from the ARPES (orange cruve) and the experimentally observed Raman spectra (blue curve) for B_{1g} (a) and B_{2g} (b) polarizations. The red and green curves are obtained from the ARPES data for the OP and IP, respectively. In the total curve IP+OP (thick orange), the contributions both from IP and OP are taken into account

2. Development of Low Temperature 6-Axis Manipulator for High-Resolution ARPES

We have developed low temperature 6-axis manipulator for high energy resolution ARPES measurements and achieved one of the lowest temperature 6-axis manipulators in the synchrotron radiation facilities in the world. To achieve lower temperature, we have started computational thermal simulation.

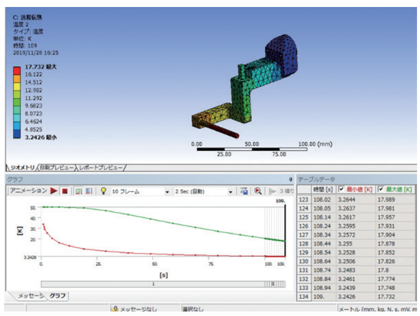

Figure 2. Modeling and simulation of thermal analysis for 6-axis manipulator.

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

Photoelectron Momentum Microscope at IMS

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research

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Senior Researcher

The demand for photoelectron micro-spectroscopy and spectro-microscopy to characterize the electronic properties of microstructures is growing rapidly. Photoelectron spectroscopy resolved in three-dimensional momentum space with a microscopic field of view is realized by combining a so-called Momentum Microscope (MM) with a soft X-ray synchrotron radiation source. We built a new MM station at BL6U,[1] an undulator-based soft X-ray beamline. This station opens the door to direct observation of the Fermi surface of μm-sized crystals, which was difficult with conventional ARPES-type hemispherical analyzers.

1. Momentum Microscope

As shown in Figure 1, the system consists of a photoemission electron microscope (PEEM) lens, a hemispherical deflection analyzer as an imaging-type energy filter, and a 2D detector with a CMOS camera. The details of the specification evaluation result are described elsewhere.[1] In brief, the energy resolution of the analyzer was estimated to be 20 meV at pass energy of 20 eV. The spatial resolution in the microscopy mode was evaluated to be about 50 nm. The momentum resolution of 0.012 Å^{-1} has been achieved. The position of sample stage facing to the analyzer is precisely controlled by a hexapod. Samples can be cooled down to 8 K and heated up to 400 K.

Since a high voltage is applied between the sample and the PEEM lens, it is essential that the sample be flat. We have developed a technique to cleanly cleave sub-mm-sized crystals in the ultra-high vacuum condition. Figure 2 shows a valence band measurement with a wide **k** acceptance of 6 Å^{-1} in diameter.

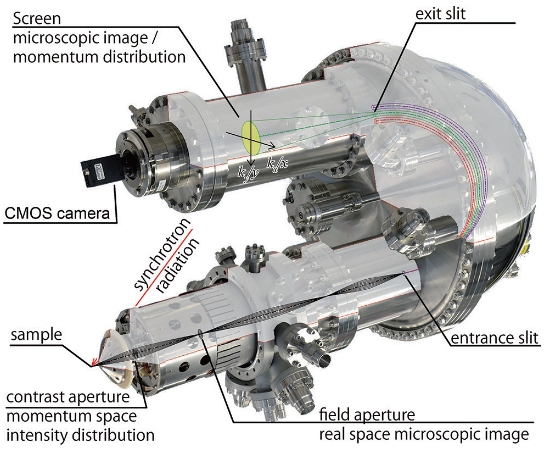

Figure 1. Schematic drawing of the momentum microscope.[1]

2. Original Electron Analyzers

Furthermore, we are aiming at highly efficient and comprehensive measurement of spin distribution as the final goal. *Omnidirectional photoelectron acceptance lens* (OPAL)[2] together with *Projector for electron spectroscopy with collimator analyzer* (PESCATORA)[3] enables photoelectron holography measurement of the full hemisphere. Moreover, we invented *Right angle deflection imaging analyzer* (RADIAN)[4] for spin vector analysis with ***k***/***r***-space resolution. We are expanding the MM system based on our original device developments.

