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

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



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

Education

Education 1987 B. E. The University of Tokyo 1992 Ph.D. The University of Tokyo	IMS Research Assistant Professor TOMITA, Takafumi
Professional Employment 1992 Research Associate, Tohoku University 2001 Associate Professor, Tohoku University 2003 Professor, Institute for Molecular Science Professor, The Graduate University for Advanced Studie 2004 Visiting Professor, Tohoku University (-2005) 2007 Visiting Professor, Tokyo Institute of Technology (-2008)	Post-Doctoral Fellow BHARTI, Vineet KUNIMI, Masaya CHAUHAN, Vikas S Visiting Scientist MATSUBARA, Takuya*
 2009 Visiting Professor, The University of Tokyo (-2011) 2012 Visiting Professor (Humboldt Awardee), University of Heidelt 2014 Visiting Professor, University of Strasbourg (-2016) Awards 1998 Award by Research Foundation for Opto-Science 	and MOREA AL TATTA, 30a DENECKER, Tom* KOCIK, Robin* BARRE, Maxence*
Technology 2007 JSPS Prize 2007 Japan Academy Medal 2008 Norman Hascoe Distinguished Lecturer, University of Connecticut, 2009 Fellow of the American Physical Society 2012 Humboldt Research Award 2017 Hiroshi Takuma Memorial Prize of Matsuo Foundation	USA TIRUMALASETTY PANDURANGA With LELA Rene
 2018 Commendation for Science and Technology by the Ministe Education, Culture, Sports, Science and Technology of Ja 2021 Medal with Purple Ribbon (by His Majesty the Emperor Japan) 	er of Secretary Ipan KAWAMOTO, Minako or of TANAKA, Ryo

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

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

ultracold rubidium (Rb) Rydberg atoms, as depicted schematically in Figure 1 and named "ultrafast quantum simulator," envisaging the quantum-classical boundary connected smoothly.

Member Assistant Professor

SUGAWA, Seiji DE LÉSÉLEUC, Sylvain



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

Bismuth," Nat. Commun. 4, 2801 (2013).

- N. Takei et al., "Direct Observation of Ultrafast Many-Body Electron Dynamics in an Ultracold Rydberg Gas," Nat. Commun. 7, 13449 (2016).
- · C. Liu et al., "Attosecond Control of Restoration of Electronic Structure Symmetry," Phys. Rev. Lett. 121, 173201 (2018).
- M. Mizoguchi et al., "Ultrafast Creation of Overlapping Rydberg Electrons in an Atomic BEC and Mott-Insulator Lattice," Phys. Rev. Lett. 124, 253201 (2020).
- Y. Chew et al., "Ultrafast Energy Exchange between Two Single Rydberg Atoms on a Nanosecond Timescale," Nat. Photonics 16, 724 (2022).

1. Development of an "Ultrafast Quantum Simulator" by Optical Control with Precisions on the Attosecond Temporal and Submicron Spatial Scales³⁻¹⁰⁾

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

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



Figure 2. Schematic of the standard hardware of the ultrafast quantum simulator.^{3,4,6,7,10}

We have also completed a readout interface of our ultrafast quantum simulator, which is the time domain Ramsey interferometry of ultracold

Rydberg atoms with attosecond precision, whose contrast is almost 100%.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.

2. Development of an Ultrafast Quantum Computer^{3,10,12)}

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



Figure 3. Examples of the world's smallest arbitrary arrays of optical traps. (a) Square lattice; (b) Kagome Lattice; (c) Hexagonal (Honeycomb) lattice.¹⁰⁾

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 in just 6.5 ns, as depicted schematically in Figure 4.¹²) This is the world's fastest controlled gate, which is the most important two-qubit gate (a fundamental arithmetic element essential for quantum computing). This high-impact result was highlighted on the front cover of the Oct 2022 Issue of Nature Photonics,¹² and by more than 200 news articles worldwide, such as in Japan, US, Europe, China, *etc.*



Figure 4. Conceptual diagram of the world's fastest controlled gate for ultrafast quantum computing. Two single atoms captured in optical tweezers (red light) with a separation of a micrometer are entangled by an ultrafast laser pulse (blue light) shone for only 10 picoseconds.¹²⁾ Image source: Dr. Takafumi Tomita (IMS)

References

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- Patent "Quantum Simulator and Quantum Simulation Method," H. Sakai (Hamamatsu Photonics K.K.), K. Ohmori (IMS) *et al.* (US 2020, JP 2021).
- 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).
- Highlighted in "Quantum-Technology Innovation Strategy" by the Cabinet Office of Japan, January 2020.
- 7) M. Mizoguchi et al., Phys. Rev. Lett. 124, 253201 (2020).
- 8) H. Katsuki et al., Acc. Chem. Res. 51, 1174-1184 (2018).
- 9) N. Takei et al., Nat. Commun. 7, 13449 (2016).
- 10)S. Sugawa et al., Solid State Physics 56, 243 (2021). (Invited Paper/Cover-Page Highlight)
- 11) D. Barredo et al., Science 354, 1021 (2016).
- 12) Y. Chew et al., Nat. Photonics 16, 724 (2022). (Cover-Page Highlight)

Award

OHMORI, Kenji; National Medal with Purple Ribbon by His Majesty the Emperor of Japan (2021).

