THE 9th INTERNATIONAL CONFERENCE
ON HARD X-Ray PHOTOELECTRON SPECTROSCOPY

HAXPES
2022

May 31 – June 3, 2022
Himeji, Japan

Conference format : In-person + Online hybrid
Place : Arcrea HIMEJI (Himeji Convention Center)

Early bird registration deadline : February 28, 2022
Abstract deadline : December 10, 2021 (Oral presentation)
January 31, 2022 (Poster presentation)

Co-chairs :
Eiji IKENAGA (Nagoya University)
Masaki OURA (RIKEN SPring-8 Center)
Akira SEKIYAMA (Osaka University)

Information and registration :http://rsc.riken.jp/haxpes2022/ **Contact :** haxpes2022@spring8.or.jp
Hosted by RIKEN SPring-8 Center & HAXPES2022 Organizing Committee

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Topic 6 : Techniques and Instrumentations
Topic 7 : New Trends and Applications

Japan (UTC+9)	5/31 (Tue.)		6/1 (Wed.)		6/2 (Thu.)		6/3 (Fri.)	
9:00			1a1-1 : Invited (On-site) **Hidenori Fujiwara**	Chair : S. Ueda	2a1-1 : Invited (On-line) **Slavomir Nemsak**	Chair : A. Gray	3a1-1 : Plenary (On-line) **Bongjin Simon Mun**	Chair : Y. Yamashita
9:30			1a1-2 : Oral (On-line) **Alexander X. Gray**		2a1-2 : Invited (On-site) **Shigenori Ueda**			
			1a1-3 : Oral (On-site) **Munetaka Taguchi**				3a1-2 : Invited (On-line) **Kelvin Zhang**	
10:00			Short break		2a1-3 : Oral (On-site) **Yoshiyuki Yamashita**			
			1a2-1 : Invited (On-line) **Eric L. Shirley**	Chair : K. Mimura	2a1-4 : Oral (On-site) **Lothar Weinhardt**		Breakout session	
10:30					2a1-5 : Invited (On-site) **Akira Yasui**			
			1a2-2 : Oral (On-site) **Hsun-Yun Chang**					
11:00			1a2-3 : Oral (On-line) **Trung-Phuc Vo**		Open discussion & Lunch		Closing Concluding remarks by **Wolfgang Drube** Next venue	
			1a2-4 : Oral (On-site) **Dirk Hauschild**					
11:30			Lunch session					
12:00								
12:30								
13:00			Company presentation	Chair : S. Yasuno				
13:30								
14:00								
			1p1-1 : Invited (On-site) **Atsushi Hariki**	Chair : M. Taguchi				
14:30					2p1-1 : Invited (On-line) **Stephen H. Southworth**	Chair : M. Oura		
			1p1-2 : Oral (On-line) **J. Matthias Kahk**					
15:00			1p1-3 : Oral **Victor Kimberg**		2p1-2 : Oral (On-line) **Minna Patanen**			
			Coffee break		2p1-3 : Oral (On-site) **Faris Gelmukhanov**			
15:30					2p1-4 : Plenary (On-line) **Marc Simon**			
			1p2-1 : Plenary (On-line) **Michael Sing**	Chair : (TBD)				
16:00								
					Coffee break			
16:30			1p2-2 : Oral (On-line) **Jan Minar**					
			1p2-3 : Oral (On-line) **Fredrik O.L. Johansson**		2p2-1 : Oral (On-line) **Natalia M. Martin**	Chair : T. -L. Lee		
17:00	Registration		1p2-4 : Oral (On-line) **Curran Kalha**		2p2-2 : Oral (On-line) **Andrew J. Naylor**			
			Short break		2p2-3 : Invited (On-line) **Sara Blomberg**			
17:30			1p3-1 : Oral (On-line) **J.-P. Rueff**	Chair : E. Ikenaga				
	Announcement		1p3-2 : Oral (On-site) **Daisuke Takegami**		2p2-4 : Oral (On-line) **Patrick Lömker**			
18:00	Opening : Tetsuya Ishikawa	Chair : W. Drube	1p3-3 : Oral (On-line) **Pierre-Marie Deleuze**		2p2-5 : Oral (On-line) **Eugenie Martinez**			
	31n1-1 : Plenary (On-line) **Gerd Schönhense**		1p3-4 : Oral (On-line) **Sunil Wilfred DSouza**		2p2-6 : Oral (On-line) **Dibya Phuyal**			
18:30			Memorial Talk Charles Fadley & Yasutaka Takata		Dinner session			
			Dinner & Poster session					
19:00	31n1-2 : Oral (On-line) **Andreas Lindblad**							
	31n1-3 : Oral (On-line) **Julius Hällstedt**							
19:30	31n1-4 : Invited (On-line) **Christoph Schlueter**							
20:00	31n1-5 : Oral (On-line) **Vladimir N. Strocov**							
	31n1-6 : Oral (On-line) **Martina Müller**							
20:30	Dinner & Night session		1n1-1 : Invited (On-site) **Marcus Bär**	Chair : R. Claessen				
21:00			1n1-2 : Oral (On-line) **Austin Irish**					
			1n1-3 : Oral (On-line) **Jay R. Paudel**					
21:30					IAB meeting			

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Akira Yasui	JASRI, Japan
Satoshi Yasuno	JASRI, Japan

Program

Tuesday, 31 May, Hotel Monterey Himeji

17:00 - 17:45 Registration
17:45 - 18:00 Announcement

18:00 - 18:15 Opening, **Tetsuya Ishikawa** (RIKEN SPring-8 Center)

Chair: Wolfgang Drube

18:15 - 19:00	Plenary talk **Gerd Schönhense** (Johannes Gutenberg University, Germany) “Momentum-resolved hard x-ray photoemission and photoelectron diffraction using time-of-flight recording” (On-line)
19:00 - 19:15	**Andreas Lindblad** (Uppsala University, Sweden) “First results from the HAXPES-lab@UU facility” (On-line)
19:15 - 19:30	**Julius Hållstedt** (Excillum AB, Sweden) “High performance X-ray sources for X-ray photoelectron spectroscopy” (On-line)
19:30 - 20:00	Invited talk **Christoph Schlueter** (Deutsches Elektronen-Synchrotron DEZY, Germany) “HAXPES at PETRA III and IV: electronic structure, operando devices and in-situ catalysis” (On-line)
20:00 - 20:15	**Vladimir N. Strocov** (Swiss Light Source, Switzerland) “k-resolved electronic structure and band offsets at the NbN/GaN superconductor/semiconductor heterojunction” (On-line)
20:15 - 20:30	**Martina Müller** (University of Konstanz, Germany) “How dopants and defects stabilize ferroelectricity in HfO_2 - a spectroscopic approach” (On-line)

17:30 - 18:00 Dinner & Night session

Wednesday, 1 June, Arcrea Himeji (Himeji Convention Center)

Chair: Shigenori Ueda

9:00 - 9:30	Invited talk **Hidenori Fujiwara** (Osaka University, Japan) “Probing spin-polarized electronic structures on half-metallic Heusler alloys using combined bulk-sensitive spectroscopies”
9:30 - 9:45	**Alexander X. Gray** (Temple University, USA) “Probing strain-engineered anion ordering via depth-sensitive and polarization-dependent X-ray spectroscopic techniques” (On-line)
9:45 - 10:00	**Munetaka Taguchi** (Toshiba Nanoanalysis Corporation, Japan) “Total reflection hard X-ray photoelectron spectroscopy for $La_{1-x}Sr_xMnO_3$ and $La_{1.9}Ce_{0.1}CuO_4$”

10:00 – 10:15 Short break

Chair: Kojiro Mimura

10:15 - 10:45	Invited Talk **Eric L. Shirley** (National Institute of Standards and Technology, USA) “Theoretical aspects of resonant Auger processes in solids” (On-line)
10:45 - 11:00	**Hsun-Yun Chang** (ULVAC-PHI Inc., Japan) “Accurate and non-destructive layer structure analysis of semiconductor materials using angle resolved HAXPES”
11:00 - 11:15	**Trung-Phuc Vo** (University of West Bohemia, Czech Republic) “Theoretical and experimental HARPES study of Weyl-semimetal TaAs: The application of machine-learning” (On-line)
11:15 - 11:30	**Dirk Hauschild** (Karlsruhe Institute of Technology, Germany) “Core-level peaks and the inelastic background in HAXPES: a depth-resolved analysis of the CdS/Cu(In,Ga)Se_2 interface for thin-film solar cells”

11:30 – 13:00 Lunch session

13:00 – 14:00 Company presentation

Chair: Satoshi Yasuno

13:00 - 13:15	**Andreas Thissen** (SPECS, Germany) “From SXPS to HAXPES – instrumentation and applications of surface and bulk chemical analysis in the laboratory”
13:15 - 13:30	**Marten Patt** (FOCUS GmbH, Germany) “NanoESCA, HAXPEEM, xTOF – An evolution in hard x-ray microscopy” (On-line)
13:30 - 13:45	**Peter Amann** (Scienta Omicron, Sweden) “Hard X-ray photoemission spectroscopy possibilities at Scienta Omicron” (On-line)
13:45 - 14:00	**Yoshio Ichii** (Jtec Corporation, Japan) ”New Optical Devices for Next-Generation Synchrotron Facilities”

14:00 – 14:15 Short break

Chair: Munetaka Taguchi

14:15 - 14:45	Invited Talk **Atsushi Hariki** (Osaka Metropolitan University, Japan) “LDA+DMFT approach to core-level x-ray spectroscopies in correlated electron systems”
14:45 - 15:00	**J. Matthias Kahk** (University of Tartu, Estonia) “Theoretical prediction of core electron binding energies using the ⊿SCF method-application to 3d transition metals” (On-line)
15:00 - 15:15	**Victor Kimberg** (KTH Royal Institute of Technology, Sweden) “Time-resolved study of recoil-induced rotation by X-ray pump - X-ray probe spectroscopy”

15:15 – 15:45 Coffee break

Chair: (TBD)

15:45 - 16:30	Plenary Talk **Michael Sing** (Julius-Maximiliians-Universität Würzburg, Germany) "Hard x-ray photoelectron spectroscopy of functional oxide films and heterointerfaces"(On-line)
16:30 - 16:45	**Jan Minar** (University of West Bohemia, Czech Republic) "Proximity-induced spin-orbit interaction in $MoSe_2$ overlayered with amorphous Pb films " (On-line)
16:45 - 17:00	**Fredrik O.L. Johansson** (Sorbonne Universite, France) "Studying anisotropic electronic structure in 2D-layered crystals using resonant Auger spectroscopy" (On-line)
17:00 - 17:15	**Curran Kalha** (University of College London, UK) "Dynamic SXPS and HAXPES study of the thermal stability of Ti_xW_{1-x}/Cu heterostructures" (On-line)

17:15 – 17:30 Short break

Chair: Eiji Ikenaga

17:30 - 17:45	**Jean Pascal Rueff** (Synchrotron SOLEIL, France) "Probing quantum materials interfaces with HAXPES at the GALAXIES beamline : results and perspectives" (On-line)
17:45 - 18:00	**Daisuke Takegami** (Max Planck Institute Chemical Physics of Solids, Germany) "$CaCu_3Ru_4O_{12}$: a high Kondo-temperature transition metal oxide"
18:00 - 18:15	**Pierre-Marie Deleuze** (Université Grenoble-Alpes, France) "Lab-Scale hard X-ray photoelectron spectroscopy for the analysis of buried interfaces in device technologies" (On-line)
18:15 - 18:30	**Sunil Wilfred DSouza** (University of West Bohemia, Czech Republic) "Metallic antiferromagnetic spintronics: Mn_2Au a case study" (On-line)

Memorial Talk

18:30 - 18:45	**Memorial Talk** **Charles Fadley & Yasutaka Takata**

18:45 – 20:30 Dinner & Poster session

Chair: Ralph Claessen

20:30 - 21:00	Invited Talk **Marcus Bär** (Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Germany) “Materials research for metal-halide Perovskite solar cells enabled by hard x-ray photoelectron spectroscopy”
21:00 - 21:15	**Austin Irish** (Lund University, Sweden) “Characterization of ferroelectric devices: HfZrO thin films on InAs” (On-line)
21:15 - 21:30	**Jay R. Paudel** (Temple University, USA) “Combining depth-sensitive X-ray spectroscopic and scattering techniques to probe interfacial magnetic phenomena in oxide superlattices” (On-line)

Thursday, 2 June, Arcrea Himeji (Himeji Convention Center)

Chair: Alexander Gray

9:00 - 9:30	Invited Talk **Slavomir Nemsak** (Advanced Light Source, USA) “Using X-ray optical effects in photoelectron spectroscopy to study heterogeneous systems” (On-line)
9:30 - 10:00	Invited Talk **Shigenori Ueda** (National Institute for Materials Science, Japan) “New dimensions of HAXPES: variable polarization, total reflection, and spin resolution”
10:00 - 10:15	**Yoshiyuki Yamashita** (National Institute for Materials Science, Japan) “Gap states at SiO_2/4H-SiC interface obtained from operando hard x-ray photoelectron spectroscopy”
10:15 - 10:30	**Lothar Weinhardt** (Karlsruhe Institute of Technology) “X-SPEC : A 70eV to 15keV undulator beamline for electron and x-ray spectroscopies”
10:30 - 11:00	Invited Talk **Akira Yasui** (Japan Synchrotron Radiation Research Institute, Japan) “Upgrading of a HAXPES beamline BL09XU of SPring-8”

11:00 – 14:30 Open discussion & Lunch

Chair: Masaki Oura

14:30 - 15:00	Invited Talk **Stephen H. Southworth** (Argonne National laboratory, USA) “Atomic inner-shell resonance, threshold, and double ionization studies” (On-line)
15:00 - 15:15	**Minna Patanen** (University of Oulu, Finland) “First HAXPES results on clusters : K-shell photoexcitation and subsequent electronic decays in Ar clusters” (On-line)

15:15 - 15:30	**Faris Gelmukhanov** (KTH Royal Institute of Technology, Sweden) “Translational and rotational recoil effects in HAXPES of free molecules”
15:30 - 16:15	Plenary Talk **Marc Simon** (CNRS & Sorbonne University, France) “Gas phase HAXPES studies” (On-line)

16:15 – 16:45 Coffee break

Chair: Tien-Lin Lee

16:45 - 17:00	**Natalia M. Martin** (Uppsala University, Sweden) “Depth-resolved HAXPES study of (Ag,Cu)(In,Ga)Se_2 thin film solar cells” (On-line)
17:00 - 17:15	**Andrew J. Naylor** (Uppsala University, Sweden) “Using hard X-ray photoelectron spectroscopy to probe rechargeable battery interfaces” (On-line)
17:15 - 17:45	Invited Talk **Sara Blomberg** (Lund University, Sweden) “Operando high pressure catalytic studies using HAXPES” (On-line)
17:45 - 18:00	**Patrick Lömker** (Stockholm University, Sweden) “Co(0001) Fischer-Tropsch observations at 550mbar (and above) using a virtual cell approach and hard x-rays” (On-line)
18:00 - 18:15	**Eugenie Martinez** (Université Grenoble Alpes, France) “Laboratory HAXPES of GaN structures for power electronics” (On-line)
18:15 - 18:30	**Dibya Phuyal** (KTH - Royal Institute of Technology, Sweden) “Strong dimensional character in layered ReS_2 and ReSe studied by complimentary X-ray photoelectron spectroscopy” (On-line)

18:30 – 21:00 Dinner session
21:30 – 22:30 International Advisory Board (IAB) meeting

Friday, 3 June, Arcrea Himeji (Himeji Convention Center)

Chair: Yoshiyuki Yamashita

9:00 - 9:45	Plenary Talk **Bongjin Simon Mun** (Gwangju Institute of Science and Technology, Korea) “Exploring surface catalytic nature with ambient pressure XPS” (On-line)
9:45 - 10:15	Invited Talk **Kelvin Zhang** (Xiamen University, China) “The electronic structure of degenerately doped Ga_2O_3 thin files studied by HAXPES” (On-line)

10:15 – 11:00 Breakout session

11:00 – 12:00 Closing
Concluding remarks by **Wolfgang Drube**
Next venue

Poster session

P01	Resonant Auger of Xe on transition metal substrates **Elin Berggren** – Uppsala University (On-line)
P02	In-plane orientation of the 4f-orbital distribution in $YbRh_2Si_2$ probed by linear dichroism in core-level HAXPES **Akane Ose** – Osaka University
P03	Magnetometry of buried Co-based nanolayers by hard x-ray photoelectron spectroscopy **Andrei Hloskovsky** – DESY (On-line)
P04	Bulk electronic state of Au-Ga-Ce and Cd-Ce quasicrystalline approximants probed by hard x-ray photoemission spectroscopy **Goro Nozue** – Osaka University
P05	Electronic structure of thermoelectric Heusler-type $Co_2MnSi_{1-x}Al_x$ compounds **Hidetoshi Miyazaki** – Nagoya Institute of Technology
P06	Valence fluctuation in thermoeletric material $YbSi_2$ probed by HAXPES **Kentaro Kuga** – Toyota Technological Institute
P07	Relationship of Ce 4f-5d coulomb repulsion on CeT_2X_2 (T=Cu,Ru; X=Si,Ge): resonant HAXPES study **Kojiro Mimura** – Osaka Metropolitan University
P08	Charge transfer energy in iridates : a hard X-ray photoelectron spectroscopy study **Liu Hao Tjeng** – Max Planck Institute Chemical Physics of Solids (On-line)
P09	Observation of ferroelectric skewed band structure in multiferroic $BiFeO_3$ **Shibei Huang** – Okayama University
P10	Dy 3d core-level photoemission lineshape and its correspondence with chemical state **Shin Imada** – Ritsumeikan University
P11	Resonant hard x-ray photoemission on valence fluctuating system α-$Yb(Al_{1-x}Fe_x)B_4$ **Takuma H. Nishioka** – Osaka University
P12	Systematic charge distribution changes in Bi, Pb-3d transition metal perovskite oxides **Takumi Nishikubo** – Kanagawa Institute of Industrial Science and Technology
P13	Electronic states of FeGa alloy studied by hard and soft x-ray photoelectron spectroscopy **Tetsuya Miyazaki** – Ritsumeikan University

Poster session

P14	Electronic structure of covalent-chain antiferromagnets $TlFeX_2$ (X=S,Se) studied by HAXPES **Tomoki Shinozuka** – Osaka Metropolitan University
P15	Electronic states of type-II Na-Ba-Si clathrete studied by hard x-ray photoelectron spectroscopy **Yuchen Chen** – Ritsumeikan University
P16	Ir 5d state of $CuIr_2S_4$ studied by Ir $2p_{3/2}$-5d resonant HAXPES **Yukihiro Taguchi** – Osaka Metropolitan University (On-line)
P17	Electronic structure of Co_2NbSn studied by hard x-ray photoelectron spectroscopy **Yuuki Shimada** – Ritsumeikan University
P18	Trial of hard x-ray photoemission electron microscopy (HAXPEEM) at SPring-8 **Satoru Hamamoto** – RIKEN SPring-8 Center
P19	Ground state symmetry of rare-earth ions and emission angle dependence in resonant x-ray photoemission **Hisashi Soma** – Osaka Prefecture University
P20	Core-level photoemission spectroscopy of 3d transition metal oxides : DFT+DMFT analysis **Keisuke Higashi** – Osaka Prefecture University
P21	Design of an in-situ, in-operando HAXPES cell for the characterization of SEI formation in Li-ion batteries **Federico Giovanni Capone** – PHENIX Lab., Sorbonne Universite
P22	APXPS study on $Pt_3Pd_3Sn_2/C$ catalyst for dimethyl ether oxidation **Jan Minar** – University of West Bohemia
P23	First HAXPES studies of photovoltaic materials at EMIL: depth-resolved structure of $(Ag,Cu)(In,Ga)Se_2$ **Regan G. Wilks** – Helmholtz-Zentrum Berlin fur Materialien und Energie GmbH
P24	HZB energy materials research at SPring-8 BL15XU 2018-2019 **Regan G. Wilks** – Helmholtz-Zentrum Berlin fur Materialien und Energie GmbH
P25	X-ray spectroscopic study of the electronic states of Pd nanoparticles **Satoshi Ogawa** – Nagoya University
P26	Spectroscopic study and photocatalytic activity of Au-Ni nanoparticles for hydrogen production reaction by water splitting **Takumi Kawaguchi** – Nagoya University

Poster session

P27	The "Two-Color" EMIL beamline at the BESSY II light source **Mihaela Gorgoi** – Helmholtz-Zentrum Berlin fur Materialien und Energie GmbH (On-line)
P28	Effects of Sample-Aperture Cone Distance on Environmental Charge Compensation in Hard X-ray Photoelectron **Satoru Suzuki** – LASTI, University of Hyogo (On-line)
P29	Hard x-ray photoelectron spectroscopy beamline designed at Shenzhen Innovation Light-Source Facility (SILF) **Tianhui Liu** – Institute of Advanced Science Facilities (On-line)
P30	Three-dimensional spatial-resolved HAXPES measurement using a 1 mm focused beam **Yasumasa Takagi** – Japan Synchrotron Radiation Research Institute
P31	The operande measurement for bias applied technique using laboratory based hard x-ray photoelectron spectroscopy with Ga Ka x-ray source **Tappei Nishihara** – Meiji University
P32	Study on the relationship between chemical structure and function of amphiphilic gel surface **Eri Ito** – Nagoya Institute of Technology

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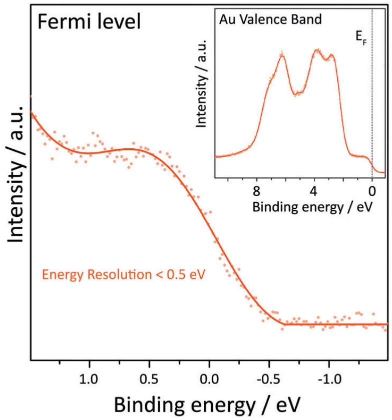
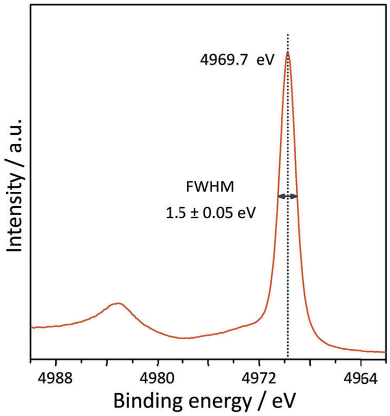
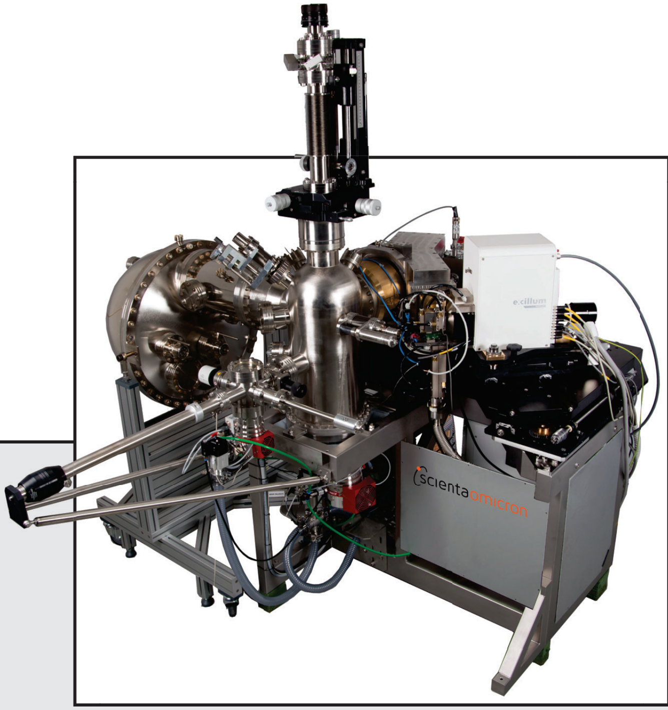
HAXPES Lab

A Window to the Bulk







HAXPES Lab Features:

- Destruction free information from deep below the surface
- Designed for fast data acquisition
- From Synchrotron to home lab: 24/7 access to HAXPES

Top left: Fermi level measurement on Au reveals fantastic instrumental energy resolution.
Bottom left: Ti 1s from a single crystal rutile TiO_2 measured with 9.25 keV Ga-source. Measurement time: 35 min.

