ICESS-16

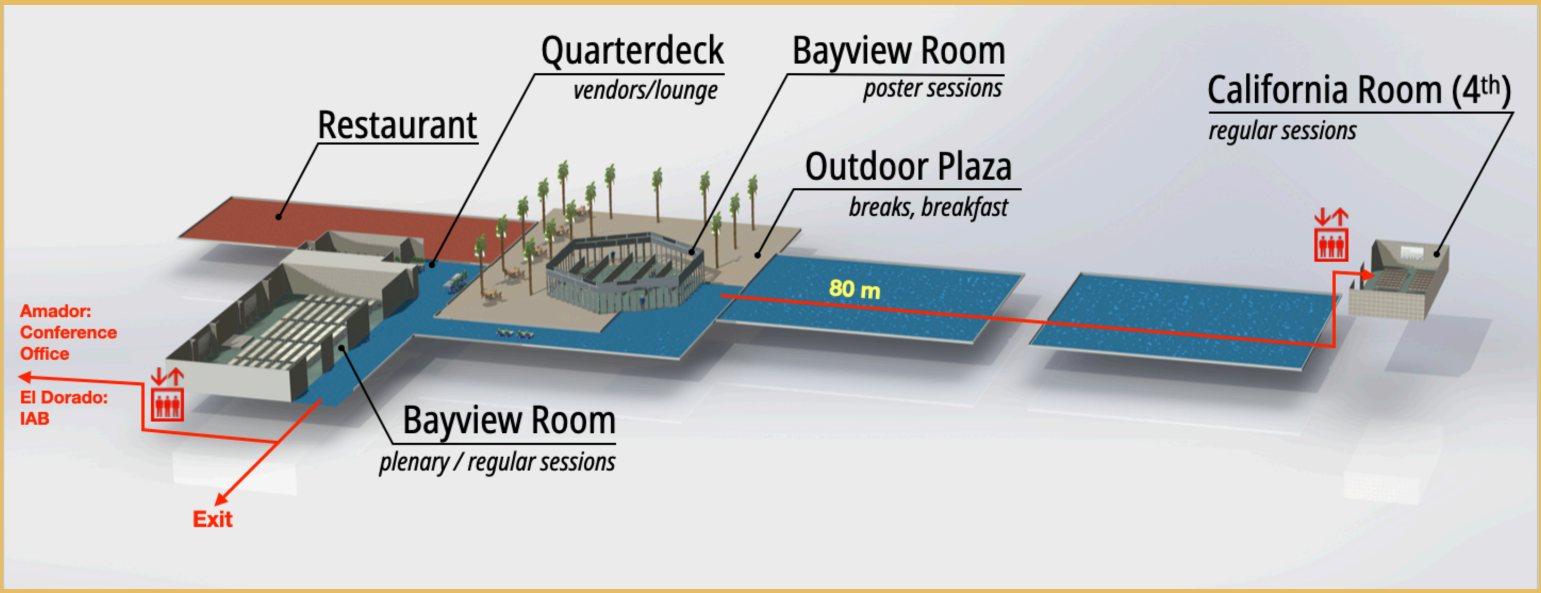
16th International Conference on Electronic Spectroscopy and Structure

Berkeley, California, USA

July 20th- July 25th 2025

Conference Program

Getting around the Conference



Within Walking Distance

Skates on the Bay (Seafood)

Hana Japanese (Steak & Seafood)

ICESS-16

16th International Conference on Electronic Spectroscopy and Structure

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Sunday 7/20

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09:15 AM

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University of Chicago
Recent progress in predicting the optical properties of defective and disordered systems

09:15 AM-
10:00 AM

MoI_02: Winter, B.
Fritz-Haber-Institut
Liquid-jet photoelectron spectroscopy of aqueous solutions: Electron scattering, electronic structure and work functions

10:00 AM-
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University of California, Davis
Photoemission spectromicroscopy: correlating local chemistry and electronic structure in quantum materials

11:00 AM-
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MoI_05: Abbamonte, P.
University of Illinois - Urbana-Champaign
Conformally invariant charge fluctuations in a strange metal

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MoI_06: Denlinger, J. D.
Lawrence Berkeley National Lab
Temperature dependent crossover behaviors in photoemission of strongly correlated f- and d-electron systems

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MoI_07: Regmi, S.
Idaho National Laboratory
Electronic structure study of a uranium-based material using angle-resolved photoemission spectroscopy

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MoII_04: Gilbert, P. U.
UW-Madison
Evolution of enamel nanocrystal misorientation before and after diet changes

11:00 AM-
11:30 AM

MoII_05: Liu, Z.
title not available

11:30 AM-
11:50 AM

MoII_06: Tomanik, L.
University of Chemistry and Technology
Chemical shifts in photoelectron spectroscopy of liquids

11:50 AM-
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MoII_07: Carniato, S.
sorbonne université, Paris
Fingerprint of the dipole orientation of water molecules in Cu2+ aqueous solution probed by x-ray photoelectron spectroscopy

12:10 PM-
02:00 PM

MoII_08
Lunch

	Session SMoI_02: Correlated Materials I		Session SMoII_02: Interfacial Energy and Charge Transfer
02:00 PM- 02:30 PM	**MoI_09: Zhou, K.** title not available	02:00 PM- 02:30 PM	**MoII_09: Roth, F.** *TU Bergakademie Freiberg* Capturing Electrons in Motion: Time-Resolved X-ray Photoemission Spectroscopy for Unraveling Charge Transfer Dynamics
02:30 PM- 02:50 PM	**MoI_10: Fan, S.** *Brookhaven National Laboratory* Polaronic state across the photoinduced transition in a magnetoresistive manganite	02:30 PM- 02:50 PM	**MoII_10: Vannucchi, N.** *Uppsala University / Sorbonne University* The impact of the oxide transport layer on the energy band alignment of the frontier orbitals in non-fullerene acceptor organic solar cells
02:50 PM- 03:10 PM	**MoI_11: Horio, M.** *The University of Tokyo* Enhanced superconducting gap in the Hg-based trilayer cuprate superconductor revealed by micro-spot ARPES	02:50 PM- 03:10 PM	**MoII_11: Lindblad, A.** *Uppsala University* Hard X-ray Core-hole Clock Spectroscopy of Adsorbates and Polymer Mixtures
03:10 PM- 03:30 PM	**MoI_12: Xu, K.** *University of California, Berkeley* Anomalous normal state gap in an underdoped n-type cuprate	03:10 PM- 03:30 PM	**MoII_12: Bandaranayake, S.** *Sandia National Laboratories* Unraveling charge and energy transfer dynamics between plasmonic matels and adsorbates

03:30 PM- 04:00 PM	**MoII_13** *Break*

	Session SMoI_03: Correlated Materials II		Session SMoII_03: Interface Dynamics and Chemistry
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04:30 PM- 05:00 PM	**MoI_15: Miao, H.** title not available	04:30 PM- 05:00 PM	**MoII_15: Dupuy, R.** title not available

Session SMoI_03: Correlated Materials II

05:00 PM-05:20 PM
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National Taiwan University
The Growing Charge-Density-Wave Order in CuTe Lightens and Speeds up Electrons

05:20 PM-05:40 PM
MoI_17: Ideura, K.
Hiroshima University
Angle-resolved photoemission spectroscopy of Dirac nodal-line superconductor ZrSi$_{1-x}$P$_{x}$Se

05:40 PM-06:00 PM
MoI_18: Blyth, J.
Monash University
Three-dimensional flat band in ultra-thin Kagome metal, Mn3Sn

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MoII_16: Perco, D.
University of Trieste
Limitations in Determining Oxidation States in Condensed Matter at the Sub-Nanometric Scale

05:20 PM-05:40 PM
MoII_17: Mora Perez, C.
Lawrence Berkeley National Laboratory
Tuning Au–TiO2 Photocatalytic Interfaces: Impact of Oxygen Vacancies and Gold Nanoparticle Size on Electronic Structure and Carrier Dynamics

05:40 PM-06:00 PM
MoII_18: Ehm, A.
Chemnitz University of Technology
Electronic Properties of Discrete NDI-T2 Oligomer Thin Films Using Photoelectron Spectroscopy and Low-energy Inverse Photoemission Spectroscopy

Tuesday 7/22

Session STu_01: Plenary Talks

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TuI_01: Rolles, D.
Kansas State University
Gas-phase photochemistry studied by time-resolved photoelectron spectroscopy with free-electron lasers

09:15 AM-
10:00 AM
TuI_02: Maiti, K.
title not available

10:00 AM-
10:30 AM
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Session STuI_01: Novel Electronic Phenomena

10:30 AM-
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TuI_04: Bansil, A.
Northeastern University
Recent surprises in quantum materials among many legacies of Einstein

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TuI_05: Zonno, M.
title not available

11:30 AM-
11:50 AM
TuI_06: Akiyama, R.
Institute of Science Tokyo
Interface-induced Weak Ferromagnetism in Altermagnetic MnTe Thin Films

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Lawrence Berkeley National Lab
Nonrelativistic spin-splitting above and below the Fermi level in a g-wave altermagnet

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10:30 AM-
11:00 AM
TuII_04: Feifel, R.
University of Gothenburg
Symmetry breaking in core-valence double ionization of allene

11:00 AM-
11:30 AM
TuII_05: Bergmann, U.
University of Milwaukee-Madison
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11:30 AM-
11:50 AM
TuII_06: Doumy, G.
Argonne National Laboratory
Nonlinear resonant core-excitations in molecules

11:50 AM-
12:10 PM
TuII_07: Nam, Y.
Argonne National Laboratory
Probing Molecular Chirality with Nonlinear X-ray Spectroscopies

12:10 PM-
02:00 PM
TuI_08
Lunch

	Session STuI_02: Spin and Orbital Textures		Session STuII_02: Spin and Magnetism
02:00 PM- 02:30 PM	**TuI_09: Plucinski, L.** *Forschungszentrum Jülich* Exploring Approaches to Probe Full Electronic Wave Functions in Solids via Photoemission	02:00 PM- 02:30 PM	**TuII_09: Moser, S.** *Julius-Maximilians-Universität Würzburg* The Dirac Semimetal RuO2 - From Topology to Functionality
02:30 PM- 02:50 PM	**TuI_10: Suen, C.** *Max Planck Institute for Solid State Research, University of British Columbia (Quantum Matter Institute)* Observing the electronic response of a Mott insulator at a current-induced insulator-to-metal transition with transport-ARPES	02:30 PM- 02:50 PM	**TuII_10: Shen, L.** *SLAC National Accelerator Laboratory* Dispersion of Spin Stripe Fluctuations in FeGd Measured by Megahertz X-ray Photon Correlation Spectroscopy
02:50 PM- 03:10 PM	**TuI_11: Donath, M.** *University of Muenster* The puzzling case of Pt(111): Spin texture of the prototypical L-gap surface state	02:50 PM- 03:10 PM	**TuII_11: Klewe, C.** *Lawrence Berkeley National Laboratory* Depth-Resolved Profile of the Interfacial Ferromagnetism in CaMnO3/CaRuO3 Superlattices
03:10 PM- 03:30 PM	**TuI_12: Schusser, J.** *University of West Bohemia in Pilsen* Assessing nontrivial topology in Weyl semimetals by dichroic photoemission	03:10 PM- 03:30 PM	**TuII_12: Pathiraja, C. S.** *University of Houston* Spin-flip Determination in CrX3 (X = Cl, Br, and I) 2D Magnets using High-resolution X-ray Scattering
03:30 PM- 04:00 PM	**TuII_13** *Break*		

	Session STuI_03: Inelastic/Elastic Scat-tering Spectroscopy		Session STuII_03: 2D Magnetism
04:00 PM- 04:20 PM	**TuI_14: Rosa, F.** *Politecnico di Milano* Tridimensional magnetism in Superconducting Infinite-Layer PrNiO2 studied with Resonant Inelastic X-ray Scattering	04:00 PM- 04:20 PM	**TuII_14: Mcchesney, J. L.** *Argonne National Laboratory* Proximity-Induced Long-Range Magnetism in Ultra-High Conductivity PdCoO2
04:20 PM- 04:40 PM	**TuI_15: Wansorra, C.** *Karlsruhe Institute of Technology (KIT)* Electronic Structure of LaxSr1−xFeO3−δ Probed by X-ray Absorption and Resonant Inelastic Soft X-ray Scattering	04:20 PM- 04:40 PM	**TuII_15: He, W.** *SLAC National Accelerator Laboratory* Dispersive Hund’s Excitons in Magnetic van der Waals Materials

Session STuI_03: Inelastic/Elastic Scat-tering Spectroscopy		Session STuII_03: 2D Magnetism	
04:40 PM- 05:00 PM	**TuI_16: Lee, G.** *Lawrence Berkeley National Laboratory* A Precise Understanding of the Cathode Redox Mechanism in Commercial Lithium-Ion Batteries Through RIXS/sXAS Analysis	04:40 PM- 05:00 PM	**TuII_16: Guo, Y.** *Rice University* Van Hove Singularities Induced Large Electron-boson Coupling in Cr2Ge2Te6

05:00 PM-
06:30 PM

TuI_17: Poster Session I with Casual Dinner

Wednesday 7/23

Session SWe_01: Plenary Talks

08:30 AM- 09:15 AM	**WeI_01: Matsuda, I.** *Institute for Solid State Physics* The rise of nonlinear X-ray spectroscopy
09:15 AM- 10:00 AM	**WeI_02: Wörner, H.** title not available
10:00 AM- 10:30 AM	**WeII_03** *Break*

Session SWeI_01: Surfaces and Interfaces		Session SWeII_01: Attosecond and Femtosecond Molecular Dynamics	
10:30 AM- 11:00 AM	**WeI_04: Regoutz, A.** title not available	10:30 AM- 11:00 AM	**WeII_04: Cryan, J.** title not available
11:00 AM- 11:30 AM	**WeI_05: Kim, C.** title not available	11:00 AM- 11:30 AM	**WeII_05: Ramasesha, K.** *Sandia National Laboratories* Discovery of New Excited State Pathways in Transition Metal Carbonyls using Ultrafast Core-Level Spectroscopy
11:30 AM- 11:50 AM	**WeI_06: Montero, I.** *CSIC. Instituto de Ciencia de Materiales de Madrid* Low Secondary Electron Emission 2D Materials for Space Electronics Applications	11:30 AM- 11:50 AM	**WeII_06: Chuan, C.** *Stanford University* Probing H_2^+ Formation in Strong-Field Ionization of Water Using Pump-Probe Spectroscopy and Covariance Mapping
11:50 AM- 12:10 PM	**WeI_07: Ha, Y.** *Lawrence Berkeley Lab* Soft X-ray Spectroscopy as a Neat Tool to Assign Electronic Structures of {FeNO} Complexes	11:50 AM- 12:10 PM	**WeII_07: Kumar, S.** *Lawrence Berkeley National Laboratory* Intramolecular particle exchange in the fragmentation of methanol upon valence photo double ionization

