



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
TAKEI, Nobuyuki
IMS Research Assistant Professor
TANAKA, Akira
Post-Doctoral Fellow
ZHANG, Yichi
MENG, Zengming
Visiting Scientist
TIRUMALASETTY PANDURANGA, Mahesh*
MARGUET, Bastien†
MATSUMOTO, Kiyoharu‡
Graduate Student
MIZOGUCHI, Michiteru
CHEW, Yee Lai
Secretary
KAWAMOTO, Minako

Keywords

Quantum-Classical Boundary, Quantum Technology, 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 a bulk solid composed of many nuclei and electrons interacting with each other, and becomes localized in space. This change, referred to as “collapse” in quantum mechanics, is often accepted as a discontinuous event, but a basic question arises: When and how the delocalized wave function becomes localized? Our dream is uncovering this mystery by observing the spatiotemporal evolution of a wave function delocalized over many particles interacting with each other. Having this dream 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 ensemble of ultracold Rydberg atoms, as depicted schematically in Figure 1, and a bulk solid, envisaging the quantum-

classical boundary connected smoothly.

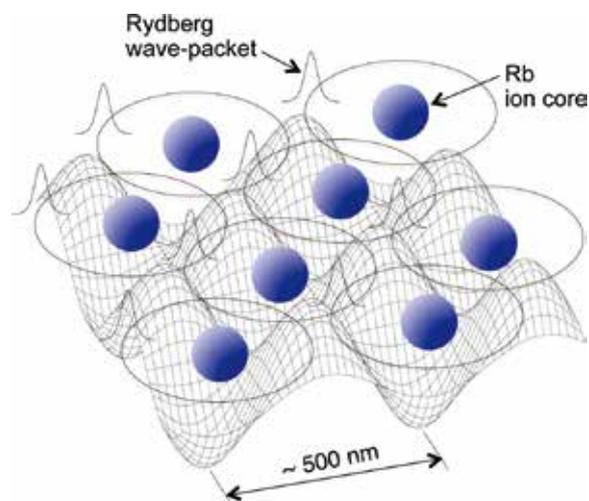


Figure 1. Schematic of the many-body system of ultracold Rydberg atoms.²⁾

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).

1. Ultrafast Coherent Control of Condensed Matter with Attosecond Precision³⁾

Coherent control is a technique to manipulate wave functions of matter with light. Coherent control of isolated atoms and molecules in the gas phase is well-understood and developed since the 1990s, whereas its application to condensed matter is more difficult because its coherence lifetime is shorter. We have recently applied this technique to condensed matter samples, one of which is solid *para*-hydrogen (*p*-H₂). Intramolecular vibrational excitation of solid *p*-H₂ gives an excited vibrational wave function called a “vibron,” which is delocalized over many hydrogen molecules in a manner similar to a Frenkel exciton. It has a long coherence lifetime, so we have chosen solid *p*-H₂ as our first target in the condensed phase. We shine a time-delayed pair of femtosecond laser pulses on *p*-H₂ to generate vibrons. Their interference results in modulation of the amplitude of their superposition. Scanning the interpulse delay on the attosecond time scale gives a high interferometric contrast, which demonstrates the possibility of using solid *p*-H₂ as a carrier of information encoded in the vibrons.

In the second example, we have controlled the terahertz collective phonon motion, called a “coherent phonon,” of a single crystal of bismuth. We employ an intensity-modulated laser pulse, whose temporal envelope is modulated with terahertz frequency by overlap of two positively chirped laser pulses with their adjustable time delay. This modulated laser pulse is shined on the bismuth crystal to excite its two orthogonal phonon modes. Their relative amplitudes are controlled by tuning the delay between the two chirped pulses on the attosecond time scale. Two-dimensional atomic motion in the crystal is thus controlled arbitrarily. The method is based on the simple, robust, and universal concept that in any physical system, two-dimensional particle motion is decomposed into two orthogonal one-dimensional motions, and thus, it is applicable to a variety of condensed matter systems.

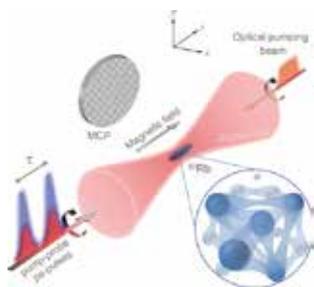


Figure 2. Schematic diagram of the experimental setup for the observation of ultrafast many-body electron dynamics in a strongly correlated ultracold Rydberg gas.³⁾

Awards

OHMORI, Kenji; Hiroshi Takuma Memorial Prize of Matsuo Foundation (2017).

OHMORI, Kenji; Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology of Japan (2018).

* IMS International Internship Program

† carrying out graduate research on Cooperative Education Program of IMS with Ecole Normale Supérieure Paris-Saclay

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

In the third example, the double-pulse interferometry used for solid *p*-H₂ has been applied to many-body electronic wave functions of an ensemble of ultracold rubidium Rydberg atoms, hereafter called a “strongly correlated ultracold Rydberg gas,” as shown in Figure 2. This has allowed the observation and control of many-body electron dynamics of more than 40 Rydberg atoms interacting with each other. This new combination of ultrafast coherent control and ultracold atoms offers a versatile platform to precisely observe and manipulate nonequilibrium dynamics of quantum many-body systems on the ultrashort time scale.

2. Attosecond Control of Electronic Structure Symmetry Restoration⁴⁾

Laser pulses can break the electronic structure symmetry of atoms and molecules by preparing a superposition of states with different irreducible representations. Here, we discover the reverse process, symmetry restoration, by means of two circularly polarized laser pulses. The laser pulse for symmetry restoration is designed as copy of the pulse for symmetry breaking. Symmetry restoration is achieved if the time delay is chosen such that the superposed states have the same phases at the temporal center. This condition must be satisfied with precision of a few attoseconds. Numerical simulations are presented for the C₆H₆ molecule and ⁸⁷Rb atom as shown in Figure 3. The experimental feasibility of symmetry restoration is demonstrated by means of high-contrast time-dependent Ramsey interferometry of the ultracold ⁸⁷Rb atom.