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Hard X-ray photoemission spectroscopy possibilities at Scienta Omicron

P. Amann[1,2], M. Masatake[3], P. Palmgren[1], T. Wiell[1], M. Lundwall[1], S. Eriksson[1]
[1] *Scienta Omicron AB, Danmarksgatan 22, 75323 Uppsala, Sweden*
[2] *Scienta Omicron GmbH, Limburger Strasse 75, 65232 Taunusstein, Germany*
[3] *Scienta Omicron, Inc., 6–17–10, MinamiOi, Shinagawa-ku, Tokyo 140–0013, Japan*

X-ray photoemission spectroscopy (XPS) using photons from Al Kα is a powerful method to investigate the chemical nature of surfaces. However, the investigation of buried interfaces occurring in, e.g., device electronics or the work with elevated pressure or solid-liquid interfaces is difficult as the energy of the created photoelectrons is not high enough and scattering inside the material bulk limit the detected signal intensity.

During the past decade, increased attention has been shown to hard X-rays in the photoelectron spectroscopy field [1]. This opens a path towards increased information depth and allows probing high binding energy core levels.

In this talk I will give an overview on the Scienta Omicron products: HAXPES Lab, BAR XPS and HiPPHAXPES Lab.

The HAXPES Lab is a high-end laboratory based setup featuring a monochromated X-ray source creating Ga Kα radiation at 9.25 keV and a dedicated electron analyser with +/- 30° wide acceptance angle [2,3]. With this novel base system, the investigation of buried interfaces becomes possible. The increased probing depth allows for destruction free investigation of device electronics making operando measurements attractive.

The BAR XPS is a dedicated ambient pressure HAXPES synchrotron end-station that integrates the virtual cell concept and allows for operation under unprecedented high-pressure conditions [4]. Grazing incidence spectroscopy allows for high surface sensitivity and the small cell volume of below ca. 0,1 mm^3 allows for fast gas exchange times at the sample allowing to investigate catalyst dynamics.

The HiPPHAXPES Lab combines the features of the HAXPES Lab and the Bar XPS in a laboratory based ambient pressure instrument. This is made possible by the small spot size of the Ga X-ray source of about 30 x 45 μm^2, which is combined with the ambient pressure HiPP-2 Analyser allowing for operation up to 10 keV.

[1] C. Kalha, N.K. Fernando, P. Bhatt, F.O.L. Johansson, A. Lindblad, H. Rensmo, L.Z. Medina, R. Lindblad, S. Siol, L.P.H. Jeurgens, C. Cancellieri, K. Rossnagel, K. Medjanik, G. Schonhense, M. Simon, A.X. Gray, S. Nemsak, P. Lomker, C. Schlueter, A. Regoutz, J. Phys. Condens. Matter 33 (2021) 233001.

[2] A. Regoutz, M. Mascheck, T. Wiell, S.K. Eriksson, C. Liljenberg, K. Tetzner, B.A.D. Williamson, D.O. Scanlon, P. Palmgren, Rev. Sci. Instrum. 89 (2018).

[3] T. Hashimoto, P. Amann, A. Regoutz, N. Barrett, L.F.J. Piper, W. Hamouda, O. Renault, M. Lundwall, M. Masatake, Vac. Surf. Sci. 64 (2021) 493–498.

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TOYAMA's Monozukuri Center Opened in April 2015.

TOYAMA's new factory, *Monozukuri* Center, has a total floor area of 12,400 m², including 1,900 m² of machining & workshop areas, 2,400 m² of assembly & test areas and 800 m² of clean room space.

***Monozukuri* is a term which is used to describe Japanese manufacturing processes. It is difficult to translate into English, literally "*mono*" is the thing that is made and "*zukuri*" is the act of making. "*Monozukuri*", however, has a deeper meaning beyond the literal which describes the craftsmanship involved in making objects with skill, artistry and continuous improvement. It might be defined as "The art, science and craft of making things".**

Assembly and Test Area

- Vacuum baking and system test
- Assembling

Machine Shop

Crane: 2.8t, Lifting range: 6m


Vertical CNC lathe


General lathe


Horizontal NC Machine

- 6 general lathes
- 8 general milling machines
- 6 machining centers
- 2 Horizontal NC Mchines
- Multitasking machine *etc.*


Wire electric discharge machine


Multitasking machine

New!

- Vertical CNC lathe
- Wire electric discharge machine

Assembly, Weld and Test Area

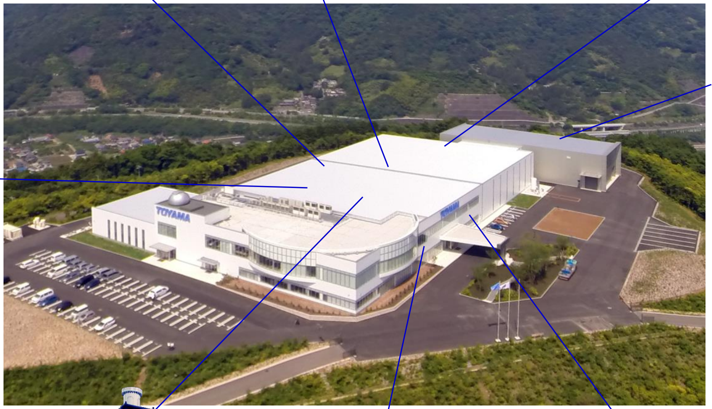

Clean room (Class 1000/10000)

Temperature Control Room


3D coordinate measuring machine

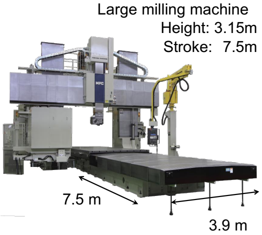

Ultra-precision machining

- 3D coordinate measuring machine
- Ultra-precision machining and measuring systems
- 3D optical profiler
- Laser interferometer
- Auto collimator *etc.*



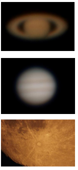
Large Machine Shop

Crane: 5t, Lifting range: 6m

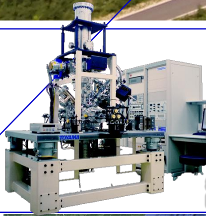


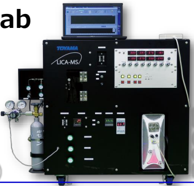
Observatory Room

40 cm Cassegrain reflecting telescope



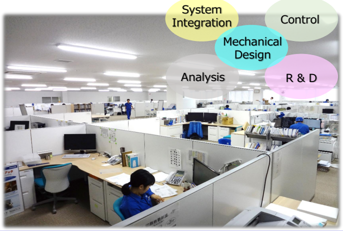
R&D Lab


FILMER (FIB-TOF-SIMS: Focused Ion Beam Time-Of-Flight Secondary Ion Mass Spectrometer)


LICA-MS (Laser Ionization Compact Analyzer Mass Spectrometer)

Engineering Room

Capacity: Over 100 seats



Main Entrance and Mt. Fuji

Vacuum Furnace Area

NanoESCA, HAXPEEM, xTOF – An evolution in hard x-ray microscopy

Marten Patt[1], Nils Weber[1], Matthias Escher[1], Michael Merkel[1]

[1]*FOCUS GmbH, 65510 Hünstetten, Germany*

The NanoESCA was initially developed as photoemission electron microscope (PEEM) for real-space imaging, combined with a powerful double-hemispherical energy band-pass filter. As such it is already a powerful tool for work function mapping and for chemical imaging of nanostructured samples with unsurpassed lateral resolution (if combined with an x-ray source) [1]. Soon after, the concept was extended to image the momentum distribution of the photoelectrons [2], generally excited by VUV sources. The combination of energy-filtered real-space and momentum-space microscopy makes it the most versatile photoemission instruments of today, addressing the questions of future device engineering with respect to their chemical states or electronic properties. Micro-ARPES becomes possible by isolating a small region of interest in real-space with an aperture while projecting the full angular distribution of electrons from this region onto the imaging detector unit.

To take advantage of the higher probing depth of HAXPES experiments, the NanoESCA was extended to work with high kinetic energy electrons. This HAXPEEM experiment allows for chemical and lateral resolved images of active layers beneath a top electrode [3], making it a promising technique for the analysis of microstructured and layered samples in material science and microelectronics [4].

An increase of efficiency of the HAXPEEM approach can be reached by combining the double-hemispherical band-pass filter of the NanoESCA with an imaging time-of-flight detector, i.e. a delay line detector. While the latter allows for parallel detection of lateral and energy coordinates, the upstream band-pass filter cuts off unwanted energies and avoids overlapping spectra generated from subsequent pulses at high repetition rates. We will present the concept and initial proof-of-principal measurements of this patented new xTOF approach [5, 6].

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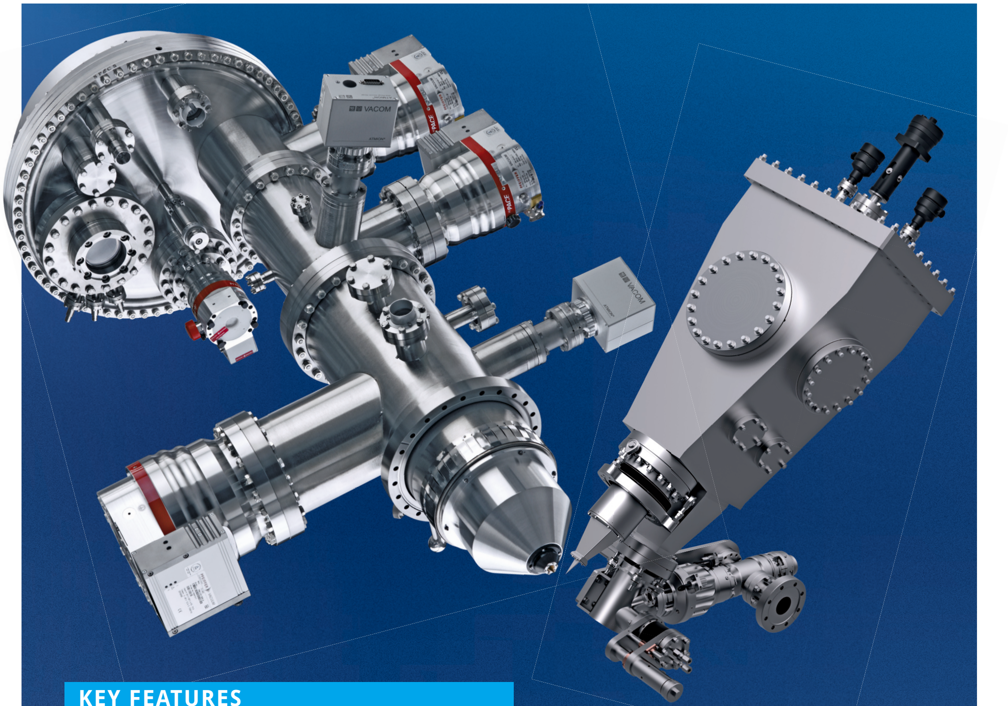

From SXPS to HAXPES – instrumentation and applications of surface and bulk chemical analysis in the laboratory

Andreas Thissen [1], Deler Langenberg [1]

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In recent years, there has been an increasing demand in hard X-ray photoelectron spectroscopy (HAXPES) depth and operand measurements in the study of devices especially the bulk and interface characteristics.

To gain access to the bulk and interface states of the material, it is necessary to increase the kinetic energy of the photoelectrons using high-energy X-rays. The photon energies of HAXPES typically range from 3 to 15 keV, and information can be obtained from depths of up to 100 Å. Therefore, it is particularly effective for extracting information on electronic and magnetic states from the inner shell spectrum in materials with greatly different surface and bulk properties, such as thin films and strongly correlated materials.

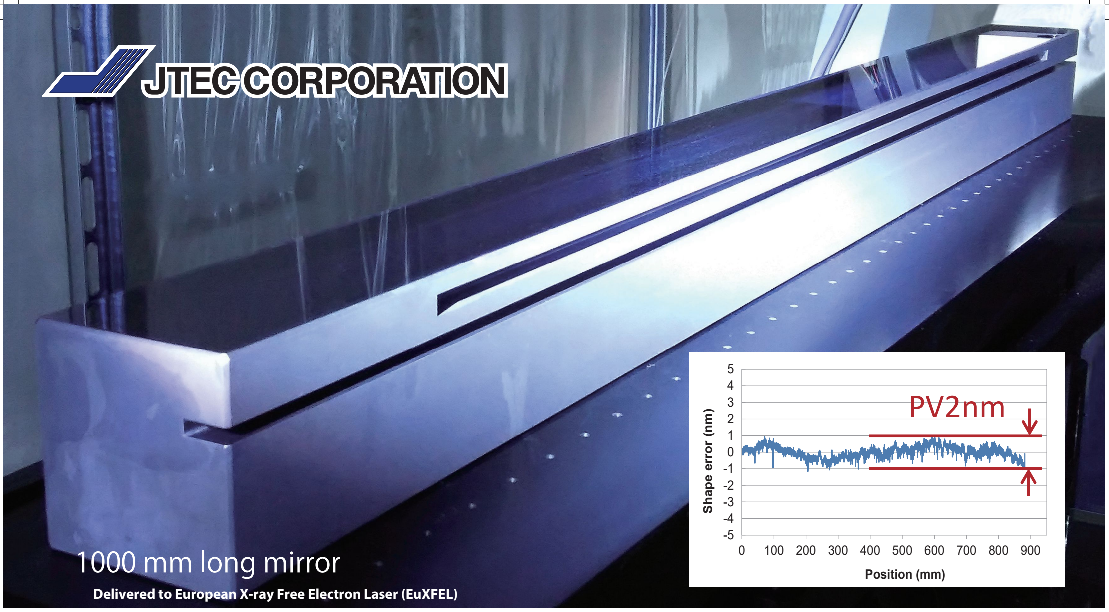
However, special consideration is required for the equipment due to the low photoionization cross section at high excitation energies. Therefore, a light source with high photon density and efficient electron detection are important.

In this announcement, we will introduce various HAXPES products from SPECS.

KEY FEATURES

- High transmission analyzer series for kinetic energies up to 7 keV, 10 keV and 15 keV
- Small spot X-ray monochromator sources with highest flux densities for any combination of Al K_α, Ag L_α and Cr K_α
- Flexible but proven system integration for any customer application
- Available for UHV (10^{-11} mbar) to Near Ambient Pressure (up to 200 mbar) conditions

***Fig.** 1: sources and analyzers for chemical composition studies of surfaces and bulk materials in laboratory environments.*



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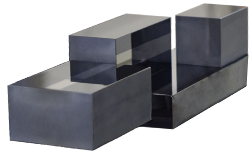

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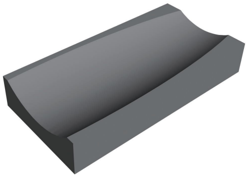

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Abstract
Oral session

Momentum-resolved hard X-ray photoemission and photoelectron diffraction using time-of-flight recording

Gerd Schönhense

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Momentum microscopy (MM) is a novel way of performing ARPES. Combined with time-of-flight (ToF) recording, its high parallelization is advantageous for photon-hungry experiments like X-ray ARPES, spin-resolved and time-resolved ARPES. Here we introduce the technique and illustrate its performance by selected examples obtained at hard X-ray energies. High-energy MM uses the same recording scheme as the low-energy variant [1], but a new electron optics enables energies up to >7keV and large k-fields-of-view comprising up to tens of Brillouin zones (BZs). Next to valence-band mapping, effective recording of XPD patterns [2] yields structural information. The new microscope can host an imaging spin filter; for details, see [3].

Fig. 1 shows an example for the Kondo system $YbRh_2Si_2$, where fast 4D mapping revealed the temperature dependence of the Fermi surface [4]. The XPD pattern (a) shows the two (110)-type Kikuchi bands (width 3.1$Å^{-1}$ given by the reciprocal lattice vector $2G_{110}$). The k-field of >12 $Å^{-1}$ corresponds to five adjacent BZs. This enables capturing the full 3D-dispersion of valence bands without changing photon energy. The perpendicular momentum component along k_z decreases with increasing parallel momentum k_x, Fig. 1(b,c). The energy conservation law forces all final states on an *energy isosphere* with radius k_f. Due to the curvature, k_z covers a range of Δk_z=0.5 G_{001} in one experimental run at fixed hν. Precise setting to hν=5297eV yields $|\boldsymbol{k}_f|$=29 G_{001}, so that in the image center the sphere intersects the 29th repeated BZ through the Γ-point. With increasing radius, k_z continuously drops toward the border of the 5th BZ, Fig. 1(c). The energy isosurfaces in (k_x,k_y,k_z) (d) have been captured at a single photon energy of hν=5297 eV [4,3]. First fs time-resolved measurements have been performed using soft X-rays at FLASH [5].

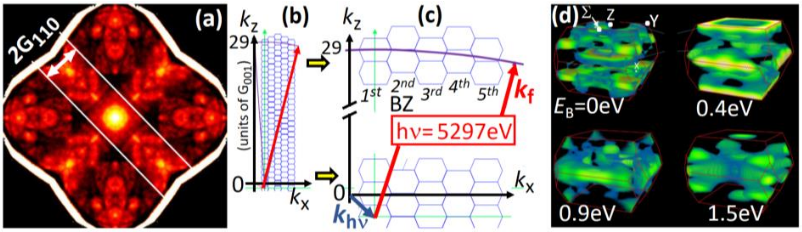

Fig. 1: *Hard-X-ray analysis of the Kondo system $YbRh_2Si_2$. (a) XPD at the Yb 4f level probes the geometric structure and provides a metric in k-space. (b,c) The final-state energy isosphere intersects adjacent BZs at different k_z, accessing the full 3D energy isosurfaces (d) without changing photon energy.*

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First results from the HAXPES-lab@UU facility

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An in-house HAXPES-lab facility built up around a liquid metal jet (Ga Kα) hard X-ray and an Al Kα soft X-ray source have been recently established at our department in Uppsala. Since the instrument is meant to study a broad range of materials physics problems – from fundamental interface properties to applied studies of *e.g.* batteries and photovoltaics *in operando*, the capabilities of the system needs to be implemented with this in mind from the outset. I will present the first results from the system together with how the facility fits with other hard/ tender X-ray PES initiatives in Sweden and our roadmap to develop the facility further with complementary capabilities.

High performance X-ray sources for X-ray photoelectron spectrocopy

Julius Hållstedt

Excillum AB, Jan Stenbecks Torg 17, 164 40 Kista, Sweden

High end X-ray photoelectron spectroscopy (XPS) relies heavily on the X-ray source performance for the analytical output. In principle there is a trade off between X-ray flux, monochromatizating and beam size which traditionally has especially for hard energy XPS (HAXPES) led to very long measurement times and poor signal to noise. In the recent years however, X-ray sources based on liquid-metal-jet technology (MetalJet) has successfully been introduced into HAXPES community in many labs across the globe. The increased power loading capability from MetalJet sources is explained by the fact the electrons are in MetalJet sources impinging on a continuously generated liquid alloy compared to a static solid anode in standard X-ray tubes, see fig 1. This significantly increase X-ray brightness and enables small X-ray spots in combination with high powers. This has enabled the electron spectroscopist with the convenience of HAXPES home lab capability in combination with excellent analytical results at short measurement times [1].

This communication will discuss the recent development with the MetalJet technology, its scalability, suitability and future potential for electron spectroscopy. We will also present recent result how total power has increased with a factor four (4) compared to first generation MetalJets [2]. Finally, the potential usage of both new high end solid and liquid anode X-ray sources for general XPS will be addressed.

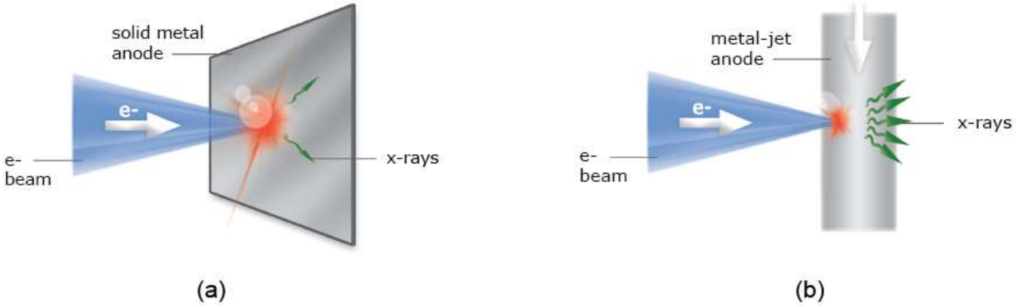

Fig. 1: The principle of a solid anode X-ray tube (a) and a liquid-metal-jet X-ray tube (b).

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HAXPES at PETRA III and IV: electronic structure, operando devices and in-situ catalysis

Christoph Schlueter[1], Andrei Gloskovskii[1], Yury Matveyev[1], Patrick Lömker[1,5], Thiago Peixoto[1], Katrin Ederer[1], Ilja Schostak[1], Michael Sing[2], Ralph Claessen[2], Carsten Wiemann[3], Claus M. Schneider[3], Katerina Medjanik[4], Gerd Schönhense[4], Peter Amann[5], Anders Nilsson[5] and Wolfgang Drube[1]

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In September 2018, the X-ray undulator beamline (P22), fully dedicated to hard X-ray photoelectron spectroscopy (HAXPES) techniques, opened to users at PETRA III. The beamline hosts four specialised experimental end stations for high-resolution studies of the electronic and chemical structure of complex materials, realistic device-like structures and catalytic interfaces. The main instrument for conventional HAXPES techniques offers sample cooling and in-situ electrical characterisation for *operando* studies. A separate instrument provides full-field, sub-µm electron spectro-microscopy (HAXPEEM). Additionally, a specialized setup for high-pressure HAXPES applications (POLARIS) recently demonstrated its capabilities at pressures >2.0 bar. Finally, a full-field *k*-microscope with time-of-flight energy discrimination delivered first fully *k*-resolved valence band structures in the HAXPES energy range (up to 7 keV). All instruments are implemented and operated in close collaboration with external user groups and as such reflect the wide range of scientific fields currently covered by our community.