12:10 PM- 02:00 PM	**WeII_08** *Lunch*

	Session SWeI_02: Surfaces and Interfaces		Session SWeII_02: Fundamental Electronic Structures and Ionization Dynamics
02:00 PM- 02:30 PM	**WeI_09: Toyoshima, R.** *The University of Tokyo* Operando Spectroscopic View for Thin Film Gas Sensors	02:00 PM- 02:30 PM	**WeII_09: Bharti, D.** *Max Plank Institut für Kernphysik, Heidelberg* Interferometric Probing of Photoionization Dynamics and Resonant Control of Photoemission Delays
02:30 PM- 02:50 PM	**WeI_10: Omondi, C. A.** *Masinde Muliro University of Science and Technology* Impact of 3D/2D Composition Ratios on the Crystallinity, Optical Properties, and Stability of Hybrid Perovskite Films via GIWAXS and Spectroscopic Measurements.	02:30 PM- 02:50 PM	**WeII_10: Schweikhard, L.** *University of Greifswald* Highly-sensitive negative ion spectroscopy in an electrostatic ion beam trap
02:50 PM- 03:10 PM	**WeI_11: Shatsala, M. E.** *Lawrence Berkeley National Laboratory* Real-Time Insights into Perovskite Evolution: In Situ GIWAXS and Optoelectronic Studies of PEACl-PPF Co-Passivated Cs0.1FA0.9PbI3 Thin Film Growth	02:50 PM- 03:10 PM	**WeII_11: Bowlan, P.** *Los Alamos National Lab* Molecular fingerprinting of f-electron molecules using a soft x-ray, HHG light Source
03:10 PM- 03:30 PM	**WeI_12: Takahiro, I.** *NUSR/Nagoya University* Electronic Structure of Solid Electrolyte Li6.5La3Zr1.5Ta0.5O12 Bulk Single Crystal Studied by Angle-resolved Photoemission Spectroscopy	03:10 PM- 03:30 PM	**WeII_12: Duraisamy Suganthi, M.** *Rice University* Characterization of 4f^13 (2F(5/2)) 5d6s(1D) 1[5/2](5/2) state for Metastable Qubit Operations in Yb+ ions
03:30 PM- 04:00 PM	**WeI_13** *Break*		

	Session SWeI_03: Surfaces and Interfaces		Session SWeII_03: Fundamental Dynamics of Photocatalysis
04:00 PM- 04:30 PM	**WeI_14: Foehlisch, A.** *Helmholtz Zentrum Berlin* Activation Mechanisms of layered MoS2 Surfaces probed by time-based and time-resolved Electron Spectroscopy	04:00 PM- 04:30 PM	**WeII_14: Techert, S.** *Deutsches Elektronen-Synchrotron DESY* Temporal Electronic and Structural Landscapes in Catalytic Water-Splitting
04:30 PM- 05:00 PM	WeI_15	04:30 PM- 05:00 PM	**WeII_15: Ingle, R.** title not available

	Session SWeI_03: Surfaces and Interfaces		Session SWeII_03: Fundamental Dynamics of Photocatalysis
05:00 PM- 05:20 PM	**WeI_16: Im, H.** *Pusan National University* Interfacial interaction between TaSe2 and bilayer graphene	05:00 PM- 05:20 PM	**WeII_16: Donnellan, Z.** *Lawrence Berkeley National Lab & UC Berkeley* Ultrafast Charge Transfer Dynamics in a Plasmonic Photocatalyst Studied by Femtosecond Time-Resolved X-ray Photoelectron Spectroscopy
05:20 PM- 05:40 PM	**WeI_17: Taylor, N. K.** *Harvard John A. Paulson School of Engineering and Applied Sciences (SEAS) and Lawrence Berkeley National Laboratory Advanced Light Source (ALS)* Synthesis and Electronic Characterization of Nd$_{2-x}$Sr$_x$NiO$_4$ thin films ($0 \leq x \leq 1.4$)	05:20 PM- 05:40 PM	**WeII_17: Sahel-schackis, S.** *SLAC National Accelerator Laboratory* Investigation of the effects of nanoscale facets on catalytic activity in photo-driven nanosystems
05:40 PM- 06:00 PM	**WeI_18: Jose, A.** *Lawrence Berkeley National Lab* Magnetic Spectroscopy Across Scales: Investigating Biological Energy Systems and Synthetic Magnetic Architectures	05:40 PM- 06:00 PM	**WeII_18: Lim, J. W.** *Nanyang Technological University, Singapore* Anisotropic Ultrafast Carrier Dynamics Observed in Bulk ReS2 Using Femtosecond Time- and Energy-Resolved Photoemission Electron Microscopy
06:00 PM- 07:30 PM	**WeII_19: Free Time**		
07:30 PM- 09:30 PM	**WeII_20** *Banquet Dinner*		

Thursday 7/24

Session STh_01: Plenary Talks

08:30 AM-
09:15 AM

ThI_01: Prisle, N. L.

University of Oulu

Shifted protonation equilibrium of atmospheric acids and bases at the aqueous surface and impacts on cloud droplet chemistry and climate effects

09:15 AM-
10:00 AM

ThI_02: Kim, K.

title not available

10:00 AM-
10:30 AM

ThII_03

Break

Session SThI_01: Ambient Pressure Spectroscopy	Session SThII_01: Electronic Control in 2D Materials
10:30 AM- 11:00 AM **ThI_04: Boscoboinik, J. A.** *Brookhaven National Laboratory* Studying Nobles Gases with Photoelectron Spectroscopy	10:30 AM- 11:00 AM **ThII_04: King, P.** *University of St Andrews* Insensitivity of charge ordering instability to screening in the excitonic insulator candidate TiSe2
11:00 AM- 11:30 AM **ThI_05: Waluyo, I.** *Brookhaven National Laboratory* Revealing The Dynamic Surface State of Cu-based Single-Atom Alloys Using Ambient Pressure X-ray Photoelectron Spectroscopy	11:00 AM- 11:30 AM **ThII_05: Gedik, N.** title not available
11:30 AM- 11:50 AM **ThI_06: Guo, J.** *Lawrence Berkeley National Laboratory* Probing electrochemical interfaces and solvation in calcium-ion battery using operando soft X-ray spectroscopy	11:30 AM- 11:50 AM **ThII_06: Allison, T. K.** *Stony Brook University* Ultrafast dynamics in 2D materials and heterostructures visualized with time- and angleresolved photoemission
11:50 AM- 12:10 PM **ThI_07: Weinhardt, L.** *Karlsruhe Institute of Technology (KIT)* Local and Symmetry-Resolved Electronic Structure of Liquid Dimethyl Sulfoxide from Resonant Inelastic Soft X-ray Scattering	11:50 AM- 12:10 PM **ThII_07: Kundu, A. K.** *Brookhaven National Laboratory* Coexistence of Dirac and flat bands in 2D material beyond geometrical frustrated lattice

12:10 PM-
02:00 PM

ThII_08

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Session SThI_02: Ambient Pressure Spectroscopy

02:00 PM-
02:30 PM
ThI_09: Eren, B.
WEIZMANN INSTITUTE OF SCIENCE
SIMULTENOUS ELECTRON SPECTROSCOPY AND X-RAY SCATTERING ON MODEL CERIA CATALYSTS

02:30 PM-
02:50 PM
ThI_10: Anderson, S. R.
University of Wisconsin - Madison / Lawrence Berkeley National Lab
Inherent Co-ion Influence on Electric Double Layer Structure and Electrochemical Reactivity in the Hydrogen Evolution Reaction

02:50 PM-
03:10 PM
ThI_11: Weng, Y.
Uppsala University
Operando HAXPES Study of Solid Electrolyte Interphase Formation in Anode-Free Solid-State Batteries with PEO:LiDFOB

03:10 PM-
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ThI_12: Paudel, J. R.
Lawrence Berkeley National Laboratory
In Situ and Operando Characterization of Electronic and Chemical States of WO_3 During Electrochemical Proton Intercalation.

Session SThII_02: 2D Systems: Chirality & Spin Order

02:00 PM-
02:30 PM
ThII_09: Katoch, J.
Carnegie Mellon University
Inoperando nanoARPES on van der Waals heterostructures

02:30 PM-
02:50 PM
ThII_10: Nishino, F.
Institute for molecular science
Observation of the effect of surface chirality induced by chiral molecular overlayer on achiral substrates

02:50 PM-
03:10 PM
ThII_11: Panaccione, G.
NATIONAL RESEARCH COUNCIL (CNR) ITALY
Depth-dependent magnetic properties in the ferromagnetic van der Waals semiconductor VI3

03:10 PM-
03:30 PM
ThII_12: Ekahana, S.
Carnegie Mellon University
Flattened Band and AA+AB on bilayer graphene heterostructure

03:30 PM-
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ThII_13
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Session SThI_03: Ambient Pressure Spectroscopy

04:00 PM-
04:20 PM
ThI_14: Jaugstetter, M.
Lawrence Berkeley National Laboratory
Insitu photoelectron spectroscopy reveals the structure of the electrical double layer

04:20 PM-
04:40 PM
ThI_15: Zhou, Q.
Fritz-Haber-Institut der Max Planck Gesellschaft
Liquid-jet Velocity Map Imaging

04:40 PM-
05:00 PM
ThI_16: Huttula, M.
University of OUlu
Scaling research: From photoelectron-ion coincidences to photocatalytic solar hydrogen production

Session SThII_03: Electronic States at Buried Interfaces

04:00 PM-
04:20 PM
ThII_14: Ueda, S.
National Institute for Materials Science
Interface electronic and magnetic states and band dispersion of insulator/ferromagnet heterostructures studied by hard x-ray angle-integrated and angle-resolved photoemission

04:20 PM-
04:40 PM
ThII_15: Middey, S.
Indian Institute of Science
Site-selective polar compensation of Mott electrons in a double perovskite heterointerface

04:40 PM-
05:00 PM
ThII_16: Choi, B.
Lawrence Berkeley National Laboratory
Direct Observation of a Thickness-Induced Mott Transition in LaTiO3 films

	Session SThI_04: Photoemission Microscopy		Session SThII_04: Crystalline Electronic Structure
05:00 PM-05:20 PM	**ThI_17: Aashwin, A.** *SLAC National Accelerator Laboratory* nanoXPEEM: Attention Based Nanoscale X-ray Photoemission Electron Microscopy	05:00 PM-05:20 PM	**ThII_17: Fukutani, K.** *Institute for Molecular Science* Translational symmetry-breaking in the quasiparticle band structure of organic semiconductor rubrene
05:20 PM-05:40 PM	**ThI_18: Sadowski, J. T.** *Brookhaven National Laboratory* Spectro-microscopy of surface magnetization in Fe3GeTe2	05:20 PM-05:40 PM	**ThII_18: Fukushima, Y.** *Science Tokyo* Observation of surface-state transport by Mn doping into a topological crystalline insulator SnTe thin film with strain
05:40 PM-06:00 PM	**ThI_19: Achinuq, B.** *Lawrence Berkeley National Laboratory* An XPEEM Study of Voltage-induced Magnetic Domain Separation in a La0.7Sr0.3MnO3 Thin Film	05:40 PM-06:00 PM	**ThII_19: Cao, Y.** *Brookhaven National Laboratory* Unveiling a Unified Mechanism for Single-Photon Emission Across the III-Nitride
06:00 PM-07:30 PM	**ThI_20: Poster Session II with Casual Dinner**		

Friday 7/25

Session SFr_01: Plenary Talks

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title not available

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10:00 AM
FrI_02: Reutzel, M.
Georg August Universität Göttingen
Excitons in space and time - a femtosecond momentum microscopy study

10:00 AM-
10:30 AM
FrII_03
Break

Session SFrI_01: Theory

10:30 AM-
11:00 AM
FrI_04: Minár, J.
University of West Bohemia, Pilsen, Czechia
Quantum Materials and Magnetic Phenomena Studied by Spin-Resolved ARPES: Theoretical perspectives

11:00 AM-
11:30 AM
FrI_05: Zhang, T.
Institute of Theoretical Physics(ITP, CAS)
Topological and chiral phonons: theory, prediction and detection

Session SFrII_01: Electronic Spectromicroscopy

10:30 AM-
11:00 AM
FrII_04: Chen, Y.
University of Oxford
Strong Inter-valley Electron-Phonon Coupling in Magic-Angle Twisted Bilayer Graphene

11:00 AM-
11:30 AM
FrII_05: Jozwiak, C.
Advanced Light Source
Tipping the length scale with ARPES

11:30 AM-
11:40 AM
FrI_06
Break

11:30 AM-
11:45 AM
FrII_06
break

11:45 AM-
12:30 PM
FrII_07: Poster Prize / Announcements / Closing Remarks

12:30 PM-
02:30 PM
FrII_08: SSRL Tour (Proposed)

Abstracts

Recent progress in predicting the optical properties of defective and disordered systems

Giulia Galli [1,2,3]

[1]*Pritzker School of Molecular Engineering, University of Chicago*
[2]*Department of Chemistry, University of Chicago*
[3]*Material Science Division, Argonne National Laboratory*

We present recent progress in predicting the optical properties of spin defects in semiconductors and insulators and of solid/liquid interfaces using advanced first principle techniques. These include quantum defect embedding theory, hybrid, spin-flip time dependent density functional theory and the Bethe-Salpeter equation. We compare our results with several experiments and also discuss open challenges in the field.

Liquid-jet photoelectron spectroscopy of aqueous solutions: Electron scattering, electronic structure and work functions

Bernd Winter

Fritz-Haber-Institut, Faradayweg 4-6, 14195 Berlin, Germany

The electronic structure of liquid water and aqueous solutions is directly accessible by liquid-jet photoelectron spectroscopy (LJ-PES). Energies of photoelectron spectral features have so far typically been referenced to the vacuum-energy level, *i.e.*, in relation to the gas phase, which per definition disregards explicit surface properties such as the work function of aqueous solutions. We discuss how the solution work function changes, as a function of solute type and concentration, can be inferred from a LJ-PES measurement, based on an explicit consideration of a solution's Fermi energy. Competing surface-charging effects contributing to energy shifts of solute and solvent spectral features are important to be quantified and will be explored. This inherently connects to the ability to extract accurate electron binding energies from aqueous solution via LJ-PES experiments.