Electronic Property of Functional Organic Materials

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

KERA, Satoshi Professor [kera@ims.ac.jp]	ducation 996 B.E. Chiba University 998 M.E. Chiba University 909 Ph.D. Chiba University 909 JSPS Research Fellow 901 Research Associate, Chiba University 903 Research Associate, Institute for Molecular Science 903 Postdoctoral Fellow, Wuerzburg University 904 Assistant Professor, Chiba University 905 Associate Professor, Chiba University 906 Visiting Associate Professor, Institute for Molecular Science 903 Visiting Associate Professor, Soochow University 904 Visiting Associate Professor, Soochow University 905 Visiting Professor, Chiba University for Advanced Studies 903 Visiting Professor, Chiba University 904 Visiting Professor, Chiba University for Advanced Studies 905 Visiting Professor, Chiba University 904 Professor, The Graduate University for Advanced Studies 905 Visiting Professor, Kyoto University, Hiroshima University 907 Visiting Professor, Tohoku University	Graduate Student NISHINO, Fumi PALASSERY ITHIKKAL, Jaseela KANAZAWA, Shingo* MIYATA, Takeshi [†] YAMAUCHI, Saki [‡] SEMA, Ryota [‡] Secretary HAGIWARA, Hisayo KAMIYA, Miho
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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 organicorganic 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

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]

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 electronphonon coupling is a central issue on our agenda.

Member Assistant Professor

FUKUTANI, Keisuke



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.

- 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. Photoemission Tomography of One Dimensional Row Structure of Flat-Lying Picene Multilayer on Ag(110)¹⁾

Photoemission tomography (PT) is a powerful technique for the detailed analysis of the shape and the energy of molecular orbitals from the photoelectron momentum distribution. In the case of planar-conjugated molecules with light atoms, the momentum map of the photoelectrons from frontier orbitals can be seen as the Fourier transform (FT) of the corresponding molecular orbital, by assuming the plane-wave final state in the photoemission process. Thus, PT is able to determine the shape of the molecular orbitals in the sample if the molecular arrangement in the sample is known, while it can also be utilized for the determination of the molecular arrangement in the specimen if the molecular orbital of the sample molecule is known.

We applied PT to a unique one-dimensional row structure of a picene multilayer realized on an anisotropic Ag(110) surface. The clearly deconvoluted experimental momentum maps were compared to the FT simulation of the molecular orbitals of picene in detail, enabling not only the evaluation of the electronic structure of the picene in the multilayer but also the quantitative determination of the molecular orientation in the multilayer within a few degrees. In addition, the PT results indicated the orientation of the molecules in all layers to be flat-lying. The successful demonstration of PT of the multilayer molecular film marks an important step toward the widerange utilization of the PT technique.



Figure 2. Scheme of photoelectron spectrum of the picene film, STM molecular arrangement, and PT images for top three molecular orbitals. The figure is after ref 1).

2. Sample-Shape Dependent Energy Levels in Organic Semiconductors²⁾

Most of the tuning of the ionization energy and electron affinity was done by changing the constituent molecules in the device. This is because the organic solids consist of organic molecules bound by weak vdW interactions, hence intermolecular interaction is considered to play a minor role. The contribution of the intermolecular interaction to the energy levels of organic solids is represented by the difference in the energy levels between the gas and solid phases.

Recent studies, however, have revealed that the energy levels can be altered as much as 1 eV by the molecular orientation in the film or the molecular mixing ratio in the binary film, owing to the intermolecular electrostatic interaction. Because of the long-range nature of Coulomb interaction, theory predicts that the electrostatic energy should depend on the sample shape. In this study, we examined the coverage-dependent energy levels of zinc phthalocyanine (ZnPc) and per-fluorinated ZnPc (F₁₆ZnPc) in the monolayer region with ultraviolet photoelectron spectroscopy and lowenergy inverse photoelectron spectroscopy. Using the procedure we reported previously, we separately evaluated the electronic polarization energy and electrostatic energy as a function of coverage. Unlike the electronic polarization, which contributes only as much as 10 meV, the electrostatic energy contributes as much as 120 meV to the coverage-dependent energy shift. We concluded that the shift in energy levels by changing the coverage is attributed to the sample shapedependent energy level, owing to the long-range nature of the charge-permanent quadrupole interaction.



Figure 3. (a) Disk model for the growing island without a changing lattice constant. (b) Clculated polarization energy and electrostatic energy for ZnPc and F_{16} ZnPc are indicated with blue circles and red squares. The figure is after ref 2).