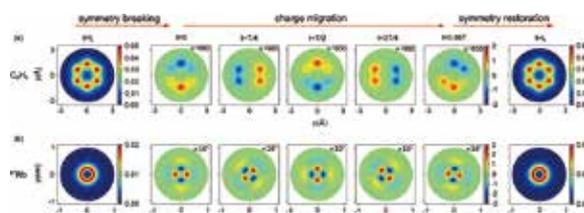


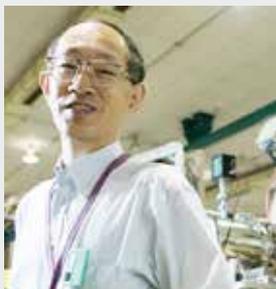
Figure 3. Numerical simulations of the evolutions of electronic densities upon laser-induced symmetry breaking, charge migration, and symmetry restoration in the (a) C₆H₆ molecule and (b) ⁸⁷Rb atom. The central panels show snapshots of the time-dependent parts of their electronic densities.⁴⁾

References

- 1) K. Tonomura *et al.*, *Am. J. Phys.* **57**, 117 (1989).
- 2) K. Ohmori, *Found. Phys.* **44**, 813–818 (2014).
- 3) H. Katsuki *et al.*, *Acc. Chem. Res.* **51**, 1174–1184 (2018).
- 4) C. Liu *et al.*, *Phys. Rev. Lett.* **121**, 173201 (2018).

Local Chemical State Analysis Using Soft X-Rays: Experiment and Theory

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



KOSUGI, Nobuhiro
Professor
(–March, 2018)
[kosugi@ims.ac.jp]

Education

1976 B.S. Kyoto University
1981 Ph.D. The University of Tokyo

Professional Employment

1981 JSPS Postdoctoral Fellow
1982 Research Associate, The University of Tokyo
1988 Lecturer, The University of Tokyo
1989 Associate Professor, Kyoto University
1993 Professor, Institute for Molecular Science
1993 Professor, The Graduate University for Advanced Studies
2018 Director, Institute of Materials Structure Science, KEK

1996 Visiting Professor, McMaster University, Canada
2008 Visiting Professor, Université P&M Curie - Paris VI, France
2011 Visiting Professor, Université Paris-Sud - Paris XI, France
2013 Visiting Professor, Freie Universität Berlin, Germany

Award

2015 The 68th Chemical Society of Japan (CSJ) Award

Member

Assistant Professor
NAGASAKA, Masanari
Post-Doctoral Fellow
YUZAWA, Hayato
Visiting Scientist
VU, Anna A.*
Secretary
INAGAKI, Itsuko

Keywords

X-Ray Spectroscopy, Local Chemical State Analysis, Quantum Chemistry

This research group has strong international network in the field of soft X-ray molecular sciences using world-leading advanced synchrotron radiation facilities for more than 20 years.

Soft X-rays cannot pass through air or bulk water due to photoabsorption of N₂, O₂, and H₂O molecules. Such strong photoabsorption can be used in highly sensitive chemical state analysis of thin or dilute samples, because soft X-rays with photon energies of 100–700 eV excite 1s inner-shell (K-shell) electrons of chemically important light elements such as C, N, and O selectively to unoccupied states of molecules. The 1s electron is localized and bound by an atom in the system but is slightly affected by surrounding atoms and chemical bonds. Therefore, we can select a 1s electron in each atom in molecules by choosing different X-ray energies with 10–100 meV accuracy, and know each atomic component and chemical environment in the unoccupied state.

In order to realize *in situ* and *in operando* chemical state analysis revealing local electronic structures and weak intermolecular interactions in molecular systems such as organic solids, liquids, aqueous solutions, and molecular clusters, we are developing and improving several kinds of thin sample cells, detection systems, and spectro-microscopic techniques in X-ray absorption spectroscopy (XAS) and X-ray photo-

electron spectroscopy (XPS). We are also using resonant and non-resonant X-ray emission spectroscopy (XES) and angle-resolved photoelectron spectroscopy (ARPES).

Sample thickness should be optimized below 1 μm to get optimal absorbance in XAS. For inhomogeneous samples, the 10 nm-scale spatial resolution is necessary. It is also important to reduce the radiation damage of samples due to strong light-matter interaction in the soft X-ray region.

Highly brilliant soft X-rays for the chemical state analysis are available on our UVSOR-III Synchrotron in IMS. In addition to quite unique experimental and instrumental developments at UVSOR-III BL3U, BL4U and BL6U, we are developing an original *ab initio* quantum chemical program package GSCF, which is optimized to calculation of molecular inner-shell processes.

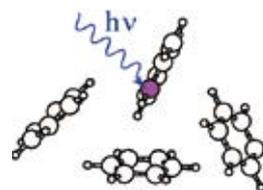


Figure 1. The C 1s excitation energy in interacting benzene molecules is dependent and selective on chemically different atomic sites.

Selected Publications

- M. Nagasaka, H. Yuzawa, T. Horigome, A. P. Hitchcock and N. Kosugi, “Electrochemical Reaction of Aqueous Iron Sulfate Solutions Studied by Fe L-Edge Soft X-Ray Absorption Spectroscopy,” *J. Phys. Chem. C* **117**, 16343–16348 (2013).
- M. Nagasaka, K. Mochizuki, V. Leloup and N. Kosugi, “Local Structures of Methanol-Water Binary Solutions Studied by Soft

X-Ray Absorption Spectroscopy,” *J. Phys. Chem. B* **118**, 4388–4396 (2014).

- H. Yamane and N. Kosugi, “Substituent-Induced Intermolecular Interaction in Organic Crystals Revealed by Precise Band Dispersion Measurements,” *Phys. Rev. Lett.* **111**, 086602 (5 pages) (2013).

1. Temperature Dependent Structural Changes in Liquid Benzene

Benzene is the simplest aromatic molecule with a possibility of several kinds of intermolecular π - π interactions. Since ordered liquids are key structures to study chemical and biological phenomena in the liquid state, ordered structures of benzene confined in nanopores have been extensively studied, whereas those in the liquid state are still unknown. In this study, we address fundamental questions whether ordered structures of benzene are formed in the liquid state by using carbon K-edge X-ray absorption spectroscopy (XAS) as a sensitive local probe.