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k-resolved electronic structure and band offsets at the NbN/GaN superconductor/semiconductor heterojunction

Tianlun Yu[1,2], John Wright[3], Guru Khalsa[3], Betül Pamuk[4], Celesta S. Chang[5], Yury Matveyev[6], Xiaoqiang Wang[1], Thorsten Schmitt[1], Donglai Feng[7,8,9], David A. Muller[5,10], Huili Grace Xing[10,11], Debdeep Jena[10,11], Vladimir N. Strocov[1]

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Electronic structure of heterointerfaces plays a pivotal role in their device functionality. Recently, ultrathin films of superconducting NbN (T_c=17K) have been integrated by MBE with the semiconducting GaN. To directly measure **k**-resolved electronic structure and band offsets at this Schottky heterointerface as well as the band-bending profile into GaN, we use soft X-ray ARPES at the ADRESS beamline of the Swiss Light Source. With different out-of-plane lattice constants of NbN and GaN, our experiment uses different photon energies to probe electronic structure at the opposite sides of the interface. We support the experimental findings with DFT calculations as well as transport and optical measurements. Most importantly, we find that the Fermi states of NbN are well separated from the GaN states in energy and **k**, excluding their crosstalk and thus poisoning of the superconductivity in NbN by the GaN electrons. This finding opens prospects of integrating NbN-based superconducting devices (single-photon detectors, Josephson junctions, etc) into semiconductor technology.

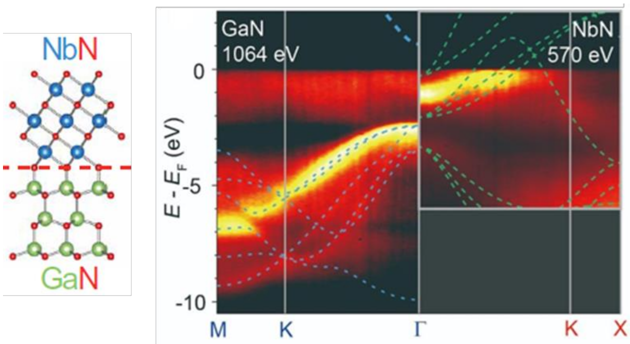

Fig. 1: *Experimental band structure of NbN matched to GaN at the interface.*

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How dopants and defects stabilize ferroelectricity in HfO_2 – a spectroscopic approach

Thomas Szyjka[1,2], Lutz Baumgarten [2], Terence Mittmann[3], Monica Materano[3], Yury Matveyev[4], Christoph Schlueter[4], Thomas Mikolajick[3], Uwe Schroeder[3], and Martina Müller[1]

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HfO_2 is a key multifunctional material for nonvolatile ferroelectric field effect transistors, memristors or novel neuromorphic computing concepts. Oxygen vacancies and metallic impurity ions are recognized as defects which both can stabilize the ferroelectric phase in undoped HfO_2. HAXPES provides a unique tool to explore the interface reactivity and tunability of the oxygen content in HfO_2-based devices. We present studies on the emerging chemical states of HfO_2-based capacitors and focus on the identification of the (mostly indirect) signatures of oxygen vacancies or rare-earth dopants in the HfO_2 layers and at the metal electrode interfaces [1].

We correlate HAXPES data recorded at the beamline P22 (DESY Hamburg, Germany) to electric field cycling experiments, and conclude on how the microscopic HfO_2 electronic structure directly relates to macroscopic ferroelectric polarization and switching properties. We show selected examples which reval the different interface oxygen exchange mechanisms in HfO_2-metal electrode structures [2], demonstrate the oxygen vacancy formation in HfO_2 layers dependent on rapid thermal annealing processes [3], as well as oxygen vacancy/nitrogen impurity interactions altered by deposition-specific parameters [4].

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1a1-1 : Invited

Probing spin-polarized electronic structures on half-metallic Heusler alloys using combined bulk-sensitive spectroscopies

Hidenori Fujiwara

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Toward the next generation spintronic application using the spin-degrees of freedom, it is essential to optimize the spin-polarized electronic structures of the spintronic materials. The Halfmetallic Heusler alloys [1] are candidates of the spintronic materials, since one of the spin-subbands is metallic and the other is semiconducting with a gap at the E_F predicted by the density functional theory (DFT). However, it is not straightforward to reveal the intrinsic electronic structures of Heusler alloys, since it is well known that the off-stoichometric effects and various disorder often change the magnetic properties and the local electronic structures.

To reveal the intrinsic spin-polarized electronic structures of halfmetallic Heusler alloys, we have conducted combined bulk-sensitive spectroscopies for high-quality single-crystalline samples of ferromagnetic Co_2MnSi, Co_2FeSi and ferrimagnetic Mn_2VAl using linearly polarized hard x-ray photoelectron spectroscopy (HAXPES), soft x-ray absorption spectroscopy (XAS), and resonant inelastic soft x-ray scattering (RIXS). HAXPES measurements have been performed at SPring-8 BL19LXU, where transmission-type phase retarders were installed [2,3]. Moreover, XAS-MCD [4,5] and RIXS-MCD [5,6] were performed at SPring-8 BL23SU and BL07LSU, respectively.

The valence band HAXPES obtained with s-polarization configuration is nicely described by the simulation based on the weighted sum of the partial density of states by taking into account the asymmetry factors of the photoionization cross-section. Moreover, the RIXS-MCD experiments under the external magnetic field probes the intrinsic spin-polarized electronic structures thanks to the spin-conserved dipole-allowed transition from the spin-polarized $2p_{3/2\ \mathrm{mj}\ =\ \pm 3/2}$ states [5,6,7]. The strong MCD contrast in the RIXS-MCD spectra for Co_2MnSi, Co_2FeSi and Mn_2VAl reflect the up and down spin contribution of the $3d$ states in the positive and negative MCD signals. These RIXS-MCD spectra are also qualitatively explained by the DFT-based simulation as well as HAXPES and XAS-MCD spectra. By using the combination of the advanced spectroscopies, we have successfully proved the halfmetallic electronic structures of Co_2MnSi and Mn_2VAl. This suggests that the multimodal investigation of the electronic structures using the bulk sensitive spectroscopies are powerful for revealing the detailed spin-polarized electronic structures of the functional magnetic materials.

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Probing strain-engineered anion ordering via depth-sensitive and polarization-dependent X-ray spectroscopic techniques

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Inducing atomic-scale ordering in functional heteroanionic materials is a promising approach for realizing new or enhanced physical properties. In this study, we demonstrate that epitaxial strain can be used to control F/O ordering in epitaxial $SrMnO_{2.5-\delta}F_{\gamma}$ thin films deposited on substrates with different in-plane lattice constants [1]. By using a complementary suite of lab-based valence-band hard x-ray photoemission (HAXPES), polarization-dependent soft x-ray resonant valence-band photoemission, x-ray linear dichroism, and density functional theory calculations, we demonstrate that F occupies the apical sites under compressive strain leading to Mn−F bonds oriented along the out-of-plane direction. In contrast, F is incorporated on equatorial sites under tensile strain, leading to Mn−F bonds parallel to the in-plane directions. Such anionic-site selective substitutions result in changes to the valence-band density of states and lead to an increased resistivity in films in which F occupies the apical sites compared to those in which F resides at the equatorial position. By demonstrating a general strategy for inducing preferential anion-site occupancy in oxyfluoride perovskites, this work lays the foundation for future materials design and synthesis efforts that leverage this greater degree of atomic control to realize new polar or quasi-two-dimensional materials. Furthermore, it highlights new possibilities for combining lab-based valence-band HAXPES with resonant and polarization-dependent synchrotron-based spectroscopic techniques for comprehensive studies of electronic structure in engineered materials.

This work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences, and Engineering Division under Award DE-SC0019297

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Total Reflection Hard X-ray Photoelectron Spectroscopy for $La_{1-x}Sr_xMnO_3$ and $La_{1.9}Ce_{0.1}CuO_4$

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Over the last decade, the HAXPES technique made a major impact in unraveling the electronic properties of solids, encompassing a large field of applications, from fundamental to applied science. In the present study, we demonstrate that total reflection hard x-ray photoelectron spectroscopy (TR-HAXPES) is a versatile method for elucidating a difference between surface and bulk electronic states of strongly correlated electron systems, complementing conventional bulk sensitive hard x-ray photoelectron spectroscopy (HAXPES). By using the HAXPES in total reflection (TR) condition, we have revealed the profound differences between surface and bulk electronic screening and correlation in $La_{1-x}Sr_xMnO_3$ and the electron doped high-Tc superconductor $La_{1.9}Ce_{0.1}CuO_4$[1]. From the incidence angle dependence of the spectral line shapes, we found that the well-screened peaks first discovered by HAXPES[2] almost disappeared at the TR condition. It is crucial that the probing depth at TR condition is several nanometers, while it is several tens of nanometers at normal condition. The results strongly demonstrate that this method allows us to measure both surface and bulk electronic states without making any changes of experimental conditions like the energy resolution, x-ray energy and the beamline.

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Theoretical Aspects of Resonant Auger Processes in Solids

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Many research opportunities exist at hard x-ray photoelectron spectroscopy (HAXPES) beamlines. These beamlines frequently involve electron analyzers collecting electrons emitted along the same direction as the incident photon polarization for linearly polarized light. X-ray absorption spectroscopy (XAS) measures photoexcitation from each core subshell, e.g., L_3, M_5. XAS sums over orbitals, and contributions by each orbital might not be revealed using XAS alone. In addition, while partial densities of unoccupied states (e.g., d states with T_{2g} or E_g symmetry) can be different, it may be hard to distinguish between excitations into various states. Resonant Auger electron spectroscopy also probes unoccupied states, because an incident x-ray photon energy can be shared by an Auger electron, a two-hole final state that follows Auger decay, and a spectator photoelectron. If a spectator photoelectron was excited with too much or too little energy to achieve its final state, an energy surplus or deficit can be carried off or paid back by the Auger electron, as revealed in kinetic-energy distributions intrinsic to the Auger line shape [1,2,3]. One can predict the rate of excitations from each core orbital as a function of its angular momentum along a photon polarization direction and/or along the Auger emission direction. This highlights a connection between partial densities of states and resonant Auger spectra based on "core-hole memory" that retains angular-momentum information. We illustrate this for a variety of x-ray edges in several noble metals.

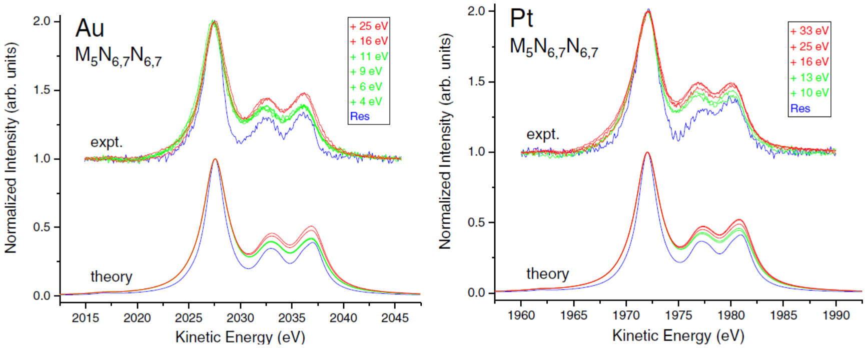

Fig. 1: *Resonant Auger emission spectra for several photon energies given relative to threshold.*

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Accurate and Non-destructive Layer Structure Analysis of Semiconductor Materials Using Angle Resolved HAXPES

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Optimizing the processes of semiconductor materials to increase the efficiency is always one of top target in photovoltaic industries. Not only surface chemical analysis is essential to examine the effect of processes to these semiconductor materials, but also the accurate layer structure analysis is critical to inspect the quality of semiconductor devices. X-ray photoelectron spectroscopy (XPS) is a common tool to reveal the surface chemical composition. In addition, by performing the angle resolved XPS (AR-XPS), the layer structure of multiple thin films can be obtained. When the total thickness of multilayer is over the detection depth, hard X-ray photoelectron spectroscopy (HAXPES) holds the potential to perform AR-HAXPES without the need of sputtering due to its greater detection depth. To compare the layer structure analysis using AR-XPS and AR-HAXPES, Al_2O_3 with 15 nm thickness is deposited on c-Si surface and the thickness of thermally grown SiO_2 passivation layer is controlled by a series of annealing temperature. To examine the thickness of interfacial SiO_2 layer, a lab type XPS system equipped with both AlKα and CrKα X-ray sources are utilized to perform AR-XPS and AR-HAXPES respectively. Because the thickness of Al_2O_3 layer is over the detection depth of AlKα X-ray, sputtering using monoatomic Ar^+ to remove a thickness of Al_2O_3 layer is performed to obtain the signal from interfacial SiO_2 layer and c-Si substrate. The result shows the thickness of SiO_2 layer increases with the annealing temperature in both AR-XPS and AR-HAXPES. However, the SiO_2 thickness of AR-XPS result is thinner than that of AR-HAXPES. The reduction of SiO_2 observed in AR-XPS spectrum suggests the thinner SiO_2 layer is caused by sputter damage. The AR-HAXPES is performed without any sputtering and the obtained thickness of SiO_2 is found to be close to the cross-sectional TEM image. Therefore, with the greater detection depth, AR-HAXPES realized the accurate thickness analysis of semiconductor materials. In the presentation, the examples of AR-HAXPES for semiconductor materials will be demonstrated.

Theoretical and experimental HARPES study of Weyl-semimetal TaAs: The application of machine-learning

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A Weyl semimetal is a new matter state possessing Weyl fermions near the Fermi level with several unique physical properties and it is confirmed by the existence of Fermi arc surface states [1]. In this work we study tantalum arsenide (TaAs) which is a prototypical Weyl semimetal compound. The electronic structure properties have been studied by soft and hard X-ray angle-resolved photoemission spectroscopy (ARPES) at energies of 440 eV and 2150 eV, respectively. For the first time, TaAs is experimentally investigated by the bulk sensitive photoemission in the hard X-ray regime. In order to interpret experimental data we performed one-step model of photoemission calculation which includes all matrix elements and final state effects [2-4]. Due to the strong photon momentum effects and uncertainty in the tilt of experimental geometry we used a so-called machine learning algorithm combined with a free-electron final-state model to find best possible experimental parameters. Our findings re-emphasize the overwhelming accuracy of hard X-ray ARPES (HARPES) compared to the traditional ultraviolet and soft X-ray one in case of bulk electronic structure, motivating further material discoveries [5].

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Core-level peaks *and* the inelastic background in HAXPES: a depth-resolved analysis of the CdS/Cu(In,Ga)Se_2 interface for thin-film solar cells

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Interfaces are crucial for multilayer-containing electronic devices (such as batteries or solar cells), generally (only) optimized *empirically*. For a further *insight-driven* optimization, a deeper understanding of the interface properties in such devices needs to be obtained. Hard x-ray photoelectron spectroscopy (HAXPES) is predestined to characterize (buried) interfaces, as it is non-destructive, chemically sensitive, and the inelastic mean free path λ of the (unscattered) photoelectrons can be tuned (e.g., up to ~16 nm for photon excitation energies up to 15 keV). In addition, the inelastic background contains information about the elemental distribution for depths of 10-20 λ, even if the corresponding (unscattered) photoelectron peak is fully attenuated.

In this contribution, we analyze both the core-level peaks *and* the inelastic background of a series of CdS/CIGSe interface samples [1]. This interface is the central component in chalcopyrite-based thin-film solar cells (with record efficiencies above 23% [2]). For this purpose, HAXPES spectra with photon excitation energies from 2.1 up to 14 keV were collected using the newly commissioned X-SPEC beamline at the KIT synchrotron [3]. We paint a depth-resolved picture of the CdS/CIGSe interface, for which the solution-grown CdS film thickness can be accurately determined as a function of the CdS deposition time. We also find that, for shorter CdS deposition times, the film thickness varies laterally. Furthermore, small amounts of Se and process-related Rb can be detected in a thin (~2 nm) surface layer of all investigated CdS films. The findings thus demonstrate the power of HAXPES to discern details of a buried interface in applied materials systems and devices.

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1p1-1 : Invited

LDA+DMFT approach to core-level x-ray spectroscopies in correlated electron systems

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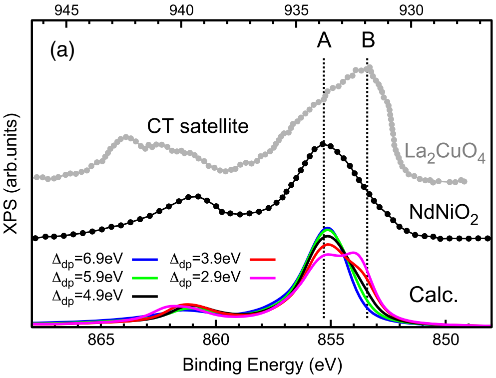
We develop an ab-initio computational method for simulating core-level x-ray spectroscopies in strongly correlated electron systems, such as 3*d* transition metal oxides [1,2]. This method builds on a combination of local density approximation (LDA) and dynamical mean-field theory (DMFT), which can be viewed as an extension of conventional cluster model, employed widely in HAXPES analysis, to incorporate the orbital hybridization with the conniuum of valence bands in the realstic lattice. This method is particulary beneficial to study nonlocal charge-transfer screening features in core-level HAXPES spectra of a series of 3*d*, 4*d*, and 5*d* transition metal oxides [1,3,4]. Besides, it provides a coherent description of HAXPES and other core-level x-ray excitation processes, e.g., x-ray absorption spectroscopy and resonant inelastic x-ray scattering [2]. In this talk, a special focus is paid on the Ni 2*p* core-level x-ray photoemission spectra of $NdNiO_2$, see figure, a mother compound of recently discovered superconducting infinite-layer nickelate. We show that a future HAXPES experiment will allow to derive the key parameters of this material class [4].



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Theoretical prediction of core electron binding energies using the ΔSCF method – applications to 3*d* transition metals

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Theoretical calculations of core electron binding energies are important for aiding the interpretation of experimental core level X-ray photoelectron spectra. Previously, it has been shown, that using the Δ-Self-Consistent-Field (ΔSCF) method, it is possible to predict accurate absolute core electron binding energies in free molecules, adsorbed molecules, and periodic solids [1,2]. However, so far, most theoretical work has focused on light elements (up to and including Cl), and relatively little is known about the performance of current theoretical methods for heavier elements, including transition metals.

In this talk, a brief overview of the ΔSCF method will be given, and the requirements for predicting accurate absolute core electron binding energies will be discussed. In addition, the application of the ΔSCF method to elements of the first transition metal series will be examined in detail [3]. It is shown that scalar-relativistic ΔSCF calculations based on density functional theory and the SCAN functional typically underestimate absolute transition metal (TM) $2p_{3/2}$ core electron binding energies in compounds of first row transition metals by ~ 0.5 – 1.0 eV. However, calculated relative core electron binding energies (≈ binding energy shifts) are still highly accurate, with a mean absolute error of 0.2 eV for a dataset of 60 compounds. Moreover, with the use of element-specific correction terms, the prediction of accurate absolute TM $2p_{3/2}$ core electron binding energies is also possible. Together with the recent work on extending the ΔSCF method to periodic solids, these results lay the foundations for the use of ΔSCF calculations for predicting core electron binding energies in a wide range of experimentally relevant systems.

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Time-resolved study of recoil-induced rotation by X-ray pump - X-ray probe spectroscopy

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Modern stationary X-ray spectroscopy is unable to resolve rotational structure. In the present paper, we propose to use time-resolved two color X-ray pump-probe spectroscopy with picosecond resolution for real-time monitoring of the rotational dynamics induced by the recoil effect. The proposed technique consists of two-steps. The first short pump X-ray pulse ionizes the valence electron, which transfers angular momentum to the molecule. The second time-delayed short probe X-ray pulse resonantly excites a 1s electron to the created valence hole. Due to the recoil-induced angular momentum the molecule rotates and changes the orientation of transition dipole moment of core-excitation with respect to the transition dipole moment of the valence ionization, which results in a temporal modulation of the probe X-ray absorption as function of the delay time between the pulses. We developed an accurate theory of the X-ray pump-probe spectroscopy of the recoil-induced rotation and study how the energy of the photoelectron and thermal dephasing affect the structure of the time-dependent X-ray absorption using the CO molecule as a case-study. We also discuss the feasibility of experimental observation of our theoretical findings, opening new perspectives in studies of molecular rotational dynamics.

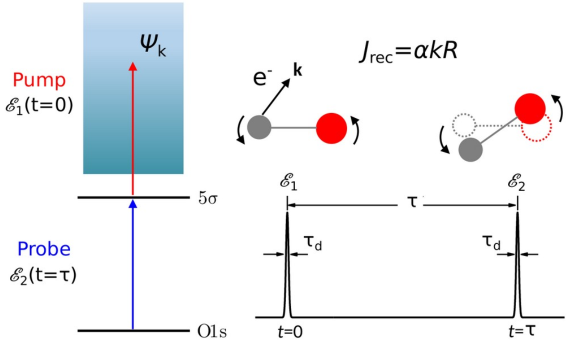

Fig. 1: *Scheme of the electronic transitions (left panel) in the studied pump-probe process with two time delayed X-ray pulses (right panel).*

Hard X-ray Photoelectron Spectroscopy of Functional Oxide Films and Heterointerfaces

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Hard X-ray photoelectron spectroscopy has now established itself as a useful method for the study of a broad class of materials and is recognized not only by experts in the field. In my talk, I will review our recent results on functional oxide films and heterointerfaces that may serve as a platform for future all-oxide electronic devices. Topics include oxygen vacancy-driven band engineering at the interface of LaAlO$_3$/SrTiO$_3$ heterostructures [1], band alignment and electronic reconstruction in the LaVO$_3$/SrTiO$_3$ system [2], determination of the bulk spin polarization of magnetite [3], and momentum mapping of the buried high-mobility two-dimensional electron system in LaAlO$_3$/KTaO$_3$ [4]. The latter two studies were performed using a time-of-flight momentum microscope equipped with a spin-filter crystal.

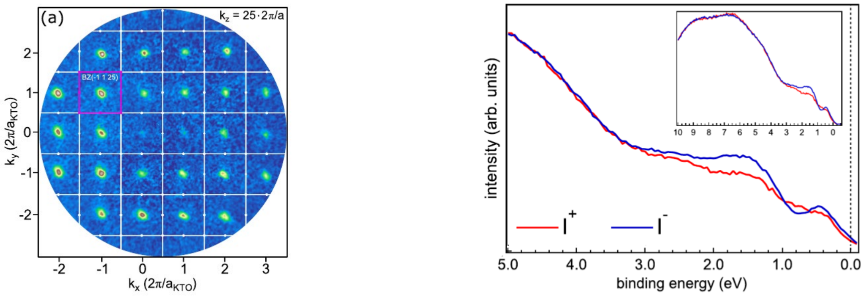

Fig. 1: *(a) Fermi surface map of the buried two-dimensional electron system in the LaAlO$_3$/KTaO$_3$ heterostructure. (b) Spin-resolved valence-band spectra of a Fe$_3$O$_4$(111) film, measured with an excitation energy of hv = 5.0 keV.*

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Proximity-induced spin-orbit interaction in MoSe2 overlayered with amorphous Pb films

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To design new functional materials for our increasing daily demands, we must abandon standard models and move towards innovative compounds. These may be based on hybrid systems and heterostructures, invoking new physical mechanisms originating from proximity effects. Spin-orbit (SO) proximity effects, in particular, are finding increasing applications across the whole spectrum of condensed matter physics. The present work focuses on the modulation of the SO interaction in 2H-MoSe2 which is induced by proximity effects at its interface with amorphous films of Pb, the high-Z element providing strong SO coupling. The key element of this research study is the formation of amorphous Pb overlayers rather than epitaxial crystalline ones, allowing us to overcome the k-space wavefunction mismatch at the interface. With an advantage of the enhanced probing depth, the use of soft x-ray angle-resolved photoelectron spectroscopy (SX-ARPES) allows reaching the interface regions where the SO interaction is modulated. k-space dependence of the proximity-induced SO interaction, as well as its dependence on the Pb-overlayer thickness will be presented. Experimental SX-ARPES data are combined with corresponding state of the art fully relativistic one-step model calculations based on the mutiple scattering Green function KKR method. The amorphous layer of Pb was simulated by the coherent potential approximation. This fundamental knowledge will pave novel approaches towards realization of spintronic devices.