3. Other Activities in UVSOR

We have conducted beamline R&D and user supports in collaboration with other universities. Experiments using photoelectron momentum microscope are developing at BL6U.^{3,4)}

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- 2) S. A. Abd-Rahman, T. Yamaguchi, S. Kera and H. Yoshida, *Phys. Rev. B* 106, 075303 (8 pages) (2022).
- Y. Hasegawa, F. Matsui and S. Kera, *e-J. Surf. Sci. Nanotechnol.* 20, 174–179 (2022).
- 4) F. Matsui, Y. Okano, H. Matsuda, T. Yano, E. Nakamura, S. Kera and S. Suga, *J. Phys. Soc. Jpn.* **91**, 094703 (6 pages) (2022).

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Light Source Developments by Using Relativistic Electron Beams

UVSOR Synchrotron Facility Division of Advanced Accelerator Research



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Education

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

Professional Employment

- 1986 Reseach Associate, National Laboratory for High Energy Physics
- 2000 Associate Professor, Institute for Molecular Science 2004 Professor, Institute for Molecular Science
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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–Eelectron Beam Interaction," *Nat. Phys.* 4, 390–393 (2008).
- M. Shimada, M. Katoh, M. Adachi, T. Tanikawa, S. Kimura, M. Hosaka, N. Yamamoto, Y. Takashima and T. Takahashi, "Transverse-Longitudinal Coupling Effect in Laser Bunch Slicing," *Phys. Rev. Lett.* 103, 144802 (2009).
- M. Katoh, M. Fujimoto, H. Kawaguchi, K. Tsuchiya, K. Ohmi, T. Kaneyasu, Y. Taira, M. Hosaka, A. Mochihashi and Y. Takashima, "Angular Momentum of Twisted Radiation from an Electron in Spiral Motion," *Phys. Rev. Lett.* **118**, 094801 (2017).
- S. Matsuba, K. Kawase, A. Miyamoto, S. Sasaki, M. Fujimoto, T.



Member Assistant Professor

FUJIMOTO, Masaki*

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

Konomi, N. Yamamoto, M. Hosaka and M. Katoh, "Generation of Vector Beam with Tandem Helical Undulators," *Appl. Phys. Lett.* **113**, 021106 (2018).

- Y. Hikosaka, T. Kaneyasu, M. Fujimoto, H. Iwayama and M. Katoh, "Coherent Control in the Extreme Ultraviolet and Attosecond Regime by Synchrotron Radiation," *Nat. Commun.* 10, 4988 (2019).
- T. Kaneyasu, Y. Hikosaka, M. Fujimoto, H. Iwayama and M. Katoh, "Electron Wave Packet Interference in Atomic Inner-Shell Excitation," *Phys. Rev. Lett.* 126, 1132202 (2021).
- T. Kaneyasu, M. Hosaka, A. Mano, Y.Takashima, M. Fujimoto, E. Salehi, H. Iwayama, Y. Hikosaka and M. Katoh, "Double-Pulsed Wave Packets in Spontaneous Radiation from a Tandem Undulator," *Sci. Rep.* 12, 9682 (2022).

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 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.

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 quasiconstant at a high beam current, 300mA, and the novel beam injection scheme with a pulsed sextupole magnet. As the result of all these efforts, now, the machine is one the brightest synchrotron light sources among the low energy machines below 1GeV in the world.

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

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



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



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

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

UVSOR Synchrotron Facility Division of Beam Physics and Diagnostics Research

FairsArrow<	 Education 2007 B.S. Nagoya University 2009 M.S. Nagoya University 2012 Ph.D. Nagoya University Professional Employment 2011 JSPS Research Fellow 2012 Research Scientist, National Institute of Advanced Industrial Science and Technology (AIST) 2018 Senior Research Scientist, National Institute of Advanced Industrial Science and Technology (AIST) 2020 Associate Professor, Institute for Molecular Science Associate Professor, The Graduate University for Advanced Studies Awards 2010 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 Researcher Best Presentation Award, Beam Physics Workshop 2015 2021 Outstanding Presentation Award, 64th Annual Meeting of the Japanese Society of Radiation Chemistry
Keywords F	ron Beams, Synchrotron Radiation, Gamma-Rays

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

Inverse Thomson (Compton) scattering is a method to generate a high energy gamma-ray by the interaction between a high energy electron and a laser. We have developed ultrashort 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 induced positron annihilation pectroscopy (GiPAS). A positron

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).

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.

Member Assistant Professor

SUGITA, Kento



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

- Y. Taira, T. Hayakawa and M. Katoh, "Gamma-Ray Vortices from Nonlinear Inverse Thomson Scattering of Circularly Polarized Light," *Sci. Rep.* **7**, 5018 (2017).
- Y. Taira and M. Katoh, "Gamma-Ray Vortices Emitted from Nonlinear Inverse Thomson Scattering of a Two-Wavelength Laser Beam," *Phys. Rev. A* **98**, 052130 (2018).
- Y. Taira, M. Fujimoto, S. Ri, M. Hosaka and M. Katoh, "Measurement of the Phase Structure of Elliptically Polarized Undulator Radiation," *New J. Phys.* 22, 093061 (2020).