The experiments were performed at soft X-ray undulator beamline BL3U at UVSOR-III Synchrotron. Figure 2(a) shows C K-edge XAS of liquid benzene at different temperatures. The energy shifts of the solid at -193 °C¹⁾ and clusters²⁾ from the gas peak are -55 meV and -70 meV, respectively. The energy shift of liquid benzene is between the gas and the solid at -193 °C. Figure 2(b) shows the energy shift of the π^* peak of liquid benzene relative to benzene gas as a function of temperature. The π^* peak of liquid benzene shows a lower energy shift with increasing temperature. Although the π^* peak of liquid benzene should be approaching that of gaseous benzene with increasing temperature, the experimentally observed energy shift of the π^* peak goes into the opposite direction.

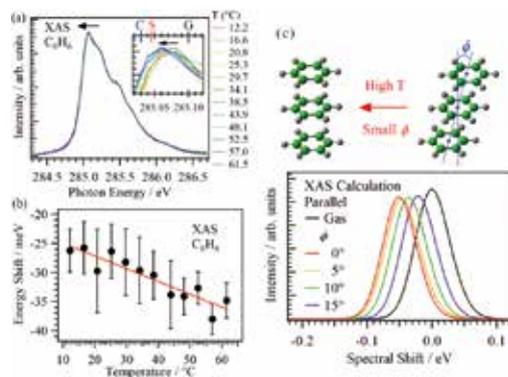


Figure 2. (a) C K-edge XAS of liquid benzene at different temperatures. (G), (S), and (C) are the peaks of gas, solid, and clusters, respectively. (b) Energy shifts of the π^* peak as a function of temperature. (c) Calculated spectral shifts of parallel structures at the different angles ϕ , which become smaller at higher temperatures (T).

For understanding the characteristic temperature behavior of the π^* peak shift in C K-edge XAS of liquid benzene, we have performed molecular dynamics simulation and inner-shell calculation based on quantum chemistry. In radial distribution functions of parallel structures in liquid benzene, the components of small intermolecular distance become abundant by increasing temperature. Figure 2(c) shows inner-shell spectra of benzene trimers at different angle ϕ in parallel structures. The π^* peaks show lower energy shifts with decreasing the tilt angle ϕ of the

Award

NAGASAKA, Masanari; Young Scientist Awards of the Japan Society for Molecular Science (2017).

* IMS International Internship Program

parallel displaced structures. Since the angle ϕ in the parallel displaced structure becomes smaller by increasing temperature, these spectral shifts suggest that parallel displaced structures in liquid benzene are changed to sandwich (parallel non-displaced) structures for the higher temperature.

2. Microfluidics of Liquid Mixtures Observed by Spatially-Resolved XAS

Microfluidics is a technique for efficient chemical reactions, interfaces, and transport processes in the liquid phase.³⁾ Although XAS is an element specific method to reveal structures of liquids by microfluidics, the application of XAS to microfluidics is difficult since soft X-rays cannot deeply penetrate a microfluidic cell. In this study, we have developed a microfluidic cell, where a T-shape microfluidic setup with the width and thickness of 50 μm is made of PDMS resin and is covered by a 100 nm thick Si_3N_4 membrane.

Figure 3(a) shows a soft X-ray fluorescence image of the T-shape microfluidic cell excited by soft X-rays at 550 eV. The spatial resolution of this image is 30×30 μm^2 . The fluorescence shows high intensity at the water part and low intensity at the pyridine part. A laminar flow of pyridine and water is observed in the mixed part of the microfluidic cell. As shown in Figure 3(b), the π^* peak of pyridine in N K-edge XAS shows a higher energy shift from the pyridine to water part across the mixed part due to the formation of hydrogen bond between pyridine and water.⁴⁾ This energy shift indicates that the phase separation in the laminar flow is clearly observed by spatially resolved XAS. This spectroscopic technique will be applicable to only to liquid mixtures but also to chemical and biological reactions.

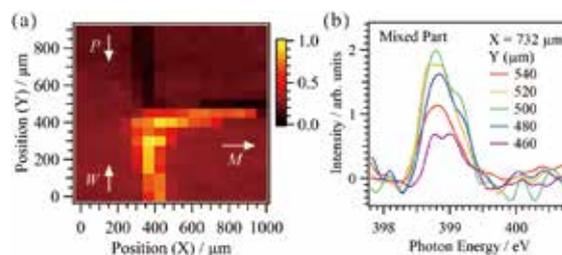


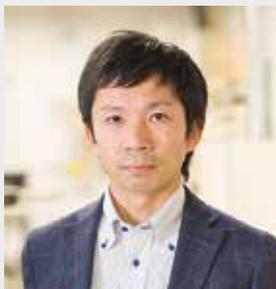
Figure 3. Soft X-ray fluorescence image of a T-shaped microfluidic cell excited by soft X-rays at 550 eV. Pyridine (P) and water (W) flow in the upper and lower sides, respectively, and the laminar flow is observed in the mixed part (M). (b) N K-edge XAS at different positions in the mixed part shown in (a). The horizontal position is fixed to $X = 732$ μm , and the vertical position is scanned.

References

- 1) R. Flesch *et al.*, *Phys. Chem. Chem. Phys.* **14**, 9397 (2012).
- 2) I. L. Bradeanu *et al.*, *Phys. Chem. Chem. Phys.* **8**, 1906 (2006).
- 3) T. Kitamori *et al.*, *Anal. Chem.* **76**, 53 (2004).
- 4) M. Nagasaka *et al.*, *Z. Phys. Chem.* **232**, 705 (2018).

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

Member

Assistant Professor
UEBA, Takahiro
Post-Doctoral Fellow
MEISSNER, Matthias
Visiting Scientist
SCHAAL, Maximilian*
METZGER, Christian*
Graduate Student
SHIRAIISHI, Ryo
YAMAGUCHI, Takuma
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.