This talk further discusses the application of LJ-PES to provide structural information of biomolecules in a complex aqueous environment, such as adenosine triphosphate interacting with metal cations. This is achieved by the simultaneous analysis of valence, core-level, and non-local autoionization electron signals. We conclude with a consideration on applying electron Velocity Map Imaging (VMI) to liquid jets, with a focus on electron scattering in solution. VMI promises to vastly increase photoelectron collection efficiency and angular range, and in particular to enable the detection of photoelectron angular distributions in a single measurement. This includes the challenging detection of photoelectron circular dichroism (PECD) from aqueous-phase chiral (bio)molecules.

Photoemission spectromicroscopy: correlating local chemistry and electronic structure in quantum materials

Inna Vishik[1,2], Journey Byland[1], Matthew Staab[1], Sudheer Sreedhar[1], Ruichao Chen[2], Andrea Gauzzi[2], Valentin Taufour[1]

[1] *University of California, Davis, CA, USA*
[2] *IMPMC, Sorbonne Université-CNRS-MNHN, Paris, France*

Angle-resolved photoemission spectroscopy (ARPES) is a surface sensitive probe of electronic band structure in crystalline solids, and electronic structure can be strongly influenced by local chemistry/composition. Examples where composition strongly affects electronic structure includes doping-controlled superconductors and topological materials where the presentation of the characteristic surface state depends on the surface termination. Surface chemistry can be measured sequentially with ARPES in the same probing volume using x-ray photoelectron spectroscopy (XPS), and micro/nanofocusing of the photon beam has enabled these techniques to tackle systems with mesoscale inhomogeneity. I will discuss my group's work correlating local surface chemistry with surface electronic structure, using high-temperature superconductors and Weyl semimetals as examples.

Keywords: microARPES, XPS, correlated electron systems, superconductors, Weyl semimetals

Acknowledgement: The work on high temperature superconductors is supported by GBMF Award ID #12957, and the work on Weyl semimetals is supported by NSF DMR 2428464

Evolution of enamel nanocrystal misorientation before and after diet changes.

Pupa U.P.A. Gilbert[1], Barat Q. Achinuq[2], Mackie C. O'Hara
[1] *UW-Madison, Madison, WI, USA*
[2] *Advanced Light Source, Berkeley, CA, USA*
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Tooth enamel is macroscopically similar across all vertebrate animals, but at the nanoscale it varied dramatically. Using PhotoEmission Electron Microscopy (PEEM)(1,2) and Polarization-dependent Imaging Contrast (PIC) mapping (3), we first revealed that the enamel nanocrystal misorientation varies gradually across modern human enamel (4). Then, we compared tooth enamel before and after 3 major dietary shifts: the introduction of meat 2 million years ago, the Agriculture Revolution 10,000 years ago, and the Industrial Revolution 200 years ago. Key findings include:

A. After the introduction of meat, the nanoscale misorientation of adjacent crystal increased significantly, by a factor of 1.5x.

B. After the Agriculture Revolution, the misorientation increased even more, by a factor of 2x.

C. After the Industrial Revolution, the misorientation did not change.

D. Comparing modern chimpanzee and modern human, we find an even greater misorientation increase, by a factor of 3x.

These findings were obtained comparing the misorientation of millions of pixels, and they are all statistically highly significant. They provide crucial, quantitative evidence of enamel adaptations to dietary changes throughout hominoid evolution, offering a new perspective on the evolution of our lineage.

Acknowledgements: European Union's Horizon 2020 programme, Marie Sklodowska-Curie grant 101026776 to MCOH. Turkana Basin Institute-Simons Foundation grant to MCOH; DOE grant DE-FG02-07ER15899 and NSF grant DMR-2220274 to PUPAG. Experiments done at Advanced Light Source, a U.S. DOE Office of Science User Facility under Contract no. DE-AC02-05CH11231.

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3. RA Metzler... PUPA Gilbert. Phys Rev B 77, 064110-064111/064119 (2008).

4. E Beniash.... PUPA Gilbert. Nat Commun 10, 4383/4381-4313 (2019).

Conformally invariant charge fluctuations in a strange metal

Peter Abbamonte[1,2], Xuefei Guo[1,2], Jin Chen[1,2], Faren Hoveyda-Marashi[1,2], Simon Bettler[1,2], Dipanjan Chaudhuri[1,2], Caitlin Kengle[1,2], John Schneeloch[3], Ruidan Zhong[3], Genda Gu[3], Tai-Chang Chiang[1,2], Alexei M. Tsvelik[3], Thomas Faulkner[1], Philip W. Phillips[1,4]

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In this talk I will give a common-sense introduction to the topic of strange metals. First observed in copper oxide high-temperature superconductors, the strange metal state is now found in a wide variety of materials, ranging from organic molecular crystals to cold atom simulators to twisted bilayer graphene. The key feature is Planckian dissipation, where the scattering rate $\tau^{-1} = k_B T / \hbar$ is determined only by fundamental constants, representing a conjectured universal limit on the degree of quantum entanglement possible in a many-body system. I will trace the history of this field, from the marginal Fermi liquid theory of the late 1980's to the development of the SYK model by Sachdev and Ye in 1993, which Kitaev showed in 2014 could be "derived" using holographic duality, drawing intriguing parallels to black holes.

Additionally, I will present new momentum-resolved inelastic electron scattering experiments on density fluctuations in the strange metal $Bi_2Sr_2CaCu_2O_{8+x}$, revealing that the density fluctuations are quantum critical and exhibit conformal invariance—a property that featured prominently in Kitaev's derivation. Finally, I will discuss evidence for a newly identified excitation, the "scramblon," which is a characteristic of the extreme dissipative nature of these weird materials.

Keywords: strange metals, EELS

Acknowledgement: This work was primarily supported by the Center for Quantum Sensing and Quantum Materials, an Energy Frontier Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under award DE-SC0021238. P.A. gratefully acknowledges additional support from the EPiQS program of the Gordon and Betty Moore Foundation, grant GBMF9452.

Interfaces: Structure & Chemistry (California)
MoII_05 - 11:00 AM - 11:30 AM

Abstract Not Available

Temperature dependent crossover behaviors in photoemission of strongly correlated f- and d-electron systems

Jonathan Denlinger[1]

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The Kondo effect in highly correlated rare-earth and actinide heavy fermion f-electron systems is well-known to exhibit a temperature-dependent crossover from high *T* fluctuating local moments to fully-screened moments at low *T*. For crystalline systems, a lattice coherence crossover behavior that is also experimentally manifested as low T downturns of the resistivity, and in the photoemission signatures of strong variations in the narrow-width *f*-peak intensity and in their band-crossing hybridization with non-*f* bands. The most prominent examples are for single *f*-electron (or *f*-hole) Ce, Sm or Yb compounds.

Another related, but different, *T*-dependent crossover behavior exists in *multi-electron multi-orbital* correlated *d*-electrons systems which arise from Hund's rules physics favoring high-spin configurations, and the interplay between different spin and orbital coherence temperature scales. Fewer examples exist of this Hund metal coherence crossover behavior in photoemission experiments, with Fe-based superconductors and ruthenate oxides being the most well-known. Here we present ARPES and spin-resolved ARPES of a triple-layer ferromagnetic strontium ruthenate that contains *two* separate narrow bands that exhibit a strong *T*-dependent crossover behavior that is unprecedentedly as strong as observed in any *f*-electron Kondo system [1]. Moreover, the two narrow *d* bands are shown to be *spin-polarized,* and with opposite-sign, highlighting the differences with Kondo screening, and are discussed in the context of a site- and orbital-differentiated Hund metal correlations in presence of magnetic order.

Keywords: Kondo effect, Hund metal, strong electron correlations, coherence.

[1] P. Ngabonziza, J. D. Denlinger, A. V. Fedorov, J. W. Allen, G. Cao, G. Gebreyesus. Author, R. M. Martin, "*Layer-dependent spin-resolved electronic structure in ferromagnetic triple layered ruthenate* $Sr_4Ru_3O_{10}$", Physical Review B (111) 115146 (2025).

Chemical shifts in photoelectron spectroscopy of liquids

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[1] *Department of Physical Chemistry, University of Chemistry and Technology, Prague, Czech Republic*
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Chemical shifts in X-ray Photoelectron Spectroscopy (XPS), also known as Electron Spectroscopy for Chemical Analysis (ESCA), provide crucial insights into the electronic environment of atoms in different chemical states. While well-established in gas-phase and solid-state studies, ESCA chemical shifts in the liquid phase remain less explored. Recent advances in liquid-jet photoelectron spectroscopy (LJ-PES),[1] particularly absolute binding energy calibration methods,[2] now allow for unprecedented precision in aqueous-phase measurements. We present the first systematic study of chemical shifts in aqueous ESCA, focusing on carbon 1s binding energy variations across key organic functional groups. We analyze trends in oxidation states, correlate measured shifts with computed partial charges, and compare liquid-phase ESCA data with gas-phase and solid-state counterparts. Additionally, we explore secondary chemical shifts propagating through adjacent bonds and discuss the complementarity of ESCA and NMR in structural analysis. Our findings establish fundamental principles for interpreting chemical shifts in solution and pave the way for broader applications of LJ-PES in molecular chemistry and materials science.

Keywords: Chemical shifts, liquid jets, ESCA, XPS, Liquid-Jet Photoelectron Spectroscopy, chemical analysis, NMR.

Acknowledgement: Work supported by the Czech Science Foundation, grant number 24-12267O.

[1] R. Signorell, B. Winter, “*Photoionization of the aqueous phase: clusters, droplets and liquid jets*”, Physical Chemistry Chemical Physics (24) 13438-13460 (2022).

[2] S. Thürmer et al., “*Accurate vertical ionization energy and work function determinations of liquid water and aqueous solutions*”, Chemical Science (12) 10558-10582 (2021).

Electronic structure study of a uranium-based material using angle-resolved photoemission spectroscopy

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The study of the electronic structure of *f*-electron strongly correlated systems is a topic of substantial research interest due to the intriguing phenomena they exhibit, such as heavy fermion behavior and unconventional superconductivity, as well as their relevance in quantum and nuclear technologies. 5*f*-electron systems, in particular, present a captivating topic of study because of the unique duality of the 5*f*-electrons. This duality has been a challenge in understanding their relation to the physical phenomena and Fermi surface topology. Angle-resolved photoemission spectroscopy (ARPES) is a state-of-the-art tool that allows direct visualization of the electronic structure of crystalline solids. Here, we present the ARPES study of a uranium-based 5*f*-electron system, supported by density-functional theory calculations. The results unveil the nature of 5*f* electrons, their hybridization with conduction electrons, and overall electronic structure of this material. This study will advance the fundamental understanding of the electronic structure of uranium-based and actinide materials in general.

Acknowledgement: This work is supported by Idaho National Laboratory's Laboratory Directed Research and Development (LDRD) program and Department of Energy (DOE) Office of Science. This study utilized the resources of the Advanced Light Source (ALS), which is a DOE Office of Science user facility.

Fingerprint of the dipole orientation of water molecules in Cu^{2+} aqueous solution probed by x-ray photoelectron spectroscopy

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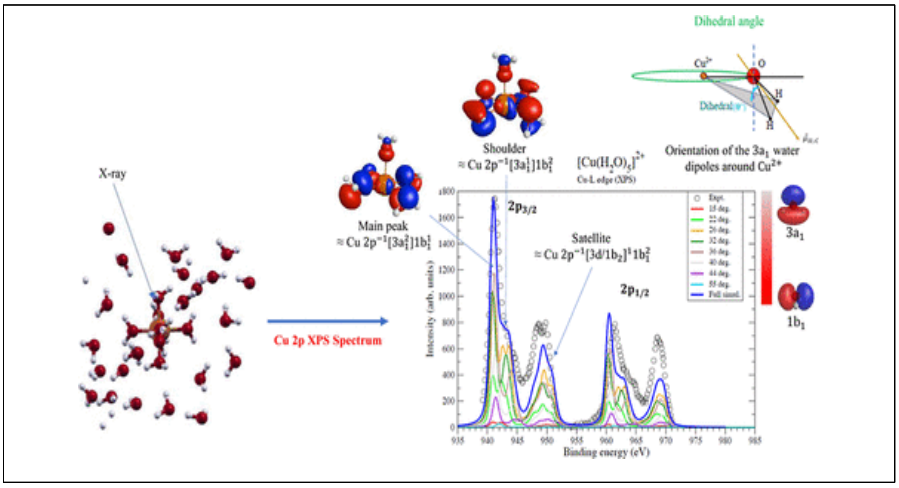
Keywords: liquid, X-ray photoelectron spectroscopy, ab-initio molecular dynamics and simulation of core-level spectra

The hydration of transition metals in aqueous solutions has long been studied due to its theoretical and experimental complexity, with much focus on the number of water molecules in the first coordination shell. Various methods, including EXAFS, NEXAFS, neutron diffraction, and computational approaches like Molecular Dynamics and Density Functional Theory, have been employed.

Recently, X-ray Photoelectron Spectroscopy (XPS) recorded the core-level spectra of copper compounds in liquid for the first time. Although typically used to identify chemical shifts and oxidation states, XPS has now been shown to reveal the orientation of neighboring water molecules through secondary excitations following Cu2p core ionization. This discovery highlights a novel use of XPS in probing non-covalently bonded systems, offering insights into dipole moment distributions. The ab-initio calculations of the Cu 2p core-level spectra presented in this context provide a comprehensive understanding of the underlying mechanisms responsible for both the main peak and satellite features.