1. Gamma 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 BaF_2 detector, and calculate the time difference distribution. One digital oscilloscope for four BaF_2 detectors is used as a pair of detection systems. The annihilation gamma-rays are generated to whole solid angle. Therefore array detectors are effective to increase the count rate of the annihilation gamma-rays and to reduce the measurement time. A detection system with eight detectors and two digital oscilloscopes was constructed (Figure 2). Time resolution is 140 ps in full width at half maximum, which is high despite the use of a 52-mm thick BaF_2 scintillator. The count rate is 20 cps.

Users can currently utilize GiPALS at BL1U in UVSOR-III. A result of defect analysis for a $Lu_3Al_5O_{12}$ scintillator was published in 2022.¹⁾



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

Positron age-momentum correlation (AMOC) is an approach for measuring the time resolved momentum distribution of an electron, which provides different information about defects compared to positron annihilation lifetime. Gamma ray-induced AMOC has been developed by using a BaF_2 detector, a germanium detector, and a 12-bit digital

oscilloscope. Typically, a multichannel analyzer is used to measure the energy spectrum of gamma rays. However, a 12-bit digital oscilloscope is employed in this experiment; thus, high-energy resolution can be obtained with the digital oscilloscope alone. The results of multiple sample measurements are summarized in the paper.

We are planning to develop other measurement technique for the annihilation gamma-rays, such as coincidence Doppler broadening and spin polarized positrons generated from circularly polarized gamma-rays.

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.

UVSOR-III also has a laser with a pulse energy of 50 mJ and has completed start-up work on the laser. Experiments on nonlinear inverse Thomson scattering will be performed after October 2022 to measure the spatial distribution of high harmonic gamma rays.

Reference

1) M. Kitaura, Y. Taira and S. Watanabe, *Opt. Mater.: X* 14, 100156 (2022).

Award

SALEHI, Elham; 4th International School on Beam Dynamics and Accelerator Tehnology, ISBA Gold award (2022).

Application of X-Ray Microscopy

UVSOR Synchrotron Facility Division of Beam Physics and Diagnostics Research



OHIGASHI, Takuji Associate Professor (Cross Appointment) [ohigashi@ims.ac.jp]

Education

- 1998 B.E. University of Tsukuba 2003 Ph.D. (Engineering) University of Tsukuba
- Professional Employment
- 2003 Lecturer, University of Tsukuba
- 2004 Research Fellow, Tohoku University
- 2005 Research Fellow, Japan Synchrotron Radiation Research Institute (JASRI)
- 2008 Postdoctral Fellow, Ritsumeikan University
- 2011 Assistant Professor, Institute for Molecular Science
- 2022 Associate Professor, KEK
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Keywords

X-Ray Microscopy, In-Situ Technique, X-Ray Absorption Spectroscopy

A synchrotron-based scanning transmission X-ray microscope (STXM) is a technique to perform 2-dimensional (2-D) X-ray absorption spectroscopy with high spatial resolution up to 30 nm. By noticing the X-ray absorption edge of the specific element, 2-D chemical state of a sample can be obtained. Since characteristics of UVSOR is suitable for using extreme ultra-violet and soft X-ray region, the STXM in UVSOR is suitable to analyze soft materials and organic materials. The unique features 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. Especially, nowadays, an in-situ/operando analysis is attracting more attentions of researchers because that is an important technique to understand intrinsic state of the samples. For example, heating and cooling of the sample, humidity control system and electrochemistry, 2-D orientation of molecules, 3-D 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 filed of science. These techniques are difficult to perform by using the other microscopic techniques.

Member Secretary

ISHIHARA, Mayumi KAMO, Kyoko



Figure 1. Schematic optical system of STXM.

Selected Publications

- T. Ohigashi, M. Nagasaka, T. Horigome, N. Kosugi, S. M. Rosendahl and A. P. Hitchcock, "Development of In-Situ Sample Cells for Scanning Tranmission X-Ray Microscopy," *AIP Conf. Proc.* 1741, 050002 (2016).
- T. Ohigashi, A. Ito, K. Shinohara, S. Tone, Y. Inagaki, H. Yuzawa and N. Kosugi, "3-Dimensional Chemical Structures of an Isolated Cell Nucleus by a Scanning Transmission X-Ray Microscope," *Microsc. Microanal.* 24, pp. 400–401 (2018).
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- J.-J. V. Véles, Y.-Y. Chin, M.-H. Tsai, O. J. Burton, R. Wang, S. Hofmann, W.-H. Hsu, T. Ohigashi, W.-F. Pong and C.-H. Chuang, "Evidence of Synergistic Elecrocatalysis at a Cobalt Oxide-Graphene Interface through Nanochemical Mapping of Scanning Transmission X-Ray Microscopy," *Chin. J. Phys.* 76, 135–144 (2022).