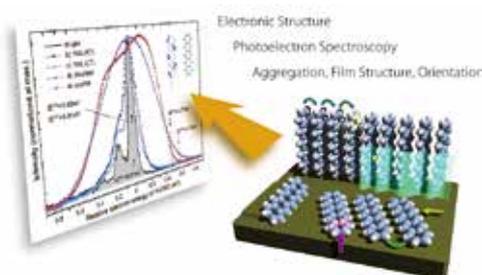


Figure 1. Scheme of a rich assortment of structures of functional molecular materials and spectral features of ultraviolet photoelectron spectroscopy (UPS) for the highest occupied molecular orbital (HOMO), measured for various structural phases (gas-phase, lying monolayers, standing monolayer, and disordered film).

Selected Publications

- S. Kera, T. Hosokai and S. Duhm, “Characteristics of Organic–Metal Interaction: A Perspective from Bonding Distance to Orbital Delocalization,” *J. Phys. Soc. Jpn.* **87**, 061008 (7 pages) (2018). [review]
- J.-P. Yang, F. Bussolotti, S. Kera and N. Ueno, “Origin and Role of Gap States in Organic Semiconductor: As the Nature of Organic Molecular Crystals,” *J. Phys. D: Appl. Phys.* **50**, 423002 (45 pages) (2017). [review]
- S. Kera and N. Ueno, “Photoelectron Spectroscopy on the Charge Reorganization Energy and Small Polaron Binding Energy of Molecular Film,” *J. Electron Spectrosc. Relat. Phenom.* **204**, 2–11 (2015). [review]

1. Evolution of Intermolecular Energy Bands in Organic Thin Films¹⁾

In organic semiconductors, the hole and electron transport occurs through the intermolecular overlaps of highest occupied molecular orbitals (HOMO) and lowest unoccupied molecular orbitals (LUMO), respectively. A measure of such intermolecular electronic coupling is the transfer integral (t), which can experimentally be observed as energy level splittings or the width of the respective energy bands. Quantum chemistry textbooks describe how an energy level splits into two levels in molecular dimers, into three levels in trimers and evolves into an energy band in infinite systems, a process that has never been observed for the LUMO or beyond dimers for the HOMO. In this work, our new technique, low-energy inverse photoelectron spectroscopy (LEIPS), was applied to observe the subtle change of the spectral line shape of a LUMO-derived feature while we used ultraviolet photoelectron spectroscopy (UPS) to investigate the occupied states. We show at first that tin-phthalocyanine molecules (SnPc) grow layer-by-layer in quasi one-dimensional stacks on graphite, and then discuss a characteristic and systematic broadening of the spectral line shapes of both HOMO and LUMO. The results are interpreted as energy-level splittings due to the intermolecular electronic couplings. On the basis of the Hückel approximation, we determined the transfer integrals for HOMO-1, HOMO, and LUMO to be ~ 15 meV, ~ 100 meV, and ~ 128 meV, respectively.

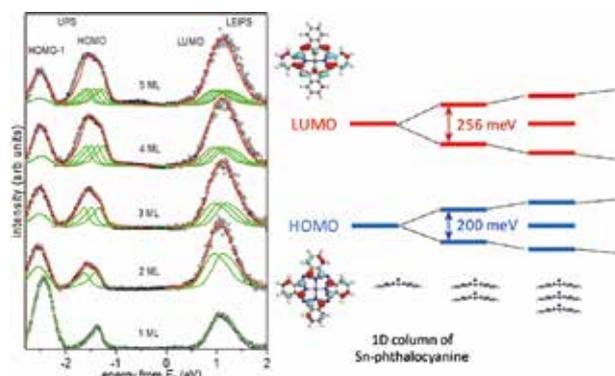


Figure 2. Left: Combined UPS and LEIPS spectra of SnPc films prepared on graphite (HOPG), with the thickness ranging between 1 and 5 monolayer (ML). The experimental data (open circles) are reproduced with the sum (red lines) of several spectral line shapes of the 1 ML peak (green lines) based on the energy levels calculated by Hückel model. Right: Energy splittings of HOMO and LUMO due to intermolecular orbital interaction. Corresponding orbital distributions are schematically shown together with quasi one-dimensional molecular stacking.

2. Efficient Organic Semiconductor Doping²⁾

Doping plays a crucial role in semiconductor physics, with n-doping being controlled by the ionization energy of the

impurity relative to the conduction band edge. In organic semiconductors, efficient doping is dominated by various effects that are currently not well understood. Here, we simulate and experimentally measure, with direct and inverse photoemission spectroscopies, the density of states and the Fermi level position of the prototypical materials C_{60} and zinc phthalocyanine n-doped with highly efficient benzimidazole radicals (2-Cyc-DMBI). We simulate the electronic states of nanoclusters of different molecules in the centre surrounded by up to 14 additional C_{60} molecules to describe the near-field dielectric screening of charge distributions, and evaluate various charging states of the molecules in the centre of the clusters and their charging energies (EA and IP), and thereby construct the DOS of the doped system. We study the role of doping-induced gap states, and, in particular, of the difference Δ_1 between the electron affinity of the undoped material and the ionization potential of its doped counterpart. We show that this parameter is critical for the generation of free carriers and influences the conductivity of the doped films. Efficient dopants should be optimized with regard to the molecular doping parameter Δ_1 , which is crucial in organic semiconductors but does not exist in traditional inorganic semiconductors because of the strong screening of electrostatic effects in inorganic materials.

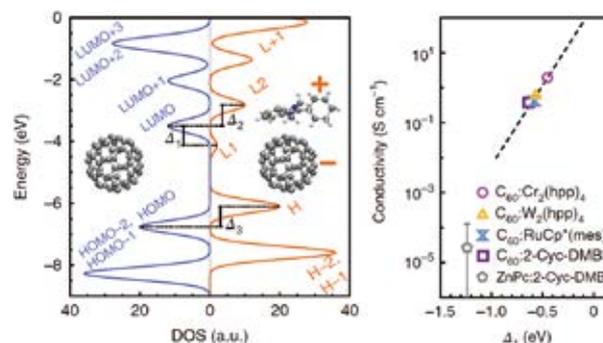


Figure 3. Left: DOS for neutral C_{60} (blue) and charged C_{60} next to a dopant (orange) in neat C_{60} bulk (14 molecules). Energy zero is aligned to vacuum and a Gaussian broadening of 0.2 eV is applied for illustration purposes. The introduction of dopants modifies the DOS of neat C_{60} considerably. While almost all levels of the anion shift upwards, the only one that shifts downwards is the occupied former LUMO of C_{60} (L1). Right: Conductivities in n-doped C_{60} at 2% concentration against Δ_1 for various molecular dopants. Tuning of Δ_1 may provide alternative strategies to optimize the electronic properties of organic semiconductors.