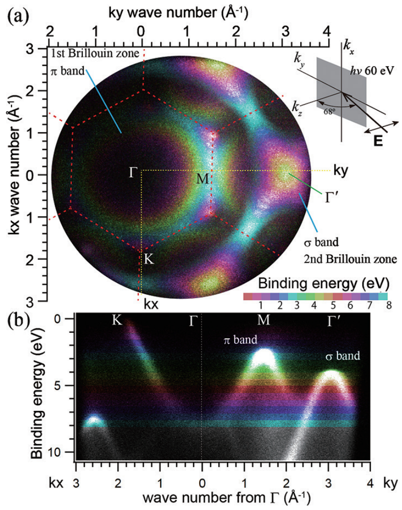

Figure 2. (a) Photoelectron angular distribution and (b) band dispersion of the cleaved graphite crystal surface.

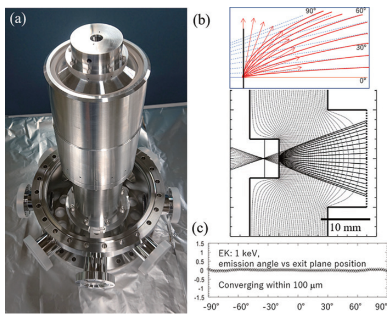

Figure 3. (a) Photograph of the omnidirectional photoelectron acceptance lens (OPAL). (b) 1-keV electron trajectory at the sample surface. (c) Performance of electrons emitted in full hemisphere.[2]

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4) H. Matsuda and F. Matsui, Patent submitted (2020.07.09)

Local Structural Analyses of Liquids by Soft X-Ray Absorption Spectroscopy

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

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Assistant Professor

Soft X-ray absorption spectroscopy (XAS) is an element specific method to reveal local structures of liquids with the K-edges of light elements (C, N, and O). We have investigated local structures of several liquids by using a liquid flow cell for XAS in transmission mode.[1] In this year, we have developed two techniques for applying XAS to the low energy region below 200 eV including K-edges of Li and B and L-edges of Si, P, S, and Cl, as described below.

1. Development of Soft X-Ray Transmission Argon Gas Window

XAS in the low energy region is difficult since transmitted soft X-rays mostly consist of high order X-rays due to the low transmission of first order X-rays. We have proposed the soft X-ray transmission argon gas window that removes high order X-rays by the absorption of Ar L-edges (240 eV).[2]

2. Development of Photoelectron Based Soft X-Ray Detector

We have developed a photoelectron based soft X-ray (PBSX) detector that removes high order X-rays.[3] In this detector, the Au 4f photoelectrons emitted by first order X-rays are separated from those by the high order X-rays using a difference in kinetic energy of photoelectrons.

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Award

NAGASAKA, Masanari; YUZAWA, Hayato; KOSUGI, Nobuhiro; Analytical Sciences Hot Article Award (2020).

Development of a Surface-Sensitive Detection Method for Scanning Transmission X-Ray Microscopy

UVSOR Synchrotron Facility
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Assistant Professor

A scanning transmission X-ray microscope (STXM) obtains 2-dimensional X-ray absorption of a sample to obtain chemical status. Generally, the STXM gives us integrated (bulk) information of the sample along a path of the X-ray. Alternative approach of the STXM is to detect secondary electron from the sample by using a channeltron. The detection of the secondary electron enables us to obtain information of near surface of the sample because of escape depth of the secondary electrons around a few nm. Additionally, this technique overcomes a main difficulty of the STXM by measuring a thick sample which cannot be penetrated by the X-rays.

A channeltron is used as a detector for the secondary electron and is placed upstream of the sample. This optical system can obtain an X-ray transmission image and a secondary electron image simultaneously. As a test sample, a thin section of blended polymer on a copper grid was measured. The energy of the X-ray was 400 eV and the dwell was 500 ms per pixel. In Figure 1(b), an empty space indicated by a dotted circle also shows higher signals of the secondary electron than those from the blended polymer. This problem is under discussion.

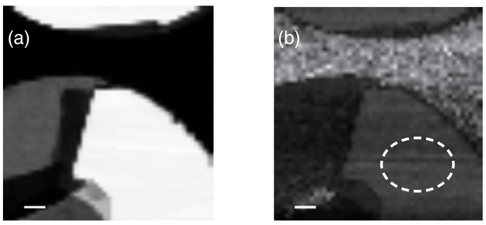

Figure 1. (a) An X-ray transmission image and (b) a secondary electron image of a thin section of blended polymer on a copper grid. Scale bars are 5 μm.