1. Analysis of Organic Materials in Returned Samples from the Asteroid *Ryugu* by STXM

In 2014, a small spacecraft Hayabusa2 was launched to an asteroid 1999 JU3, named Ryugu, 280,000,000 km away from the earth. Hayabusa2 accumulates 5.4 g of pebbles and sands from surface (stored in a chamber A) and subsurface (in a chamber C) of Ryugu and the sample chambers came back to the earth in the end of 2020.1) Compared to ordinally meteorites, those samples from Ryugu are expected to retain pristine information of the asteroid since they did not suffer from heating by the atmosphere and contamination by terrestrial materials. Moreover, the samples in the chamber C are likely to have less space weathering than those of the chamber A. Therefore, the samples in the chamber C could be one of the standards of nature of extraterrestrial material. Our team, Phase2 curation Kochi team (collaborative team among JAXA, JAMSTEC, NIPR, SPring-8 and IMS), has developed "a linkage analytical system" to analyze the sample by using various apparatus, such as synchrotron-based X-ray diffraction (XRD), computed tomography (CT), focused-ion-beam (FIB) process, transmission electron microscopy (TEM), NanoSIMS and STXM, without exposing to the air through whole process and a protocol for analysis with minimum destructive process and damages.²⁾ Ryugu is primarily considered as a carbonaceous asteroid so that abundant of organic material is expected. Main concerns of Phase2 Kochi team are water and organic materials. Therefore, the STXM at BL4U in UVSOR is a promising tool to analyze localized organic materials with high spatial resolution around 30 nm. In BL4U, a sample transfer system, a special sample cell, an FIB grid (namely Kochi grid), and a container to transfer between facilities, which enable to transfer the sample from a glovebox to a main chamber of the STXM without exposing to air, have been developed for the analysis of the Ryugu samples.

First of all, 3-dimensional structure and crystallography of a grain C0068 in the chamber C were confirmed by CT and XRD in SPring-8 to determine a region of interest (ROI) without any destructive process. The ROI was roughly cut out by using a diamond saw and was finished as an ultra-thin section sample by using FIB process in JAMSTEC as dimension of $25 \times 25 \times 0.1$ (thick) μ m³ on the Kochi-grid.²) That ultrathin section sample is used for high spatial resolution analyses by TEM, NanoSIMS and STXM. The procedures including the sample preparation, measurement and transportation between research institutes were performed under grade 1 nitrogen gas condition.

Figure 2(a) shows representative NEXAFS spectra of

aromatic (C=C)-rich (plotted in red), C-H bonding-rich regions (green), matrix (blue) of the sample C0068,25 and Murchison meteorite (gray) around C K-edge. From this comparison, the peak at 287.5 eV, assigned as C-H bonding including aliphatic, is remarkable feature of the Ryugu samples. Their distributions are depicted in Figure 2(c) as RGB-color composite mapping. In regard to the distributions, the aromatic-rich region (shown in red) is less than the other components. Figure 2(b) shows spatial distribution of carbon-related materials. Then, the dark area has no (or much less) carbon-related component. Detailed analyses of dashed line squares in Figures 2(b) and 2(c) were performed by using TEM (shown in Figures 2(d) and 2(e)). The corresponding dark spot areas are confirmed as pyrrhotite and pentlandite. A large nanoglobule is consisted of amorphous silicate as a core and aromatic-rich organic materials around that.

Further analysis of the Ryugu samples is in progress. As one of the topics, we notice a potential relationship between space weathering and evolution of organic materials.



Figure 2.³⁾ (a) NEXAFS spectra around C K-edge normalized to 292 eV of aromatic (C=C)-rich regions (red), C–H bonding-rich regions (green) and matrix (blue). The grey line is a spectrum of Murchison insoluble organic matter. (b) An STXM image dominated by carbon. (c) An RGB mapping with aromatic-rich (C=C) areas (red), C–H bonding-rich areas (green) and matrix (blue). (d) C–H bonding-rich organics concentrated in coarse-grained phyllosilicates in an area enlarged from the white dashed boxes in (b) and (c). (e) A large nanoglobule extracted from the dashed white boxes in (b) and (c) where Po and Pn are pyrrhotite and pentlandite, respectively.

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Establishing Advanced Photoemission Methodologies for Novel Spin Materials Science

UVSOR Synchrotron Facility Division of Advanced Solid State Physics



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Education

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

Professional Employment

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

Awards

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

Keywords

Photoelectron Spectroscopy, Momentum Microscope, Electronic Spin Structure

Electrons in material are excited by photons and emitted into the vacuum as photoelectrons. Interestingly, the angular distribution of these photoelectrons reveals 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 can 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 μ m-sized area. The combination of domain-resolved photoelectron microscopy and μ m-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

Selected Publications

- F. Matsui, Y. Okano, H. Matsuda, T. Yano, E. Nakamura, S. Kera and S. Suga, "Domain-Resolved Photoelectron Microscopy and µm-Scale Momentum-Resolved Photoelectron Spectroscopy of Graphite Armchair Edge Facet," *J. Phys. Soc. Jpn.* **91**, 094703 (2022).
- F. Matsui and S. Suga, "Coupling of k_z-Dispersing π Band with Surface Localized States in Graphite," *Phys. Rev. B* 105, 23526 (2022).
- F. Matsui, 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).

