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

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

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

Education
1987 B. E. The University of Tokyo
1992 Ph.D. The University of Tokyo

Professional Employment
1992 Research Associate, Tohoku University
2001 Associate Professor, Tohoku University
2003 Professor, Institute for Molecular Science
Professor, The Graduate University for Advanced Studies
2004 Visiting Professor, Tohoku University (–2005)
2007 Visiting Professor, Tokyo Institute of Technology (–2008)
2009 Visiting Professor, The University of Tokyo (–2011)
2012 Visiting Professor, University of Heidelberg
2014 Visiting Professor, University of Strasbourg (–2016)

Awards
1998 Award by Research Foundation for Opto-Science and Technology
2007 JSPS Prize
2007 Japan Academy Medal
2009 Fellow of the American Physical Society
2012 Humboldt Research Award
2017 Hiroshi Takuma Memorial Prize of Matsuo Foundation
2018 Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology of Japan

Member
Assistant Professor
SUGAWA, Seiji
DE LÉSÉLEUC, Sylvain
IMS Research Assistant Professor
TOMITA, Takafumi
Post-Doctoral Fellow
TANAKA, Akira
ZHANG, Yichi
BHARTI, Vineet
KUNIMI, Masaya
Graduate Student
MIZOGUCHI, Michiteru
CHEW, Yee Lai
TIRUMALASETTY PANDURANGA, Mahesh
MATSUMOTO, Kiyoharu*
Secretary
KAWAMOTO, Minako

Keywords Quantum-Classical Boundary, Quantum Technology, Attosecond Coherent Control

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 in an optical lattice, as depicted schematically in Figure 1, envisaging the quantum-classical boundary connected smoothly.

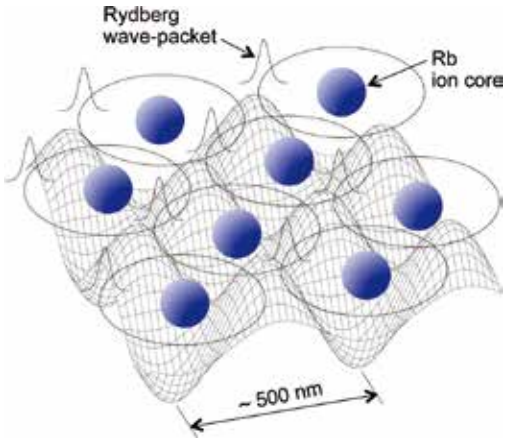


Figure 1. Schematic of the many-body system of ultracold Rydberg atoms.[2]

Selected Publications

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

1. Development of an "Ultrafast Quantum Simulator" by Optical Control with Precisions on the Attosecond Temporal and Submicron Spatial Scales[3–5)]

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"[6)] and "a strongly-correlated ultracold Rydberg gas."[7)]

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 laser pulse, as schematically illustrated in Figure 2.[3,4)] 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. We expect that the overlap between their Rydberg orbitals could give rise to an exotic metal-like phase, where the electron could delocalize over the lattice.

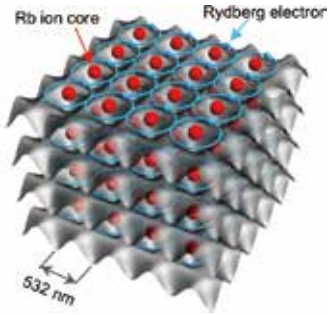


Figure 2. Schematic of the hardware of the ultrafast quantum simulator.[3,4)]

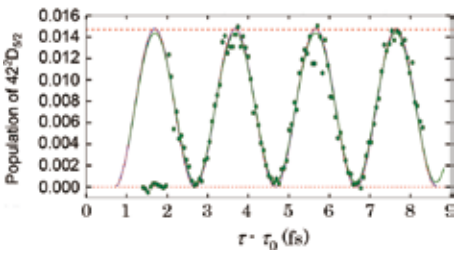


Figure 3. Time domain Ramsey interferometry of ultracold ^{87}Rb atoms with attosecond precision to be used as a readout interface of the ultrafast quantum simulator. Population of the $42^2D_{5/2}$ Rydberg state is plotted as a function of the delay τ between two laser pulses, where $\tau_0 \sim 50$ ps. Adopted from Ref. 5).

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.