[1] Mosaferi et al., *J. Am. Chem. Soc.* 2024, 146, 14, 9836-9850

Correlated Materials I (Island)
MoI_09 - 02:00 PM - 02:30 PM

Abstract Not Available

Capturing Electrons in Motion: Time-Resolved X-ray Photoemission Spectroscopy for Unraveling Charge Transfer Dynamics

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The study of excitations and their dynamics is of great significance in nature and applied technologies. A multitude of fundamental aspects of our world, including chemical processes and transport phenomena, are based on dynamic reaction chains. At the core of numerous emerging concepts concerning the conversion of sunlight into fuel and electricity are interfacial processes that necessitate an optimised, concerted flow of charge and energy at the molecular level. In order to gain a comprehensive understanding of the fundamental dynamics and scaling laws that enable molecular, interfacial, and macroscopic charge and energy transport, it is necessary to connect processes that evolve on spatial and temporal scales spanning orders of magnitude. As interface properties undergo changes during operation, for instance, under the influence of applied electrochemical stimulus or external photoexcitation, and due to the coexistence and competition of multiple bulk and interface processes, detailed in operando characterisation is imperative. Concurrently, interfaces present a challenge in technologies that rely on charge transfer processes. These processes require sophisticated experimental probes to elucidate the underlying physical and chemical mechanisms. In this context, soft X-ray spectroscopy techniques have emerged as a particularly effective means of monitoring the electronic and chemical states of matter, exhibiting the requisite elemental site-specificity and chemical sensitivity to facilitate rigorous testing and enhancement of our fundamental understanding of interfacial chemistry and photophysics in complex systems. Here we present an overview of the diverse experiments conducted in recent years using synchrotron radiation sources and free-electron lasers. The results of these experiments demonstrate that time-resolved XPS provides a unique opportunity to investigate the photo-induced interfacial charge transfer dynamics with exceptional site-specificity and temporal resolution in the picosecond to femtosecond regime across a range of sample systems. Access to the element and chemically specific core-levels provides a unique perspective of the dynamic charge evolution in the direct vicinity of the atom from which the core electron is emitted. This unique capability will enable previously unattainable insights into underlying microscopic mechanisms and thus pave the way for a better understanding of emerging photovoltaic and photocatalytic frameworks.

Keywords: Charge transfer dynamics, Time-resolved XPS, Synchrotron radiation, Free-electron laser (FEL), Energy conversion

Acknowledgement: This work was supported within the program “Structure of Matter” of the Helmholtz Association and by the BMBF (Grant No. 05K22OF2 within ErUM-Pro).

Polaronic state across the photoinduced transition in a magnetoresistive manganite

Shiyu Fan[1], Feng Jin[2], Taehun Kim[1], Umesh Kumar[3], Zixun Zhang[2], Vivek Bhartiya[1], Jiemin Li[1], Brandon Yalin[1], Yanhong Gu[1], Mingqiang Gu[4], Wen Hu[1], Claudio Mazzoli[1], G. Lawrence Carr[1], Osor. S. Bariˇsi´c[5], Andrey. S. Mishchenko[5], Valentina Bisogni[1], Sobhit Singh[6], Wenbin Wu[2], and Jonathan Pelliciari[1]

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Creating non-ergodic ‘hidden’ phases beyond thermodynamic equilibrium is a key topic in condensed matter physics. Femtosecond laser pulses can induce long-lived, reversible transitions into such states, but their microscopic interactions remain elusive due to experimental limitations. Here, we combine femtosecond laser excitation with ultrahigh-resolution Resonant Inelastic X-Ray Scattering (RIXS) to probe elementary excitations in $La_{2/3}Ca_{1/3}MnO_3$ (LCMO). Ultrafast photoexcitation at 1.2 eV suppresses the Jahn-Teller distortion by renormalizing Mn^{3+} e_g electron occupancy, softening polaron energy, and stabilizing a metastable ‘hidden’ phase distinct from the bulk ferromagnetic metallic state. By correlating polaron energy with resistivity, together with the analysis of Jahn-Teller distortion, we reveal that polaronic states of the antiferromagnetic insulating (AFI), ferromagnetic metallic (FMM), paramagnetic insulating (PMI) and photoinduced phases in LCMO have distinct origins, highlighting the complexity of electron-lattice interactions in manganites. This laser-RIXS approach paves the way for exploring photoinduced ‘hidden’ phases in quantum materials with sensitivity to multiple electronic degrees of freedom.

Keywords [optional]: Polaron, RIXS, ultrafast laser, manganites, photoinduced `hidden’ phase

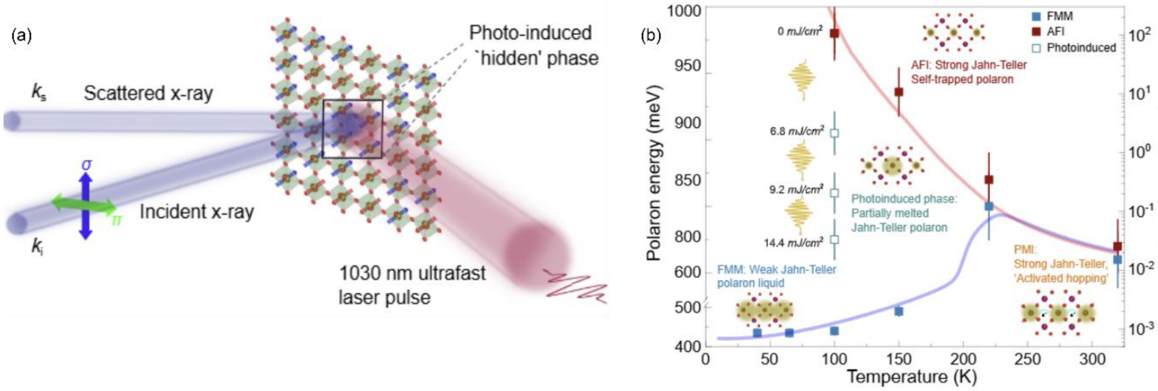

Figure 1: (a) Schematic view of the laser-RIXS experimental setup at the 2ID-SIX beamline in Brookhaven National Laboratory. (b) Polaron phase diagram of LCMO. Square data points are polaron energies measured by RIXS and the solid curves are electric resistivity. The cartoons illustrate the different Jahn-Teller polaronic states corresponding to the AFI, FMM, photoinduced, and PMI phases, respectively.

The impact of the oxide transport layer on the energy band alignment of the frontier orbitals in non-fullerene acceptor organic solar cells

Noemi Vannucchi[1,2], Fredrik O.L. Johansson[2], Mariam Ahmad[3,4], Tanya Kumari[3], Chun Yuen Ho[3], Kun Wang[3], Cathy Vang[1], Hervé Cruguel[1], Morten Madsen[3,4], Andreas Lindblad[2] and Nadine Witkowski[1]

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[2]Division of X-ray Photon Science, Uppsala University, Uppsala, Sweden
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The transition from fullerene to non-fullerene acceptors (NFAs) has drastically increased the power conversion efficiency (PCE) of organic photovoltaics, reaching PCEs above 20% [1]. The PM6:Y7 polymer donor:NFA binary blend is one of the most promising systems [2] thanks to its broad absorption range, which favors high photocurrents in such based devices [3]. Frontier orbital band alignment between the active layer and the oxide charge transport layers is a key factor affecting the device's efficiency. This aspect remains unexplored in the PM6:Y7 blend system, especially with varying donor-acceptor ratios. This work investigates the impact of the charge transport layers ZnO and MoO_3 on the electronic structure and the band alignment of spin-coated PM6:Y7 thin films as a function of the donor-to-acceptor mass ratio. To map this energetic landscape, we employed synchrotron techniques such as X-ray and UV photoemission spectroscopy to access the core level electronic structure, the work function, and the HOMO onset, and X-ray absorption spectroscopy for determining the LUMO onset. We observed that the optimal charge transport preventing recombination at both oxide interfaces is obtained for a 1:2 donor-to-acceptor mass ratio blend film, corresponding to the lowest bandgap (Fig. 1). Moreover, when films are prepared on ZnO substrate, a reversed p-n junction for charge extraction is observed, driven by interface dipoles at the oxide/organic interface. In conclusion, our work has shed light on the impact of oxide transport layers in shaping band alignment within an organic solar cell stack— a crucial factor in achieving optimal device efficiency.

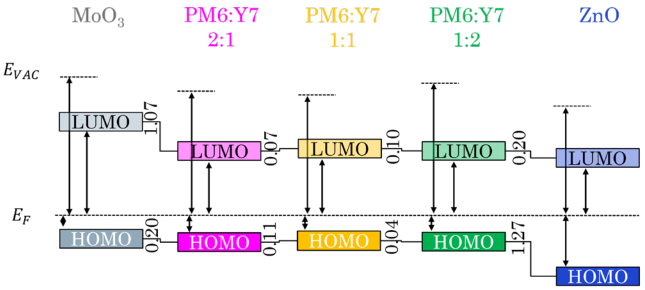

Figure 1. Frontier orbital band alignment at the interface between the organic blend PM6:Y7 with different mass ratios and the oxide transport layers

Acknowledgement: We thank the Synchrotron SOLEIL, the Helmholtz-Zentrum Berlin für Materialien und Energie, and the ISA Centre for Storage Ring for the allocation of synchrotron radiation beamtimes.

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[3] R. Yu, et al., "*Realization of high performance for PM6:Y6 based organic photovoltaic cells*", Journal of Energy Chemistry (61) 29-46 (2021).

Enhanced superconducting gap in the Hg-based trilayer cuprate superconductor revealed by micro-spot ARPES

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Yu Murano[1], Jacek Osiecki[4], Balasubramanian Thiagarajan[4], Craig Polley[4], Chul-Ho Lee[3],
Taichiro Nishio[2], Hiroshi Eisaki[3], Iwao Matsuda[1]

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The record of the highest superconducting transition temperature (T_c) at ambient pressure has been held by the Hg-based trilayer cuprate $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Hg1223) [1] for more than 30 years. Even though its electronic-structure investigation is of primary importance, angle-resolved photoemission spectroscopy (ARPES) studies have been hampered by the limited availability of high-quality single crystals as well as by the difficulty in exposing a flat surface with large area by cleaving. Unlike the well-studied Bi-based counterpart $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi2223) with a lower T_c, Hg1223 lacks natural cleavage planes, and hence cleaving should yield a disordered surface. Recently, Mino *et al.* [2] have established methodology to grow single crystals of the Hg-based trilayer cuprates with high reproducibility through partial Re substitutions, and single-crystalline $(Hg,Re)Ba_2Ca_2Cu_3O_{8+\delta}$ [(Hg,Re)1223] samples with T_c's exceeding 130 K have been reproducibly synthesized. While inhomogeneity created by cleaving would be inevitable, one could purify ARPES signals by exploiting a tightly focused incident beam.

We have performed micro-spot ARPES measurements of (Hg,Re)1223 at the Bloch beamline of MAX IV [3], where the photon beam is focused down to $10\mu m \times 10\mu m$. The quality of the ARPES spectra varied significantly over space, but Fermi surfaces and superconducting gaps were successfully captured by pinpointing an appropriate spot. The prominent feature of the trilayer cuprate is the existence of two inequivalent CuO_2 planes in a unit cell. Compared to the trilayer cuprate Bi2223 with a lower T_c, the superconducting gap of (Hg,Re)1223 was enhanced selectively for one of those CuO_2 planes, suggesting the reinforced Cooper paring on that CuO_2 plane as a key ingredient of the highest T_c of the Hg-based trilayer cuprate.

Keywords: high-temperature superconductors, cuprates, micro-spot ARPES.

Acknowledgement: Work supported by Japan Society for the Promotion of Science (JSPS).

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Hard X-ray Core-hole Clock Spectroscopy of Adsorbates and Polymer Mixtures

Andreas Lindblad[1], Elin Berggren[1], Yi-Chen Weng[1], Fredrik O. L. Johansson[1], U. Cappel
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Resonantly exciting a core electron promotes it to an unoccupied electronic state. The core-excited state decays with the core hole filled by an electron. Excess energy may be emitted by another electron in a resonant Auger decay. In solids and adsorbates, the core-excited electron can tunnel away, which then leads to a normal Auger decay of the core hole at energies below the ionization potential.

Analysing spectral components' relative intensities in *e.g.* the KLL Auger kinetic energy region with photon energies near the resonance reveals insights into the electron's propensity to tunnel away as it is dependent on the energy landscape. This information aids the study of charge transfer processes in materials [1-3]. The tunnelling must occur within the core-hole lifetime to affect the spectral content, hence acting as a "core-hole clock".

In Xenon adsorbed on metal surfaces[1], polymer heterojunctions for organic electronics[2], and quantum dots (PbS) [3] the low excess energy part vary depending on composition whereas higher energy excitations share the same tunneling behavior. For adsorbed Xe core-excitation highlight channels present only in the core-excited state. It turns out that the material compositions that give best macroscopic performance with nanoparticles and in the polymer case conductivity in organic electronics built from these materials also have the swiftest charge transfer times measured via the core-hole clock method. The amount of molecular alignment also impact the charge transfer time.

An outlook with recent results on bilayer and heterojunction polymer systems will be given with an emphasis on evolution of the core hole clock spectrum upon X-ray exposure.

Keywords [optional]: Core hole clock spectroscopy; synchrotron based hard X-ray photoelectron spectroscopy; polymers; adsorbate

Acknowledgement [optional]: Work was supported by the Swedish Research Council.

[1] Fredrik O. L. Johansson, *et al.* "Resonant Auger spectroscopy on solid xenon on gold, silver, and copper substrates." Physical Review A **107**:3 (2023): 032802.

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Anomalous normal state gap in an underdoped n-type cuprate

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The cuprate superconductors host a mysterious pseudogap phase, where the density of states near the Fermi level is partially suppressed. While the microscopic origin of the pseudogap phase in the p-doped cuprates is not yet agreed upon, the pseudogap of around a few hundred meV in the n-type cuprates is thought to arise from short-ranged antiferromagnetism order. This antiferromagnetic pseudogap evolves into the full antiferromagnetic gap when long-range order is achieved below about x = 0.12 in $Nd_{2-x}Ce_xCuO_4$ (NCCO). Here, we show through angle-resolved photoemission spectroscopy that underdoped NCCO with long-range antiferromagnetic order hosts another distinct normal state gap that is not compatible with antiferromagnetism and charge order [1]. This gap opens on the reconstructed small Fermi pockets centered at the (0, π) points in the Brillouin zone, and its doping dependence smoothly evolves into the (0, π) superconducting gap near optimal doping. These observations suggest that the ground state of the underdoped n-type cuprates are not conventional antiferromagnetic metals, but possibly hosting incoherent Cooper pairs with an energy scale up to ~40 meV and a temperature scale up to ~150 K.

Keywords: Angle-resolved photoemission spectroscopy, high Tc superconductors, pseudogap

Acknowledgement: Work was supported by the Department of Energy, Office of basic energy science

[1] K.-J. Xu et al., “Anomalous normal-state gap in an electron-doped cuprate”, *Science* **385**, 796-800 (2024).