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

Member Secretary

> ISHIHARA, Mayumi KAMO, Kyoko



Figure 1. Photoelectron momentum microscope at BL6U of UVSOR synchrotron facility together with valence photoelectron holograms and dispersions of TaS_2 and Bi_2Se_3 . Charge density wave phase transition and topological nature can be directly studies in detail.

- F. Matsui and H. Matsuda, "Projection-Type Electron Spectroscopy Collimator Analyzer for Charged Particles and X-Ray Detections," *Rev. Sci. Instrum.* **92**, 073301 (2021).
- F. Matsui, 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).
- F. Matsui, H. Nishikawa, H. Daimon, M. Muntwiler, M. Takizawa, H. Namba, and T. Greber, "The 4π k_z Periodicity in Photoemission from Graphite," *Phys. Rev. B* 97, 045430 (2018).

1. Single Graphite Step Visualized

Graphite is an incredibly important, versatile mineral, with uses spanning industries. Graphite is an essential component of many batteries, including lithium-ion batteries, and demand is only increasing as new technology is developed. Even though graphite has been thoroughly researched for decades, there is still more to be uncovered. Surprisingly, no photoelectron spectroscopic studies have so far accurately measured the electronic states of the surface and the edge of graphite from a microscopic point of view. It has been "common knowledge" that the electronic structure of graphite is six-fold symmetric, but local observations using microscopy capabilities¹⁾ have revealed the existence of two three-fold symmetric domains that are mirror symmetric by the termination of the alternating stacking structure at the topmost surface (Figure 2).²⁾ Whereas conventional measurements look at the sum of both, photoelectron momentum microscopy reveals a step-edge structure at the boundary of two terraces of monoatomic layers of graphite with mirror symmetry with respect to each other.



Figure 2. (a) Atomic structure of the cleaved graphite surface. (b) Iso-energy cross section of the graphite surface π band. (c) Graphite single atomic layer step was imaged with different contrast owing to the difference in the π band binding energies in two different terraces as shown in (d).²⁾

2. Embed Valence Band in Auger Electrons

Resonant photoelectron spectroscopy highlights certain

elemental components of the valence band by adjusting the photon energy to a core-level excitation threshold. However, most of the investigations to date has focused on angleintegrated spectral analysis for elucidating the element-specific density of states. Thus, we explored the condition for the transition of the valence band dispersion information to the Auger electrons by momentum-resolved measurements with a wide-range and high-resolution and realized a new photoelectron spectroscopy with the specificity of elemental and atomic orbitals in band structure analysis.

We performed momentum-resolved resonant photoelectron spectroscopy measurements of graphite crystals using soft X-ray. We identified four different types of resonant pathways at the C K-shell absorption threshold (Figure 3).³⁾ Fano-resonance-like behavior was confirmed for photoelectron emission from the π band dispersion. The π band dispersion disappeared just below the absorption threshold, and was strongly enhanced at the π^* absorption resonance peak photon energy. In addition, two types of resonant Auger electron emission involving the Dirac cone shake-up process were observed. Furthermore, we discovered a peculiar dispersion structure embedded in the normal Auger electron energy region. This phenomenon has also been confirmed with monolayer graphene and adsorbed aromatic molecular species.⁴) This resonant valence excitation technique provides a versatile means for characterizing valence band and molecular orbital with element specification.



Figure 3. Momentum-resolved valence photoelectron and Auger electron spectra along the direction excited at the photon energy of (a) 284.01 eV and (b) 285.40 eV. Black and red dotted lines indicate the valence band and Auger electron dispersions, respectively. (c) Isoenergy momentum-resolved Auger electron intensity distribution at the kinetic energy of 266.5 eV. (d) Schematic of resonant Augerelectron emission for the pathway S3.³⁾

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- F. Matsui, S. Makita, H. Matsuda, E. Nakamura, Y. Okano, T. Yano, S. Kera and S. Suga, *J. Phys. Soc. Jpn.* **90**, 124710 (2021).
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Angle-Resolved Photoemission Study on Strongly Correlated Electron Materials

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Education

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- 2005 Ph.D. The University of Tokyo

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Keywords

Strongly Correlated Electron System, Synchrotron Light, Photoemission

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

Selected Publications

- K. Tanaka *et al.*, "Distinct Fermi-Momentum-Dependent Energy Gaps in Deeply Underdoped Bi2212," *Science* **314**, 1910–1913 (2006).
- K. Tanaka *et al.*, "Quantitative Comparison between Electronic Raman Scattering and Angle-Resolved Photoemission Spectra in Bi₂Sr₂CaCu₂O_{8+δ} Superconductors: Doping Dependence of Nodal and Antinodal Superconducting Gaps," *J. Phys. Soc. Jpn.* 88, 044710 (2019).