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.[8)] 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,[8)] in tight collaborations with Hamamatsu Photonics K. K.[3)] Their examples are shown in Figure 4, where we used a convex lens for clear visualization of the trap arrays. We have recently replaced this convex lens by an objective lens to realize the world's smallest arbitrary trap array with its nearest neighbor distance now only 1.06 micron, which used to be typically ~4 micron in previous works.[9)]

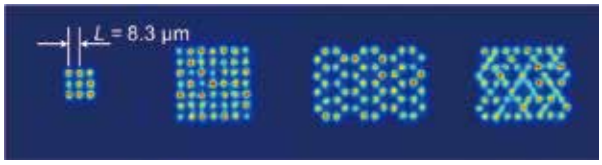


Figure 4. Examples of arbitrary optical trap arrays generated with a low NA convex lens (L = 8.3 μm ; wavelength = 633 nm ; NA = 0.0446), corresponding to L = 640 nm with NA = 0.75 at the wavelength 820 nm.[3)]

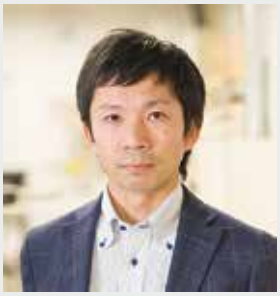
References

1) K. Tonomura *et. al.*, *Am. J. Phys.* **57**, 117 (1989).
2) K. Ohmori, *Found. Phys.* **44**, 813–818 (2014).
3) Patent Publication: US 2018/0292786 A1; JAPAN 2018-180179, "Quantum simulator and quantum simulation method," H. Sakai (Hamamatsu Photonics K.K.), K. Ohmori (IMS) *et al.*, Oct. 11, 2018 (US); Nov. 15, 2018 (JAPAN).
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) H. Katsuki *et al.*, *Acc. Chem. Res.* **51**, 1174–1184 (2018).
7) N. Takei *et al.*, *Nat. Commun.* **7**, 13449 (2016).
8) A. W. Glaetzle *et al.*, *Nat. Commun.* **8**, 15813 (2017).
9) D. Barredo *et al.*, *Science* **354**, 1021 (2016).

* carrying out graduate research on Cooperative Education Program of IMS with Nara Institute of Science and Technology

Electronic Property of Functional Organic Materials

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



KERA, Satoshi
Professor
[kera@ims.ac.jp]

Education
1996 B.E. Chiba University
1998 M.E. Chiba University
2001 Ph.D. Chiba University

Professional Employment
1998 JSPS Research Fellow
2001 Research Associate, Chiba University
2003 Research Associate, Institute for Molecular Science
2003 Postdoctoral Fellow, Wuerzburg University
2004 Assistant Professor, Chiba University
2007 Associate Professor, Chiba University
2009 Visiting Associate Professor, Institute for Molecular Science
2013 Adjunct Lecturer, The Open University of Japan
2013 Visiting Associate Professor, Soochow University
2014 Professor, Institute for Molecular Science
Professor, The Graduate University for Advanced Studies
Visiting Professor, Chiba University
2019 Visiting Professor, Kyoto University, Hiroshima University

Member
IMS Fellow
HASEGAWA, Yuri
Post-Doctoral Fellow
LEE, Dooyong
Visiting Scientist
HAGENLOCHER, Jan*
YANG, Jinpeng*
Secretary
MASUDA, Michiko

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 were sometimes challenging 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 key to precisely investigate the electronic structure at various interfaces, including organic–organic and organic–inorganic (metal/semiconductor) contacts. In these systems, the weak electronic interaction appears as small intensity modulations of fine features 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, weak band dispersion, and dynamic electronic polarization. To elucidate what really impacts 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 would be 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.

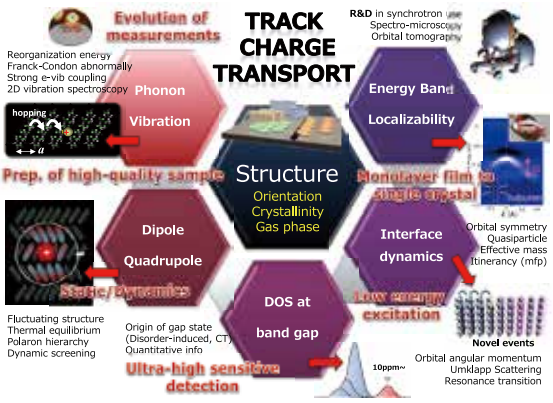


Figure 1. Overview of our agenda. A rich assortment of surface and interface structures of FOM to provide complicated spectral features of ultraviolet photoelectron spectroscopy.