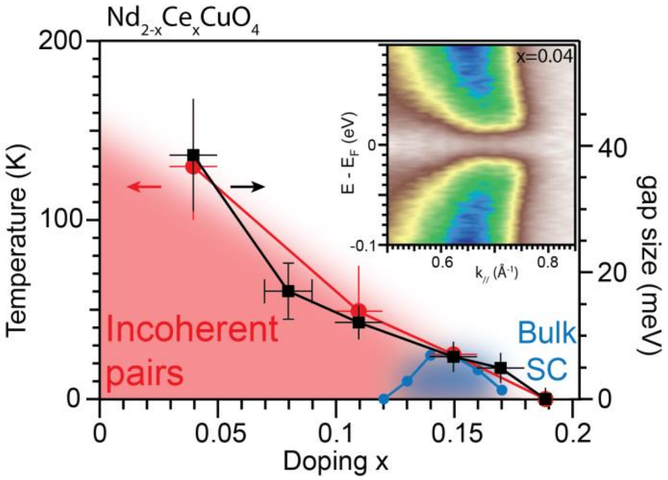

Figure 1. Simplified phase diagram of $Nd_{2-x}Ce_xCuO_4$. Bulk superconductivity, as defined by resistivity, is in blue. The normal state gap phase consistent with incoherent pairs is in red. Black data point indicate the gap size at the Brillouin zone boundary, and red data points indicate the gap filling temperature. Inset is the symmetrized ARPES spectra showing the normal state gap at *x*=0.04. Not shown are AF and charge order at higher temperature scales.

Unraveling charge and energy transfer dynamics between plasmonic metals and adsorbates

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Plasmonic metals are potential candidates for photocatalysis due to their tailorable light-matter interactions, which allows absorption of a significant portion of the solar energy spectrum with large cross-sections. The reactants and intermediates are adsorbed onto the plasmonic metal surface, and following the photoexcitation, the charge and/or energy transfer to the adsorbate drives the chemical transformation during photocatalysis. In this presentation, I will discuss our efforts to unravel ultrafast charge and energy transfer mechanisms using a model plasmonic metal-adsorbate photocatalyst: a thin gold film with a self assembled monolayer of 4-mercaptobenzoic acid (MBA). Ultrafast UV-visible transient spectroscopy was utilized to study photo-induced carrier dynamics of gold and MBA-gold upon interband (400 nm) and intraband (800 nm) excitations. The results suggests an elongation of electron-phonon (el-ph) and phonon-phonon (ph-ph) scattering rates when MBA is adsorbed onto gold compared to pristine gold. It is likely that el-ph scattering rate is slowed by hot electron trapping at the delocalized, lowest unoccupied molecular orbital (LUMO) in MBA-Au, mediating carrier recombination and that ph-ph lifetimes are extended due to the steric strain by MBA on Au atoms. Furthermore, ongoing theoretical investigations using real-time time-dependent density functional theory aim to interpret and support these experimental results.

Keywords: Ultrafast spectroscopy, plasmonic metals, photocatalysis

Universal electronic structure of layered nickelates via oxygen-centered planar orbitals

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In a series of groundbreaking discoveries, superconductivity has recently been demonstrated in $La_3Ni_2O_7$ up to 91 K under moderate pressure in bulk crystals, and up to 48 K at ambient pressure in thin films grown under compressive strain. Key questions remain open regarding the crystal structure and low-energy electronic states that support superconductivity in these compounds. Here we take advantage of the natural polymorphism between bilayer (2222) or alternating monolayer-trilayer (1313) stacking sequences that arises in bulk $La_3Ni_2O_7$ crystals to identify universal features in this family of materials. Employing angle-resolved photoemission spectroscopy (ARPES) we observe the fingerprint of a spin-density wave (SDW) instability, strong and coherent enough to modify the electronic structure[1]. We demonstrate that this feature is a 'translated' β Fermi surface associated with a scattering vector $Q_{t\beta}$ which matches the Q_{SDW} detected by neutron and x-ray scattering experiments. This observation provides an important link between surface and bulk probes, and demonstrates a universal connection between magnetism and fermiology in $La_3Ni_2O_7$ as well as $La_4Ni_3O_{10}$. Using an effective tight-binding model, we simulate the spectral weight distribution observed in our ARPES dichroism experiments and establish that the low-energy electronic phenomenology is dominated by oxygen-centered planar orbitals, which – upon moving along the Fermi surface away from the Ni-O-Ni bond directions – evolve from the $d3x^2-r^2$ and $d3y^2-r^2$ symmetry characteristic of 3-spin polarons to the familiar dx^2-y^2 Zhang-Rice singlets that support high-temperature superconductivity in cuprates. Despite the multiorbital nature of the nickelates, our work establishes an empirical correspondence between the low-energy electronic structure of cuprates and nickelates, thus suggesting a common origin for their unconventional superconductivity[1].

[1] Christine C. Au-Yeung, X. Chen, S. Smit, M. Bluschke, et al., arXiv:2502.20450 (2025).

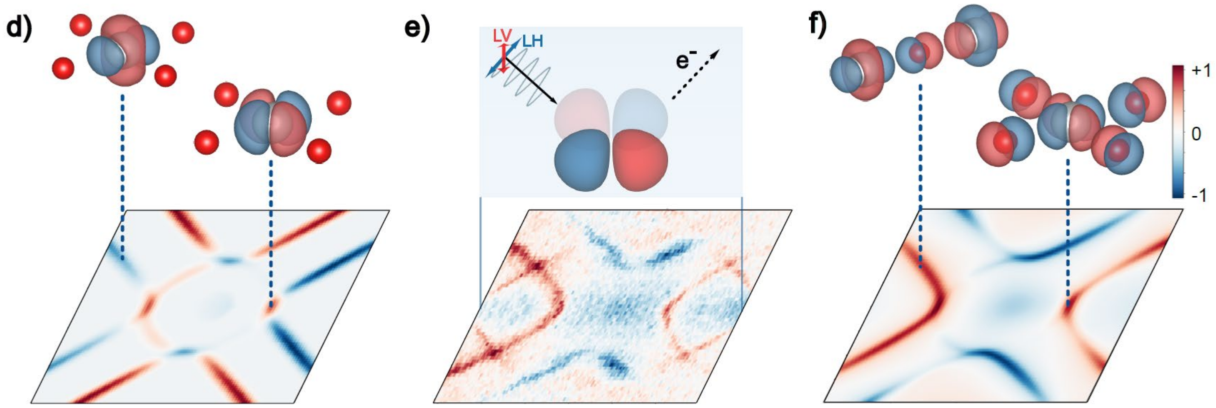

Figure 1. (d,f) (top) Schematic representation of wavefunctions at the indicated momenta along the FS with relative phases in red and blue (O and Ni sites in red and silver, respectively), along with (bottom) the simulated ARPES dichroism (LV-LH). (e) (top) Experimental geometry, showing the polarization vector of the light with respect to the in-plane orbitals, along with (bottom) the experimental dichroism measured by ARPES on LNO327.

Monitoring Uptake, Release and Reaction of Gases at the Liquid-Vapor Interface.

Tillmann Buttersack[1]

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Multiphase reactions are omnipresent in nature, industrial applications. The direct observation of reactions at the liquid-vapor interface requires spectroscopic techniques that are surface specific and chemically sensitive to detect low concentrations (~1 mM), e.g., photoelectron spectroscopy (XPS). Furthermore, the sample delivery method must allow sufficient time for an interface reaction to proceed. These complex challenges require individual approaches for each system of interest.

One example for a multiphase process is the reaction between liquid alkali metal (NaK) and water vapor, which is extremely fast (Fig. 1). We used a slow droplet train of NaK in a wet atmosphere and observed the formation of golden aqueous solutions with metallic properties with XPS.[1]

An example with relevance for atmospheric chemistry is the formation and the release of sulfur dioxide (SO_2) from aqueous sulfite solutions due to acidification. We demonstrated that dissolved gases can be detected with XPS even though their concentration is only about 1 mM.[2]

[1] P. Mason, C. Schewe, T. Buttersack, V. Kostal, M. Vitek, R. McMullen, H. Ali, F. Trinter, C. Lee, D. Neumark, S. Thürmer, R. Seidel, B. Winter, S. Bradforth, P. Jungwirth: "*Spectroscopic evidence for a gold-colored metallic water solution*", Nature 595, 673-676 (2021).

[2] T. Buttersack, I. Gladich, S. Gholami, C. Richter, R. Dupuy, C. Nicolas, F. Trinter, A. Trunschke, D. Delgado, P. Coral Arroyo, E. Parmentier, B. Winter, L. Iezzi, A. Roose, A. Boucly, L. Artiglia, M. Ammann, R. Signorell, H. Bluhm: "*Direct observation of the complex S(IV) equilibria at the liquid-vapor interface*", Nat. Commun. 15, 8987 (2024).

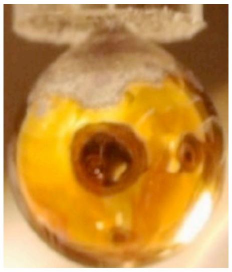

Figure 1. Gold colored metallic aqueous solution formed by the multiphase reaction between gaseous water and liquid alkali metal.

Correlated Materials II (Island)
MoI_15 - 04:30 PM - 05:00 PM

Abstract Not Available

Interface Dynamics and Chemistry (California)
MoII_15 - 04:30 PM - 05:00 PM

Abstract Not Available

The Growing Charge-Density-Wave Order in CuTe Lightens and Speeds up Electrons

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Charge density waves (CDWs) are pervasive orders in solids that usually enhance the effective mass (m*) and reduce the Fermi velocity (v_F) of carriers. Here, we report on the inverse — a reduced m* and an enhanced v_F correlated with the growth of the CDW order in CuTe with gapped, practically linearly dispersing bands — reminiscent of emergent CDW-gapped topological semimetals. Using momentum-dependent electron energy-loss spectroscopy (q-EELS, Fig. 1), we simultaneously capture m* and v_F of the CDW-related, practically linearly dispersing electrons by plasmon dispersions across the transition (335 K, T_{CDW}), with m* of 0.28 m_0 (m_0, the electron rest mass) and of ~0.005*c* (*c*, the speed of light) at 300 K [1]. With the growth of the CDW order-parameter strength toward 100 K, the electrons become lighter and move faster by ~20%. Thorough inspections below T_{CDW} unveil the essential underlining factor of the decreasing light-electron density by the increasing opening of the CDW gap. The quantum analogy of the reducing electron density in graphene diminishes electronic screening, yielding renormalized conical-linear bands with lighter, faster electrons and hinting on our discoveries. CuTe urges explorations on the CDW-correlation notion across versatile CDW-gapped quantum matters with q-EELS as a prominent probe for the subject.

Keywords: effective mass, Fermi velocity, topological quantum matters, plasmons, electron energy-loss spectroscopy.

[1] I.-T. Wang, T.-L. Chou, C.-E. Hsu, Z. Lei, L.-M. Wang, P.-H. Lin, C.-W. Luo, C.-W. Chen, C.-N. Kuo, C. S. Lue, C.-H. Chen, H.-C. Hsueh, M.-W. Chu, "*The Growing Charge-Density-Wave Order in CuTe Lightens and Speeds up Electrons*", Nature Communications (15) 9345 (2024).

Interface Dynamics and Chemistry (California)
MoII_16 - 05:00 PM - 05:20 PM

Limitations in Determining Oxidation States in Condensed Matter at the Sub-Nanometric Scale

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Oxidation state is a fundamental concept used to predict the behavior of compounds in chemical reactions[1,2]. Determining the oxidation state becomes even more important but complex when dealing with nanoclusters, where the properties of solid matter strongly depends on the number of atoms[3]. In the present work, we highlight the challenges in determining the oxidation state of size-selected clusters using synchrotron-based High-Resolution X-ray Photoelectron Spectroscopy, a technique commonly recognized for providing unambiguous fingerprints of the different oxidation state[4]. We investigated the oxidation of W_n (n=13, 25) nanoclusters deposited on epitaxial graphene at 40 K monitoring in-situ the evolution of W 4f spectral components growing at higher binding energies (BEs) compared to metallic W, attributed to the formation of tungsten oxide species. To shed light on the formation of these new components, we performed density functional theory simulations including calculations of core-electron BEs in the final-state approximation. The lack of crystalline order leads to a broad distribution of core levels that can be grouped in six different families, defined by the number of O atoms bonded to a specific W atom in the clusters. We found a strong linear relationship between valence, computed using Pauling's formula[5], and the calculated W $4f_{7/2}$ core levels. However, when the number of oxygen atoms bonded becomes large (n>3), the large variability of W-O interatomic distances causes the core levels of different families to overlap, making difficult to precisely assess W oxidation state, whose assignment should be reviewed in the case of nanostructures.

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[2] Jablonka, K. M. et al., *Using collective knowledge to assign oxidation states of metal cations in metal–organic frameworks*, Nat. Chem. 13, 771–777 (2021)
[3] Tyo, E. et al., *Catalysis by clusters with precise numbers of atoms*, Nature Nanotech. 10, 577–588 (2015)
[4] Morn, R. et al. *The oxidation of Platinum under wet conditions observed by electrochemical X-ray Photoelectron Spectroscopy*, J. Am. Chem. Soc. 141, 6537-6544 (2019).
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Angle-resolved photoemission spectroscopy of Dirac nodal-line superconductor $ZrSi_{1-x}P_xSe$

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H. Kito[6], I. Hase[6], S. Ishida[6], H. Fujihisa[6], Y. Gotoh[6], A. Iyo[6], H. Ogino[6],
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The coexistence of Dirac nodal-line (DNL) and superconductivity is expected to reveal exotic physical properties such as the Majorana fermion. ZrSiSe is a semimetal with Dirac nodal lines at the Fermi energy[1]. On the other hand, ZrPSe is a superconductor, but its nodal line is far from the Fermi energy[2,3]. In this study, we performed angle-resolved photoemission spectroscopy (ARPES) on single crystal samples of $ZrSi_{1-x}P_xSe$ ($x = 0.2, 0.45, 0.72, 1$). Figure 1 (a) shows the Fermi surface at $x = 0.72$. Two large Fermi surfaces (α, β) are observed in parallel. As the energy moves away from the Fermi energy, the α and β approach each other and form a DNL at -1.15 eV. On the other hand, the Fermi surface at $x = 0.2$ shown in Figure 1 (b), the α and β approach each other, and a DNL is formed at -0.2 eV. This observation indicates that the energy of the DNL changes continuously with the change in the substitution amount. Our findings will pave the way for the development of novel topological superconductors.

Keywords : Superconductor, Dirac nodal-line, Angle-resolved photoemission spectroscopy

Acknowledgement : The experiment was carried out at BL-1 of HiSOR (Proposal Nos. 24AG042, 23BG015). This work was financially supported by JSPS KAKENHI Nos. 23K03328, 24H01670, 25H00743.