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. Spinresolved ARPES is one of the most powerful experimental techniques to investigate the magnetic properties of such materials.

Member Graduate Student

> HOSHINA, Takumi* SUGIMOTO, Takafumi*



- S. Ideta, K. Tanaka *et al.*, "Experimental Investigation of the Suppressed Superconducting Gap and Double-Resonance Mode in Ba_{1-x}K_xFe₂As₂," *Phys. Rev. B* **100**, 235135 (7 pages) (2019).
- S. Ideta, K. Tanaka *et al.*, "Hybridization of Bogoliubov Quasiparticles between Adjacent CuO₂ Layers in the Triple-Layer Cuprate Bi₂Sr₂Ca₂Cu₃O_{10+δ} Studied by Angle-Resolved Photoemission Spectroscopy," *Phys. Rev. Lett.* **127**, 217004 (6 pages) (2021).

1. Hybridization of Bogoliubov Quasiparticles between Adjacent CuO₂ Layers in the Triple-Layer Cuprate $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}{}^{1)}$

It has been 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 CuO₂ planes (*n*). T_c generally increases from single-layer (n =1), double-layer (n = 2), to triple-layer (n = 3) and then decreases for $n \ge 4$. Although several mechanisms have been proposed to explain the *n* dependence of T_c , it is still not clear because of the lack of detailed knowledge about the electronic structure of the multi-layer ($n \ge 3$) cuprates. In this study, we performed ARPES of optimally doped triple-layer Bi₂Sr₂Ca₂ Cu₃O_{10+ δ} (Bi2223, $T_c = 110$ K) in the superconducting states at UVSOR BL7U.¹)

Figures 1(a)-(h) show hybridization of Bogoliubov quasiparticles (BQPs) between the outer CuO2 plane (OP) and the inner CuO2 plane (IP). The OP and IP are hybridized with each other and open a gap at the crossing point (an anticrossing gap between the two BQP bands) as shown in Figure 1(i). We noticed that the magnitude of the gap $(\Delta_{t\perp}(k))$ gradually increases as one goes away from the node toward the antinode and the momentum dependence of $\Delta_{t\perp}(k)$ can be reproduced by assuming the interlayer single-particle hopping parameter $t \perp \sim 56$ meV using the tight-binding model for coupled CuO₂ planes. In order to reproduce the high energy kinks for the OP and IP bands of the measured ARPES spectra, we have performed model calculation including the coupling to several oxygen-derived phonons. Taking into account the contribution of acoustic phonons, the flatness of the top of the IP band seen in the experiment is reproduced to some extent.

The most intriguing and important question relevant to the present study is how much the interlayer single-particle hopping and the obtained $\Delta_{t\perp}(k)$ contribute to the enhancement of T_c in Bi2223. In order to discuss a possible mechanism for the T_c enhancement in triple-layer cuprates, a simple four-well

model calculation is performed assuming contributions from four bosonic modes, namely, the acoustic phonon, the *c*-axis buckling phonon, the in-plane breathing phonon, and spin fluctuations. The present model calculation suggests that the electron–phonon coupling alone may not be sufficient to cause the observed T_c in the triple-layer cuprate, and the combination of spin fluctuations and phonons is essential (not shown).

As another candidate of the anticrossing gap, the effect of interlayer Cooper-pair hopping could not be isolated from the present data, but might be important to increase the T_c of Bi2223. To answer the question of how interlayer interactions, *i.e.*, single-particle hopping versus Cooper-pair hopping, contribute to the enhancement of superconductivity, further systematic studies are needed both experimentally and theoretically. The momentum-dependent hybridization gap, which is proportional to the square of the SC order parameter, would be a key piece of information to solve the T_c -enhancement mechanism of the multilayer cuprates.

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

Our group is developing a new high-efficient spin-resolved ARPES system with multi-channel detection (we call "imagespin" detection) in beamline BL5U at UVSOR. We successfully obtained spin-resolved signal of Au(111) surface and achieved 100 times better efficiency and several times better momentum resolution than the current synchrotron-based spinresolved ARPES systems in the world. In 2021, we installed a spin-manipulator lens system, where the direction of the spin can be changed to any directions. The calibration of the lens parameters will be done in 2022.

Reference

1) S. Ideta et al., Phys. Rev. Lett. 127, 217004 (6 pages) (2021).



Figure 1. ARPES spectra of the OP and IP bands in Bi2223. (a)–(d): *E-k* plots near $E_{\rm F.}$ (e)–(h): Energy-distribution curves (EDCs) corresponding to (a)–(d). (i): EDCs at momenta where the OP band and the IP band cross extracted from panels (e)–(h). (j): Momentum dependence of $\Delta_{t\perp}(k)$ plotted against the *d*-wave order parameter. (k): Schematic illustration of the hybridization between the OP and IP BQP bands in the off-nodal region.