References

- 1) Y. Kashimoto, K. Yonezawa, M. Meissner, M. Gruenewald, T. Ueba, S. Kera, R. Forker, T. Fritz and H. Yoshida, *J. Phys. Chem. C* **122**, 12090–12097 (2018).
- 2) C. Gaul, M. Schwarze, S. Schellhammer, F. Bussolotti, S. Kera, Z. Bao, G. Cuniberti, K. Leo and F. Ortmann, *Nat. Mater.* **17**, 439–444 (2018).

Light Source Developments by Using Relativistic Electron Beams

UVSOR Synchrotron Facility Division of Advanced Accelerator Research



KATOH, Masahiro
Professor
[mkatoh@ims.ac.jp]

Education

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

Professional Employment

1986 Research 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

Member

Assistant Professor
FUJIMOTO, Masaki
Post-Doctoral Fellow
GUO, Lei
Graduate Student
TAKAHASHI, Kazuyoshi*
HAMADA, Ryo*

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. This is the result of the continuous effort on improving the machine. Our research group has been developing accelerator technologies toward producing bright and stable synchrotron light, such as high brightness electron beam optics, novel insertion devices or state-of-the-art beam injection technique. We have been also developing novel light source technologies toward producing photons with novel properties, such as free electron laser, coherent synchrotron radiation, optical vortices and laser Compton gamma-rays. We are also investigating beam physics which would be the basis of the novel light source technologies. We are exploring a future plan for 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–Electron Beam Interaction," *Nat. Phys.* **4**, 390–393 (2008).
- M. Shimada, M. Katoh, M. Adachi, T. Tanikawa, S. Kimura, M. Hosaka, N. Yamamoto, Y. Takashima and T. Takahashi, "Transverse-Longitudinal Coupling Effect in Laser Bunch Slicing," *Phys. Rev. Lett.* **103**, 144802 (2009).
- E. Roussel, C. Evain, C. Szwaj, S. Bielawski, J. Raasch, P. Thoma, A. Scheuring, M. Hofherr, K. Ilin, S. Wunsch, 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 developed novel 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 for developing 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 succeeded in producing such novel photon beams from undulators for the first time. We are exploring their applications in collaboration with Saga Light Source and Toyama Univ.

We have been investigating laser Compton scattering at BL1U to produce 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 proposed several upgrade plans since 2000 and



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

have carried out them successfully. We designed a special electron beam optics intended to higher brightness. We designed necessary accelerator components, reconstructed and commissioned the machine. 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 1 GeV 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 future upgrade plan, we are considering to replace some of the undulators to fit the changes of the users' requirements on the wavelength. As a long term plan, we have designed new accelerator systems such as a linear accelerator based free electron laser or 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 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
VINCINI, Giulio*
IKEMOTO, Masafumi†
KONTANI, Syouta†

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.

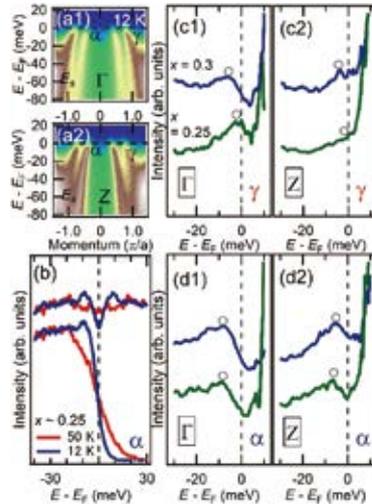


Figure 1. The superconducting (SC)-gap distribution on the hole FSs of K-Ba122 ($x \sim 0.25$). (a1)–(b2) The photoemission intensities taken along two momentum cuts near the BZ center with 21 and 32 eV photons. (b) Temperature dependence of ARPES spectra on K-Ba122 ($x \sim 0.25$) of k_F position at the Γ point. (c1), (c2) EDCs of the γ band at k_F position

divided by Fermi-Dirac (FD) distribution function for K-Ba122 ($x \sim 0.25, 0.3$). (d1) and (d2) are the same as in panel (c1) and (c2) of the α band. The gray circles in (c1)–(d2) are guides to the eyes for the value of the superconducting gap.

Selected Publications

- K. Tanaka, T. Yoshida, A. Fujimori, D. H. Lu, Z.-X. Shen, X.-J. Zhou, H. Eisaki, Z. Hussain, S. Uchida, Y. Aiura, K. Ono, T. Sugaya, T. Mizuno and I. Terasaki, “Effects of Next-Nearest-Neighbor Hopping t' on the Electronic Structure of Cuprates,” *Phys. Rev. B* **70**, 092503 (4 pages) (2004).
- 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).
- E. Uykur, K. Tanaka, T. Masui, S. Miyasaka and S. Tajima, “Coexistence of the Pseudogap and the Superconducting Gap Revealed by the c -Axis Optical Study of $\text{YBa}_2(\text{Cu}_{1-x}\text{Zn}_x)\text{O}_{7-\delta}$,” *J. Phys. Soc. Jpn.* **82**, 033701 (4 pages) (2013).