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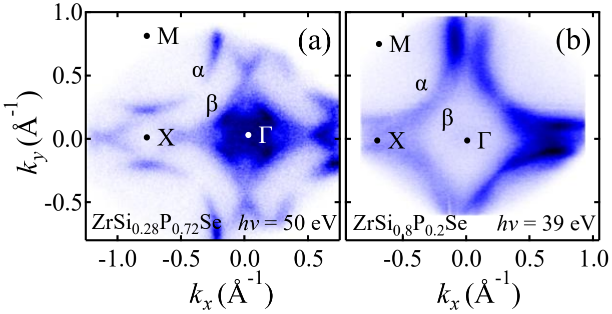

Figure 1. Fermi surface of $ZrSi_{1-x}P_xSe$, (a) $x = 0.72$ ($h\nu = 50$ eV), (b) $x = 0.2$ ($h\nu = 39$ eV).

Tuning Au–TiO_2 Photocatalytic Interfaces: Impact of Oxygen Vacancies and Gold Nanoparticle Size on Electronic Structure and Carrier Dynamics

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Gold (Au) integrated onto titanium dioxide (TiO_2) surfaces offers a promising strategy to enhance the photocatalytic efficiency of the semiconductor system. This work investigates how oxygen vacancy defects and structural changes in the anatase TiO_2 (101) surface influence the electronic properties and carrier dynamics of the Au–TiO_2 heterojunction. To this end, we compare the pristine surface with one modified by a single surface oxygen vacancy and examine how gold nanoparticle size affects these interactions by studying Au_4, Au_{19}, and Au_{20} clusters. Ground-state density functional theory (DFT) calculations demonstrate that the oxygen vacancy dramatically modifies the interfacial electronic structure, with density of states (DOS) analysis revealing the formation of a trap state for the isolated TiO_2 slab model. Upon formation of the Au–TiO_2 heterojunction, the defect site results in a larger perturbation to the DOS of the closed-shell Au clusters (Au_4, Au_{20}) TiO_2 heterojunction compared to the open-shell Au_{19} cluster TiO_2 heterojunction. Meanwhile, ab initio molecular dynamics (AIMD) simulations at 300 K further reveal distinct differences in the atomic motion of the AuNPs and the TiO_2 surface between pristine and oxygen-vacancy-modified systems under experimentally relevant conditions. Furthermore, nonadiabatic molecular dynamics (NAMD) simulations were employed to calculate carrier lifetimes associated with electron injection and electron–hole recombination processes across the interface. These findings underscore the critical role of defect engineering and interfacial structural dynamics in tailoring the performance of Au–TiO_2 photocatalytic systems.

Keywords: Gold, Oxides, DFT, AIMD, NAMD, Injection, Recombination

Correlated Materials II (Island)
MoI_18 - 05:40 PM - 06:00 PM

Three-dimensional flat band in ultra-thin Kagome metal, Mn_3Sn

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[2]*Australian Synchrotron, Clayton, VIC, Australia*

Flat bands (FBs) can give rise to strongly correlated electronic and topological phases because of their narrow energy dispersion, especially when located at the Fermi level. Whilst FBs have been experimentally realized in two-dimensional (2D) twisted van der Waals heterostructures, they are highly sensitive to twist angle, necessitating complex fabrication techniques. Geometrically frustrated lattices, such as the kagome lattice, have emerged as an attractive platform as they can natively host FBs due to destructive interference. While FBs have been observed in bulk Kagome metals, an outstanding experimental question is whether flat bands can be realized in the atomically-thin regime, with opportunities for stronger electron-electron interactions and the emergence of nontrivial topological phases.

Here we use angle-resolved photoelectron spectroscopy (ARPES), scanning tunnelling microscopy and band structure calculations to show that ultra-thin films of the kagome metal Mn_3Sn host a robust, dispersionless flat band with a bandwidth below 150 meV[1]. Furthermore, we demonstrate chemical tuning of the flat band to near the Fermi level via manganese defect engineering. The realization of tunable kagome-derived flat bands in an ultra-thin kagome metal, represents a promising platform to study strongly correlated and topological phenomena, with potential applications in quantum computing, spintronics and low-energy electronics.

Keywords: Kagome metal, Mn3Sn, flat band, ultra-thin film, strong correlation

[1] J. Blyth, M. Zhao, G. L. Causer, *et. al.* S., “*Three-dimensional flat band in ultra-thin Kagome metal Mn_3Sn film*”, arXiv: 503.05544 (2025).

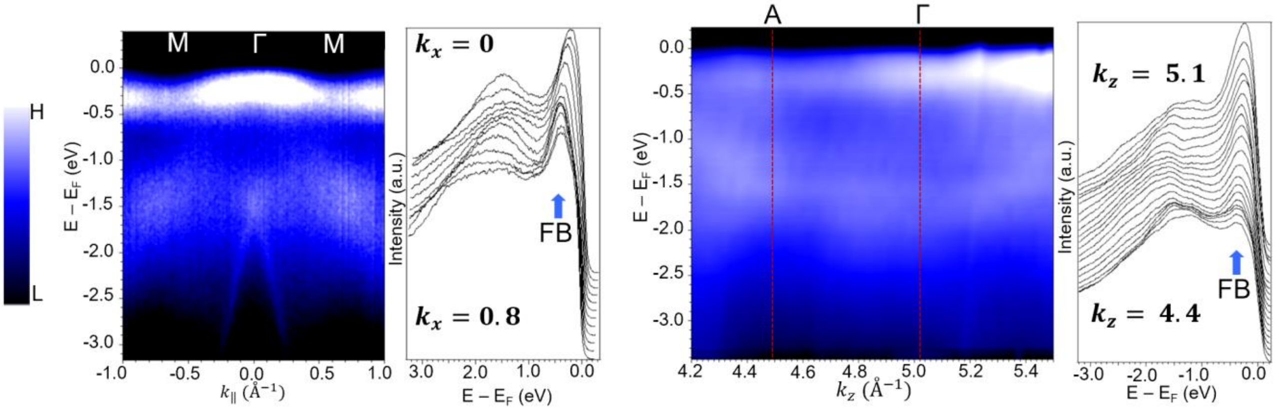

Figure 1. Observation of 3D flat band in Mn_3Sn ultra-thin films. (left panel) ARPES spectrum taken at hv = 104 eV along the ΓM direction. A high intensity band at ~ -300 meV persists across the entire momentum space with minimal dispersion (<150 meV) as shown in the single energy-distribution cuts. (right panel) out-of-plane (k_z) dispersion of the flat band extracted from photon-energy-dependent measurements where the variation in the flat band maximum is below 100 meV, indicating it is a 3D flat band.

Electronic Properties of Discrete NDI-T2 Oligomer Thin Films Using Photoelectron Spectroscopy and Low-energy Inverse Photoemission Spectroscopy

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The combination of photoelectron spectroscopy (PES) and low-energy inverse photoemission spectroscopy (LEIPES) is a valuable tool for the characterization of the density of occupied and unoccupied states in (organic) semiconductors. Their combination allows the transport or single particle bandgap to be determined. Especially for organic semiconductors, the possibility of such a measurement is crucial, as the transport bandgap is not only less accessible, but can significantly differ from the optical bandgap by 0.1 – 1 eV due to the correspondingly large Frenkel exciton binding energy that limits the charge separation in these materials. The low kinetic energy employed in LEIPES helps to avoid degradation due to the electron beam irradiation[1].

In this work, we combine PES and LEIPES with optical measurements, in particular spectroscopic ellipsometry to compare the transport and optical bandgaps of semiconducting discrete oligomers based on naphthalene diimide (NDI) and bithiophene (T2)[2] spin-coated on Ag substrates at different film thicknesses. Their thin films show optically anisotropic characteristics due to a preferential molecular orientation[3]. Here, for example in thin films of $T2\text{-}(NDI\text{-}T2)_2$, we observe an optical bandgap of 1.6 eV and a transport bandgap of 1.8 eV, suggesting an exciton binding energy of 0.2 eV. We reveal a transport bandgap narrowing effect at ultra-low film thicknesses, approaching the value of the optical bandgap, due to an energy shift in the onset position of the highest occupied molecular orbital. The potential roles of the substrate and the molecular orientation that lead to the results obtained are discussed.

Keywords: organic semiconductor, photoemission spectroscopy, low-energy inverse photoemission spectroscopy, transport bandgap, exciton binding energy, molecular orientation

Acknowledgement: The work was supported by the German Research Foundation (DFG) Research Unit FOR 5387 "POPULAR".

[1] H. Yoshida, "*Near-ultraviolet Inverse Photoemission Spectroscopy Using Ultra-low Energy Electrons*", Chemical Physics Letters (539) 180 (2012).
[2] R. Matsidik, H. Komber, M. Brinkmann, K. S. Schellhammer, F. Ortmann, M. Sommer, "*Evolution of Chain-length Dependent Properties of Discrete n-type Oligomers Prepared via Scalable Direct Arylation*", J. Am. Chem. Soc. (145) 8430 (2023).
[3] A. Ehm, E. Bortchagovsky, R. Matsidik, M. Sommer, D. R. T. Zahn, "*Uniaxial Molecular Alignment in Thin Films Composed of Discrete NDI-T2 Oligomers Investigated by Variable-Angle Spectroscopic and Imaging Ellipsometry*", Phys. Status Solidi B 2400202 (2024).

Gas-phase photochemistry studied by time-resolved photoelectron spectroscopy with free-electron lasers

Daniel Rolles[1]

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Time-resolved photoelectron spectroscopy (TRPES) is a powerful tool to study electronic de-excitation and chemical changes during photochemical reactions. However, when using visible or ultraviolet (UV) probe pulses, the observation window is often limited to low-binding-energy excited states near the Franck-Condon region. This limitation can be overcome when probing the reaction with extreme ultraviolet (XUV) pulses produced, e.g., from a free-electron laser (FEL) or a high harmonic generation (HHG) source. This allows for the entire reaction pathway to be probed, including any structural rearrangement or dissociation occurring on the electronic ground state. I will present two examples of XUV TRPES experiments performed at the FERMI FEL that take advantage of these capabilities: an investigation of the UV-(266-nm)-induced ring-opening reaction of the prototypical heterocyclic molecule thiophenone (C4H4OS)[1], and a study of the UV-(200-nm)-induced ring-reconfiguration of the quadricyclane-norbornadiene (C7H8) photo switch[2]. I will also compare the findings to results obtained for the same molecules with other ultrafast techniques to highlight the strengths and weaknesses of each method and the power of a multi-modal probing approach[3,4]. Finally, I will present results from a time-resolved inner-shell photoelectron spectroscopy experiment studying intramolecular charge transfer by taking advantage of the site-specificity of inner-shell photoemission.

Acknowledgement: Work supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, US Department of Energy, grants no. DE-FG02-86ER13491 and DE-SC0020276, and by the National Science Foundation grants no. PHYS-2409365 and PHYS-1753324, as detailed in the corresponding papers listed below.

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Plenary Talks (Island)
TuI_02 - 09:15 AM - 10:00 AM

Abstract Not Available

Recent surprises in quantum materials among many legacies of Einstein

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I will discuss several surprises we have found in investigating quantum materials that generally involve relativistic effects and novel light-matter interactions. Examples drawn from our recent work include [1-5]: (i) Presence of strong axion coupling in $MnBi_2Te_4$, where the antiferromagnetic order can be reversed with external electric field as well as with circularly polarized light. The material also supports Axion quasiparticles, which are a leading candidate for dark matter. (ii) An unusual light-matter coupling in a reconstructed surface of $SrTiO_3$, which leads to an intense, coherent beam of *secondary* photoemitted electrons, an effect which cannot be understood within the existing theory of photoemission. (iii) How concepts of ‘light cones’ and ‘event horizons’ familiar from Einstein’s special theory of relativity yield a much richer causality driven structure in quantum materials. And (iv) how the interplay of quantum geometry and topology can yield unique non-linear Hall and other effects. I will comment on these and related issues.

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[2] C. Y. Hong et al., *Nature* 617, 493 (2023).
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Symmetry breaking in core-valence double ionization of allene

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Richard J. Squibb[1], Rafael C. Couto[5], Leif Karlsson[3], Gunnar Nyman[6], Majdi Hochlaf[2],
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Conventional electron spectroscopy is an established one-electron-at-the-time method for revealing the electronic structure and dynamics of photoionized systems. By combining an electron-electron coincidence technique with the use of soft X-radiation we have measured a double ionization spectrum of the allene molecule in which one electron is removed from a C1s core orbital and one from a valence orbital, obtaining much more detailed information on doubly ionized systems compared to Siegbahn's Electron-Spectroscopy-for-Chemical-Analysis method. This core-valence double ionization spectrum shows the effect of symmetry breaking in an extraordinary way[1], when the core electron is ejected from one of the two outer carbon atoms. To explain the spectrum, we utilized a new theoretical approach combining the benefits of a full self-consistent field approach with those of perturbation methods and multi-configurational techniques, thus establishing a powerful tool to reveal molecular orbital symmetry breaking on such an organic molecule, going beyond Löwdin's standard definition of electron correlation. In view of the broken symmetry and the attosecond timescale on which the core-valence process must take place, we note the relevance of this work to pure electron-correlation-driven charge migration in ionized molecules as currently hunted for in experimental investigations.

Keywords: Electron-electron correlation, core-valence double ionization, symmetry breaking.

Acknowledgement: This work has been financially supported by the Swedish Research Council (VR, grant no. 2018-03731) and the Knut and Alice Wallenberg Foundation (grant no. 2017.0104), Sweden. We thank the Helmholtz Zentrum Berlin for the allocation of synchrotron radiation beam time. The research leading to these results has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement no. 730872. This work was carried out as part of M.H.'s Waernska Guest Professorship at the University of Gothenburg. The computations involved the Swedish National Infrastructure for Computing (SNIC) at the Chalmers Centre for Computational Science and Engineering (C3SE) partially funded by VR through grant no. 2018-05973. The authors also thank the Swedish National Infrastructure for Computing (SNIC 2022/3-34) at the National Supercomputer Centre of Linköping University (Sweden) partially funded by VR through grant no. 2018-05973.