Selected Publications

- 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. Widely Dispersed Intermolecular Valence Bands of Epitaxially Grown Perfluoropentacene on Pentacene Single Crystals[1)]

Strong intermolecular electronic coupling and well-ordered molecular arrangements enable efficient charge transport in semiconducting π-conjugated molecular solids. Molecular heteroepitaxy to form crystallized donor–acceptor molecular interfaces potentially leads to a novel strategy for creating efficient organic optoelectronic devices. In the present study, the crystallographic and electronic structures of a hetero-epitaxial molecular interface, perfluoropentacene (PFP, $C_{22}F_{14}$) grown on pentacene single crystals (Pn-SCs, $C_{22}H_{14}$), were determined by means of grazing-incidence X-ray diffraction (GIXD) and angle-resolved ultraviolet photoelectron spectroscopy (ARUPS), respectively. GIXD revealed that PFP uniquely aligned its primary axis along the [1$\bar{1}$0] axis of crystalline pentacene to form well crystallized overlayers. Valence band dispersion (at least 0.49 eV wide) was successfully resolved by ARUPS. This indicated a small hole effective mass and a significant transfer integral between the frontier molecular orbitals of the nearest-neighbor PFP molecules, ensuring the presumable occurrence of efficient band-like transport in the epitaxial PFP crystallites.

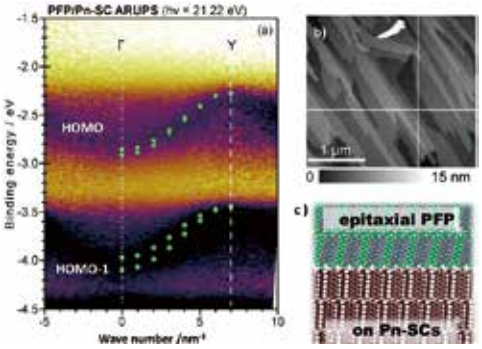


Figure 2. (a) ARUPS intensity map of the 4nm thick PFP overlayer on Pn-SC sample taken in the Γ–Y directions plotted on the E–$K_{||}$ plane. The theoretical band structures predicted by Yoshida *et al.* [*Phys. Rev. B* **92**, 075145 (2015)] are plotted as circles. (b) AFM image of a PFP (5nm)/Pn-SC substrate. The figure is after ref 1) with permission.

2. Molecular Parameters Responsible for Thermally Activated Transport in Doped Organic Semiconductors[2)]

Doped organic semiconductors typically exhibit a thermal activation of their electrical conductivity, whose physical origin is still under scientific debate. We disclose relationships between molecular parameters and the thermal activation energy (E_A) of the conductivity, revealing that charge transport is controlled by the properties of host–dopant integer charge transfer complexes (ICTCs) in efficiently doped organic semiconductors. At low doping concentrations, charge transport is limited by the Coulomb binding energy of ICTCs, which can be minimized by systematic modification of the charge distribution on the individual ions. The investigation of a wide variety of material systems reveals that static energetic disorder induced by ICTC dipole moments sets a general lower limit for E_A at large doping concentrations. The impact of disorder can be reduced by adjusting the ICTC density and the intramolecular relaxation energy of host ions, allowing an increase of conductivity by many orders of magnitude. The observed correlation between E_A and the density of ICTCs indicates that the minimum achievable E_A is generally limited by the electrostatic disorder of ICTCs, which can be reduced by using large molecules or ICTCs with small dipole moments.

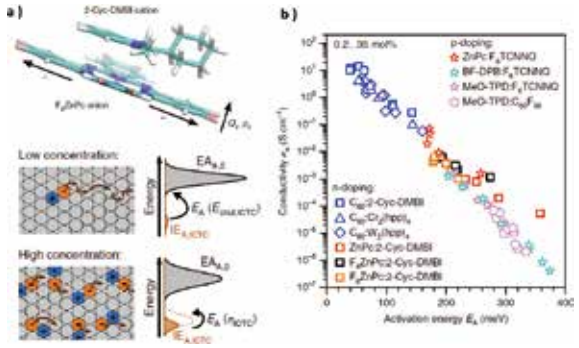


Figure 3. a) Simplified sketch of charge transport in different concentration regimes. At low doping concentrations, electron transport requires a dissociation of ICTCs into separated charges, resulting in an increase of E_A. At large doping concentrations and small distances between ICTCs, transport can be described ideally by electron transport between different ICTC configurations whose local energies change continuously and where multi-electron hops may also play a role. b) The change of conductivity with concentration goes along with a change of E_A. Starting from low doping concentrations, all material combinations investigated show a decrease of E_A with increasing doping concentration, explaining the increase of conductivity. 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. Introduction of novel momentum microscope launched at BL6U is started.

References

1) Y. Nakayama, R. Tsuruta, N. Moriya, M. Hikasa, M. Meissner, T. Yamaguchi, Y. Mizuno, T. Suzuki, T. Koganezawa, T. Hosokai, T. Ueba and S. Kera, *J. Phys. Chem. Lett.* **10**, 1312–1318 (2019). [Selected for Journal Cover]

2) M. Schwarze, C. Gaul, R. Schol, F. Bussolotti, A. Hofacker, K. S. Schellhammer, B. Nell, B. D. Naab, Z. Bao, D. Spoltore, K. Vandewal, J. Widmer, S. Kera, N. Ueno, F. Ortmann and K. Leo, *Nat. Mater.* **18**, 242–248 (2019).