Studying anisotropic electronic structure in 2D-layered crystals using resonant Auger spectroscopy

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Two-dimensional (2D) materials beyond graphene have shown promise in various applications, e.g., nanoelectronics and rechargeable batteries. One class of these materials is the transition metal dichalcogenides (TMD) a family of layered materials weakly bonded through van der Waals forces. The layered structure of these materials imply that its electronic properties are anisotropic in the layers versus between the layers and this has been studied recently [1,2]. We have used polarization-dependent x-ray absorption in the tender x-ray regime at the S K edge and maps of the resonant Auger spectra in the S KLL Auger kinetic energy range to study the anisotropy in- versus out of plane in SnS_2, as seen in Fig. 1 [1]. A close sibling of SnS_2 is SnS which also exhibits anisotropic charge transfer in- versus out of plane [3]. Compared to SnS_2 which is hexagonal, SnS is orthorhombic and thus also exhibit intraplane anisotropy along the layers two primary directions, the zig-zag and armchair directions. We show that resonant Auger spectroscopy is a method that we can achieve directional and orbital specificity with when studying in- and out of plane characteristics of layered materials.

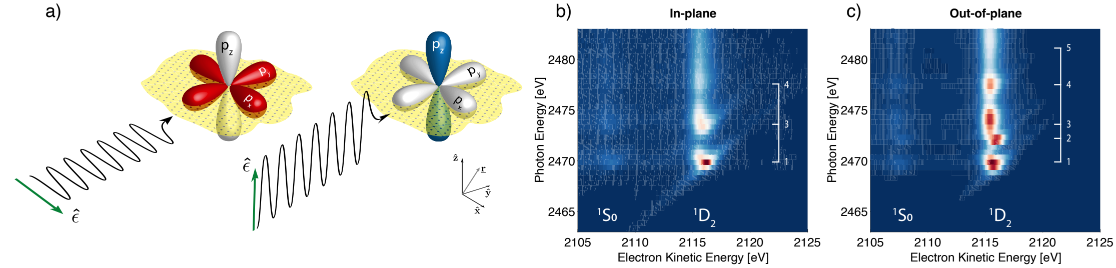

Fig.1 a) shows the principle of using the polarization direction to excited electrons into specific orbital, b) and c) shows the distinctly anisotropic resonant structure in- and out of plane of SnS_2*. Adapted from [1]*

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Dynamic SXPS and HAXPES study of the Thermal Stability of Ti_xW_{1-x}/Cu Heterostructures

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For power semiconductor devices, a binary alloy of titanium-tungsten (Ti_xW_{1-x}) is often used as a diffusion barrier to isolate the copper metallisation interconnects from the silicon substructure. A diffusion barrier is necessary as Cu rapidly diffuses into Si at low temperatures (<200°C), which can lead to premature device failure.[1] However, the progressive miniaturisation of feature sizes in microelectronic devices has increased the probability of higher local power densities and temperatures. Exposure to such conditions has been shown to induce a segregation of Ti out of the TiW and an interdiffusion into the adjacent Cu metallisation. This loss of Ti can ultimately lead to the degradation and failure of the diffusion barrier, and allow for the interaction between Si and Cu.[2]

Here, device-relevant Si/SiO_2/TiW(300 nm/Cu(25 nm) thin film stacks were characterised using a combination of soft and hard X-ray photoelectron spectroscopy (SXPS and HAXPES) at beamline I09 of the Diamond Light Source, using X-ray photon energies of 1.5 keV and 5.9 keV, respectively. The combination of SXPS and HAXPES provides the opportunity to non-destructively probe across multiple depths, including the buried TiW/Cu interface and bulk TiW.

To study the thermal stability of TiW, two experiments were conducted. The first involved a static measurement, where samples were ex-situ annealed for varying durations at 400°C under an inert atmosphere to simulate temperature stress on the device stacks. SXPS/HAXPES measurements were then conducted across the TiW/Cu interface. The second involved a dynamic measurement, where the same as-deposited samples were annealed in-situ within the analysis chamber of I09 to 400°C, and core level spectra were continuously measured to track the evolution of the Ti enrichment at the Cu surface during the annealing process.

During annealing, Ti was shown to segregate out of the TiW barrier and diffuse across Cu at temperatures as low as 250°C. However, the ex-situ interface measurements suggest that despite the out-diffusion of Ti, the diffusion barrier remains functional at 400°C as no intermetallic phases are observed and the downward diffusion of copper is successfully suppressed. Overall, the combined ex-situ and in-situ SXPS/HAXPES experiment systematic approach provides a clear picture of the influence of temperature and annealing duration as experience during device manufacturing or operation, on the stability of the TiW film and TiW/Cu interface.

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Probing quantum materials interfaces with HAXPES at the GALAXIES beamline : results and perspectives

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The functionality of novel quantum materials derives from the competition between the internal degrees of freedom (electrons, spin, orbital, topology, spin-orbit coupling) most notably observed at interfaces. An intense research activity has been set off for fabricating and manipulating interfacial properties of quantum materials leading to a frenzy of scientific discoveries including 2DEG, interfacial superconductivity, etc. HAXPES appears as a major probe of electronic interfacial behavior due to its superior probing depth and sensitivity to the local atomic properties. Here, we will review recent HAXPES results obtained at the GALAXIES beamline, SOLEIL Synchrotron [1] regarding interfacial properties of metal oxides, including CMO / YCO superlattices [2] specially when combined to standing wave approach. Extension of this work to investigating materials properties in operando conditions or characterizing ultrafast charge dynamics as recently obtained in GeSe [3] will be discussed. These results will be put in the context of the 4th generation (DLSR) synchrotron sources with much reduced H source size which offer new possibilities for in-depth interfacial characterization.

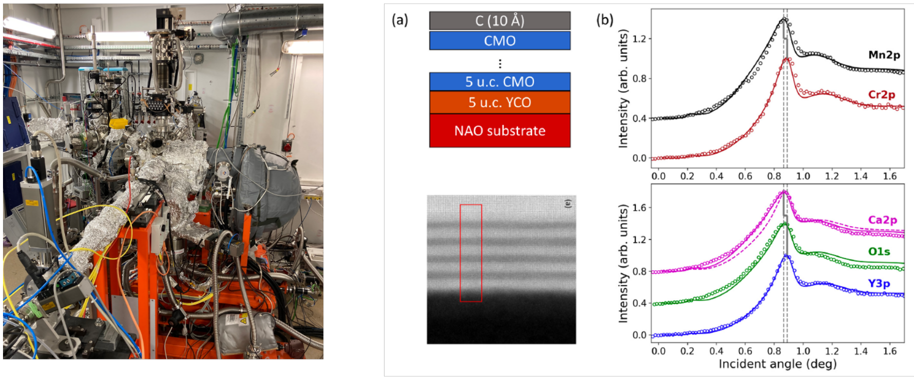

Fig. 1: *(left) HAXPES endstation at the GALAXIES beamline; (right) depth profiling of CMO / YCO superlattice by SW-HAXPES (from Ref. [2])*

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$CaCu_3Ru_4O_{12}$: a high Kondo-temperature transition metal oxide

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We present a comprehensive study of $CaCu_3Ru_4O_{12}$ using bulk sensitive hard and soft x-ray spectroscopy combined with local-density approximation (LDA) + dynamical mean-field theory (DMFT) calculations. Correlation effects on both the Cu and Ru ions can be observed. From the Cu *2p* core level spectra we deduce the presence of magnetic Cu^{2+} ions hybridized with a reservoir of itinerant electrons. The strong photon energy dependence of the valence band allows us to disentangle the Ru, Cu, and O contributions and thus to optimize the DMFT calculations. The calculated spin and charge susceptibilities show that the transition metal oxide $CaCu_3Ru_4O_{12}$ must be classified as a Kondo system and that the Kondo temperature is in the range of 500-1000 K. The high Kondo temperature is the key to reconcile the contradictory conclusions of the existing studies (conducted at moderate temperatures) on this material. Our work [1] brings *de-facto* the Kondo physics from 4f/5f intermetallic systems to also transition metal oxides. The perovskite material class with the $ACu_3B_4O_{12}$ structure provides a new platform for integrating competing quantum phenomena in strongly correlated electron systems.

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Lab-scale Hard X-ray Photoelectron Spectroscopy for the analysis of buried interfaces in device technologies

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Photoelectron spectroscopy plays a key role in the characterization of surfaces and interfaces which is of primary importance in the field of device technology. However, instruments traditionally available in the lab are usually equipped with Al Kα radiation (1486.6 eV), which limits the probing depth and therefore prevents from analyzing buried interfaces deeper than a few nanometers without resorting to destructive methods such as sputtering, causing strong chemical modifications.

However, in many modern technologies, the interfaces of interest are most of the time deeply buried underneath metal electrodes or oxides layers, requiring deeper analysis. This can be achieved by using higher energy photons. Hard x-ray photoelectron spectroscopy (HAXPES) have long been available at synchrotron facilities but its limited access lessen its interest as process development requires routine measurements and high reactivity. The recent advent of commercial lab-scale instruments opens new possibilities for non-destructive buried interface analysis [1-2].

In this presentation, we present the possibilities offered by a novel Cr Kα (5414.8 eV) lab-scale HAXPES instrument (*Quantes*, Physical Electronics Inc.). The first part is devoted to the chemical analysis of buried interfaces in TiN/HfO_2 stacks intended for oxide-resistive random access memories (OxRAMs). The influence of a metallic Ti layer deposited at the interface is especially examined. Results reveal that the addition of a Ti layer promotes the reduction of hafnium oxide thus creating oxygen vacancies, which in turn greatly determine the device properties. The second part focuses on quantitative aspects on this new instrument and in particular describes an approach to determine relative sensitivity factors. The accuracy of the empirically derived sensitivity factors is confirmed by the quantitative analysis of binary and ternary compounds.

Acknowledgements:

This work was performed at the Platform For NanoCharacterization (PFNC) of CEA-Grenoble with support of the Recherche Technologique de Base program of French National Research Agency (ANR).

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1p3-4 : Contributed

Metallic Antiferromagnetic Spintronics: Mn_2Au a case study

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The electronic structure of bulk Mn_2Au has been investigated by first-principles density-functional theory calculations based on the Green's function technique. Existence of significant out-of-plane magnetic anisotropy combined with the large strength of short range antiferromagnetic exchange interactions between Mn atoms located at two dfiferent Wyckoff positions results in the stabilization of the antiferromagnetic ground state. Two dimensional plots of constant energy surfaces in the Γ-X-Σ plane of the Brillouin zone exibits a 4-fold to 2-fold symmetry breaking as a function of the binding energy at 0.00 eV and 0.25 eV below the Fermi surface. We find that such a symmetry breaking in Mn_2Au is arising due to the degeneartion of the electronic bands in the presence of external magnetic field indicating a strong spin-orbit coupling interaction. Our results describes the tuning of the magnetic and electronic properties of Mn_2Au for spintronic applications.

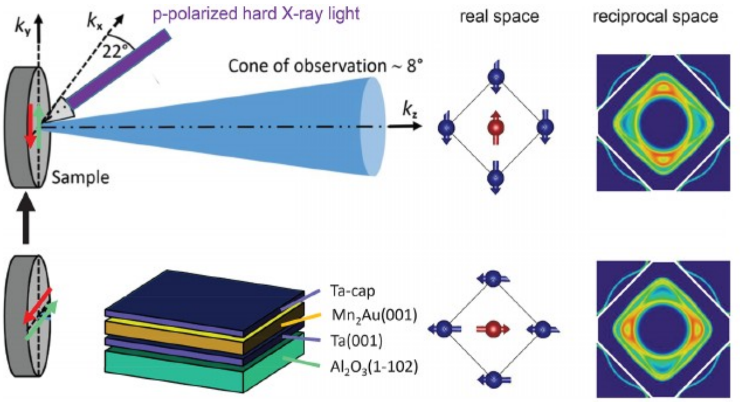

Fig. 1: *Sketch for the HARPES experiment geometry. Calculated kx-ky maps at* E_B*=0.25 eV highlighting asymmetries related to Neel vector orientation. [1]*

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Materials Research for Metal-Halide Perovskite Solar Cells Enabled by Hard X-ray Photoelectron Spectroscopy

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The multitude of layers, interfaces, elements, impurities, defects, etc. contributing to the structure, function, and performance of solar cells means that characterization and fundamental understanding of the chemical and electronic structures of each component, as well as their interactions at interfaces – also upon external stimuli – are crucial to support technical progress. HAXPES can reveal crucial information and enable studies that would not be possible employing soft X-rays, opening new routes to insight-driven strategies to improve device performance.

To showcase the capabilities, I will present HAXPES-derived chemical and electronic structure information of the (currently) most prominent absorber material for solar cells: Metal-halide perovskites – corresponding devices have impressed by an unprecedented performance increase over the past decade and are now on the verge of revolutionizing commercial PV. The presentation will include some examples focussing on near-surface bulk and interface characteristics and how these properties are influenced by illumination, bias application, or exposure to humidity.

Characterization of Ferroelectric Devices: HfZrO thin films on InAs

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High-k hafnium oxides such as HfZrO have amassed interest due to their dielectric superiority over silicon. Compatible with atomic layer deposition, exploitation of their ferroelectricity promises thin film memory technologies which are poised to invigorate electronics. Sophisticated control and characterization remain vital as structural size and complexity are pushed to their nanometric limits. Exploiting the brilliance of synchrotron-based hard x-ray photoelectron and absorption spectroscopies, we get a deep and realistic look inside next-generation memories. After thorough ex-situ analysis, we push further with operando studies for insight into actual device functioning and performance. The result is unparalleled access to the origin and inner workings (ex. wake-up, switching and endurance) of hafnia-based ferroelectricity.

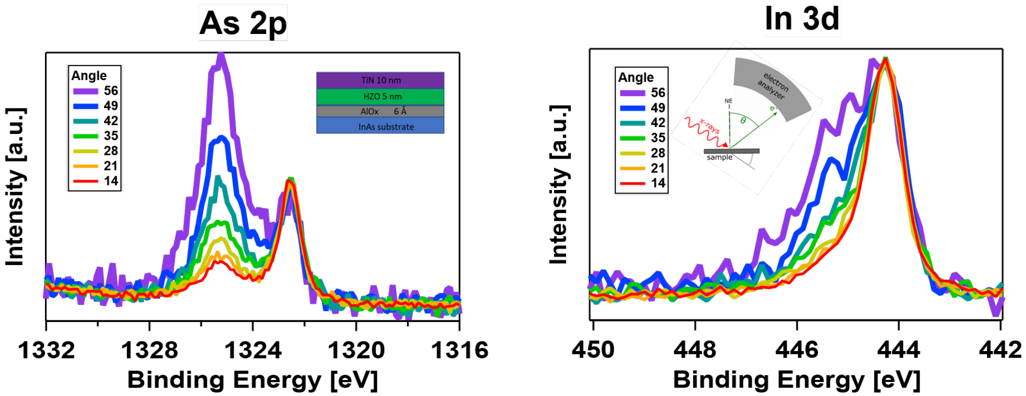

Fig. 1: *Angle-resolved hard X-ray photoelectron spectroscopy is able to penetration functioning device material stacks more than ten nanometers thick. Looking through the top electrode and the ferroelectric dielectric, we probe the bottom high-k/III-V interface and monitor operando chemical changes.*

Combining depth-sensitive X-ray spectroscopic and scattering techniques to probe interfacial magnetic phenomena in oxide superlattices

Jay R. Paudel[1], Michael Terilli [2], Ivana Vobornik[3], Pasquale Orgiani[3], Giancarlo Panaccione[3], Christoph Klewe[4], Padraic Shafer[4], Vladimir Strocov[5], Andrei Gloskovskii[6], Christoph Schlueter[6], Jak Chakhalian[2] and Alexander X. Gray[1]

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Emergent interfacial ferromagnetism in $CaMnO_3/CaRuO_3$ and $CaMnO_3/LaNiO_3$ heterostructures has been investigated for nearly two decades. Several experimental and theoretical studies suggest that this phenomenon is stabilized by the charge transfer across the interface and suggest different thicknesses of the resultant ferromagnetic layers [1-4]. In this comprehensive study [5], we have utilized bulk-sensitive hard x-ray photoemission spectroscopy (HAXPES) and soft x-ray standing-wave photoemission spectroscopy in both first- and second-order Bragg geometries to probe the depth-dependent chemical profile and interfacial charge reconstruction in $CaMnO_3$. Furthermore, we have carried out polarization-dependent x-ray absorption spectroscopy and x-ray resonant magnetic scattering to probe depth-resolved magnetic profile at the interfaces in these material systems. Finally, high-resolution angle-resolved photoemission spectroscopy with in-situ pulsed-laser deposition synthesis were used to track the topology of the Fermi surface of $LaNiO_3$ in the superlattice across the thickness-dependent metal-insulator transition. The results were compared to the first-principles theoretical calculations.

This work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences, and Engineering Division under Award DE-SC0019297

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2a1-1 : Invited

Using X-ray optical effects in photoelectron spectroscopy to study heterogeneous systems

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Due to its relatively large information depth, HAXPES has become a popular technique to study chemical and electronic properties of bulk materials and buried interfaces. By using X-ray optical effects in near total reflection or in Bragg geometry (related to periodic samples, such as multilayer mirrors), one can also achieve high depth selectivity. A few recent mini reviews were dedicated to high resolution profiling of emergent phenomena at interfaces of complex oxide materials [1,2], and high temperature applications [3]. In this talk, I will briefly introduce the X-ray optical effects in play and give an overview of the standing-wave photoelectron spectroscopy of multilayer samples. Later I will discuss applications to a hybrid multiferroic system BFO/LSMO [4] and extreme UV lithography photoresists [5,6].

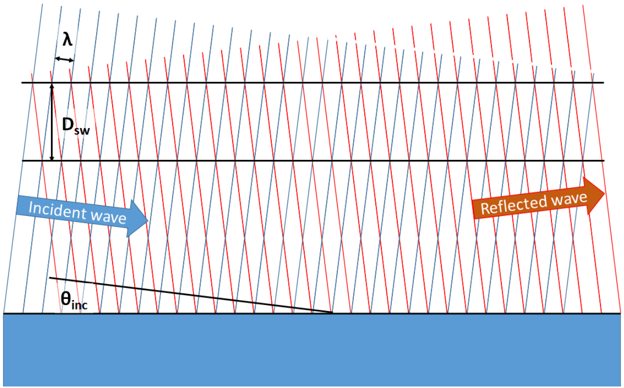

Fig. 1: *Schematics showing the formation of the X-ray standing wave over a reflective sample.* Θ_{inc} *is incident angle of X-rays,* λ *is the wavelength, and* D_{sw} *is a standing wave period. Nodes and antinodes of the standing wave are present in planes parallel to the reflective surface.*

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New Diemensions of HAXPES: Variable Polarization, Total Reflection, and Spin Resolution

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Hard X-ray photoemission spectroscopy (HAXPES) with variable X-ray polarization is useful tool for studying the bulk-sensitive electronic states of solids. Particularly, by utilizing the X-ray polarization-dependent photoionization cross-sections in the valence electrons, one can probe atomic orbital dependent electronic structures by X-ray polarization-dependent HAXPES [1]. We briefly show how the X-ray polarization affects the valence band HAXPES spectral shapes of 3*d*, 4*d*, and 5*d* transition metals [1-3]. The tendency of photoionization cross-section in the 3*d*, 4*d*, and 5*d* transition metals helps ones to understand the valence band spectra of multinary compounds [1-3]. Since HAXPES is a bulk-sensitive probe of electronic states due to the large inelastic mean-free-path of photoelectrons, it is used to detect the electronic states of not only bulk but also buried interfaces [4]. To enhance the sensitivity to buried interfaces, take-off angle dependent HAXPES measurements are generally performed. Instead of take-off angle dependent HAXPES, we will show a method for probing the buried interface and bulk electronic structures of magnetic thin films under MgO or AlO_x layers obtained by HAXPES combined with X-ray total reflection [5]. We will also present the result of spin-resolved valence band HAXPES spectra for buried Fe films obtained by using an ultracompact Mott-type spin filter [6]. This spin filter can be mounted on a sample carrier, and would be compatible with most HAXPES apparatuses without modifying an electron analyzer [6]. Although the angle-resolved PES for band dispersion measurements is difficult to perform with hard X-rays, spin-resolved HAXPES, which gives Brillouin-zone (or momentum) averaged information due to a low Debye-Waller factor in HAXPES, can be a powerful tool for studying bulk-derived spin-dependent electronic states of magnetically ordered materials.

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Gap States at SiO_2/4H-SiC interface obtained from Oprando Hard X-ray Photoelectron Spectroscopy

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SiC is attracting a great attention for high temperature, high frequency, and high voltage operation power devices. Although SiC has a great advantage for the preparation of MOS structures, one of big issues on SiC is the formation of high density of gap states at SiO_2/SiC interface. This causes a poorer channel mobility than predicted. Therefore, control of the gap states is crucial for fabrication of the high quality SiC based MOS devises. However, the gap states densities are usually below 0.1% interface atomic density, which hardly ever detect them directly. Recently, we developed a new spectroscopic method to observe the energy distribution of gap states. This method is based on operando hard x-ray photoelectron spectroscopy (OP-HAXPES). In the present study, we investigated the energy distribution of the gap states at the SiO_2/4H-SiC(0001) interface using OP-HAXPES.

The OP-HAXPES measurements were performed using BL15XU in SPring-8. An incident photon energy was 5.95 keV while a total energy resolution was set to 240 meV.

The binding energy of the SiC substrate in Si 1s was shifted by applying a bias voltage between top electrode and rear of the SiC substrate. These shifts were attributed to electron occupation and electron vacancy in the gap states at SiO_2/4H-SiC(0001) interface. Therefore, analysis of these core-level shifts as a function of bias voltage enables us to obtain the energy distribution of the gap states at SiO_2/4H-SiC(0001) interface, which is shown in Fig. 1. For the gap states, continuous states in the whole gap and the state with the discreate energy level near the CBM are observed. The detail discussions will be performed in the conference.