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Novel Electronic Phenomena (Island)
TuI_05 - 11:00 AM - 11:30 AM

Abstract Not Available

X-ray Multiple Ionization and Nonlinear Spectroscopy (California)
TuII_05 - 11:00 AM - 11:30 AM

Abstract Not Available

Interface-induced Weak Ferromagnetism in Altermagnetic MnTe Thin Films

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Altermagnetic materials exhibit spin-compensated magnetic order like antiferromagnets while breaking time-reversal symmetry like ferromagnets [1]. MnTe, which has a NiAs-type structure, has been identified as an altermagnet with in-plane magnetic moments, as evidenced by nodal spin splitting observed in angle-resolved photoemission spectroscopy (ARPES) [2] and oscillatory x-ray magnetic circular dichroism (XMCD) signals [3]. In our previous study, we demonstrated that embedding 4 layers of MnTe into a topological insulator Bi_2Te_3 (BT) results in the formation of a $Mn_4Bi_2Te_7$/BT heterostructure, where the MnTe layers adjacent to the BT interface adopt a NaCl-type structure and the system exhibits out-of-plane ferromagnetism [4]. Building up on these findings, we consider embedding multilayer MnTe into the minimal unit of BT, a single quintuple layer (1QL- BT), which should allow the growth of MnTe thin films with surfaces and substrate interfaces terminated by Bi and Te atomic layers. However, the influence of such termination modifications on the altermagnetic properties of MnTe thin films remains unclear and requires investigation.

Therefore in this study, we fabricated MnTe thin films terminated with Bi and Te atomic layers both at the surface and the interface to the Si substrate and compared them with pure MnTe films directly grown on Si. Scanning transmission electron microscopy (STEM) revealed that while these films predominantly exhibit the NiAs-type structure, NaCl-type stacking appears near the Si interface, resembling our previously reported $Mn_4Bi_2Te_7$/BT heterostructure. XMCD measurements confirmed the coexistence of altermagnetic and out-of-plane ferromagnetic signals. ARPES measurements showed that upon cooling, the nodal splitting emerges in association with the altermagnetic transition. Furthermore, an exchange splitting at the Γ point, likely induced by ferromagnetism, was also observed by further cooling as shown in the Figure. These findings indicate that the structural change at the interface cants the magnetic moment to the out-of-plane direction, resulting in the coexistence of altermagnetism and ferromagnetism in this system.

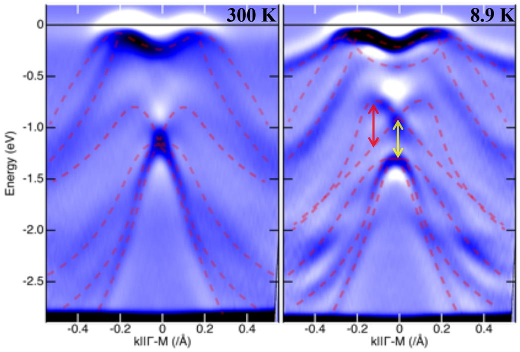

Fig.) Band structures of MnTe terminated with Bi and Te, measured along the Γ-M direction with $h\nu = 19$ eV. (Left) Paramagnetic state ($T = 300$ K). (Right) Coexistence of altermagnetic and ferromagnetic states ($T = 8.9$ K). Red and yellow arrows indicate altermagnetic and ferromagnetic band splitting, respectively.

[1] Phys. Rev. X **12**, 031042 (2022).   [2] Phys. Rev. Lett. **132**, 036702 (2024).
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Nonlinear resonant core-excitations in molecules.

Gilles Doumy[1], Eetu Pelimanni[1], Adam E. A. Fouda[1], Phay J. Ho[1], Thomas M. Baumann[2], Sergey I. Bokarev[3,4], Alberto De Fanis[2], Simon Dold[2], Gilbert Grell[5,4], Iyas Ismail[6], Dimitrios Koulentianos[1], Tommaso Mazza[2], Michael Meyer[2], Maria-Novella Piancastelli[6], Ralph Püttner[7], Daniel E. Rivas[2], Björn Senfftleben[2], Marc Simon[6], and Linda Young[1,8]

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The resonant interaction of x-rays with matter is central for exploiting site-selectivity in molecular systems. However, this interaction remains largely unexplored when subjected to intense x-ray pulses, which can trigger subsequent processes like stimulated emission and stimulated x-ray Raman scattering [41], as well as create induced resonances and multiple resonant excitations. We demonstrated experimentally that fully resonant sequential multi-core excitations to exotic neutral multi-core excited states ($K^{-1}K^{-1}V^{2}$) are feasible[1]. Notably, the resonant transition energies are only slightly affected by a preceding resonant excitation at a different atomic site in the molecule (~ 1-2 eV), in contrast to sequential ionization energies (≥10 eV).Thus, multiple resonances at different atomic sites of the same element can in fact be covered within the spectral content of a single XFEL pulse. We have experimentally confirmed the formation of double-core-excited molecule in the Nitrogen molecule at the SQS endstation of the EuXFEL, by comparing electronic decay spectra obtained at high and low x-ray intensity. The high intensity signal included distinct peaks corresponding to the unique decay channels where one or both of the core-excited electrons take an active role (so-called participator-spectator and participator-participator decays). The observed features are in very good agreement with calculated decay spectra obtained using a more realistic approach for continuum states involved in the processes.

Keywords [optional]: XFEL, nonlinear resonant excitation, Auger spectroscopy.

Acknowledgement [optional]: This work was supported by the US Department of Energy, Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division under award # DEAC02-06CH11357. We acknowledge European XFEL in Schenefeld, Germany, for the provision of X-ray free-electron laser beamtime at Scientific Instrument SQS (Small Quantum Systems).

[1]E. Pelimanni, A. E. A. Fouda, P.J. Ho, T.M. Baumann, S.I. Bokarev, A. de Fanis, S. Dold, G. Grell, I. Ismail, D. Koulentianos, T. Mazza, M. Meyer, M.-N. Piancastelli, R. Puettner, D. Rivas, B. Senfftleben, M. Simon, L. Young and G. Doumy '*Observation of molecular resonant double-core excitation driven by intense X-ray pulses*', Communication Physics, **7**, 341 (2024)

Non-relativistic spin splitting above and below the Fermi level in a g-wave altermagnet

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Nonrelativistic spin splitting (NRSS) challenges conventional wisdom about antiferromagnets by allowing spin-split electronic bands even in collinear orders with zero net magnetization. This sub-class of antiferromagnets, recently dubbed "altermagnets," enforces distinctive spin textures via spin-group symmetries in the crystal. However, direct experimental evidence for such symmetry-driven magnetism remains scarce. Here, we combine first-principles calculations, symmetry analysis, and two spin-resolved spectroscopies—angle-resolved photoemission (spin-ARPES) and our newly developed spin- and angle-resolved electron reflection spectroscopy (spin-ARRES)—to achieve the first complete momentum-resolved mapping of NRSS in $CoNb_4Se_8$. By probing both the occupied (spin-ARPES) and unoccupied (spin-ARRES) electronic states in a single experiment, we uncover a momentum-dependent spin splitting that switches sign under sixfold rotations and persists far above and below the Fermi level. Crucially, the observed collapse of NRSS near the Néel temperature confirms a genuine magnetic phase transition. Our work demonstrates, for the first time, the combined power of spin-ARPES and spin-ARRES in capturing the full spin texture across an extended energy range, positioning $CoNb_4Se_8$ as a prototype for exploring spin-group-based phenomena.

Acknowledgement: This work was primarily supported by the Laboratory Directed Research and Development Program of LBNL under the U.S. Department of Energy (DoE) Contract No. DE-AC02-05CH11231.

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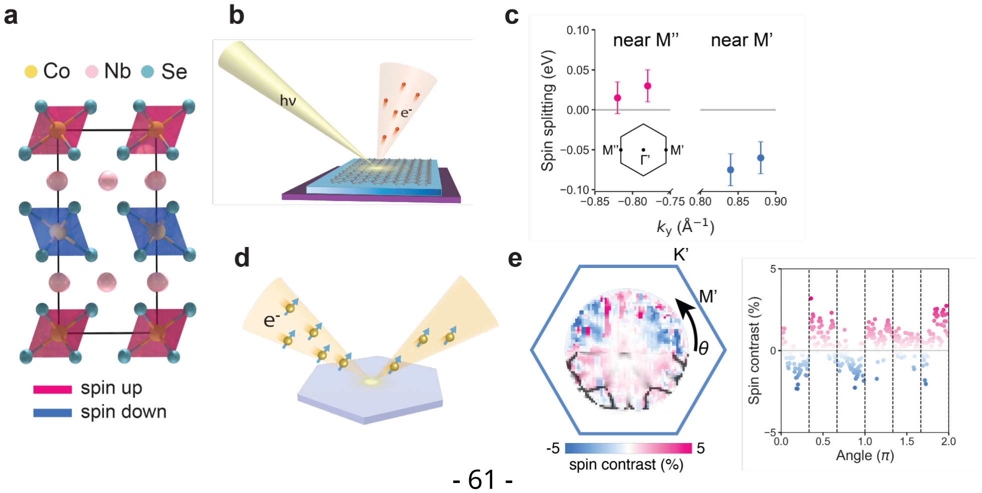
Figure 1. Direct Probes of NRSS in $CoNb_4Se_8$. (**a**) Magnetic structure of $CoNb_4Se_8$ along the b-axis. (**b,c**) spin-ARPES probes alternating spin-texture in occupied states. (**d,e**) spin-ARRES probes alternating spin-texture in unoccupied states



Probing Molecular Chirality with Nonlinear X-ray Spectroscopies

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Recent advancements in synchrotron and X-ray free-electron laser (XFEL) light sources have opened new avenues for exploring molecular chirality through element-sensitive nonlinear spectroscopies. While traditional techniques like circular dichroism (CD) and sum- and difference-frequency generation (SFG/DFG) have been widely used in the optical regime, extending them to the X-ray domain provides deeper insights into local structural asymmetries by exploiting the element specificity of core orbitals. In this work, we simulate X-ray absorption spectroscopy (XAS), X-ray circular dichroism (XCD), and nonlinear optical/X-ray SFG and DFG (OX SFG/DFG) signals for two prototypical chiral molecules, fenchone and cysteine. Our multi-reference simulations successfully reproduce available experimental data and highlight how novel X-ray spectroscopies enhance sensitivity to molecular asymmetry. We find that XCD spectra exhibit strong asymmetries at chiral centers, while contributions from distant atoms are significantly weaker. Additionally, OX SFG/DFG signals, under fixed resonant optical excitation, show a strong dependence on core transitions and valence excitation energies, enabling the introduction of a two-dimensional chirality-sensitive valence-core spectroscopy. This approach provides new insights into interplay between valence orbitals and local molecular asymmetry. Finally, we estimate the feasibility of such non-linear experiments at XFELs using realistic laser and X-ray pulse parameters. Our results suggest that nonlinear X-ray spectroscopies represent a promising tool for probing the geometric and electronic structures of chiral molecules with unprecedented sensitivity.

Keywords: Chirality, Nonlinear spectroscopy, Fenchone and Cysteine.

Acknowledgement: This work was supported by the U.S Department of Energy (DOE), office of Science, Basic Energy Science (BES), Chemical Sciences, Geosciences and Biosciences Division (CSGB) under Contract No. DE-FOA-0003176 for the study on linear signals (XAS, XCD) and Contract No. DE-AC02-06CH11357 for the study on nonlinear signals (SFG/DFG). MC acknowledges support of the European Research Council via the Advanced Grant CHIRAX (ID 101095012).

[1]Y. Nam, M. Chergui, L. Young, and J. R. Rouxel, "Linear and nonlinear X-ray spectra of chiral molecules: X-ray Circular Dichroism, Sum and Difference-Frequency Generation of fenchone and cysteine", arXiv:2501.14671 [physics.chem-ph]

Exploring Approaches to Probe Full Electronic Wave Functions in Solids via Photoemission

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Access to the full momentum-dependent electronic wave function of solids enables access to the quantum geometric tensor, imaginary part of which is the Berry curvature. In principle, circular-dichroic (CD) and spin-polarized angle-resolved photoemission (spin-ARPES) allow access to these wave functions, however, it is indirect. In this work, we discuss strategies on how to extract the initial state wave functions from the photoemission spectra.

On the example of graphene, we will discuss various contributions to CD-ARPES [1,2] which include phase shifts of the participating partial waves [3], the interatomic phase shifts [4], and the CD due to elastic scattering of an excited electron [5]. Subsequently, we perform similar analysis for WSe_2, a material where orbital characters are relatively well-defined.

Finally, a simple interatomic interference model that qualitatively explains asymmetric spin-ARPES spin texture from WTe_2 single crystal surface [4] is presented.

Further examples on $PtTe_2$ [6] and selected R166 Kagome compounds will be presented if time allows.

This study aims to explore how CD-ARPES and spin-ARPES techniques can enhance the understanding of quantum materials.

Keywords: Berry curvature, spin-polarized ARPES, circular-dichroic ARPES, micro-ARPES.

Acknowledgements: I gratefully acknowledge the insightful collaborations with J. Minar, J. Schusser, and J. Henk on one-step model. I would like to thank G. Bihlmayer and Y. Mokrousov for collaboration on density-functional-theory methods. I am thankful for the support received at the MAESTRO beamline at ALS, the NanoESCA beamline at Elettra, the PHELIX beamline at Solaris, and other synchrotron beamlines. I acknowledge the support and contributions of my group in Jülich.

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[2] Krüger and Matsui, JESRP 258, 147219 (2022)
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The Dirac Semimetal RuO_2 - From Topology to Functionality

Simon Moser[1,2], Philipp Keßler[1,2], Patrick Härtl[1,2], Vedran Jovic[3], Andreas Feuerpfeil[2,4], Hendrik Hohmann[2,4], Matteo Dürrnagel[2,4], Armando Consiglio[2,4], Jonas Erhardt[1,2], Michael Sing[1,2], Ronny Thomale[2,4], Matthias Bode[1,2], Ralph Claessen[1,2]

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We will discuss the synthesis, electronic properties, and magnetic structure of thin-film RuO_2, a Dirac semimetal[1] and a leading catalyst for oxygen evolution in electrochemical water splitting.[2,3] While recent claims of altermagnetism[4,5] are currently challenged,[6,7] the debate has rekindled interest in electron correlation phenomena in RuO_2. Angle-resolved photoemission spectroscopy (ARPES) reveals strong Fermi surface nesting, suggesting a propensity for electron correlations. At the RuO_2 (110) surface, these correlations appear to promote a charge density wave instability that can be directly probed via scanning tunneling microscopy (STM).

Keywords: ARPES, STM, RuO_2, Flat Surface State, Altermagnetism, Charge Density Wave.