* JSPS Summer Program

Light Source Developments by Using Relativistic Electron Beams

UVSOR Synchrotron Facility
Division of Advanced Accelerator Research

KATOH, Masahiro
Professor
(–March, 2019)
Project Professor
(April, 2019–)
[mkatoh@ims.ac.jp]

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
Professor, The Graduate University for Advanced Studies
2019 Professor, Hiroshima University
Project Professor, Institute for Molecular Science

Member
Assistant Professor
FUJIMOTO, Masaki
Post-Doctoral Fellow
GUO, Lei
Graduate Student
MATSUNAGA, Yukihiro*
KIMURA, Keigo*

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).
- E. Roussel, C. Evain, C. Szwaj, S. Bielawski, J. Raasch, P. Thoma, A. Scheuring, M. Hofherr, K. Ilin, S. Wünsch, M. Siegel, M. Hosaka, N. Yamamoto, Y. Takashima, H. Zen, T. Konomi, M. Adachi, S. Kimura and M. Katoh, "Microbunching Instability in Relativistic Electron Bunches: Direct Observations of the Microstructures Using Ultrafast YBCO Detectors," *Phys. Rev. Lett.* **113**, 094801 (2014).
- M. Katoh, M. Fujimoto, H. Kawaguchi, K. Tsuchiya, K. Ohmi, T. Kaneyasu, Y. Taira, M. Hosaka, A. Mochihashi and Y. Takashima, "Angular Momentum of Twisted Radiation from an Electron in Spiral Motion," *Phys. Rev. Lett.* **118**, 094801 (2017).
- Y. Taira, T. Hayakawa and M. Katoh, "Gamma-Ray Vortices from Nonlinear Inverse Thomson Scattering of Circularly Polarized Light," *Sci. Rep.* **7**, 5018 (2017).
- S. Matsuba, K. Kawase, A. Miyamoto, S. Sasaki, M. Fujimoto, T. Konomi, N. Yamamoto, M. Hosaka and M. Katoh, "Generation of Vector Beam with Tandem Helical Undulators," *Appl. Phys. Lett.* **113**, 021106 (2018).

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 beam-line. In these years, we are focusing on generation of structured light, such as vortex beams and vector beams from undulators, in collaboration with Hiroshima Univ. and Nagoya Univ. We have already succeeded in producing such novel photon beams and are exploring their applications in collaboration with Saga Light Source and Toyama Univ.

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. We have reconstructed the resonator free electron laser to produce more intense gamma-rays through intra-cavity inverse Compton scattering. Moreover, 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 Synchrotron Light Source and Free Electron Laser

We have carried out several upgrade plans successfully since 2000. We designed a special electron 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 installed and commissioned. Besides these major upgrades, 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 of the brightest synchrotron light sources among the low energy machines below 1GeV in the world.

Currently, the storage ring is stably operated for many of the users, however, the requirements from the users for the stability is getting higher and higher. 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. As a long-term plan, we have carried out some design studies on new accelerator systems such as a linear accelerator based free electron laser or a diffraction limited storage ring light source. We are going to continue improving these plans.

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. Optical Cavity for Resonator Free Electron Laser.

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

Angle-Resolved Photoemission Study on Strongly Correlated Electron Materials

UVSOR Synchrotron Facility
Division of Advanced Solid State Physics

TANAKA, Kiyohisa
Associate Professor
[k-tanaka@ims.ac.jp]

Education
2000 B.S. The University of Tokyo
2005 Ph.D. The University of Tokyo

Professional Employment
2005 Postdoctoral Fellow, Stanford University and Lawrence Berkeley National Laboratory
2008 Assistant Professor, Osaka University
2013 Associate Professor, Osaka University
2014 Associate Professor, Institute for Molecular Science
Associate Professor, The Graduate University for Advanced Studies

Member
Assistant Professor
IDETA, Shin-ichiro
Graduate Student
IKEMOTO, Masafumi*
KONTANI, Syouta*
HAYASHI, Naoki*
NABEHIRA, Naoki*

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), a powerful tool in studying the electronic structure of complex materials, based on synchrotron radiation.