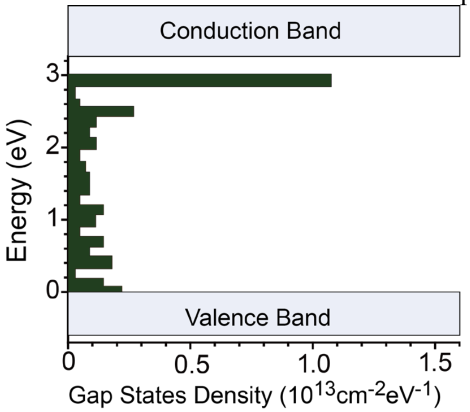

Fig. 1: *Energy distribution of the gap states at SiO_2/4H-SiC(0001) interface.*

X-SPEC: A 70 eV to 15 keV undulator beamline for electron and x-ray spectroscopies

Lothar Weinhardt[1,2,3], Ralph Steininger[1], Dagmar Kreikemeyer-Lorenzo[1], Stefan Mangold[1], Dirk Hauschild[1,2,3], David Batchelor[1], Thomas Spangenberg[1], and Clemens Heske[1,2,3]

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Electron and x-ray spectroscopies are highly valuable techniques for material characterization, both in fundamental and applied studies. Applied material systems highly profit from a combination of several of these techniques, offering different information depths with complementary information content. In particular, x-ray emission spectroscopy (XES), resonant inelastic x-ray scattering (RIXS), and hard x-ray photoelectron spectroscopy (HAXPES) have seen a significant development over the last years and create a unique method combination.

In this contribution, we report on the X-SPEC beamline [1], which was recently built, commissioned, and is now operated at the KIT light source. X-SPEC is a high-flux spectroscopy beamline for electron and x-ray spectroscopy, featuring a uniquely wide photon energy range. Using a combination of soft and hard x-ray undulator sources and optics, X-SPEC can cover an energy range from 70 eV up to 15 keV. The flux of the beamline is maximized by optimizing the magnetic design of the undulator, minimizing the number of optical elements, and optimizing their parameters.

The beam can be focused into two experimental stations that are designed for measuring solid samples under UHV conditions, as well as for *in situ* and *operando* studies under ambient conditions, respectively. Measurement techniques include x-ray absorption spectroscopy (XAS), extended x-ray absorption fine structure (EXAFS), low-energy photoelectron spectroscopy (PES) and HAXPES, as well as XES and RIXS. The wide energy range of the beamline enables us to combine very surface-sensitive synchrotron PES measurements, using electron energies at the minimum of the inelastic mean free path, with more bulk-sensitive HAXPES at energies up to 15 keV.

We will discuss the design considerations and the resulting beamline layout, describe the performance of the beamline, and show first spectroscopic results.

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Upgrade of a HAXPES beamline BL09XU of SPring-8

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SPring-8 is conducting extensive upgrades of beamlines so as to introduce novel scientific capabilities and to reinforce key functions as a research infrastructure. In 2021, an X-ray undulator beamline BL09XU, which had been used for hard X-ray photoelectron spectroscopy (HAXPES) and nuclear resonant scattering (NRS) applications, was converted to a beamline dedicated for HAXPES with merging the relevant activities at BL47XU. The key points of the upgrade are summarized as follows [1]:

1) Two types of analyzers, high-voltage-resistance (up to 12 kV) and wide-acceptance-angle (±32°) analyzers, are installed for the experimental hutch (EH) 1 and 2, respectively.
2) Different types of high-resolution X-ray monochromators (HRMs), as well as double phase retarders for polarization control, are installed into the optics hutch (OH). A high tunability of the X-ray beam performance over a wide photon energy range is achieved.
3) A Wolter focusing mirror is installed in EH1 so as to provide a brilliant micron-sized X-ray beam with high stability and reproducibility.
4) It is planned that a nano focusing device will be installed for EH2 for conducting nano HAXPES analysis.

One of the specific targets at BL09XU is the resonant HAXPES analyses that we recently developed. In the conventional HAXPES, the photon energies are fixed at discrete values, i.e., 6, 8, and 10 keV, by using high-resolution post–monochromator of Si 333, 444, and 555, respectively, with a fixed Bragg angle. In the resonant HAXPES, however, we have made the incident photon energy sweeping available around an absorption edge of a specific element, providing unique opportunities for extracting bulk electronic states associated with specific elements and orbitals. The upgraded beamline will expand the applicable targets of resonant HAXPES from fundamental physics research to applied materials development, such as studies of photocatalytic, steel, and spintronic materials.

In this presentation, I will show the beamline characteristics, the commissioning results, and examples of pilot experiments.

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Atomic Inner-Shell Resonance, Threshold, and Double Ionization Studies

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We are using high-resolution electron spectroscopy and hard x rays for studies of atomic inner-shell photoelectron and Auger electron dynamics. In experiments at the Advanced Photon Source (APS), we studied L-shell Auger electron generation across the Kr K-edge at 14327 eV. Kr 1s hole states have high (64%) radiative decay probabilities. We recorded the 1611-eV L_2-$M_{4,5}N_{2,3}$ Auger line as x-ray absorption excites the Kr 1s-np Rydberg series below threshold and the 1s continuum above threshold. The Auger line is only weakly generated by direct L_2 photoionization and strongly enhanced by K-L_2 x-ray emission. Consequently, the yield of the L_2 Auger line follows the K-edge absorption cross section, including the 1s-np Rydberg series, resulting in resonant Auger lines. Above threshold, energy shifts of the Auger line result from post-collision-interaction (PCI) with 1s photoelectrons. Due to the 2p-1s fluorescence time delay [1], modeling PCI requires combining the 0.24-fs 1s lifetime with the 0.50-fs $2p_{1/2}$ lifetime.

Double photoionization of He is a dynamical process driven by electron correlation. It has motivated theory and experiments on heavier atoms that use K^{-2} hypersatellites in x-ray emission and Auger electron spectra to determine KK/K cross-section ratios [2,3]. Using high-resolution electron spectroscopy and 2.3-keV x rays at the GALAXIES beam line of SOLEIL, Goldsztejn *et al.* recorded rich structure in the Auger electron spectrum of Ne resulting from $K^{-2}V$ and $K^{-1}L^{-1}V$ shakeup states as well as K^{-2} hypersatellites [4]. In order to determine how the electron correlation states and the KK/K ratio varies with x-ray energy, we recorded the high-resolution Auger electron spectrum of Ne at several energies over 2.3-6.5 keV at the GALAXIES beam line. Using an identical electron spectrometer at the APS, we extended the Ne measurements to 8.5 keV. Our goals for this study are to determine how the $K^{-1}L^{-1}V$ and K^{-2} probabilities vary with x-ray energy both experimentally and in comparison with theory.

This work was supported by the US Department of Energy, Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences, and Bioscience Division

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First HAXPES results on clusters: K-shell photoexcitation and subsequent electronic decays in Ar clusters

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The access to the hard X-ray regime has opened new avenues in atomic and molecular physics [1], and HAXPES has brought new insights for studies of liquid and solid materials [2]. While these fields are now well-established, after pioneering photoabsorption studies already in early 90's by Rühl *et al.* [3], gas-phase clusters have been an uncharted territory in the landscape of HAXPES research. Clusters are small atomic or molecular aggregates, which bridge the gap between isolated species and condensed phase. We report here a study of K-shell excitation (~3.2 keV) and subsequent Auger decays in Ar clusters. The experiment was performed at GALAXIES beamline (Synchrotron SOLEIL, Saint Aubin, France) [4]. The clusters were formed via adiabatic expansion of Ar (backing pressure 8 bars) through a 25 μm nozzle in room temperature leading to mean size of about 30 atoms per cluster. The high photon flux and high-resolution electron analyser allowed us to record 2D maps in the region of the Ar 1s resonances, following evolution of KLL, KLM, and LMM Auger spectra across the resonances to the region dominated by EXAFS oscillations. Reference measurements for atomic Ar facilitated disentangling collective and environment induced effects appearing in Ar clusters. These experiments serve as a starting point for investigations of different types of clusters (e.g. hydrated species, core-shell clusters) bringing important insight for e.g. fundamental charge transfer processes.

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Translational and Rotational Recoil Effects In HAXPES of Free Molecules

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The translational and rotational recoil effects together with related Doppler effect in X-ray spectroscopy being natural fingerprint of nuclear motion gives deep insight in molecular nuclear dynamics in bound and dissociative states [1]. The Doppler labeling of the fragments of the dissociation together with the frequency controlled duration of X-ray scattering provides direct and detailed information about the potentials and dynamics in repulsive states. The rotational Doppler effect [1] in X-ray ionization gives site selective information about the valence molecular orbitals[1]. The Doppler splitting of X-ray resonances [1] gives also an inique tool to study the nonequibrium distributions over translational and rotational degrees of freedom. The Doppler control of the amount of which path information allowed us recently to attack the background problem of the wave particle duality in Einstein-Bohr recoiling double-slit gedanken experiment [1,2]. We demontrate at first time the dynamical rotational Doppler effect in high energy Auger spectra of CO and show the quantum correlation of rotational and translational degrees of freedom [1,3]. This was performed by hard X-ray C1s ionization of carbon monoxide accompanied by spinning up the molecule via the recoil "kick" of the emitted fast photoelectron. To visualize the molecular rotation, we use the dynamical rotational Doppler effect and an X-ray "pump- probe" device offered by nature itself. The time information in our experiment [8] originates from the natural delay between the C1s photoionization initiating the rotation and the ejection of the Auger electron. We overview here our recent results devoted to the role of translational and rotational recoil effects on nuclear dynamics of molecules and clusters in hard X-ray region (>10 keV) including recoil-induced dissociation [1,4]. We also overview our recent results in direct time-resolved studies of the recoil-induced rotation.

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Gas phase HAXPES studies

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Gas phase HAXPES is a technic receiving a growing interest. The first results (see ref. 1 for a recent review) have been obtained at SOLEIL between 2.3 and 13 keV of photon energy. This technic has been successfully developed at Spring8 (Japan) [2] and APS (US) (see invited talk of Stephen SOUTHWORTH) taking advantage of the scientific opportunities offered by the hard x-ray domain.

There are several good reasons to use this technic:

- A high-resolution monochromator combined with high resolution hemispherical analyzers allow to perform measurements with a resolving power of about 100000!
- Double Core Hole dynamics [3-7] are measured with a very good signal/noise ratio,
- Post Collision Interaction [8-9], much stronger than in the soft X-ray domain, are leading to interesting effects.
- Strong translational [10], vibrational [11] and rotational [11, 12] recoils occurring at high photon energy can finely observed.
- Photoionization dynamics of clusters can be obtained (see talk of Minna PATANEN).

After showing recent results, I will discuss some scientific perspectives of this technic.

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Depth-resolved HAXPES study of (Ag,Cu)(In,Ga)Se_2 thin film solar cells

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Thin film solar cells based on Cu(In,Ga)Se_2 (CIGS) absorbers have shown high and stable efficiencies for both laboratory cells and industrial modules with record cell efficiencies of 23.4%. To reach closer to the theoretical maximum efficiency of around 30%, further reductions of optical and electrical losses are needed. Recent works show that silver (Ag) alloying in CIGS to form (Ag,Cu)(In,Ga)Se_2 (ACIGS) leads to higher device efficiencies as compared to similar CIGS devices without Ag. Moreover, Ag incorporation increases the band gap, which can be exploited to pave the way for high band gap applications such as tandem solar cells. Further, a postdeposition treatment (PDT) based on alkali metal fluorides after absorber formation has been studied recently and is known to improve the efficiency in CIGS solar cells. The exact role of the silver and/or alkali elements is intensively discussed, but it is clear that interface and grain boundary effects are important in (A)CIGS solar cells. It is likely that a redistribution of the absorber elements near the surface region may occur during the alkali PDT and some works show interface formation in CIGS solar cells subjected to a metal fluoride post deposition treatment.

In this work we employed HAXPES at varying photon energies to study the near surface region of PDT-ACIGS absorbers, which have been exposed to different metal fluorides during preparation, and the interface with a CdS buffer layer. Two different alkali PDT are compared, RbF and CsF, to investigate how the choice of the metal fluoride treatment affects the buffer/absorber interface. In particular, the chemical and electronic properties of CdS/ACIGS interfaces in terms of intermixing, energy band alignment and composition for a set of device relevant samples have been studied. The results show that the electronic structure at the CdS/ACIGS interface does not change for the different PDTs. However, the ACIGS composition at the near surface region seems to change depending on the applied PDT process. Possible reasons for this will be discussed. The results give insights into how selective alkali PDT could change the ACIGS surface composition, which may influence the solar cell behavior.

Using Hard X-ray Photoelectron Spectroscopy to Probe Rechargeable Battery Interfaces

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Battery ageing often begins at the interfaces between electrodes and electrolyte through phenomena such as phase transformation, surface layer (interphase) formation, cationic/anionic redox reactions and metal dissolution. By employing advanced characterisation techniques, we can gain a better fundamental understanding, on the atomic level, of the interfacial processes occurring within modern rechargeable batteries. Hard X-ray photoelectron spectroscopy (HAXPES) is perfectly suited to determining composition, oxidation states, and bonding environments within the surface regions (~50 nm depth) of battery materials. Through tuning of the incident X-ray energy, a non-destructive depth profiling experiment can offer a true surface *vs.* bulk picture. Correlation of such data from advanced characterisation with electrochemical cycling performance allows for greater understanding of ageing mechanisms and informs strategies to stabilise interfaces for ultra-high performance batteries.

I will present examples of our research from the Ångström Advanced Battery Centre and how HAXPES depth profiling plays a large role in many of our studies of Li-, Na-, and K-ion batteries. For Li-rich cathode materials (e.g. $Li[Li_{0.2}Ni_{0.2}Mn_{0.6}]O_2$, Li_2VO_2F), we have characterised cationic and anionic redox activity through the use of HAXPES, as well as determined degradation mechanisms, which can result in rapid capacity fading [1]. Strategies including material doping, particle coatings and electrolyte additives have been developed to improve performance. For Na-ion batteries, we are developing novel fluorine-free and non-flammable electrolytes; HAXPES is employed to study the composition and stability of interphase layers formed on hard carbon electrodes [2]. Finally, for K-ion batteries we have demonstrated the poor stability of interphase layers formed on graphite electrodes through HAXPES depth profiling [3]; the challenge remains to develop high-performance electrolytes for such systems.

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2p2-3 : Invited

Operando high pressure catalytic studies using HAXPES

Blomberg Sara

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Performing fundamental operando catalysis studies under realistic conditions is key to further develop and increase the efficiency of industrial catalysts. To gain knowledge on an atomistic level, simplified model systems are often used and studied at well-controlled conditions at low pressure using surface-sensitive techniques. These conditions differ significantly compared to industrial applications, where the catalysts are operating at atmospheric pressure and above. There is therefore an ongoing debate, on whether the results achieved at low pressures are relevant also for real applications. The discussion introduced the concept “pressure gap” between fundamental research and applications and inspired research to attempt to bridge this gap. Over the last decades, a great effort has been made to develop surface-sensitive techniques to operate at more realistic conditions. Ambient Pressure X-ray Photoelectron Spectroscopy (AP-XPS) is a powerful tool in the field of surface science and catalysis with the capability of probing both gas phase adjacent to the catalysis surface simultaneously as the chemical composition of the catalyst. By using grazing incidence HAXPES, surface-sensitive *operando* studies at high pressure can be performed.

We have used *operando* HAXPES to study model catalysts in a wide pressure range to achieve a complete understanding of the catalytic process at surfaces under industrial conditions. Recently we performed an *operando* CO oxidation study at 1 bar total pressure using a novel XPS setup at Petra III, DESY in Germany, aiming to bridge the pressure gap (Fig 1)[1].

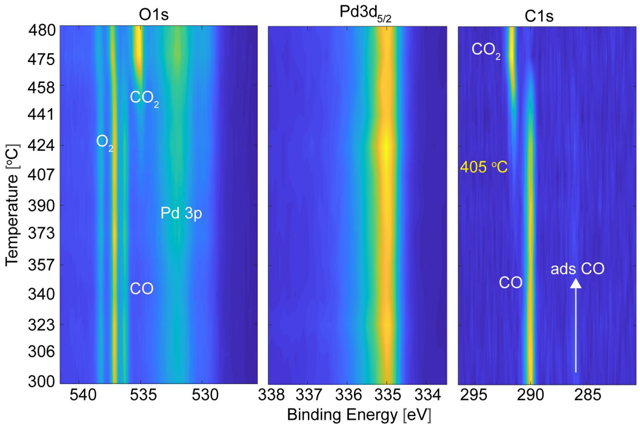

Fig. 1: *The light-off region for CO oxidation using a Pd(100) model catalysts at a total pressure of 1 bar The* CO_2 *gas-phase peak presence is a clear marker of the light-off of the catalyst, which is around 405 °C. The intensity of the spectra is color-coded, ranging from dark blue to yellow, where the latter indicates high intensity.*

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Co(0001) Fischer-Tropsch observations at 550mbar (and above) using a virtual cell approach and hard X-rays

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Catalytical reactions are both societally important and difficult topics for fundamental research. The importance can be easily demonstrated by recalling the numerous products (fertilizer, fuels, plastics …). Especially in combination with the need to move towards CO_2 neutral procedures, long standing paradigms need to be challenged. Fischer-Tropsch reactions in particular can be run to produce very different distributions of valuable hydrogenated carbons, that can be used in fuels and energy distribution schemes of the future completely based on regenerative resources.

Despite knowing that this catalytic reaction exists, available literature detailing the debated reaction mechanisms is lacking [1, 2]. Indeed, only indirect measurements (e.g. by QMS), observations far away from reaction conditions (XPS < ~10mbar) or after the reaction can be found. As seen by many publications XPS can address the reaction mechanisms in question. However, combining ultra-high vacuum (UHV), as it is required to transfer photoelectrons without scattering and high pressures (above 1 bar), as it is required for the catalytic reaction, has posed significant technical challenges and caused a pressure gap above the ~10mbar range.

The POLARIS setup (Stockholm University) is in operation at P22, PETRA III [3] and can readily be employed above this pressure gap by utilizing a virtual cell concept. Here we set out with the goal of observing the FT reaction at pressures as close as possible to the pressures applied in industrial catalysis and compare isobars at 150, 550 and 1000 mbar of undiluted reactants.. Onto the single crystalline cobalt we direct mixtures of $CO:H_2$, $CO_2:H_2$ and $CO:CO_2:H_2$. We obtain Co2p, C1s and O1s core level spectra and quantify the adsorbed species under these conditions. Our results reflect recently published results from operando STM[4] and show significant differences to findings from post-reaction XPS[5].

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Laboratory HAXPES of GaN structures for power electronics

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High-electron-mobility transistors (HEMTs) based on AlGaN/GaN heterostructures are being developed for high power electronics. The presence of a two-dimensional electron gas (2DEG) at the AlGaN/GaN interface enables reaching high electron mobility (>1500 $cm^2.V^{-1}.s^{-1}$). For safety reasons, “normally-off” HEMTs are requested and can be obtained by the full recess approach, based on the removal of the AlGaN layer underneath the gate. The dielectric (Al_2O_3) and metal gate are deposited on the etched GaN surface to create a metal-insulator-semiconductor (MIS) structure, giving rise to the so-called MIS-HEMTs [1].

The final device properties strongly depend on the quality of the buried Al_2O_3/GaN interface [2]. Advanced chemical characterization of this critical interface is thus mandatory. Laboratory HAXPES has recently appeared and its use is quickly rising for optimizing technological devices [3]. It is here a promising technique to investigate a more realistic Al_2O_3/GaN interface, e. g. with a thick Al_2O_3 layer. We use it to investigate the impact of Al_2O_3 thickness, growth process and post-deposition anneal on the GaN surface oxidation and stoichiometry.

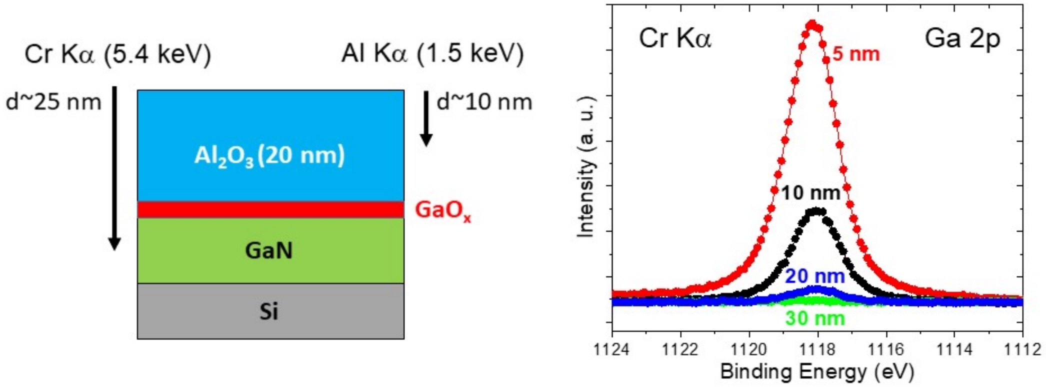

Fig. 1: *Comparison of the sampling depth for the Al_2O_3(20 nm)/GaN stack with Cr Kα and Al Kα sources and Ga 2p spectra measured at 45° with a Cr Kα source through varying Al_2O_3 thicknesses.*

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Strong Dimensional Character in layered ReS$_2$ and ReSe studied by complimentrary x-ray photoelectron spectroscopy

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Using complimentary x-ray photoelectron spectroscopy techniques, we study the electronic structures of two-dimensional van der Waals (vdW) compounds ReS$_2$ and ReSe$_2$. The in-plane and out-of-plane valence band character are studied by varying the polarization of incoming photons, we show the out-of-plane transition metal dz^2 and chalcogen p_z orbitals do not contribute significantly to the top of the valence band, supporting a weak interlayer coupling in this class of vdW materials. Furthermore, ReS$_2$ shows two distinct Re sites that are linked to strong intra-plane Re-Re metallic bonds. Resonant Auger Spectroscopy (RAS) taken across the S *K*-edge for ReS$_2$ is employed to obtain electron delocalization times across the conduction band down to the attosecond time scale. The intrinsic electron dynamics of ReS$_2$ shows a significant difference in delocalization times between *intra* and *inter*-plane orbitals. Our results are supported by density functional theory calculations that provide insight into the layer dependent properties of rhenium dichalcogenides and highlight their place among two-dimensional crystals.

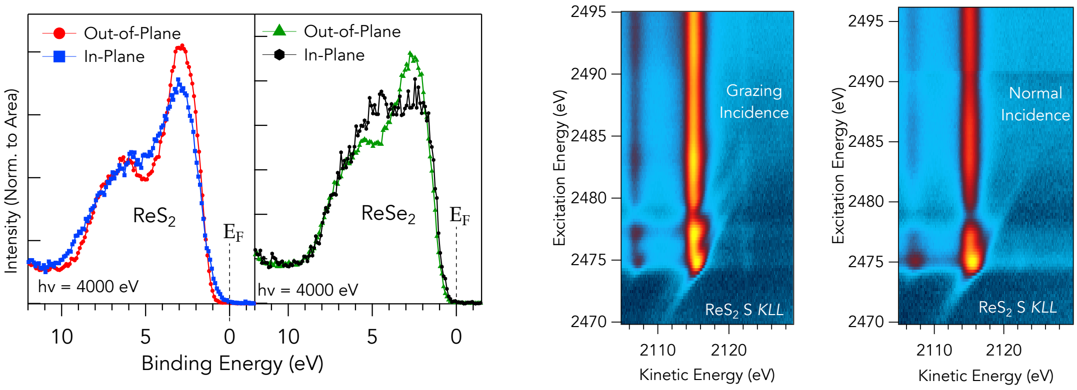

Fig. 1: *(left) Hard -x-ray photoemission spectra of the valence band (VB) for ReS$_2$ and ReSe$_2$ with different light polarizations to study the inter and intra-plane valence states. ReS$_2$ shows a valence band maximum that changes for in-plane and out-of-plane states. (right) Resonant Auger maps of S KLL for ReS$_2$ that show different delocalization pathways for in-plane and out-of-plane states.*

Exploring surface catalytic nature with ambient pressure XPS

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A synchrotron-based ambient pressure XPS (AP-XPS), first reported from ALS in 2001, has made a significant impact on the community of surface science over the years. By exploring the nature of surface under elevated pressure condition, many stimulating results have been reported, clearly putting a new road map on the surface science. While the application of AP-XPS covers the various fields of surface science, e.g. from traditional gas molecule interaction on solid surface to liquid/solid interface in electrochemistry, the community of surface catalysis has gained much benefits from the progress of AP-XPS.