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

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

1. Suppression of Superconducting Gap on $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ Observed by Angle-Resolved Photoemission Spectroscopy

Iron-based superconductors have a complex phase diagram with the antiferromagnetic, structural, and superconducting (SC) transition phases as well as high- T_c cuprates superconductors.¹⁾ Recently, nematicity, defined as broken rotational symmetry [a trigonal (C_4)-to-orthorhombic (C_2) structural transition], has shed light on the understanding of the mechanism on the iron-based superconductivity in the underdoped regime. In hole-doped BaFe_2As_2 (Ba122) system, thermal expansion, specific heat, and neutron diffraction measurements of $\text{Ba}_{1-x}\text{Na}_x\text{Fe}_2\text{As}_2$ and $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ (K-Ba122) at a certain hole concentration have revealed the magnetic order without C_4 symmetry breaking in the underdoped regime.²⁾ Besides, these materials show that the SC transition temperature (T_c) is suppressed between the SC phase and the C_4 -magnetic phase by $\sim 20\text{--}50\%$. Although these intriguing physical properties have been come into the open in the hole-doped Ba122 system universally, the microscopic mechanism of the C_4 -magnetic phase and the T_c suppression has been unclear yet. In this study, we performed ARPES study on underdoped K-Ba122 ($x \sim 0.25$, $T_c \sim 26$ K) with the C_4 -magnetic phase and T_c suppression, to elucidate the mechanism of superconductivity.

Figures 1(a1) and 1(a2) show the ARPES intensity plot as a function of energy and momentum taken at $h\nu = 21$ eV and 32 eV, corresponding to the Γ and Z points, respectively. We observe at least two hole bands for Γ using s polarization, indicating that the degenerated d_{yz} , d_{xz} bands and the d_{xy} band are observed at the Γ point. Hereafter, hole bands at the BZ center are designated as α , β , and γ for the inner, middle, and outer hole FSs, respectively.

In order to elucidate the character of the SC gap on the hole FSs, Figure 1(b) shows the temperature dependence of the near- E_F ARPES spectrum measured across T_c (~ 26 K) at Fermi momentum (k_F) for the α band. The EDC shows a SC peak in the SC state (12 K) while that disappears in the normal state (50 K). To eliminate the effect of the Fermi-Dirac (FD) distribution function, we have symmetrized the ARPES spectra measured

above and below T_c as shown in Figure 1(b). On decreasing temperature, the spectral weight near E_F is transferred to high binding energy below T_c and the symmetrized EDC shows a peak at ~ 10 meV, suggesting the opening of a SC gap. FD-function-divided EDCs obtained from K-Ba122 ($x = 0.25$ and 0.3) at the k_F 's of the α and γ bands are shown in Figures 1(c1)–2(d2) for comparison. While the SC peak ($x = 0.3$) is far from the E_F , indicating that the SC gap is observed on the α and γ FSs, we found that the SC peak of $x \sim 0.25$ moves towards E_F , which indicates the decrease or almost close of SC gaps for the γ band.

Figure 2 gives the detailed SC gap distribution of the electron bands designated as ϵ and δ for the inner and outer electron FSs, respectively. To precisely determine the SC gap size and its momentum dependence, we have performed an ARPES study at several k_F and k_z points of the ϵ and δ bands. For the k_z direction, while the SC gap for the δ band shows an isotropic gap [Figure 2(g)], the EDC peak position reflecting the SC gap for the ϵ band does not seem to be constant, namely, the peak position as shown by a gray circle decreases in going from the A to M points [Figure 2(i)]. This indicates that an anisotropic SC gap is exhibited along the k_z direction for the ϵ FS.

The present ARPES study indicates that the T_c suppression of K-Ba122 ($x \sim 0.25$) corresponds to the suppressed SC gap on the γ FS and the k_z dependent SC gap on the ϵ FS. According to the previous ARPES study, the SC gap size on hole FSs was almost identical in optimally doped K-Ba122, which was interpreted by the s^{++} -wave SC gap symmetry due to orbital fluctuations. Though SC gaps on the α and β FSs of K-Ba122 ($x \sim 0.25$) are identical, that on the γ FS seems not to be explained directly by the orbital fluctuation. The SC gap might be suppressed in the presence of the C_4 -magnetic fluctuation, assuming that the presence of the C_4 symmetry of the electronic structure with disappearance of the orbital order. We also find that the ARPES intensity plots of K-Ba122 ($x = 0.21, 0.3$) at the BZ center show the band folding of the electron-like band, which is known to be observed below the SDW transition temperature in the underdoped regime, but disappears in $x \sim 0.25$ sample (not shown). Probably recently reported antiferroic electronic instability is suppressed around $x \sim 0.25$ because of the C_4 -magnetic phase fluctuation. Thus, the experimental evidence indicates that the SC gap on the γ FS of K-Ba122 ($x \sim 0.25$) is strongly influenced by the C_4 -magnetic phase.

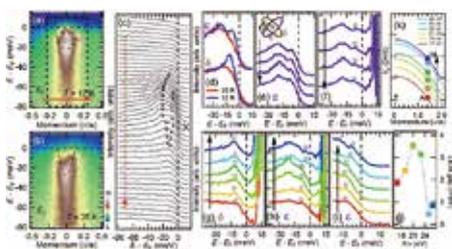


Figure 2. The SC-gap distribution on the electron FSs of K-Ba122 ($x \sim 0.25$). (a), (b) The photoemission intensities

taken at the momentum cut at the BZ corner with 23 eV photons below and above T_c , respectively. (c) Raw spectra of the data in (a) enclosed by a dotted line. (d) Temperature dependence of ARPES spectra for ϵ and δ bands at k_F . (e), (f) Raw spectra at k_F and that divided by the Fermi-Dirac function on the ϵ band taken at 23 eV photons. (g), (h) k_z dependence of the spectra divided Fermi-Dirac function measured on the ϵ and δ electron bands. (i), (j) The expansion of the spectra on the ϵ FS in the vicinity of E_F and the leading-edge midpoint for each EDC of (i), respectively. Measured k_z 's are taken at 17 eV–26 eV photons, corresponding to the k_z of the Γ -Z points shown in (k). The gray circles on the spectra are guides to the eyes for the peak position, reflecting SC gaps.

2. Development of Micro-Focused Beam ARPES

A soft X-ray beamline BL5U has been open for users from 2016 and used as high energy resolution ARPES beamline. By introducing a final focusing mirror close to the sample position (~ 50 mm), the synchrotron light whose original size was 400 (H) \times 120 (V) is successfully focused to 23 (H) \times 40 (V) μm . ARPES study on small samples or inhomogeneous samples is now available.