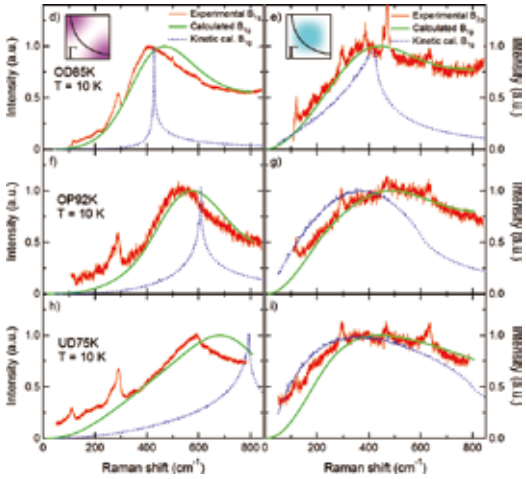


Figure 1. Comparison between experimental ERS spectra (red), calculated ERS spectra from ARPES (green), and kinetic theory (blue) for different carrier concentration of Bi2212 and different momentum space.

Selected Publications

- K. Tanaka, W. S. Lee, D. H. Lu, A. Fujimori, T. Fujii, Risdiana, I. Terasaki, D. J. Scalapino, T. P. Devereaux, Z. Hussain and Z.-X. Shen, "Distinct Fermi-Momentum-Dependent Energy Gaps in Deeply Underdoped Bi2212," *Science* **314**, 1910–1913 (2006).
- W. S. Lee, I. M. Vishik, K. Tanaka, D. H. Lu, T. Sasagawa, N. Nagaosa, T. P. Devereaux, Z. Hussain and Z.-X. Shen, "Abrupt Onset of a Second Energy Gap at the Superconducting Transition of Underdoped Bi2212," *Nature* **450**, 81–84 (2007).
- A. Takemori, T. Hajiri, S. Miyasaka, Z. H. Tin, T. Adachi, S. Ideta, K. Tanaka, M. Matsunami and S. Tajima, "Change of Fermi Surface States Related with Two Different T_c-Raising Mechanisms in Iron Pnictide Superconductors," *Phys. Rev. B* **98**, 100501(R) (2018).
- T. Adachi, S. Ideta, Z. Tin, H. Usui, K. Tanaka, S. Miyasaka and S. Tajima, "Electronic Structure of $Sr_{1-y}Ca_yFe_2(As_{1-x}P_x)_2$ (x = 0.25, y = 0.08) Revealed by Angle-Resolved Photoemission Spectroscopy," *J. Phys. Soc. Jpn.* **88**, 084701 (8 pages) (2019).
- 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).

1. Quantitative Comparison between ARPES and ERS on High-T_c Cuprate Superconductor[1)]

Both of ARPES and electronic Raman scattering (ERS) are powerful techniques which can obtain momentum-selected electronic structure. In the study of high-T_c cuprates superconductors, both techniques revealed two energy scales for the gap in different momentum spaces. However, the interpretations were different and the gap values were also different in two experiments. In order to clarify the origin of these discrepancies, we have directly compared experimental ARPES and ERS by using new calculation method of ERS through the Kubo formula.

It is well known that ARPES spectrum is a function of matrix element, Fermi Dirac function and a spectral function A(k,ω). On the other hand, ERS spectrum has been understood by simple model (so-called kinetic theory) for long time, where delta-function density of states along the Fermi surface is assumed. Therefore, quantitative analysis and discussion of ERS spectrum have been difficult. Using the Kubo formula, we noticed that the ERS spectrum can be written by square of the imaginary part of Green's function. This means that ERS spectrum can be calculated by ARPES spectrum, since the imaginary part of Green's function corresponds to the spectral function A(k,ω) in ARPES.

In this study, we have performed ARPES and ERS measurements on the same sample to directly compare the results. From ARPES spectra, we obtained A(k,ω) and calculated ERS spectra and compared to the experimental ERS spectra. For the samples, we prepared $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) with three doping levels, namely, underdoped (UD75K: T_c = 75 K), nearly optimally doped (OP92K: T_c = 92 K), and overdoped (OD85K: T_c = 85 K) samples.

Figure 1 shows the calculated ERS spectra. Here, it should be noted that the intensity distribution of A(k,ω) along the Fermi surface is an unknown parameter and we only showed the best fitted result. Compared to the conventional ERS spectral calculation based on kinetic theory (blue), our new calculation results (green) reproduced spectral features much better. Especially B_{2g} spectra, which is sensitive to the nodal region in the momentum space, were well reproduced. Doping dependence of the intensity distribution of A(k,ω) shows that the spectral function confined in the nodal region distributes to the antinodal region as the doping level increases (not shown).