In this presentation, various surface catalytic reactions on metal surfaces will be explored with the use of AP-XPS, i.e. CO_2 dissociation on Rh (111), CO oxidation on $Pt_3Ni(111)$ and Pt_3Co (111). [1,2] An ambient pressure scanning tunneling microscope (AP-STM) is accompanied to deliver the information of surface morphology under reaction condition, adding a new dimension in analyzing surface chemical reactions in real time. In addition, the pro and cons of using hard x-ray in AP-XPS is also presented.

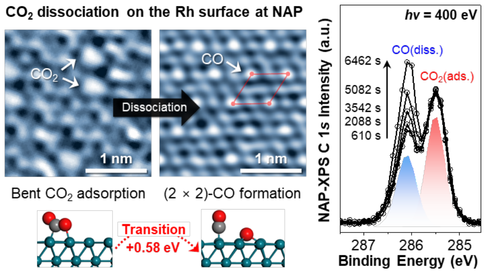

Fig. 1: *Time-lapse in situ observations on the Rh(111) surface in 0.1 Torr* CO_2 *using NAP-STM and NAP-XPS at 300 K.*

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3a1-2 : Invited

The Electronic Structure of Degenerately doped Ga_2O_3 Thin Films Studied by HAXPES

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Ga_2O_3 is emerging as a promising wide bandgap semiconductor for high-power electronics and solar-blind ultraviolet photodetector. It is highly desirable to dope it with controllable carrier concentrations for different purpose of applications. This talk reports the realization of degenerate doping of Ga_2O_3 with Si (SGO) thin films with carrier concentrations ranging from 5×10^{18} up to 2.6×10^{20} cm^{-3}. A record high conductivity of 2520 S/cm and mobility of 60 cm^2/V s was achieved. The SGO films show a high transparency over a wide spectrum from visible to deep UV. Hard x-ray photoelectron spectroscopy (HAXPES), optical spectroscopy and DFT calculations were also performed to elucidate insights on the optical and electronic structures. HAXPES at the valence band observed the widening of bandgap as a result of occupation of the conduction band (CB) and the shrinkage of bandgap associated with many-body interaction. Supplemented with DFT, we demonstrate that the bandgap shrinkage mainly result from the change of CB induced by mutual electrostatic-interaction. Because the Si energy level is sitting 3 eV above the CB bottom, there is little hybridization between Si orbital and Ga 4s derived CB, leaving the CB edge unperturbed and a small electron effective mass maintained though the doping level is high.

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Abstract
Poster session

Resonant Auger of Xe on transition metal substrates

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A study of the Auger decay process is presented for solid Xe on 3d transition metals with varied work function. Resonant Auger spectroscopy have shown to be a valuable method to study charge transfer and electronic structures of solids using the core-hole clock method [1]. In this work, the Xe L_3 edge was studied on a Xe monolayer adsorbed on metal surfaces using hard X-ray spectroscopy. Using core excitations from Xe $2p_{3/2}$ level we have recorded the resonant Auger spectra with electron kinetic energies in the LMM kinetic energy region. This was done for several excitation energies and the partial Auger (decays from charge transfer states where the electron in the excited state have tunneled away) and Raman (decays from states where the excited electron has remained on the atom with the core-hole) yields were extracted as well as the charge transfer time of core-excited states. We compare the results from the condensed case with that of Xe Resonant Auger from a gas-phase sample [2]. For instance, the charge transfer channel is not available in the isolated atom case.

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P2

In-Plane Orientation of the 4*f*-Orbital Distribution in YbRh$_2$Si$_2$ Probed by Linear Dichroism in Core-Level HAXPES

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Rare-earth Ce and Yb compounds exhibit various intriguing physical properties such as non-Fermi-liquid behavior and unconventional superconductivity near the quantum critical point (QCP). The Yb (Ce) 4*f*-orbital holes (electrons) with the $4f^{13}$ ($4f^{1}$) configurations are responsible for these properties. Such 4*f* electrons often show the dual nature of the localization and itinerancy by the hybridization with the conduction electrons. The low-temperature 4*f* electronic phases as a function of the hybridization strength and temperature is often represented by the Doniach phase diagram [1]. It has been recognized that the 4*f* spatial distribution and its orientation determined by the local crystalline electric fields (CEF) in the localized picture is important for clarifying the physics of the strongly correlated 4*f* electron systems [2].

YbRh$_2$Si$_2$ with the tetragonal ThCr$_2$Si$_2$-type crystal structure shows the non-Fermi-liquid behavior as demonstrated by the electrical resistivity and specific heat [3][4]. Therefore, YbRh$_2$Si$_2$ is considered to be located near QCP where the RKKY interaction and the Kondo effect are competed. Its 4*f*-orbital symmetry, especially the in-plane orientation of the 4*f* charge distribution, has not been rigorously clarified yet.

Here we show the 4*f* ground-state symmetry of YbRh$_2$Si$_2$ revealed by the linear dichroism (LD) in angle-resolved hard X-ray photoelectron spectroscopy (HAXPES) at BL19LXU of SPring-8 with the two-axis angle manipulation [5]. Contrary to the previous LD-HAXPES study with the imperfect experimental condition (lack of the azimuthal angle dependence), we have found the CEF-split 4*f* ground state of YbRh$_2$Si$_2$ as the Γ_7 symmetry with the mixture of $|J_z = \pm 5/2\rangle$ and $|\mp 3/2\rangle$, where J_z stands for the z component of the total 4*f* angular momentum $J = 7/2$ [6]. We have also revealed that the 4*f* charge distribution of YbRh$_2$Si$_2$ as well as valence-fluctuating YbNi$_2$Ge$_2$ is elongated along the transition-metal sites while that of YbCu$_2$Si$_2$ is expanded to the Si sites [6][7]. We also discuss the qualitative difference of the in-plane orientation of the 4*f* distribution in the ThCr$_2$Si$_2$-type crystal structure from that in the Ce compounds, of which the similarity with respect to the electron-hole symmetry between the Ce and Yb systems has been often pointed out.

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Magnetometry of Buried Co-based Nanolayers by Hard X-ray Photoelectron Spectroscopy

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The intensity and shape of photoelectron lines of magnetic materials depend on the relative orientation of the sample magnetization, the X-ray beam polarization and the spectrometer axis, i.e. the electron emission direction. In the hard X-ray regime, the beam polarization can be conveniently modified utilizing the phase shift produced by a diamond phase plate in the vicinity of a Laue or Bragg reflection. A single-stage in-vacuum phase retarder is installed and commissioned in 2020 at the HAXPES beamline P22 at PETRA III (Hamburg, Germany) [1].

The electronic and magnetic properties of CoFe and Co-based Heusler nanolayers were studied using the linear and circular magnetic dichroism in the angular distribution of photoelectrons. The layers were remanently magnetized in-situ and Co $2p_{1/2}$ and $2p_{3/2}$ core levels were probed at room temperature [2-3]. Both the polarization-dependent spectra and the dichroism indicate that the lines of the multiplet extend over the entire spectral range. In particular, the dichroism does not vanish between the two main parts of the spin–orbit doublet.

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Bulk Electronic State of Au-Ga-Ce and Cd-Ce Quasicrystalline Approximants Probed by Hard X-ray Photoemission Spectroscopy

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Strongly correlated rare-earth based quasicrystals and approximants attract much attention due to unique quantum critical phenomena [1]. Among them, Ce-based 1/1-quasicrystalline approximants $Au_{60.3}Ga_{26.1}Ce_{13.6}$ and $Au_{59.2}Ga_{25.7}Ce_{15.1}$ have recently been developed, of which the strongly correlated Ce 4*f* electronic states could be mutually different. In the strongly correlated 4*f*-electrons systems, revealing localized or itinerant character of 4*f* electrons is important to understand the physical properties like a quantum critical phenomena. We have performed hard X-ray photoemission spectroscopy (HAXPES) for the Au-Ga-Ce approximants to find out the character of 4*f* electrons.

HAXPES of the quasicrystalline approximants was carried out in BL19LXU of SPring-8 with energy resolution of 250 meV at $h\nu$ = 7.9 keV. In the Ce 3*d* core-level photoemission spectra, the $3d^9 4f^0$ final-state spectral weight indicating the itinerant 4*f* character is negligible for both compounds. This result indicates that the 4*f* electrons are highly localized in the Au-Ga-Ce approximants. On the other hand, the $3d^9 4f^1$ final-state multiplet structure suggesting the ionic-like localized 4*f* electronic state is strongly suppressed or smeared out in the spectra of Au-Ga-Ce approximants. It is known that there is the chemical disorder (interchange of Au and Ga sites) in the Au-Ga-Ce approximants. Therefore, we have also performed the Ce 3*d* core-level HAXPES for a Cd-Ce approximant without the chemical disorder and found the clear $3d^9 4f^1$ multiplet structure in the Ce 3*d* spectrum of the Cd-Ce approximant. Our result suggests that the chemical disorder strongly influences the electronic state of the Au-Ga-Ce approximants.

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Electronic structure of thermoelectric Heusler-type $Co_2MnSi_{1-x}Al_x$ compounds

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The half-metallic Heusler-type Co_2MnSi compound is expected to be used as a thermoelectric material for high-temperature applications because of its high power factor at high temperatures.[1] In the $Co_2MnSi_{1-x}Al_x$ compounds, the *sp* electrons, which are many-spin electrons, become the main carriers, and a rigid-band shift of the Fermi level to the half-metal gap edge may result in a dramatic change in the Seebeck coefficient.[2] In this study, we report the results of thermoelectric properties and electronic structure in $Co_2MnSi_{1-x}Al_x$ compounds.

The $Co_2MnSi_{1-x}Al_x$ compounds was prepared by arc melting in Ar atmosphere and sintering at 1173 K for 5 min by pulsed current sintering under pressure of 50 MPa. The photoelectron spectroscopy measurements were performed at BL47XU of SPring-8 using an excitation photon energy ($h\nu$) of 7940 eV.

Fig. 1 shows the valence band photoemission spectra of $Co_2MnSi_{1-x}Al_x$ compounds. In the range of $x = 0$ to 0.5, where the power factor increases with increasing Al substitution, the shape of the photoemission spectra does not change significantly, and the electronic structure changes in a rigid band model, shifting to the lower binding energy side with deceasing carrier. In the range of 0.5, the shape of the photoelectron spectrum does not change significantly. On the other hand, for $x > 0.5$, where the power factor decreases abruptly, the photoelectron spectrum becomes broadened due to the decrease in the structural ordering. The present study shows that the rigid band model control of the electronic structure is effective for material design even in systems with a half-metal-like electronic structure.

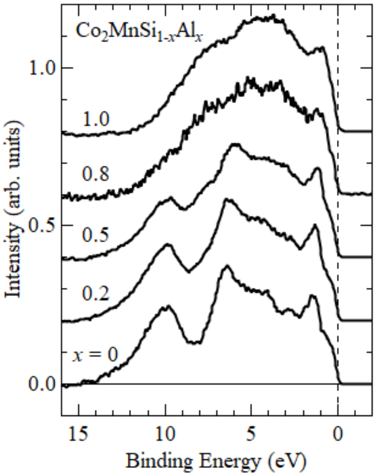

Fig. 1: *Valence band photoemission spectra of $Co_2MnSi_{1-x}Al_x$ compounds with $h\nu = 7940$ eV.*

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Valence fluctuation in thermoelectric material $YbSi_2$ probed by HAXPES

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$YbSi_2$ is a high-performance thermoelectric material with the second highest Peltier coefficient (S/ρ, where S: Seebeck coefficient, ρ : electrical resistivity) among the reported materials and the highest thermoelectric power factor (S^2/ρ) among the Yb-based materials. The thermopower is determined by the density of state near the Fermi energy and it is important to investigate the electronic state. Because Yb $4f$ electron can have both itinerant and localized feature, core level HAXPES will help us to deeply understand the electronic state of Yb $4f$ electron.

Figure 1(a) shows the Yb $3d_{5/2}$ core level HAXPES spectrum at 20 K. Yb^{2+} and Yb^{3+} $3d_{5/2}$ peaks are well separated which indicate the mixed valence state. Figure 1(b) shows the valence band spectrum at 20 K. Yb^{2+} $4f_{7/2}$ peak locates just below the Fermi energy, suggesting the Kondo resonance peak and the contribution of $4f$ electron to the transport property. These results indicate valence fluctuating state which is similar to valence fluctuating materials $YbAl_3$ and α-$YbAlB_4$ with high thermoelectric power factors [1,2]. In the presentation, we will also show the SXPES spectra and explain in detail.

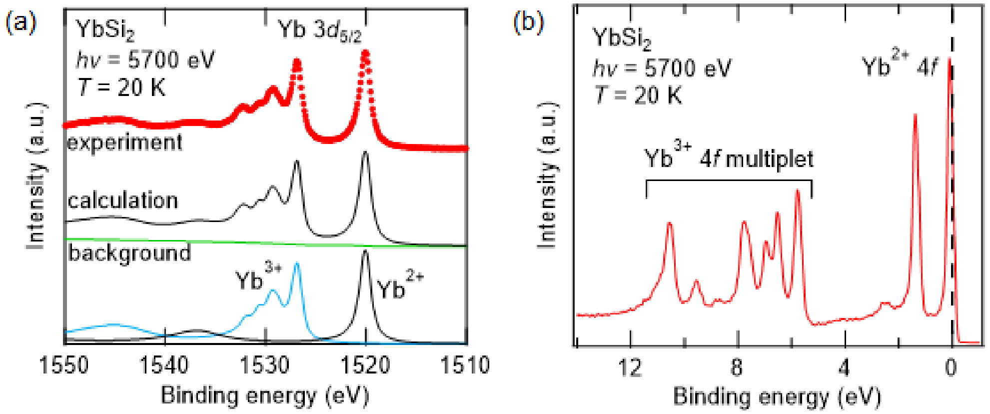

Fig. 1: *Yb $3d_{5/2}$ core level (a) and valence band (b) HAXPES spectra.*

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Relationship of Ce 4*f*-5*d* Coulomb Repulsion on CeT_2X_2 (T = Cu, Ru; X = Si, Ge): Resonant HAXPES Study

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Electronic structure of $CeCu_2Ge_2$ (Kondo temperature: T_K ~5 K) [1,2], $CeCu_2Si_2$ (T_K = 10 K) [3-6], and $CeRu_2Si_2$ (T_K = 20 K) [7] has been investigated by resonant hard x-ray photoemission spectroscopy (rHAXPES) to discuss a relation between Ce 4*f*-5*d* Coulomb repulsion (U_{fd}) [8] and Quantum critical phenomena on the Doniach phase diagram. The rHAXPES experiments around the Ce L_3 absorption region at 20 K were performed at BL09XU of SPring-8 [9]. Ce 3*d* spectra of all the three compounds consist of three features attributed to the $4f^0$, $4f^1$, and $4f^2$ final states. The photon-energy ($h\nu$) distribution of photoemission intensity, that is, constant initial state (CIS) spectrum, of each final state shows distinct resonance enhancement. In particular, the peak positions of CIS spectra depend on compounds. A theoretical analysis based on the single impurity Anderson model was performed to reproduce the $h\nu$ dependence of Ce 3*d* rHAXPES and CIS spectra. The resonance enhancement with $h\nu$ of all the three compounds is well explained by U_{fd}, together with hybridization strength between 4*f* and conduction electrons and charge transfer energy Δ equivalent to the energy position of bare 4*f* level. We found that the U_{fd} becomes larger in order of $CeRu_2Si_2$ (1.70 eV), $CeCu_2Si_2$ (3.00 eV), and $CeCu_2Ge_2$ (3.80 eV). This trend suggests that U_{fd} is one of intrinsic physical parameters to understand the Quantum critical phenomena, even if the phenomena stem from the magnetic fluctuation.

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Charge transfer energy in iridates: a Hard X-ray Photoelectron Spectroscopy study

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We have investigated the electronic structure of iridates in the double perovskite crystal structure containing either Ir4+ or Ir5+ using hard x-ray photoelectron spectroscopy. The experimental valence band spectra can be well reproduced using tight binding calculations including only the Ir 5d, O 2p and O 2s orbitals with parameters based on the downfolding of the density-functional band structure results. We found that regardless of the A and B cations, the A_2BIrO_6 iridates have essentially zero O 2p to Ir 5d charge transfer energies. Hence, double perovskite iridates turn out to be extremely covalent systems with the consequence being that the magnetic exchange interactions become very long-ranged, thereby hampering the materialization of the long-sought Kitaev physics. Nevertheless, it still would be possible to realize a spin-liquid system using the iridates with a proper tuning of the various competing exchange interactions

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Observation of ferroelectric skewed band structure in multiferroic $BiFeO_3$

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Ferroelectric skewed band structure is understood as the combination of band structure and electric polarization, which exhibits behavior as the electron energy level shifts according to the depth in the polarization direction. By using angle-resolved hard X-ray photoelectron spectroscopy (AR-HAXPES), the skewed band structure of ferroelectric $BaTiO_3$ thin film was observed successfully [1]. For further study to clarify the semiconductor physical properties of multiferroics materials, in this research, we prepared single domain $BiFeO_3$ (BFO) thin films with downward polarization vertically oriented to the $SrRuO_3$(~3 nm)/$SrTiO_3$(111) substrate. For the direct observation of ferroelectric band skewing structure, we performed AR-HAXPES of multiferroic BFO thin films. As shown in Fig. 1, the depth profiles of Bi-4*f* atomic orbital consists of two regions: the potential slope according to depth and polarization in the deeper region (FEBS: ferroelectric band skewing), and the slope exhibits flat on the surface (SBS: surface band skewing). Similar behavior is also observed in Fe-2*p* and the valence band.

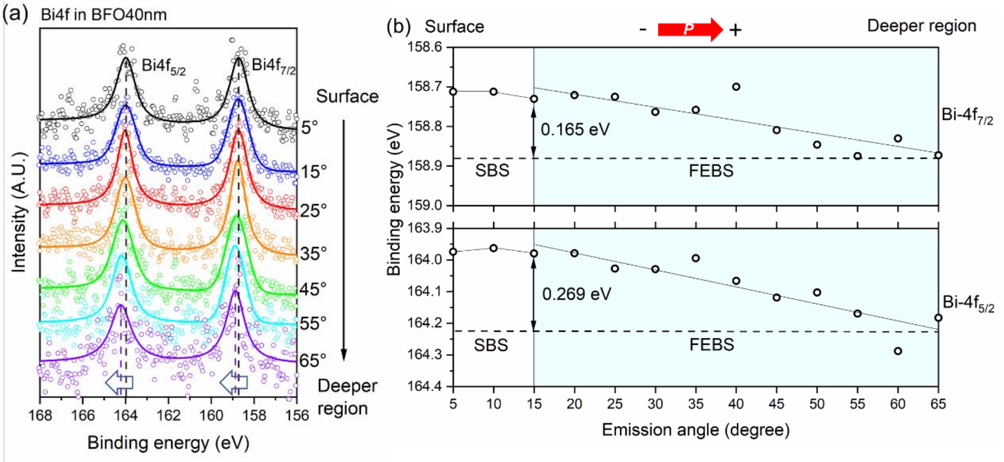

Fig. 1: *(a) AR-HAXPES spectra observed from emission angle 5 ° to 65 ° of Bi-4*$f_{5/2}$* and Bi-4*$f_{7/2}$* in the 40nm thick BFO. Black curve and dots are from the surface, purple curve and dots are from the deeper region. (b) Depth dependence of binding energies of Bi-4*$f_{5/2}$* and Bi-4*$f_{7/2}$* in the 40nm thick BFO. Red arrows indicate the inherent direction of the electric polarization for as-grown film.*

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Dy 3d Core-Level Photoemission Lineshape and Its Correspondence with Chemical State

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There are a variety of Dy (dysprosium) compounds that have been attracting strong interest such as Dy-doped Nd-Fe-B permanent magnet. In the study of chemical state and electronic state of Dy in such compounds, Dy 3d core-level photoemission (Dy 3d XPS) is important. Since the binding energy of the Dy 3d core level is around 1300 eV, photoelectron energy is as low as 200 eV when Al K α X-ray source is used. Then, the effect of the secondary electrons background is quite serious and also the spectrum is quite surface sensitive. Therefore, only very small number of experimental studies have been performed. In other words, hard X-ray photoemission is expected to be a powerful tool.

Another aspect of Dy 3d XPS is that multiplet structures, due to exchange interaction between 3d core and 4f orbital, is expected. Such multiplet structures may sometimes be mistakenly assigned to different chemical states. In Eu^{2+} compounds, for example, the structure in the larger binding energy region of 3d XPS turned out to be nothing but a part of multiplet structures [1].

In this study, Dy mental and Dy oxide have been studied by X-ray photoelectron spectrometer (PHI Quantes, Dual Scanning X-ray Photoelectron Microprobe) equipped with hard X-ray (Cr Kα) and soft X-ray (Al Kα) monochromatized sources at SR Center, Ritsumeikan University.

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Resonant hard X-ray photoemission on valence fluctuating system α-Yb($Al_{1-x}Fe_x$)B_4

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The strongly correlated 4*f*-electron system β-$YbAlB_4$ exhibits unconventional superconductivity at ambient pressure with zero field below 80 mK whereas α-$YbAlB_4$ shows Fermi liquid behavior [1,2]. However, by doping with 1.4 % Fe in the Al sites, α-YbAlB4 becomes an anomalous metallic state and shows quantum criticality like β-$YbAlB_4$ [3]. When Fe is doped above 1.4 %, the valence of Yb increases rapidly, and thus the critical valence fluctuation is considered to play an important role in the mechanism of the anomalous properties [4]. This critical valence fluctuation is theoretically attributed to the Coulomb interaction U_{fd} between 4*f* electrons and conduction electrons [5]. Therefore, we have attempted to evaluate U_{fd} experimentally by Yb 2*p*-5*d* resonant hard X-ray photoemission spectroscopy (rHAXPES) for α-$YbAlB_4$ doped with Fe. The experiment was carried out at BL09XU of SPring-8 at a temperature of 20 K and an energy resolution of 250 meV.

U_{fd} is estimated by the difference in the center-of-gravity energy and resonance energy of the Yb^{2+} and Yb^{3+} components of the Yb $3d_{5/2}$ core-level photoelectron spectrum. As shown in Fig. 1(a), the Yb $3d_{5/2}$ rHAXPES spectra of α-$YbAlB_4$ shows a resonance enhancement. Constant Initial Sate (CIS) spectra are extracted from spectrum at each peak. CIS spectra shown in Fig. 1(b) give different resonance energies depending on the structure and composition. From this result, we found that U_{fd} shows a composition dependence.