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

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



NAGASAKA, Masanari Assistant Professor

Soft X-ray absorption spectroscopy (XAS) observes local structures of liquids with different light elements. We have developed liquid cells and devices with precise absorbance control and observed several chemical processes in solution by using *operando* XAS.^{1,2)} In this year, we have investigated the process of hydrophobic cluster formation in aqueous ethanol

solutions by using XAS.³⁾

1. Hydrophobic Cluster Formation in Aqueous Ethanol Solutions

Hydrophobic cluster structures in aqueous ethanol solutions at different concentrations have been investigated by XAS.³⁾ In the O K-edge XAS, we have found that hydrogen bond structures among water molecules are enhanced in the middle concentration region by the hydrophobic interaction of the ethyl groups in ethanol. On the other hand, in the C K-edge XAS, the lower energy features arise from a transition from the terminal methyl C 1s electron to an unoccupied orbital of 3s Rydberg character and show characteristic four concentration regions. From the comparison of C K-edge XAS with the inner-shell calculations, we have revealed the intermolecular interactions of ethanol with water at different concentration regions and found that ethanol clusters are easily formed in the middle concentration region due to the hydrophobic interaction of the ethyl group in ethanol, resulting in the enhancement of the hydrogen bond structures among water molecules.

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Twisted Structure Analysis of Soft Matters by Resonant Soft X-Ray Scattering

UVSOR Synchrotron Facility Division of Advanced Photochemistry



IWAYAMA, Hiroshi Assistant Professor

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. We developed a new resonant soft x-ray scattering method which has various kind of selectivities such as elements, groups and molecular alignments.¹⁾

1. Twisted Structure of Helical-Nanofilament

Twisted structure can be often observed in soft matter. However, since electron density modulations resulting from the twisted structures are weak, conventional small angle x-ray scattering method is difficult to observe it. The resonant soft x-ray scattering measurements can probe twisted structures and obtain pitch of twisted structure. This is because a resonant process strongly depends on an angle between molecule and polarization vector of incident soft x-ray. In this year, we performed resonant soft x-ray scattering experiments at UVSOR BL3U for a helical-nanofilament of liquid-crystal matter.

We successfully obtained diffraction images at the wavelength of 4.34 nm (285 eV), which correspond to C 1s-to- π^* core excitations. From the analysis of images, we found a resonant enhancement of diffractions corresponding to a twist pitch of 80 nm. We also found that twist pitch depends on the sample compositions and its temperatures.

Reference

1) B. A. Collins and E. Gann, J. Polym. Sci. 60, 1199 (2022).

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 (RT) by forming an active surface when heated in ultra-high vacuum (UHV). When NEG is deposited on the inner wall of a vacuum vessel, the vacuum vessel will evacuate the residual gases just by baking, and UHV can be maintained without electric power for long time. Therefore, the development of NEG will

contribute to CO_2 emission reduction and Sustainable Development Goals (SDGs). However, the activation temperature of NEG (the temperature required to create an active surface by heating in UHV) needs to be lowered if NEG is to be widely used in various vacuum-related industries. Recently we have developed a new NEG, Pd overcoated on Ti thin film with a purity higher than 99.95% (oxygen-free Pd/Ti hereafter), which evacuates H₂ and CO at RT after baking at 133 °C for 12 hours. NEG pumps using oxygen-free Pd/Ti deposition has been commercialized in 2019 and are widely used in synchrotron radiation facilities such as SPring-8, UVSOR, and Photon Factory.



Visiting Associate Professor FUKUHARA, Takeshi (from RIKEN)

Single-Atom-Resolved Imaging of Quantum Gases in 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 various fundamental models in solidstate physics. Key technologies for this research are the detection and manipulation of such gases at the single-atom level. We have prepared quantum gases in triangular optical lattices for the study of frustration

physics and successfully realized single-atom-resolved detection using fluorescence imaging. Raman sideband cooling has been utilized for the detection because the sample of ultracold atoms is heated and destroyed due to photon scattering. Several parameters, such as intensities and frequencies of the cooling lasers, are required to be tuned for successful imaging. We automatically adjusted the parameters using Bayesian optimization, which is a machine learning method. Now we are improving the scheme for the optimization method to enhance the imaging fidelity.



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

Epitaxially-Grown Single-Crystalline Organic Molecular Semiconductors

While epitaxial growth of single-crystalline (inorganic) semiconductor materials is one of the most essential technologies for modern electronic applications, current organic semiconductor electronics are mostly built on heterojunctions composed of polycrystalline or amorphous molecular solids. On the other hand, 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. Our group has been working on single-crystalline heterojunctions of organic molecular semiconductors by epitaxial growth techniques. Recently, we discovered as a collaborative work with IMS groups that a methyl- and trifluoromethyl-substituted derivative of rubrene forms high-quality single-crystalline junctions on the single-crystal surface of (unsubstituted) rubrene in a "quasi-homoepitaxial" manner. Electronic band measurements on this quasi-homoepitaxial molecular junction by means of angle-resolved photoelectron spectroscopy are one of our next targets.