The peak energies of the calculated ERS spectra were plotted in Figure 2 together with the experimental Raman, ARPES and STM data. From B_{1g} spectra, which is sensitive to the antinodal region in the momentum space, we found that the ARPES antinodal gap is always larger than the experimental B_{1g} peak energy. Since the difference increases with underdoping, this difference is possibly caused by the pseudogap. In Figure 2, we also plotted the pseudogap energy determined by ARPES Bi2212 data taken at 100 K. The pseudogap increases rapidly with underdoping and it seems that the superconducting gap in ARPES is enhanced by underlying- high-energy-pseudogap.

The present results give us the following important messages. First, Raman spectrum can be well reproduced by ARPES spectrum, and Raman and ARPES can be understood with the same gap profile. Namely, the nodal slope of gap profiles is doping independent, as reported by ARPES. The apparent doping dependence of the B_{2g} peak energy is caused by the change of spectral weight of A(k,ω) along the Fermi surface. Second, the antinodal gap of ARPES is a superconducting gap that is strongly affected by the pseudogap, whereas the Raman B_{1g} gap is moderately affected. This probe-dependent effect of the pseudogap is the main source for the difference between the Raman B_{1g} gap and the ARPES aninodal gap energies. Third, while the spectral weight of A(k,ω) is confined into the nodal region in the underdoped sample, the antinodal region gains spectral weight with doping and contributes to superconductivity. Although this is similar to the "Fermi arc" picture reported before, the Fermi surface area contributing to superconductivity is larger than that estimated from the normal state ARPES as a Fermi arc. All these findings reflect the unusual electronic states where superconductivity and coexist even at the lowest temperature.

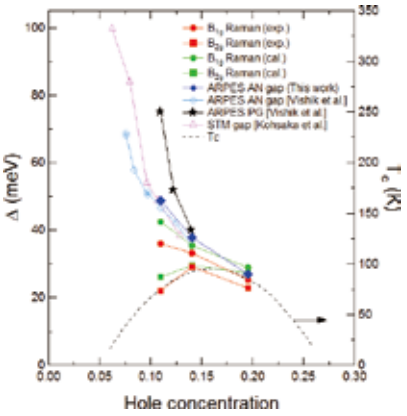


Figure 2. Doping dependence of the peak energies in Bi2212 obtained from ERS calculations in comparison to the experimental data from Raman, ARPES and STM measurements.

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

To perform high energy resolution ARPES measurements, the temperature of samples is important. We developed low temperature 6-axis manipulator by ourselves and achieved 4.5 K with tilting angle −15 ~ 55 deg. and azimuth angle ±120 deg. This is one of the lowest temperature 6-axis manipulators in the synchrotron radiation facilities in the world.

Reference

1) K. Tanaka, N. Hieu, G. Vincini, T. Masui, S. Miyasaka, S. Tajima and T. Sasagawa, *J. Phys. Soc. Jpn.* **88**, 044710 (2019).

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

Resonant Photoemission: Selective Access to Surface Atomic and Molecular Orbitals

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research

MATSUI, Fumihiko
Senior Researcher

The electronic properties and chemical reactivities of materials are closely related to the behavior of valence electrons near the Fermi level. Momentum-resolved valence-band photoelectron spectroscopy is a powerful technique to characterize such electrons. Furthermore, by tuning the excitation energy to the core-level absorption edge, valence electron can be resonantly selected and analyzed. In order to establish the reliability of this method, comprehensive measurement and understanding of the photoelectron emission process are important.[1] BL6U at UVSOR is a beamline dedicated to valence band dispersion mapping with a practical photon energy range of 40 to 600 eV. Wide-acceptance-angle acquisition system enables measurement of full set of valence band dispersion data over several Brillouin zones.[2]

1. Graphite Valence Band Dispersion in Resonance Auger Electron Spectra

Figure 1 shows a comparison of the angular distribution of graphite valence band between below and above the C 1s absorption threshold. The intensity of the π band with parabolic dispersion is enhance at the π* resonance due to "participant Auger electron process." Until now, there was a common understanding that the angular distribution of Auger electron intensity is due to diffraction, not band dispersion. Here, we found that the resonant Auger electrons *do* carry band dispersion information. The red dashed curves show that momentum is conserved by the electrons involved in the "spectator Auger electron process."

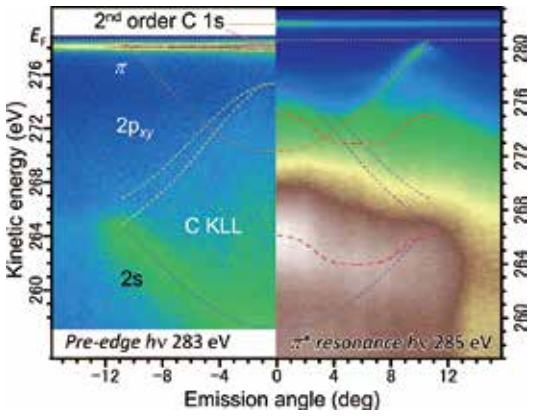


Figure 1. Angle-resolved photoelectron spectra below (left) and above (right) the C K edge threshold. Red dashed lines in the right panel are the newly found participant Auger electron dispersion features.