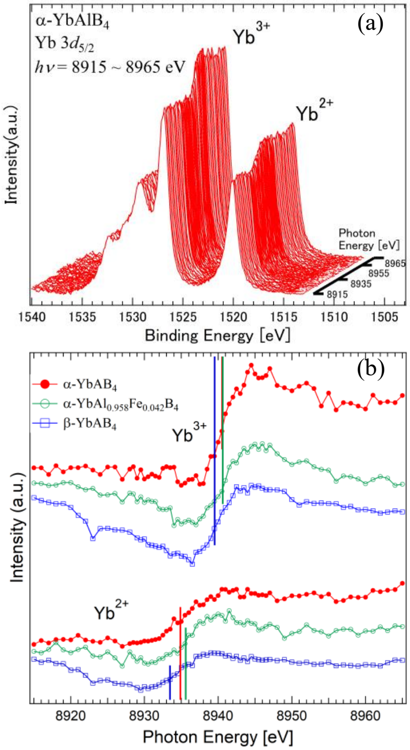

Fig. 1: *(a) rHAXPES spectra of Yb $3d_{5/2}$ for α-$YbAlB_4$. (b) CIS spectra of Yb^{2+} and Yb^{3+}. The vertical lines indicate resonance energy*

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Systematic charge distribution changes in Bi, Pb-3d transition metal perovskite oxides

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Bi and Pb have a unique $6s^0$ and $6s^2$ electron configuration that creates charge degrees of freedom. Due to the lack of this $6s^1$ electron configuration, a property called valence skipper, Bi takes 3+ and 5+ and Pb takes 2+ and 4+. In particular, for perovskite compounds containing Bi or Pb at the A site, the valence state changes according to the depth of the d-orbitals of the transition metal ions corresponding to the order in the periodic table of the elements due to the close relationship between the 6s level of Pb or Bi and the 3d level of the 3d transition metal ions. For example, for $BiMO_3$, M= Cr, Mn, Fe, Co, the state is $Bi^{3+}M^{3+}O_3$, while $BiNiO_3$ has a specific valence state of $Bi^{3+}{}_{0.5}Bi^{5+}{}_{0.5}Ni^{2+}O_3$. $PbMO_3$ has $Pb^{2+}{}_{0.5}Pb^{4+}{}_{0.5}MO_3$ for M= Ti and V, $Pb^{2+}{}_{0.5}Pb^{4+}{}_{0.5}M^{3+}O_3$ for M= Cr and Fe, $PbCoO_3$ has $Pb^{2+}{}_{0.25}Pb^{4+}{}_{0.75}Co^{2+}{}_{0.5}Co^{3+}{}_{0.5}O_3$, $PbNiO_3$ has $Pb^{4+}Ni^{2+}O_3$. In $BiNiO_3$ and $PbMO_3$ (M = Cr, Fe and Co), Bi and Pb become charge disproportionated in the $6s^0$ and $6s^2$ states, and temperature- and pressure-induced elimination of charge disproportionation and charge transfer phase transitions occur. The valence of the transition metal M is changed in this process, which leads to unique properties such as metal-insulator transition and negative thermal expansion. In this study, we evaluated the electronic states of these materials by HAXPES, and the crystal structure distortions caused by these electronic states were investigated by crystal structure analysis using synchrotron radiation X-ray diffraction and local structure analysis using atomic pair distribution function (PDF) obtained from synchrotron radiation X-ray total scattering patterns.

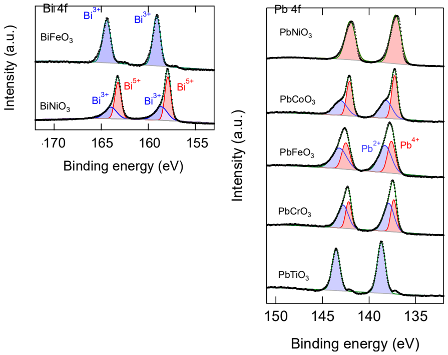

Figure. Bi4f and Pb 4f HAXPES spectra for $BiMO_3$ (M= Ni and Fe) and $PbMO_3$ (M= Ti, Cr, Fe, Co, and Ni)

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Azuma, M.; Sakai, Y.; Nishikubo, T.; Mizumaki, M.; Watanuki, T.; Mizokawa, T.; Oka, K.; Hojo, H.; Naka, M. *Dalton Transactions* **2018**, *47* (5), 1371–137

Electronic States of FeGa Alloy Studied by Hard and Soft X-ray Photoelectron Spectroscopy

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FeGa alloys are one of the Fe-based magnetostrictive materials. These alloys have different crystal structures such as disordered A2, ordered B2, and $D0_3$ structures depending on temperature and composition. Magnetostriction is a phenomenon in which magnetization and elongation occurs in same time, it also called Joule effect. Large magnetostriction over 200 ppm was obtained at room temperature in Fe-Ga bcc single crystal structure with a Ga concentration of 17 at% by Clark et al [1]. On the other hand, magnetization change when external force is applied to the material. The phenomenon is inverse magnetostriction, also it called Villari effect. Because of these phenomenon, FeGa alloys are expected to be applied in not only actuator material but also Power generation device as magnetostrictive material.

In this research, we studied Fe82％Ga18％ single crystals by X-ray photoelectron spectrometer (PHI Quantes, Dual Scanning X-ray Photoelectron Microprobe) equipped with hard X-ray (Cr Kα) and soft X-ray (Al Kα) monochromatized sources at SR Center, Ritsumeikan University.

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Electronic Structure of Covalent-Chain Antiferromagnets $TlFeX_2$ (X = S, Se) Studied by HAXPES

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Electronic structure of the covalent-chain antiferromagnets $TlFeX_2$ (X = S, Se) [1-4], having a monoclinic structure of space group $C2/m$, has been investigated by polarization dependent HAXPES. The polarization dependent HAXPES experiments with incident photon energy ($h\nu$) of 7.05 keV were done at BL09XU of SPring-8. The samples were arranged so that infinite chains, built along c-axis of FeX_4 tetrahedra (β modification), are parallel (perpendicular) to the electric field vector in the p- (s-) polarized configuration. Valence band HAXPES spectra show distinct polarization dependence as shown in Fig. 1. For both compounds, the valence band spectra at the p-polarized configuration correspond to calculated density of state (DOS) except for slight discrepancies around 5 eV, as is the result observed at $h\nu$ = 6 keV [5]. These discrepancies suggest the competition between localized and delocalized characters of Fe 3d electrons. On the day, we are planning to compare the spectra at s-polarized configuration with the calculated DOS and to discuss about the Fe 3d state in $TlFeX_2$ in detail.

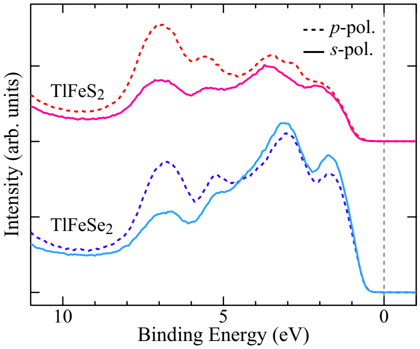

Fig. 1: *Polarization dependence of valence band HAXPES spectra for $TlFeX_2$ (X = S, Se) taken at 300 K.*

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Electronic States of type-II Na-Ba-Si Clathrate Studied by Hard X-Ray Photoelectron Spectroscopy

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Yuri Murakami[2,4], Natsumi Tanaka [2,4], Yuina Kanai-Nakata[1,2],
Hidenori Fujiwara[2,3], Satoru Hamamoto[2], Atsushi Higashiya[2,5],
Atsushi Yamasaki[2,4], Akira Sekiyama[2,3], Kenji Tamasaku[2], Makina Yabashi[2],
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The Seebeck effect is the electromotive force that develops across two points of a semiconductor or metal when there is a temperature difference between them, which has been applied to thermoelectric conversion. In order to improve the performance of thermoelectric properties, the electrical resistivity and thermal conductivity must be reduced. A material that transmits electricity well like crystals and does not transmit heat like glass would be ideal.

The $Na_{16}Ba_8Si_{136}$ belongs to the type II clathrate. Its crystal structure consists of 16 dodecahedron and 8 hexakaidecahedron cages. The Ba and Na atoms occupy the 8b and 16c sites in the cage, respectively. The atoms in the cage can move around inside the cage. By measuring the core-level spectra of the three elements, the electronic states can be understood. Experiments have been carried out at BL19LXU in SPring-8 using MBS A-1 analyzer.

Typically, there is only one peak per core-level, however, peak splitting was observed in the core-level spectrum of the clathrate compound. The fact that the atoms inside the cage can move around may be one of the reasons for the peak splitting. In order to know how the electronic states of the material changes at different temperatures, we have also measured the core-level spectra and the valence band spectra by x-ray photoelectron spectroscopy at 5K, 100K, 180K, and 260K.

Ir 5d State of CuIr$_2$S$_4$ Studied by Ir 2p$_{3/2}$-5d Resonant HAXPES

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CuIr$_2$S$_4$ exhibits structural, metal-insulator, and paramagnetic-diamagnetic transitions around 226 K [1]. The Ir 5d state of CuIr$_2$S$_4$, which plays an important role in the transitions, has been investigated by Ir 2p$_{3/2}$-5d resonant HAXPES at 300 and 150 K. Figure 1 shows the on- (solid curve) and off- (dotted) resonant valence-band (VB) photoemission spectra at 300 K along with difference (open circles) between the two spectra. Resonant enhancement of the Ir 5d photoemission is not so strong as was expected, possibly because of the itinerant nature of the Ir 5d state to some extent. Owing to the photoionization cross sections, however, the Ir 5d-derived shoulder and peak are clearly observed at 0.5 and 1.6 eV, respectively, in contrast to the VB spectrum taken with $h\nu$=1.3 keV [2]. The shoulder and the peak closely resemble those predicted theoretically [2]. The Ir 5d state is redistributed by cooling below the transition temperature. Change in the difference spectrum is observed around 3, 5.5, and 10.5 eV as indicated by arrows.

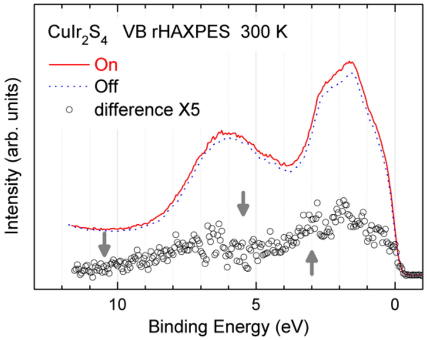

Fig. 1: *On- (solid curve) and off- (dotted) resonant valence-band spectra of CuIr$_2$S$_4$ in the Ir 2p$_{3/2}$-5d absorption region around 11.2 keV. Open circles are difference between the two spectra multiplied by 5.*

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Observation of electronic structure of Co_2NbSn by hard x-ray photoelectron spectroscopy

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Goro Nozue[2,3], Hayato Hashizume[2,3], Akane Ose[2,3], Miwa Tsutsumi[2,3] ,
Kanzi Miyazaki[2,4], Yuri Murakami[2,4], Natsumi Tanaka [2,4],
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Heusler alloys are typical ordered alloys with the molecular formula of X_2YZ.
Co2NbSn exhibits a structural phase transition (martensite transition) that is closely related to shape memory alloys among Heusler alloys.
During the martensitic transition, various properties such as crystal structure, magnetic properties, and electrical resistivity are changed.
The structural phase transition temperature and Curie temperature of Co2NbSn differ between samples. The sample undergoes a structural phase transition from cubic to orthorhombic at 191K, and from paramagnetic to ferromagnetic at 106K. Since Co2NbSn contains Sn as an impurity phase, we focused on the Co inner shell.
In view of these two points, the present measurements were aimed at clarifying whether the Co2p peak changes its spectrum with temperature by moving back and forth between 240 K and 150 K.
Also, Heusler alloys are generally itinerant electronic states. However, the 3d orbitals of transition metals are also thought to be localized. Therefore, because of the 3d magnetic moment, the 3s orbitals of the inner-shell electrons are observed as a multiplet splitting.
Therefore, the 3s orbitals were measured at 240K and 150K to confirm the multiplet splitting.
The measurements were carried out at BL-19LXU in Spring-8 using hard X-rays with an energy of 7900 eV.
As a result, the temperature dependence at 240 K and 150 K was confirmed.
However, the analysis of the multiplet structure of Co3s and the HAXPES-MCD analysis are still in progress.
As for the temperature dependence, it is thought that the energy degeneracy was solved by changing the structure. This is thought to be due to the band-Jahn-Teller effect.

Trial of hard X-ray photoemission electron microscopy (HAXPEEM) at SPring-8

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Photoemission microscopy with hard X-rays, HAXPEEM, is a promising method for real-space electronic/magnetic states analysis of electronic devices under operation, due to its bulk-sensitivity. We brought a PEEM apparatus (FOCUS PEEM with imaging energy filter) into the hard X-ray beamline BL19LXU at SPring-8 and performed HAXPEEM observations, as a first feasibility test. Figure 1 shows XAS- and XMCD-PEEM images and spectra of a $Gd_{26}Fe_{66}Co_{8}$ thin film, whose surface is irradiated by femtosecond laser pulses prior to the PEEM observations to create random magnetic domains. To examine the bulk sensitivity, half area of the sample surface is capped with 100 nm-thick Au. The local XAS spectrum (Fig. 1 (a)), chemical contrast (Fig. 1 (b)) and XMCD contrast (Fig. 1 (c)) at Gd L_3-edge were clearly seen on the bare GdFeCo area. As for the area capped by the Au, although the XAS spectra (Fig. 1 (a)) and XMCD contrast (Fig. 1 (c)) were under detection limit, faint but noticeable chemical contrast (artificial ditches (dotted circles in Fig. 1 (b))) were found. In the presentation, we will discuss more detailed results as well as future prospective of application to actual device research.

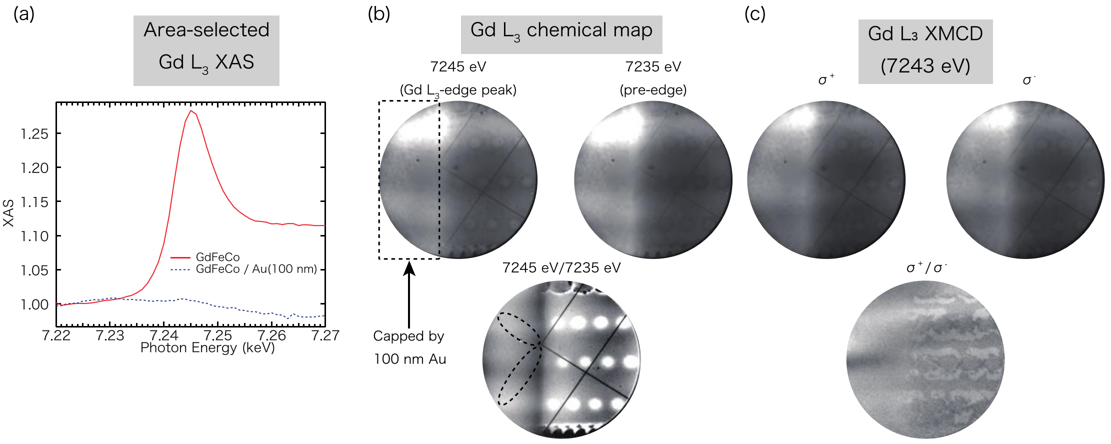

Fig. 1: *HAXPEEM data at Gd L_3-edge of GdFeCo thin film irradiated by femtosecond laser pulses. 100 nm-thick Au capping layer is deposited at the right half region of the field of view. (a) Area-selected XAS spectra (b) Chemical mapping (c) XMCD-PEEM.*

Ground State Symmetry of Rare-earth Ions and Emission Angle Dependence in Resonant X-ray Photoemission

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4f rare-earth compounds exhibit a variety of physical properties and have been widely studied due to their interest in materials science. Core-level X-ray spectroscopy is a powerful technique to investigate the electronic state of such strongly correlated electronic systems.

The purpose of this study is to clarify the relationship between the ground state (GS) symmetry of the rare-earth (RE) ions and the resonant X-ray photoemission (RXPS), by focusing on the dependence on both the incident photon polarization and the emission angle of photoelectrons. This will provide us useful information for GS character of RE ions, including multipole states.

In this study, phase diagrams for RE ions under a weak cubic crystal field are systematically calculated, and the RXPS at the 2p→5d resonant excitation are calculated for each phase of typical RE ions by considering the ionic full-multiplet interactions, especially paying attention on the peak-intensity change around the azimuth angles of the sample surface. The figures show the results for Er^{3+} ion, as an example, where the RXPS peak-intensity patterns for azimuth angle (right) are shown for each GS phase in the phase diagram (left) and for the sample surface normals.

In the presentation, we also report the peak-intensity patterns calculated for resonant X-ray emission spectroscopy (RXES) at the 2p→4f excitation, and the relationship between the GS symmetry and the peak-intensity patterns of RXPS and RXES are discussed.

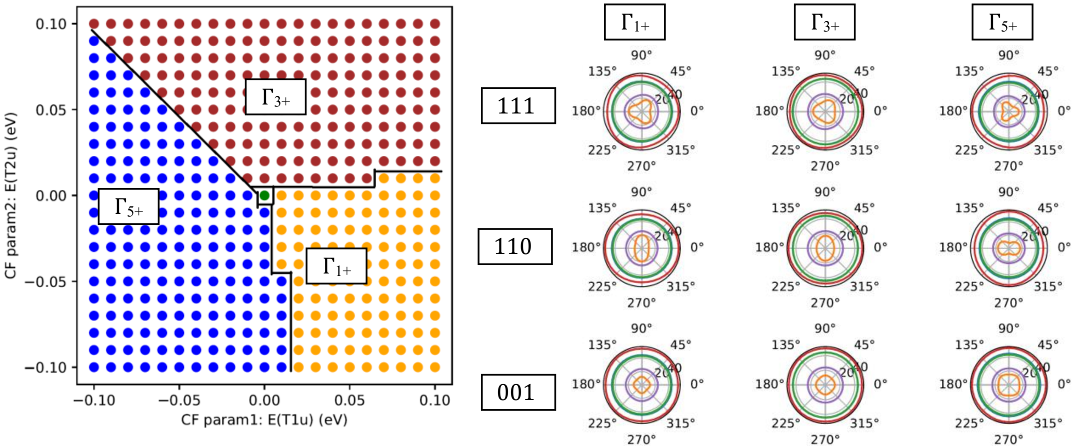

Fig. 1: *Phase diagram for Pr^{3+} under cubic crystal field (left), and the RXPS peak-intensity patterns of Er^{3+} for azimuth angle around the sample surface normals.*

Core-level x-ray photoemission spectroscopy of 3*d* transition metal oxides: DFT+DMFT analysis

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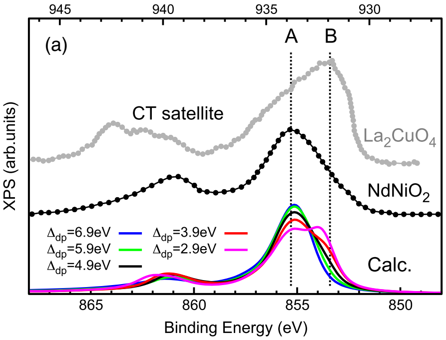
Core-level hard x-ray photoemission spectroscopy (HAXPES) is a powerful probe to study electronic properties of 3*d* transition-metal oxides (TMOs). In TM 2*p* HAXPES, a 2*p* core hole created by x-rays lowers the 3*d* level energies on the excited site via the electron-hole interaction, leading an electron charge transfer (CT) from surrounding ions to the empty *d* states. Besides local screening from nearest-neighboring ligands, nonlocal screening (NLS) from other TM sites affects the core-level line shape [1,2]. The NLS often provides rich information on the electronic properties of 3*d* TMOs, such as metal-insulator transition and long-range magnetic ordering.

We study the NLS effect in a series of 3*d* TMOs using a computational method based on density functional theory (DFT) combined with dynamical mean-field theory (DMFT), which was introduced recently [2,3]. This method describes both realistic valence bands with a strong electronic correlation and the local electron-hole interaction (core-hole potential) in the HAXPES process. Thus, it allows us to model the core-level spectra considering long-distant hybridization effect, beyond a conventional impurity model using e.g., MO_6 cluster model.

We analyze experimental data in early to late TMOs, paying attention to the NLS effect: 1) for $NdNiO_2$, a mother compound of recently-discovered superconducting nickelate, NLS is present, see figure, and can be used to determine the key model parameters [4], 2) for Sr_2FeMoO_6 and $LaFeO_3$, Fe 2*p* NLS features, reported recently [5], are closely related to the pattern of the magnetic ordering, and 3) $SrTiO_3$ and TiO_2, even for trivalent Ti (3d^0), a long-distant electron hopping is needed to reproduce the CT excitations in the HAXPES spectra. This study indicates that the NLS effect is important for the interpretation of core-level HAXPES in a series of 3*d* TMOs.



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Design of an in-situ, in-operando HAXPES cell for the characterization of SEI formation in Li-ion batteries

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Lithium ion batteries (LIBs) have gain tremendous importance recently as a stable energy storage system [1]. During the first cycles, a thin layer is formed on the electrode due to the reduction of the electrolyte components at the anode. This Solid Electrolyte Interface (SEI) leads to an initial capacity loss but also prevents degradation of the battery [2] and help to improve life expectancy and safety [3]. XPS is a very powerful technique for studying the SEI, but its surface sensibility limit the access to the SEI [4]. HAXPES appears as a powerful alternative to XPS to obtain reliable in-operando, in-situ depth profiling: thanks to the high kinetic energy electrons, it is possible to probe up to several tenths of nm into the SEI depth [5].

Here, we will present a new compact cell to perform in-situ operando HAXPES for batteries materials. In standard ex situ XPS, electrodes are removed from the battery and must be rinsed from the excess of electrolyte. Moreover, ex situ characterization may miss some phenomena related to metastable species. The cell design is inspired by developments in electrocatalysis [6]. In this new cell (Fig.1) SEI will be probed through a nanometer-thick membrane, supported on a silicon nitride membrane. The thin membrane separates the liquid electrolyte from the UHV while being partly transparent to the outgoing HAXPES photoelectrons. The cell can fit onto a standard sample plate compatible with usual sample transfer system. The cell is currently under fabrication and will be tested shortly at the GALAXIES beamline at SOLEIL. We believe the new cell will provide new insights into the SEI formation mechanism and batteries interfaces [7] allowing to study a system very close to the real one in-operando.

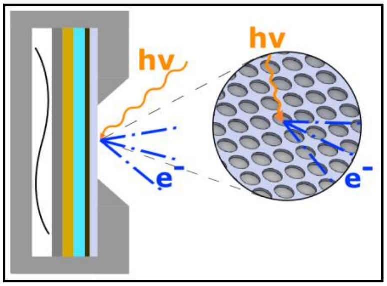

Fig. 1 : schematic design of the in-operando cell [7]

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APXPS study on $Pt_3Pd_3Sn_2/C$ catalyst for dimethyl ether oxidation

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Recently, direct methanol fuel cells, have drawn lots of attention from researchers due to their potential applications with a target to replace lithium batteries. In this work we focused on fuel cells using dimethyl ether (DME) oxidation reaction on platinum and its alloys such as PtPdSn, which shows higher specific power density. DME has several advantages over other fuels, including high energy density, pumpless fuel delivery, liquefied storage, or low toxicity. Because of the possible near-surface effects occurring in the catalysts during activation or operation we investigated the materials using various surface techniques such as ambient pressure X-ray photoelectron spectroscopy (APXPS) or X-ray absorption spectroscopy (XAS) which help us provide valuable information about the role of each element in the reaction mechanism of DME oxidation.