Resonant Soft X-Ray Scattering Measurements for Liquid-Crystal Materials

UVSOR Synchrotron Facility
Division of Advanced Photochemistry

IWAYAMA, Hiroshi
Assistant Professor

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

1. Development of Resonant Soft X-Ray Scattering Measurements

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

In this year, we developed an equipment for resonant soft x-ray scattering measurements. Since soft x-rays are strongly absorbed by air, all soft x-ray paths should be in high vacuum. In addition, a thickness of sample must be less than 1 micrometer to obtain transmitted scattered lights.

We were successful in developing the equipment and now confirmed vacuum test and sample holder test. Our equipment can probe structures in the range from 62 Å to 170 Å. A first sample will be liquid-crystal materials which show a twisted structure. The experiments will be performed at BL3U of UVSOR in September 2020.

Visiting Professors

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

Ionic Liquid/Organic Semiconductor Interfaces for Efficient Carrier Transport

Local analyses of electrolyte/organic semiconductor electrode interfaces at controlled electrode potentials are of fundamental importance to understanding the origin and properties of the electric double layer (EDL) at the interfaces, which is necessary for their application to EDL-organic field effect transistors (OFETs). Ionic liquids (ILs) gated EDL-OFETs can be operated with ultralow voltage (~0.1 V), however, ILs sometimes cause operational instability due to their unusual interface structuring. By using an IL (EMIM-FSA) and ruburene crystal, IL-derived bias stress was observed, which increased operational voltage of the EDL-OFET by 33% in 2 h. Electrochemical FM-AFM and molecular dynamics (MD) simulation revealed that the formation of structured IL layer on the surface of hole-injected rubrene; anions in the IL monolayer probably trapped hole carriers by orienting their polar parts. Application of higher magnitude of OFF-state gate voltage immediately reset the IL-derived bias stress by separating the anion–hole pairs, but the same shift occurred in the same time scale by the local structural change of the interface.

Visiting Professor
SHIMADA, Kenya *(from Hiroshima University)*

High-Resolution Angle-Resolved Photoemission Study of Correlated Materials

In order to understand the physical properties of solids, we are studying the electronic structures by means of high-resolution angle-resolved photoemission spectroscopy (ARPES) using synchrotron radiation. We have also developed an ARPES system using an ultraviolet laser (hν = 6.36 eV) to pursuit ultimate spatial, energy, and angular resolutions (<10μm, <1meV, <0.05°). By combining synchrotron radiation and laser ARPES measurements as well as extensive density functional theory (DFT) calculations, we could elucidate the termination dependent electronic structures of an antiferromagnetic topological insulator $MnBi_4Te_7$. The topological surface state is gapped for quintuple-layer (QL) termination but gapless for the septuple-layer (SL) termination. The spin texture is expected to be different for both terminations. While the DFT calculations reasonably reproduce the s-p electron bands, it is still challenging to predict magnetic 3d electronic bands. In the case of oxygen adsorbed Fe(100) surface states, we found significant deviations from the DFT results due to momentum- and orbital-dependent electron correlation effects.

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

Coherent Control in Condensed Systems

My research is focused on the ultrafast dynamics and coherent control in the condensed systems, especially in strongly coupled systems. We have recently demonstrated the ultrafast visible-pump THz-probe measurement of the carrier dynamics in a thin film of $CH_3NH_3PbBr_3$. As the pump photon energy changes around the bandgap, we can observe the drastic change of the THz transmission intensity. The results are analyzed based on the efficiency of the Auger-like process which is only observable for free carriers, and it is shown that the thermal excitation of the excitons to generate free carriers in the conduction band is not efficient in this material.

We are interested in a vibrational strong-coupling system in which a mid infrared photon is mixed with molecular vibrational quanta and forms a polaritonic quasi-particle. This phenomenon accompanies the local deformation of the potential surface, which can modulate the wave packet motion on the potential. Our final goal is the control of the wave packet motion and the photochemical reaction based on the modulation of the potential surface.