These experiments can only be performed on soft x-ray beamlines with low carbon contamination. In cooperation with the UVSOR technical team, we removed carbon from the beamline optics by O_2 photochemical etching and successfully experimented with this resonant photoelectron spectroscopy. The spectator Auger process at the C absorption edge was actively explored in the past to try to break specific chemical bonds by selecting antibonding orbitals as the transition destination of 1s electrons. The present success of resonance Auger electron experiments at the C K absorption edge is the basis for expanding the range of application to molecular adsorption systems.[3,4]

2. Momentum Microscope

Conventionally, azimuthal and polar scans of sample orientation were required for the angle-resolved photoelectron spectroscopy and diffraction measurements. Aforementioned wide-acceptance-angle acquisition system combines an optimized-structure mesh for gathering photoelectrons emitted into wide solid angle and a mechanical lens deflector for two-dimensional data acquisition. Alternatively, display-type analyzers enable the direct observation of wide-solid-angle photoelectron intensity distribution from a selected point without changing the angles of incident light or the sample orientation. By combining a photoelectron emission microscope column and two hemispherical deflection analyzers, *i.e. momentum microscope*, iso-energy photoelectron intensity k_x–k_y distribution can be obtained with high-momentum, energy, and spatial resolutions. A project is underway to install a state-of-the-art momentum microscope in UVSOR and a comprehensive photoemission experiment station.

References

1) F. Matsui *et al.*, *J. Phys. Soc. Jpn.* **87**, 061004 (2018).
2) H. Yamane, F. Matsui *et al.*, *Rev. Sci. Intrum.* **90**, 093102 (2019).
3) D. Usachov, F. Matsui *et al.*, *2D Materials* **6**, 045046 (2019).
4) D. Usachov, B. Senkovskiy and F. Matsui, *UVSOR activity reports* (2018).

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

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

NAGASAKA, Masanari
Assistant Professor

Soft X-ray absorption spectroscopy (XAS) is an element specific method to reveal local structures of liquids with the K-edges of C, N, and O. We have developed a liquid flow cell for XAS of liquids in transmission mode, where liquid thickness is controllable from 20 to 2000 nm.[1)] Local structures of several liquid samples have been investigated from the precise energy shift analyses of XAS peaks with the help of inner-shell calculations.[2,3)]

1. Microheterogeneity in Aqueous Acetonitrile Solution

In microheterogeneity (MH), two liquids are mixed in macroscopic scale but are inhomogeneous in microscopic scale. We have investigated one of the simplest MH systems, aqueous acetonitrile solution, by using XAS. Molecular interactions of acetonitrile were measured in C and N K-edges, and those of solvent water were separately observed in O K-edge. The energy shifts of the C≡N π^* peaks in C K-edge XAS show three concentration regions and especially a phase transition-like behavior. By comparing the energy shifts of XAS peaks with the inner-shell calculations, we have revealed that the MH state emerges when small acetonitrile clusters are formed by surrounding water with dipole interactions. By increasing the molar fraction of water, the MH state is diminished when the small acetonitrile clusters are taken into hydrogen bond network of water.

References

1) M. Nagasaka *et al.*, *J. Electron Spectrosc. Relat. Phenom.* **224**, 93–99 (2018).
2) M. Nagasaka *et al.*, *Z. Phys. Chem.* **232**, 705–722 (2018).
3) M. Nagasaka *et al.*, *J. Phys. Chem. Lett.* **9**, 5827–5832 (2018).

Award

NAGASAKA, Masanari; The 8th Young Scientist Award of National Institutes of Natural Sciences.

Spectro-Microscopic Analysis of Lithium in an Electrode of a Lithium-Ion Battery

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research

OHIGASHI, Takuji
Assistant Professor

Nowadays, lithium is an important element especially used for a secondary battery. To analyze chemical state and distribution of lithium in the battery, a scanning transmission X-ray microscope (STXM) will be a promising tool but an absorption edge in low energy, 55 eV, makes it difficult. Then, main issues are contamination of spectra by a lot of higher harmonic lights from a monochromator and extremely short focal length of an optical element, a Fresnel zone plate (FZP). Recently, we overcame these issues by designing a new FZP and succeeded to have an access to Li K-edge by STXM at BL4U. Parameters of the new FZP are as follows; diameter of 240 μm, outermost zone width of 60 nm, gold pattern of 100 nm thick on a silicon substrate of 200 nm thick. The new FZP works as not only a focusing device of the X-rays but also as a filter for higher harmonics above 100 eV by Si L-edge (99 eV) absorption. From evaluation of the new FZP, intensities of the higher harmonics light from 2nd to 5th orders for Li K-edge range are suppressed less than 1% of the 1st order light. Then, spatial resolution is below 85 nm at 90 eV.