In our experiments $Pt_3Pd_3Sn_2/C$ nanoparticles were synthesized and drop casted on carbon fiber paper and silicon substrates. The oxidation states and binding energies of each element were then studied in various environment such as H_2, DME or DME + H_2O.

First HAXPES Studies of Photovoltaic Materials at EMIL: Depth-Resolved Structure of (Ag,Cu)(In,Ga)Se_2

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The Energy Materials In-situ Lab Berlin (EMIL) is a joint facility of the Helmholtz-Zentrum Berlin (HZB) and the Fritz-Haber Institute of the Max Planck Society (FHI). The HZB operates a large-scale interconnected UHV system dedicated to the study of energy materials and devices. This system connects deposition and treatment chambers to advanced analytics, which include two lab-based photoemission systems in addition to a dual-color, wide-energy-range (80 eV-10 keV) beamline and an X-ray spectroscopy endstation, providing for multiple measurement techniques, serving as a materials science “tool chest.”

This contribution will illustrate the capabilities of the system by focusing on an depth-dependent photoemission study of (Ag,Cu)(In,Ga)Se_2 thin-film solar cell absorbers. Such chalcopyrite materials – traditionally Cu(In,Ga)$(S,Se)_2$ – have a bandgap which can be tuned by varying the stoichiometry of the film. Alloying Cu(In,Ga)Se_2 with Ag to produce (Ag,Cu)(In,Ga)Se_2 can additionally reduce structural defects and improve opto-electronic properties, as well as provide a further means of tuning the film’s properties. This flexibility makes them extremely useful as light-absorbing layers for solar cells, as it allows controlled variation of electronic properties throughout the thickness of the thin (on the order of 1-2 μm) layer, optimizing charge carrier creation and extraction. While the composition variation on this depth scale can be coarsely determined by destructive methods (e.g., secondary ion mass spectrometry), the near-surface (i.e., uppermost 10s of nm) region of the absorbers also shows considerable changes in composition and properties. The nature of this region can strongly influence the operating parameters of the resulting solar cell, because it ultimately forms the interface with the transparent front contact (generally via an additional buffer layer) and thus can impede charge carrier extraction if not designed carefully. In this contribution, we use the wide range of excitation energies at EMIL to probe this critical near-surface region of the (Ag,Cu)(In,Ga)Se_2 absorber to understand the relationship between Ag-alloying, Cu-deficiency, and electronic structure tuning of the film, with the goal of establishing a structure-function relationship within the system.

HZB Energy Materials Research at SPring-8 BL15XU 2018-2019

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For several years, the Department of Interface Design at Helmholtz-Zentrum Berlin für Materialien und Energie GmbH performed beamtime at the NIMS contract beamline BL15XU [1] at SPring-8, obtained through partnerships and through the "Nanotechnology Platform: Microstructure Characterization Platform" (Nano-PF) [2], to study a number of energy-related materials systems.

These studies of materials from photovoltaics, batteries, fuel cells, etc. have played crucial roles in the research efforts of the group and have contributed to numerous PhD theses, conference presentations, and joint publications. In light of the recent decommissioning of this beamline, we will present an overview of this energy-related materials research, highlighting some of the most important findings, with a focus on the study of buried interfaces as well as on the electronic/chemical structure of thin-film photovoltaic materials.

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X-ray spectroscopic study of the electronic states of Pd nanoparticles

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Palladium nanoparticles (Pd NPs) are attractive material in many scientific and industrial fields. The remarkable point is the difference from the bulk one such as high catalytic activity and anomalous hydrogen storage property [1]. It seems that these properties originate from the electronic states around Fermi level, but specifics still unknown. In this study, we have investigated the electronic state of Pd NPs by soft and hard X-ray spectroscopies. Soft and hard X-ray photoelectron spectroscopy (SXPS and HAXPES) reveals the electronic state of Pd NPs at lateral surface and inside of the NPs, respectively. Moreover, near edge X-ray absorption fine structure (NEXAFS) of Pd NPs has been observed to clarify the unoccupied electronic states.

Pd NPs were fabricated by the gas evaporation method using inert He gas [2]. The Pd NPs possess the clean surfaces because any surfactants or polymers have not been used during the fabrication process. SXPS and HAXPES have been done at BL23SU and BL47XU in SPring-8, respectively. Pd L_3-edge NEXAFS was measured at BL6N1 in Aichi SR. All samples were fabricated and transferred without exposure to the air to avoid the surface oxidation.

Fig. 1 shows valence band (VB) spectra of Pd NPs and Pd bulk obtained by HAXPES. DOS calculated based on DFT are also shown in Fig. 1. The DOS of Pd bulk represent sufficiently the feature of VB spectrum of Pd bulk. The shape of the VB of Pd NPs is different from that of Pd bulk. This difference originates from the huge specific surface area of NPs. The features of Pd NPs are similar to DOS of Pd(111) lateral surface.

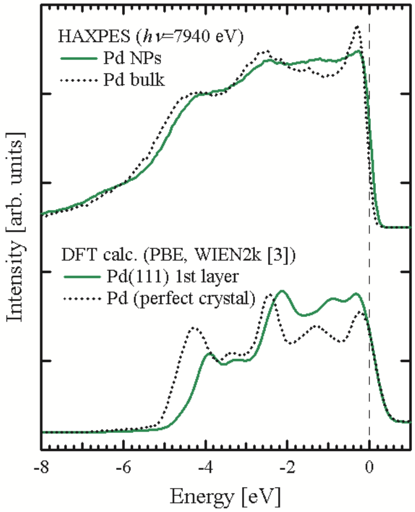

Fig. 1: *Valence band spectra of Pd NPs and Pd bulk obtained by HAXPES. Back ground has been removed by iteration method [4]. DOS of Pd(111) and Pd crystal are also shown.*

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Spectroscopic study and photocatalytic activity of Au-Ni nanoparticles for hydrogen production reaction by water splitting

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Au-Ni nanoparticles are remarkable co-catalysts for photocatalytic reactions of hydrogen production by water splitting [1]. However, it is difficult to prepare Au-Ni well-mixed nanocomposites via wet chemical method because of their immiscibility. In this research, Au-Ni nanoparticles were prepared by solution plasma process which is suitable for rapid co-evaporation and aggregation (1 μs order) of Ni and Au atoms. The composition of Au and Ni were determined by the combination of Energy Dispersive X-ray Spectroscopy and Scanning Electron Spectroscopy. Soft X-ray photoelectron spectroscopy (SXPS) was performed for surface analysis of Au-Ni nanoparticles and hard X-ray photoelectron spectroscopy (HAXPES) in SPring-8 BL09XU was performed for analysis of inside of Au-Ni nanoparticles.

Fig. 1A shows Au 4f SXPS spectra of Au-Ni nanoparticles. As Ni composition increased, the peak component appears on the lower binding energy side compared with metallic component at 84 eV. Moreover, from Ni 2p SXPS spectra of Au-Ni nanoparticles (not shown here), it is found that $Ni(OH)_2$ state is dominant on the surface of Au-Ni nanoparticles. Fig. 1B shows Au 4f HAXPES spectra of Au-Ni nanoparticles. These results indicate that metallic Au exists inside of Au-Ni nanoparticles. Fig. 1C shows Ni 2p HAXPES spectra of Au-Ni nanoparticles. NiO and $Ni(OH)_2$ components increased as Ni composition increased. However, those components decrease after $Au_{0.26}Ni_{0.74}$. It is also found that, Ni^0 state exists in all samples. From above results, it is suggested that the Au-Ni nanoparticles have the core-shell structures as shown in Fig. 1D (Au rich condition) and Fig. 1E (Ni rich condition).

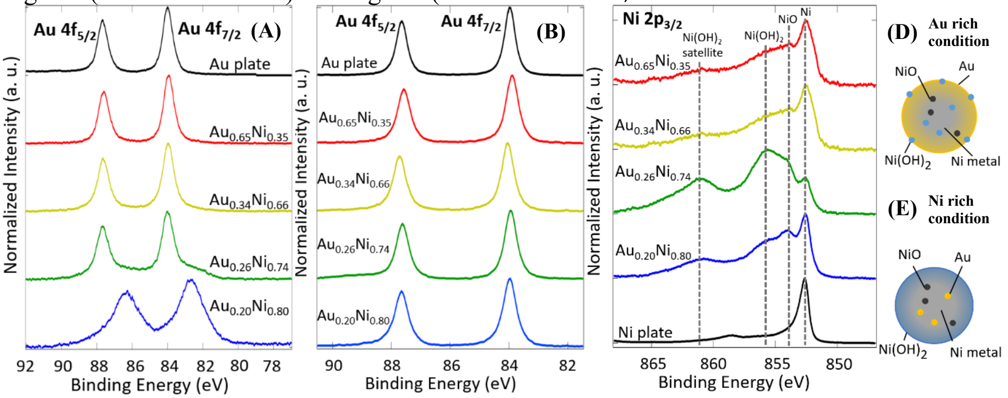

Fig. 1. SXPS spectra of Au 4f (A) and HAXPES spectra of Au 4f (B) and Ni 2p (C).The suggested structure of the nanoparticles is shown as (D), (E).

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The "Two-Color" EMIL Beamline at the BESSY II Light Source

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The Energy Material In-situ Laboratory Berlin (EMIL), a joint project of the HZB, the Fritz-Haber Institute of the Max-Planck Society and the MPI for Chemical Energy Conversion, is a unique infrastructure that focuses on the study of energy conversion, efficiency, and storage materials by combining industry-scale sample production facilities and various spectroscopic and microscopic techniques that fully exploit the 80 eV to 10 keV "two-color" beamline. EMIL is a state-of-the-art surface and interface analysis laboratory that provides access to a large variety of synchrotron methods PES, HAXPES, XAS, XES, RIXS, XRF, and STXM with the photoemission and photon-in-photon-out techniques being operable under near-ambient or true-atmospheric pressure conditions, respectively. The wide range of photon energies provides access to surface and (buried) interface chemical and electronic structures with information depths from a few Å to several µm.

The "two-color" EMIL beamline at BESSY II combines two undulator branchlines delivering soft and hard X-rays. The soft X-ray UE48 and the hard X-ray in-vacuum cryo-undulator U17 are providing high brilliance across a continuous energy range between 80 eV and 10 keV. The branchlines are designed to focus the radiation at the same spatial point in any of the three end-stations SISSY-I, SISSY-II, and CAT. Two other foci, one soft and one hard, are available to STXM and PINK end-stations, respectively. The branchlines can be operated alternatively providing X-rays to either the same analytics system or simultaneously to different analysis chambers. The EMIL beamline system contains 26 optical elements and three monochromators (two PGM and one DCM) all installed in an area of 2 meters width and 65 meters length.

With both, the UE48 soft X-ray and U17 hard X-ray branches now in full user operation, in this contribution, we will give a detailed update of the status and performance of the EMIL beamline(s). We will report on energy resolution, photon flux, and focus size as key performance indicators.

Effects of Sample-Aperture Cone Distance on Environmental Charge Compensation in Hard X-ray Photoelectron Spectroscopy

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One major problem of photoelectron spectroscopy is the charging effect in measuring bulk insulator samples. It is known that the charging effect is reduced to some extent by introducing a gas into an analysis chamber up to ~500 Pa (environmental charge compensation) [1]. Here, we demonstrate almost complete charge compensation by gas introduction into a near-ambient pressure hard X-ray photoelectron spectroscopy (NAP-HAXPES) apparatus at BL24XU, SPring-8, equipped with a HiPP-2 electron energy analyzer (Scienta Omicron). The charging of a glass slide (thickness: 1 mm) was successfully compensated by introducing nitrogen gas up to a pressure of 2500 Pa at the sample-aperture cone distance d (Fig. 2, inset) of 0.3 mm (our normal geometry), as shown in Fig. 1. More interestingly, the pressure required for charge compensation was found to strongly depend on d, as shown in Fig. 2. At d=2.1 mm, the charging was compensated at only 250 Pa. Possible reasons for the strong d dependence will be discussed.

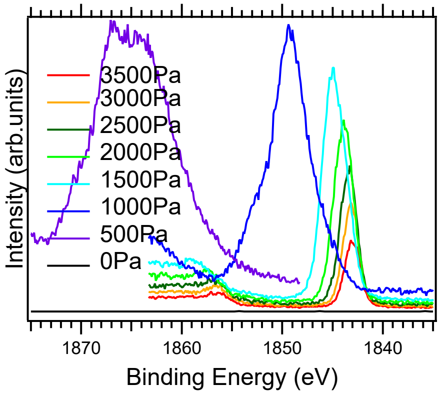

Fig. 1: *Si 1s spectra of a glass plate under various nitrogen pressures obtained at d=0.3 mm.*

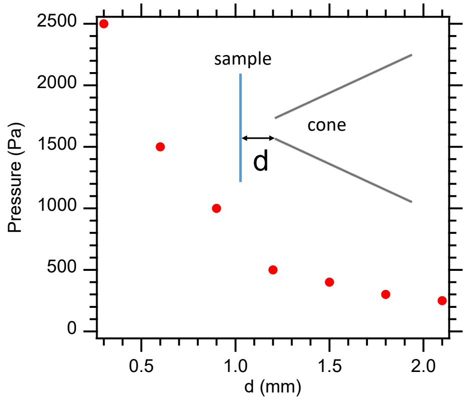

Fig. 2: *Relationship between d and the pressure required for charge compensation. The inset shows the definition of d.*

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Hard X-ray Photoelectron Spectroscopy Beamline Designed at Shenzhen Innovation Light-source Facility (SILF)

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Shenzhen Innovation Light-source Facility (SILF) is a newly proposed fourth-generation synchrotron light source located in Shenzhen, China. It is currently in the planning stage and will be constructed by the Institute of Advanced Science Facilities, Shenzhen (IASF). A 3 GeV diffraction-limited storage ring with a circumference of 696 m has been designed for a low emittance of <100 pm·rad and a beam current of 200-300 mA. Thus, a brilliant light source with a brightness in the order of 10^{21} phs/sec/mm^2/$mrad^2$/0.1BW will be built. This new synchrotron light source will provide an innovative research platform for scientists and industries in China's Greater Bay Area and even from around the world.

We will introduce the current progress of our light source facility, as well as the design of a high-flux undulator beamline for HAXPES. This beamline is equipped with a double-crystal monochromator, a high harmonics suppression mirror and Kirkpatrick–Baez mirror pairs, providing 3.4-18 keV hard X-ray with a focused spot size of 15μm^2. It will be dedicated to high energy resolution fluorescence detected X-ray absorption spectroscopy (HERFD-XAS), X-ray Emission Spectroscopy (XES), resonant inelastic X-ray scattering (RIXS) and hard X-ray photoelectron spectroscopy (HAXPES) measurements for advanced materials research.

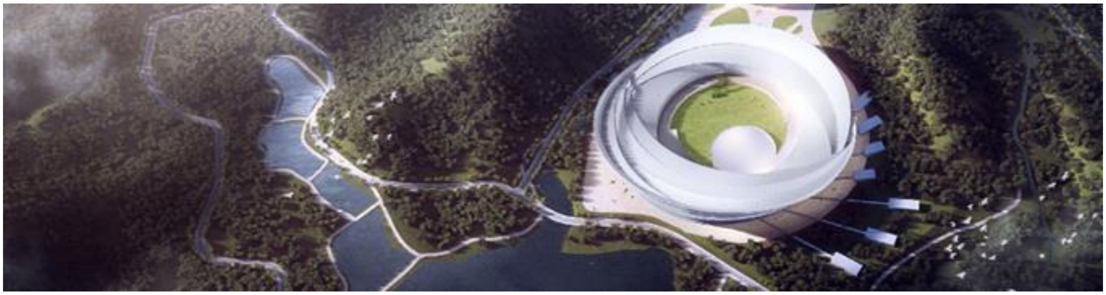

Fig. 1: *An architect's illustration of the Shenzhen Innovation Light-source Facility.*

Three-Dimensional Spatial-Resolved HAXPES Measurement using a 1 µm Focused Beam

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BL09XU at SPring-8 has been upgraded as a beamline dedicated for hard X-ray photoelectron spectroscopy (HAXPES) experiments in 2021. The HAXPES system with a wide-acceptance-angle (±32°) analyzer [1], which had been installed at BL47XU, was moved to the experimental hutch (EH) 2. One of the key issues in the upgrade was the introduction of the Si 311/Si 220 double channel-cut monochromator, which provides a more intense beam than that of BL47XU with a channel-cut monochromator with Si 444 reflection. Furthermore, a long Kirkpatrick-Baez (KB) mirror at EH 2 significantly reduced the beam loss compared to that with a short KB mirror at BL47XU. These improvements enabled us to increase the intensity of the incident beam, and to reduce the beam size by limiting a front-end slit aperture. The flux with a focused beam size of 1.5 (V) × 1 (H) μm^2 at BL09XU is 1.5 times larger than that with a size of 1(V) × 4 (H) μm^2 at BL47XU. By combining the 1 µm focused beam and an angle-resolved measurement with a wide-acceptance-angle analyzer, we were able to investigate the depth-dependent chemical states in a local region. We performed selective depth-resolved HAXPES measurements on a sample of Au patches of various sizes on a SiO_2 substrate.

Figure 1 shows two-dimensional photoelectron intensity distributions for Au 4f and Si 1s photoelectron, which were obtained by scanning the sample. We found that the distribution of Au and Si was clearly resolved in the two-dimensional maps. In addition, we performed HAXPES analysis to a local area by irradiating the beam only onto a 10 µm square SiO_2 region, and confirmed complete suppression of the Au signal from this region. By depth-resolved HAXPES measurement, the thickness of the SiO_2 film in the region was estimated to be about 1 nm

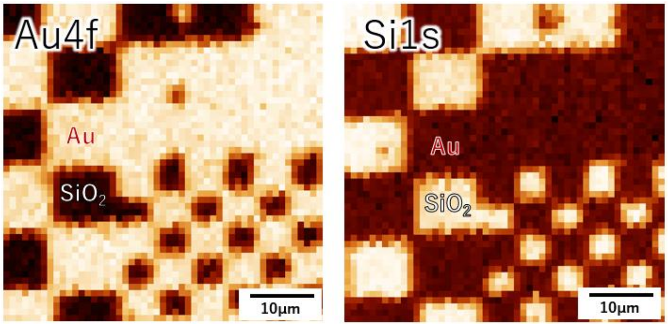

Fig. 1: *Two-dimensional photoelectron intensity distribution images of Au patches on SiO_2 acquired with Au 4f (left) and Si 1s (right). 50 × 50 μm^2*

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The Operande Measurement for Bias Applied Technique Using Laboratory Based Hard X-ray Photoelectron Spectroscopy with GaKα X-ray Source

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The performance of LSI devices is improving rapidly, and the progress is being made every day toward the realization of the IoT society. In particular, the interface is the key to the performance of the LSI devices [1, 2]. To evaluate the interfacial state, capacitance-voltage (C-V) measurements are used electrically, photoelectron spectroscopy (XPS, HAXPES) using soft and hard X-rays is used for chemical bonding states, and transmission electron microscopy (TEM) is used to evaluate the physical structure, respectively. In addition to the above evaluation techniques, knowledge of the carrier behavior and the electronic states at the interface during device operation is key to further improving performance. Therefore, HAXPES with applied bias has been attracting attention as a method to evaluate the interface state during bias operation. In this paper, we report the results of bias applied HAXPES measurements using the HAXPES_Lab. [3].

In this experiment, we deposited TiN and ZrO_2 films on Si substrates at 20 and 4 nm, respectively. The applied voltages on the sample back surface were in the range of ± 2 V, while the sample front surface was grounded. We measured Si 1s, Ti 1s, Ti 2p, O 1s and Zr 3p core level photoelectron spectra, respectively.

Figure 1 shows the dependence of the binding energy values of the Si-Si and Ti-N chemical bonds on the applied voltage, calculated from the Si 1s and Ti 2p spectra. The Si-Si bond originating from the Si substrate was found to vary linearly with the applied voltage, with a slope of 0.46 calculated by linear approximation. This indicates that the voltage drop occurs not only at the TiN/Si interface but also inside the TiN and ZrO_2 films. The Ti-N chemical bond showed an arc-shaped behavior against applied bias. This result suggests the possibility of carrier trapping at the TiN/Si interface and the TiN film defects. HAXPES_Lab. was successfully used for the measurement of the operand measure with applied voltage, and the behavior of the near the interface and in the film was successfully clarified.

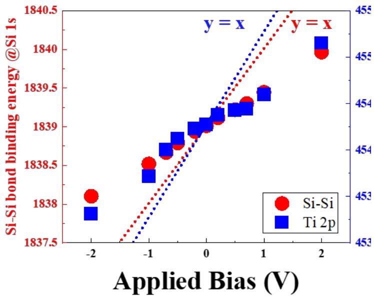

Fig. 1 Si-Si and Ti-N binding energy dependence on the applied bias. Dashed line shows the binding energy shift equal to the applied bias.

This work was partially supported by NEDO.

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Study on the Relationship between Chemical Structure and Function of Amphiphilic Gel Surface

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More than 60 years have passed since gel materials were first utilized in soft contact lens materials. The current mainstream material is Silicone Hydrogel (SiHy)[1].

SiHy, which uses silicone compounds, has higher oxygen permeability and poorer biocompatibility than general gel material. Therefore, various surface hydrophilization approaches have been tried on SiHy[2], but the mechanism of functional change has not been fully explained. In this study, we attempted to explain the surface chemical structure and functional change of SiHy by analyzing the difference between SiHy before and after plasma treatment utilizing hard X-ray photoelectron spectroscopy (HAXPES) and neutron reflectometry (NR).

From the NR measurements, the changes in the scattering length density (SLD) profile before and after the surface treatment and with the change in humidity environment are shown in Fig 1. The surface treatment decreased the film thickness by about 20 nm and increased the SLD by about 15 nm near the surface in the dry sample. These results suggested the etching of the elements from the surface, especially removing the atoms having smaller values of SLD (In this case, removal of the CH_3 groups plausibly happened). Furthermore, the SLD near the surface increased with raising humidity (D_2O), indicating the process of D_2O absorption by the SiHy.

In addition, the chemical shift of Si_{1s} after hydrophilic treatment was observed from the measurement of Si_{1s} by HAXPES shown in Fig. 2. The chemical shift of Si_{1s} (oxidation) strongly occurred in the surface layer of 20 nm, which corresponded to the depth where the change in the SLD was especially pronounced in the NR measurement.

This study can explain the relationship between the surface chemical structure and the water absorption of SiHy due to the surface treatment by utilizing HAXPES and NR. The complementary use of HAXPES and NR techniques is valuable and powerful to elucidate the microscopic changes (chemical and functional) in the SiHy surface.

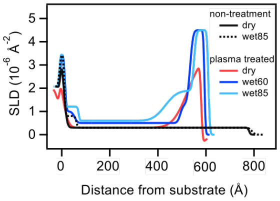

Figure1 The behaviour of SLD before and after surface treatment for SiHy and the behaviour of SLD with change of water absorption state due to humidity environment by neutron reflectometry

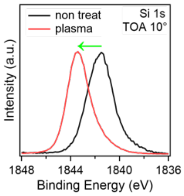

Fig.1 The chemical shift of Si $_{1s}$ spectra on SiHy before and after surface treatment observed by HAXPES

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