References

- 1) 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, *Science* **314**, 1910–1913 (2006).
- 2) R. M. Fernandes, A. V. Chubukov and J. Schmalian, *Nat. Phys.* **10**, 97–104 (2014).

Comprehensive Photoelectron Diffraction and Spectroscopy for Surface Science

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research



MATSUI, Fumihiko
Senior Researcher

Visualization of three-dimensional atomic arrangements of surfaces, interfaces, nanostructures, and molecular adsorbates is essential in materials science and engineering. This is accomplished by holographic reconstruction of element-specific photoelectron diffraction from the localized core level. Furthermore, the electronic properties and chemical reactivity of materials are closely related to the behavior of valence electrons near the Fermi level. Momentum-resolved valence-band (VB) photoelectron spectroscopy is a powerful technique to characterize such electrons. We have been working on the development of wide-acceptance-angle electron energy analyzers for photoelectron angular distribution measurement. In order to establish the reliability of these methods, comprehensive measurement and understanding of the photoelectron emission process are important.¹⁾

1. Valence Band Atomic Orbital Analysis of Layered Compounds

BL6U at UVSOR is a beamline dedicated to VB dispersion mapping with a practical photon energy range of 45 to 600 eV. Wide-acceptance-angle acquisition system enables measurement of full set of VB dispersion data over several Brillouin zones.²⁾ The cross sections of VB from MoSe₂ along

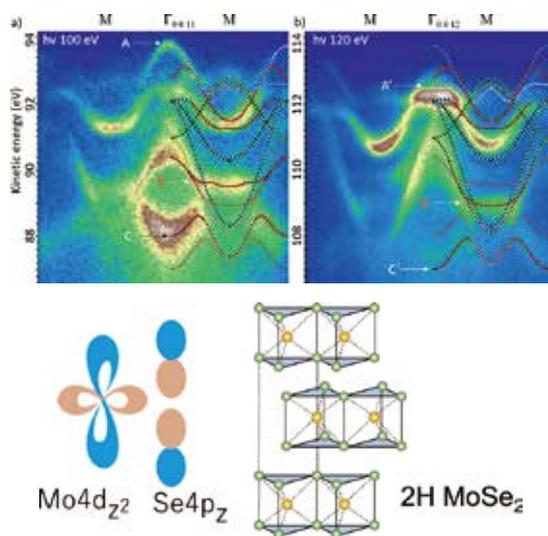


Figure 1. Cross section of MoSe₂ VB along the (a) $\Gamma_{00} 11M$ and (b) $\Gamma_{00} 12M$ directions together with ab initio simulation. Pairs of black and red dotted curves appear alternatively in the Brillouin zones along the k_z direction. Mo4d_{z²} is the key-player orbital.

the $\Gamma_{00} 11M$ and $\Gamma_{00} 12M$ directions are shown in Figure 1(a) and (b), respectively, together with the ab initio simulations. The interlayer interaction induces the splitting of the band degenerated at A point into two at Γ point. Band A, B, and C appeared intense at the $\Gamma_{00} 11M$ plane, while their counterparts, Band A', B', and C' appeared intense at the $\Gamma_{00} 12M$ plane. We found that the two split bands appear alternatively in the Brillouin zones along the k_z direction. This 4π -periodic k_z oscillation was also observed for graphite³⁾ and was explained by considering the photoelectron interference from atomic orbitals within the unit cell, *i.e.* photoelectron structure factor. We expanded this concept to the layered compound and investigated the bonding characters of these bands.

2. Momentum Microscope & PESCATORA

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 a special 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. My mission is to install state-of-the-art momentum microscope to UVSOR and establish a comprehensive photoemission experimental station.

Furthermore, we have recently invented a new type of display-type analyzer called **Projector for ESCA to Real space Analysis (PESCATORA)** using an electrostatic lens making the trajectory of photoelectrons emitted within 1π steradian into parallel rays and a fine tube array plate as a collimator for energy and angular distribution analysis, realizing the detection of photoelectron diffraction with a high angle resolution.⁴⁾

References

- 1) F. Matsui, T. Matsushita and H. Daimon, *J. Phys. Soc. Jpn.* **87**, 061004 (2018).
- 2) H. Yamane *et al.*, in preparation.
- 3) F. Matsui, H. Nishikawa, H. Daimon, M. Muntwiler, M. Takizawa, H. Namba and T. Greber, *Phys. Rev. B* **97**, 045430 (2018).
- 4) F. Matsui and H. Matsuda, Patent PCT/JJP2016/070744.

Photoelectron Spectroscopy of Ionic Liquids and Thin Films Made of Fullerenes and Their Application to Photoelectrochemistry

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



KATAYANAGI, Hideki
Assistant Professor

Electrodes made of fullerenes and their derivatives and electrolytes containing ionic liquids (ILs) are used in the photovoltaic devices such as dye-sensitized solar cells (DSSCs). Both electronic structures and mesoscopic mixing schemes of the materials are key issues which affect their efficiency and lifetime. These structures of materials can be investigated using a combined study of spectroscopic and thermodynamic measurements.

1. Fabrication of LB Film Electrodes of Sulfonated Fullerenes and Evaluation of Their Feasibility for the DSSCs

Carbon electrodes are candidates of substitutes for the Pt electrodes which are used in the most of present DSSC cells. To produce the carbon electrodes suitable for DSSC, we utilize the Langmuir-Blodgett (LB) technique and sulfonated fullerenes as building blocks. We first used stearic acid and

oleic acid to find out suitable conditions for producing thin and homogeneous LB films. We then fabricated the LB monolayers and multilayers of fullerene/fatty acid mixtures and the sulfonated fullerenes. We found that even the LB monolayers of sulfonated fullerenes have electrochemical reactant selectivity. We are trying to produce homogeneous multilayer films and examine the suitability for DSSC.

2. Stimulation Induced Phase Transition of Crystals of Salicylideneaniline Derivatives

Some crystals have solid phase transition induced by small energy such as mechanical forces and light irradiation. We have measured the Raman spectra of the crystals of salicylideneaniline derivatives before and after the phase transition. The Raman spectra were found to change gradually during the period of the phase transition. We will analyze the change of the Raman spectra and elucidate mechanisms of the transition. Knowledge on the phase transition induced by the small energy in three-dimensional space gives us clue to understand a key to control self-organization in two-dimensional films.