An ultra-thin section of an electrode of a lithium-ion battery was measured around Li K-edge. The X-ray absorption spectra of $LiCO_3$ were obtained.

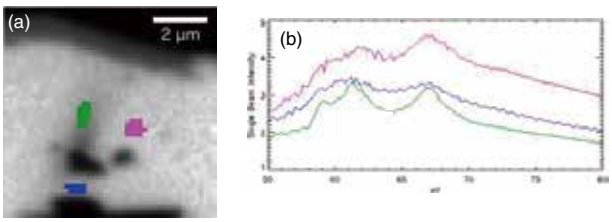


Figure 1. (a) Optical density image of an electrode of a lithium ion battery and (b) X-ray absorption spectra.

Reference

1) T. Ohigashi, in preparation.

Soft X-Ray Absorption Spectroscopy for Soft Matter

UVSOR Synchrotron Facility
Division of Advanced Photochemistry

IWAYAMA, Hiroshi
Assistant Professor

Recently we started to study soft matter with soft x-ray, whose energy region covers K-edge energies of light elements such as carbon, nitrogen and oxygen. Local chemical environments of liquid sample in a liquid cell[1)] was investigated by x-ray absorption spectroscopy (XAS).

1. Soft X-Ray Absorption Spectroscopy for Liquid-Crystal Materials

The liquid-crystal material is one of the most important soft matter. In addition to gas, liquid and solid phases, liquid-crystal materials have liquid-crystal phase between solid and liquid phases. In the case of nematic liquid-crystal phase, the rod-shaped organic molecules have no position order, but they self-align to have long-range directional order with their long axes roughly parallel. To investigate a change of local chemical environments caused by a phase transition, we measured XAS spectra of liquid-crystal materials at carbon and nitrogen K edge in solid, liquid-crystal and liquid phases.

In XAS spectra at carbon and nitrogen K edges, we find several peaks which are corresponding to core excitations such as 1s-to-π^* core excitations. We assigned these peaks to each core excitations with the help of a quantum chemical program, StoBe. The peak intensities and structures of XAFS spectra are slightly different for each phase of sample. We obtained results suggesting that π–π interactions of two molecules become weak after a phase transition from solid to liquid-crystal phases.

Reference

1) M. Nagasaka, H. Yuzawa and N. Kosugi, *J. Spectrosc. Relat. Phenom.* **200**, 293 (2015).

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 reset the IL-derived bias stress immediately 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 Associate Professor
KANEYASU, Tatsuo *(from SAGA Light Source)*

Generation and Application of Structured Light in Synchrotron Light Sources

Novel undulators in synchrotron light sources can provide energy-tunable structured light at short wavelengths. We have been investigating the generation and application of structured light, such as vortex beams and vector beams, in the extreme ultraviolet (XUV) wavelength region in the UVSOR synchrotron facility. In exploring new applications of the structured light, a fundamental understanding of the light-matter interaction is crucial. We have investigated the characteristics of the structured light by using interference measurements and have applied it to the photoionization and photoexcitation study of rare-gas atoms. In addition, we started to study the potential of undulator radiation as longitudinally coherent wave packets suitable for the coherent control at short wavelengths. We are currently performing the coherent control experiments in photoexcitation of atoms using the undulator radiation in the XUV to soft x-ray range.

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

Coherent Control in Condensed Systems

Coherent control is a technique to manipulate quantum states of a target system utilizing the interference of wavefucntions. My research is focused on the coherent control in organic crystals and in strongly coupled systems. In the first case, we have recently demonstrated the control of multiple THz intramolecular oscillatory modes in a rubrene crystal. We are planning to apply this technique for the electronic excited states in which the electron–phonon interaction plays a key role in various physical properties such as carrier transportation and photoemission. In the second case, a typical example of the strong coupled system is a cavity exciton–polariton system, which is a mixture of cavity photons and atomic/molecular excitons. Cavity exciton–polaritons have gathered much attention due to its spontaneous formation of quantum coherence in the lowest energy ($k = 0$) state and succeeding coherent emission called polariton lasing, when the excitation fluence is above the threshold. We are now trying to artificially regulate the polariton flow towards the $k = 0$ state utilizing the stimulated scattering process.