Observation of 3-Dimensional Chemical Nano-Structures of a Cell Nucleus

UVSOR Synchrotron Facility
Division of Beam Physics and Diagnostics Research



OHIGASHI, Takuji
Assistant Professor

Computed tomography (CT) is a powerful technique to observe 3-dimensional (3D) structure of a sample without any destructive process. In regard to relationship between spatial resolution and transmittance, CT by using a scanning transmission soft X-ray microscope (STXM) is a suitable combination. Moreover, this combination also enables to perform 3D spectro-microscopy by changing energies of incoming X-rays.¹⁾

3D distributions of DNA and protein in an isolated cell nucleus of a HeLa S3 at stage 0 (*i.e.* normal cell) were observed. The cell nucleus was fixed by glutaraldehyde and critical point drying without any additional staining. Since X-ray absorption spectra of DNA and protein show remarkable difference in N K-edge, 50 images stacks around N K-edge were acquired with rotating the sample 3.6° each (180° in total). The 3D distributions of DNA and protein were reconstructed after fitting each reference X-ray absorption

spectra to the energy stacks. A projection image of 3D distribution of DNA and cross sectional images of distributions of DNA and protein are shown in Figure 1.²⁾ Several cell nucleoli with high density and framework-like structures are clearly observed in Figure 1(b).

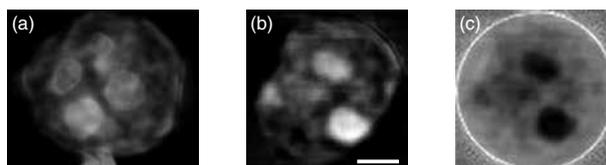


Figure 1. (a) A projection image of 3D distribution of DNA and cross sectional images of distributions of (b) DNA and (c) protein. Scale bar is 2 μm .

References

- 1) T. Ohigashi, Y. Inagaki, A. Ito, K. Shinohara and N. Kosugi, *J. Phys.: Conf. Ser.* **849**, 012044 (2017).
- 2) T. Ohigashi, A. Ito, K. Shinohara, S. Tone, Y. Inagaki, H. Yuzawa and N. Kosugi, *Microsc. Microanal.* **24**, 400–401 (2018).

Decay and Dissociation Dynamics of Core Excited Atoms, Molecules and Clusters

UVSOR Synchrotron Facility
Division of Advanced Photochemistry



IWAYAMA, Hiroshi
Assistant Professor

Core excited molecules are highly unstable and relaxes by Auger electron emission for light elements. The stability and dissociation dynamics of the Auger final states depend on their charge states and electronic states. We have investigated them by using an Auger-electron-photoion coincidence technique.

ing radiation. Recently, it was proposed that emission site and energy of the electron released during this process can be controlled by coupling the ICD to a resonant core excitation.¹⁾

We investigated ICD process of core excited nitrogen molecular clusters by using Auger-electron ion coincidence technique.²⁾ Since ICD process lower the double ionization threshold, we measured binding energy of Auger states corresponding to double ionizations. Compared to isolated nitrogen molecules, we observed lowering of double ionization threshold for nitrogen molecular clusters.

1. Intermolecular Coulombic Decay of Core Excited Nitrogen Molecular Clusters

Intermolecular Coulombic decay is an efficient electronic relaxation of excited molecules placed in a loosely bound chemical system (such as a hydrogen bounded or van-der-Waals-bounded cluster). This decay process ionize neighboring molecules to excited one and eject low-energy electrons, which play an important role in DNA damage induced ioniz-

References

- 1) K. Gokhberg *et al.*, *Nature* **505**, 661 (2014).
- 2) H. Iwayama *et al.*, *J. Chem. Phys.* **145**, 034305 (2016).

Award

IWAYAMA, Hiroshi; The 19th Young Scientist Awards of the Atomic Collision Society of Japan (2018).

Visiting Professors



Visiting Professor

KANAI, Kaname (*from Tokyo University of Science*)

Electronic Structure of Donor and Acceptor Interface in Organic Photovoltaics

It has been reported that microscopic structure at donor and acceptor (D/A) interface in organic photovoltaics has a key impact on the device performance. Especially, it has been believed that mixture inhomogeneity of donor and acceptor molecules at the interface significantly influences the efficiencies of free-charge generation and recombination. However, effects of the mixture inhomogeneity on the D/A interface remains elusive yet. Recently, we have been trying to figure out how the mixture inhomogeneity of donor and acceptor molecules affects the interface electronic structure at the D/A interface using photoemission spectroscopy (PES) at BL2B beamline of the UVSOR synchrotron facility. We found that the relatively strong intermolecular interaction between sexithiophenes (6T) and perfluorinated Cu-phthalocyanines ($F_{16}CuPc$) is key to have disordered molecular arrangements at the D/A interface by STM measurements. Now, we are trying to catch the subtle changes in the energy distribution of the molecular levels of 6T and $F_{16}CuPc$, respectively, in the mixed films using PES compared with isolated 6T and $F_{16}CuPc$'s films.



Visiting Associate Professor

KANEYASU, Tatsuo (*from SAGA Light Source*)

Generation and Application of Vortex Beams from Synchrotron Light Source

A vortex light beam having a helical wavefront carries orbital angular momentum (OAM) as well as the spin angular momentum associated with its circular polarization. Recently, it was found that the harmonic radiation from a helical undulator carries an OAM. This novel feature of the undulator radiation will allow us to utilize vortex beam at short wavelengths, and may open up new applications in synchrotron radiation research. In exploring new applications of the vortex beams, a fundamental understanding of the role of the OAM in the light-matter interaction is crucial. We have investigated the characteristics of the vortex beam in the extreme ultraviolet (XUV) region and applied it to the photoionization study of rare-gas atoms. To explore the vortex-atom interaction which is hardly observable in the conventional gas-phase experiment, we are developing a new experimental setup based on the combined use of laser and synchrotron light sources. In addition we plan to study the XUV vector beam which has space-varying polarization, and its interaction with atoms.