Acknowledgement: Work supported by the German Research Foundation under Germany's Excellence Strategy through the Würzburg-Dresden Cluster of Excellence on Complexity and Topology in Quantum Matter ct.qmat (EXC 2147, Project 390858490) and through the Collaborative Research Center SFB 1170 ToCoTronics (Project No. 258499086), as well as from the New Zealand Ministry of Business, Innovation and Employment (MBIE, Grant C05X2004)

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Spin and Orbital Textures (Island)
TuI_10 - 02:30 PM - 02:50 PM

Observing the electronic response of a Mott insulator at a current-induced insulator-to-metal transition with transport-ARPES

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The quasi-two-dimensional Mott insulator Ca_2RuO_4 exhibits a convergence of spin-orbit coupling and electron correlations that leads to exciting quantum phenomena, including a rare insulator-to-metal transition (IMT) induced by a DC current [1]. While structural changes have been tracked by diffraction [2, 3], the associated electronic changes had not been observed. Here we report angle-resolved photoemission spectroscopy (ARPES) results under DC current, which show a substantial reduction of the Mott gap, along with a change in the Ru t_{2g} band dispersion [4]. We have also captured the high temperature Fermi surface, whose spectral features are unique from the current-induced metallic Fermi surface. In conjunction with a free energy analysis, our results demonstrate that the current-induced phase, albeit thermodynamically equivalent, is electronically distinct from the high-temperature zero-current metallic phase.

Combining ARPES with transport, i.e. transport-ARPES, has been rare given the complexity of disassociating real field- or current-driven physics from the effect of stray electric and magnetic fields on the outgoing photoelectron trajectory. By taking advantage of the micron-sized beam spot at the MAESTRO beamline (7.0.2) at the Advanced Light Source and careful core level spectrum analysis, we show that transport-ARPES can be extended to the study of any ARPES-suitable material.

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Dispersion of Spin Stripe Fluctuations in FeGd Measured by Megahertz X-ray Photon Correlation Spectroscopy

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Spontaneous fluctuations play a critical role in understanding phase transitions, yet their dispersion remains largely unexplored – unlike well-characterized quasiparticle excitations such as magnons and phonons, which are routinely studied using inelastic neutron or x-ray scattering. In this work, we investigate the spontaneous fluctuations of spin stripes in a FeGd magnetic superlattice (Fig. 1) using X-ray Photon Correlation Spectroscopy (XPCS) at a megahertz (MHz) repetition-rate x-ray free-electron laser. Enabled by the significant enhancement in counting statistics at MHz rates, we demonstrate the ability to resolve the dispersion of spin stripe dynamics across at least one Brillouin zone. Our results establish MHz-XPCS as a powerful and unique tool for probing spontaneous fluctuations and the emergent phenomena they drive in complex materials.

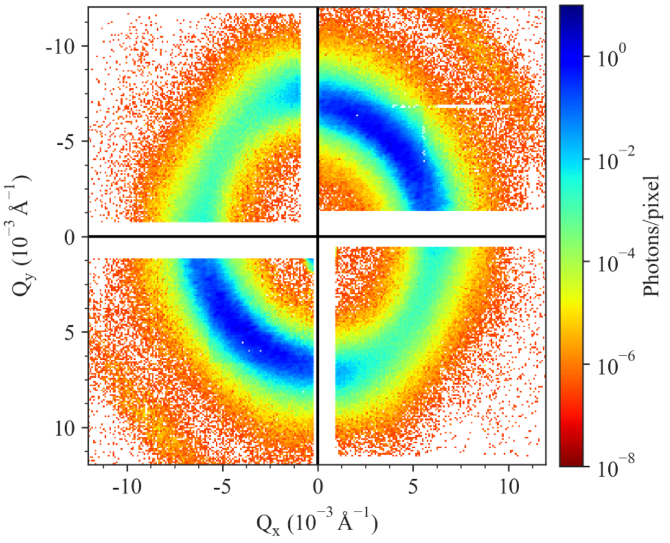

Figure 1. Resonant X-ray diffraction profile of the spin stripes in FeGd.

Spin and Orbital Textures (Island)
TuI_11 - 02:50 PM - 03:10 PM

The puzzling case of Pt(111): Spin texture of the prototypical *L*-gap surface state

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This year, we celebrate the 50th anniversary of the first experimental detection of a metallic surface state [1]: the prototypical Shockley-type *L*-gap surface state on Cu(111). Already in the very first paper by Shockley on the occurrence of surface states associated with a periodic potential [2], it became clear that the energetic position of such states depends critically on the interatomic distance, i.e., on the lattice constant. Detailed studies on various other fcc(111) surfaces revealed, among other things, the influence of spin-dependent interactions, namely exchange and spin-orbit interaction. While the surface state becomes exchange-split for Ni, contributing to the surface magnetic moment [3], the spin degeneracy is lifted by the Rashba effect for the heavy element Au, leading to a **k**-dependent spin splitting [4].

The equivalent surface state on Pt(111) had been slighted so far. Only one scanning-tunneling-spectroscopy study reported on the energy vs momentum dispersion [5]. Theoretical predictions based on DFT vary depending on the particular functionals used. To clarify this issue, we investigated the atomic structure of Pt(111) by low-energy electron diffraction and the unoccupied electronic structure by spin- and angle-resolved inverse photoemission [6]. The experimental results are backed by theoretical studies using different functionals, which show that the characteristics of the surface band depend critically on the lattice constant. We found that the surface resonance at Pt(111) is a showcase for a delicate interplay of atomic structure, hybridization with *d* bands, and the influence of spin-orbit interaction.

Keywords: Two-dimensional electron states, surface states, exchange interaction, spin-orbit interaction, spin-resolved (inverse) photoemission.

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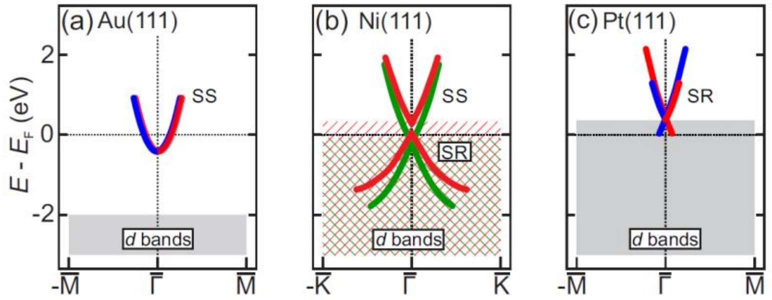

Figure 1. Simplified sketches of the *L*-gap surface states and *d* bands at (a) Au(111), (b) Ni(111), and (c) Pt(111).

Spin and Magnetism (California)
TuII_11 - 02:50 PM - 03:10 PM

Depth-Resolved Profile of the Interfacial Ferromagnetism in $CaMnO_3$/$CaRuO_3$ Superlattices

Jay R. Paudel[1], Aria Mansouri Tehrani[2], Michael Terilli[3], Mikhail Kareev[3], Joseph Grassi[1], Raj K. Sah[1], Liang Wu[3], Vladimir N. Strocov[4], Christoph Klewe[5], Padraic Shafer[5], Jak Chakhalian[3], Nicola A. Spaldin[2], and Alexander X. Gray[1]

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Interfacial magnetism in complex oxides presents a rich platform for tuning electronic and magnetic properties, with implications for spintronic and data storage applications. In this study, we investigate the emergence of interfacial ferromagnetism in oxide superlattices containing antiferromagnetic $CaMnO_3$ and paramagnetic $CaRuO_3$ using a combination of synchrotron-based X-ray spectroscopy and X-ray Resonant Magnetic Reflectivity (XRMR) measurements to uncover the depth profile of the interfacial magnetism. Our findings reveal that the emergent interface ferromagnetism exhibits an asymmetric distribution between the top and bottom interfaces and may extend beyond the immediate interface into the nominally antiferromagnetic $CaMnO_3$ layers, suggesting more complex interface interactions than previously assumed.

To understand the microscopic origin of this behavior, we employed density functional theory calculations, which indicate that the observed interfacial ferromagnetism is driven by a double exchange mechanism facilitated by charge transfer from Ru to Mn ions. Furthermore, defect chemistry, such as the presence of oxygen vacancies, may influence the local magnetic moments at the interface and contribute to the asymmetry in the ferromagnetic depth profile.

These results highlight the role of charge transfer and defect engineering in tailoring emergent interfacial properties. By providing a deeper understanding of how interfacial magnetism can be controlled at the atomic level, this work offers new strategies for designing functional oxide heterostructures with tunable magnetic properties [1].

[1] *J. Paudel* et al., Nano Lett. **24** (48), 15195-15201 (2024).

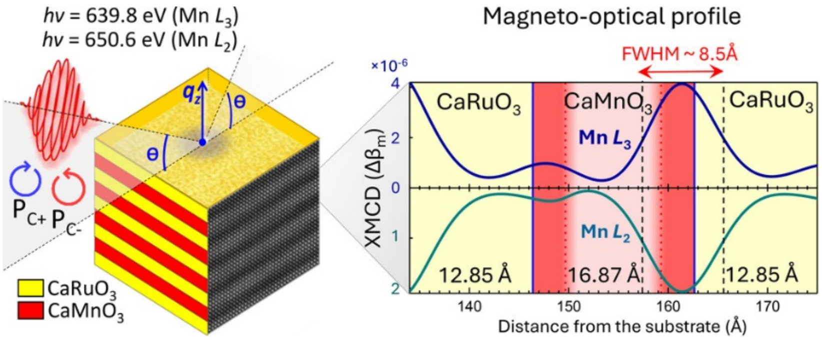

Figure 1. XRMR measurement geometry of the $CaMnO_3$/$CaRuO_3$ superlattices (left) and the derived magnetic depth profile of a single $CaMnO_3$ layer sandwiched between $CaRuO_3$ (right).

Assessing nontrivial topology in Weyl semimetals by dichroic photoemission

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Dichroic photoemission as a tool to identify topological materials and access initial state information By performing angle-resolved photoemission spectroscopy (ARPES) and state-of-the-art SPR-KKR photoemission calculations on the paradigmatic Weyl semimetals Ta(As,P) [1] we show the spectroscopic manifestation of topological features and Weyl physics beyond the simple photointensity over a broad range of excitation energies from the vacuum ultraviolet to the soft X-Ray regime and compare the surface to the bulk band structure [2]. We further show the drawbacks of the existing spectroscopic techniques used to determine whether the given material has non-zero Chern number and discuss an improved approach for identifying Fermi arcs using differential ARPES measurements, their relation to orbital angular momentum (OAM) as well as the proper final state description. Consequently, a more realistic description of the final state is needed to explain dichroism by modeling the photoemission matrix element.

Keywords: photoemission, ARPES, Topology, Weyl, SPR-KKR

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Spin-flip Determination in CrX_3 (X = Cl, Br, and I) 2D Magnets using High-resolution X-ray Scattering

Chamini Pathiraja[1], Jayajeewana N. Ranhili[1], Deniz Wong[2], Christian Schulz[2], Yi-De Chuang[3], Yu-Cheng Shao[4], Di-Jing Huang[4], Hsiao-Yu Huang[4], Amol Singh[4], Byron Freelon[1]

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Abstract:

Chromium tri-halides CrX_3 (X = Cl, Br, and I) exhibit low-temperature, layer-dependent magnetism that can be manipulated by an electric field, which makes them important candidates for spintronics applications. Their magnetic ground states depend keenly on electronic parameters such as spin-orbit coupling (SOC), Hund's coupling (J_H), $p-d$ covalency, and interorbital Coulomb interactions. Therefore, accurately determining these parameters is paramount for understanding the CrX_3 physics. We have used resonant inelastic x-ray scattering (RIXS) spectroscopy facilitated by ligand field multiplet calculations in C_3 symmetry to simulate experimental RIXS spectra. Tanabe-Sugano-like energy level diagram calculations facilitated the determination of detailed electronic structure parameters in CrX_3. These methods provide the most detailed description of CrX_3 magneto-optical and electronic energetic (terms) to date. The determined $10Dq$ values are in good agreement with the spectrochemical series, the Racah B parameter follows the expected Nephelauxetic effect, and the crystal field distortion parameters $D\sigma$ and $D\tau$ are calculated for the first time. Moreover, high-resolution RIXS spectra reveal a clear energy separation between spin-allowed quartet and spin-forbidden doublet states in CrX_3, showcasing the potential of this technique in determining materials' electronic parameters. This study validates the role of SOC in Cr *2p* spin-flip excitations. Such precise measurements offer insights into the energy design of spintronic devices that utilize quantum state tuning and the effect of halides in determining spin-flip excitation energies in 2D magnetss.

Tridimensional magnetism in Superconducting Infinite-Layer $PrNiO_2$ studied with Resonant Inelastic X-ray Scattering

F. Rosa,[1] N.B. Brookes[2], D. Di Castro[3], G. Merzoni[1,4], D. Preziosi[5], H. Sahib[5], M. Salluzzo[6] and G. Ghiringhelli[1]

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Infinite-layer (IL) nickelates have been reported as superconductive in 2019, coming to the forefront as one of the most promising class of cuprate-akin compounds[1,2]: the general interest in their electronic structure has pointed out both analogies and differences with copper oxides[3,4]. In this context, particular attention has been addressed to spin order: recent RIXS works[5,6,7] have reported the presence of magnetic excitations both in hole-doped and undoped compounds, with the former showing a much larger damping of the (para)magnon peak due to progressive hindering of magnetic correlations by the doping holes. Because p-doping is, at present, a necessary ingredient to obtain superconductivity in these compounds, this has been interpreted as a sign of intrinsic competition between magnetism and superconductivity. In this work, we question this interpretation by taking advantage of the recent report of superconductivity caused by self-doping in nominally undoped $PrNiO_2$ (PNO)[8]. We performed extensive RIXS measurements on the very same PNO samples at a temperature close to the SC onset, exploiting both the momentum and polarization resolution provided by the technique. Our results shown a rather sharp magnon dispersion over the whole Brillouin zone. The presence of a gap at the Γ point revealed an interlayer exchange coupling between the NiO_2 planes, showing the inadequacy the bidimensional Linear Spin Wave models previously employed in literature[3,5,7]. We fitted the dispersion with a tridimensional model explicitly including such coupling, and compared the result to the Infinite-Layer cuprate $CaCuO_2$. An exchange coupling of few *meV* in the direction orthogonal to the planes was then retrieved for both compounds. Our results have a double valence: (1) they hint at the possible coexistence between superconductivity and magnetism in PNO, and (2) they demonstrate the tridimensional nature of the latter, providing an estimate of the interlayer exchange coupling.

Keywords: magnetism, nickelates, RIXS, cuprate

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[9] Peng, Y. Y., et al., *Nature Physics* 13.12 (2017): 1201-1206.

Proximity-Induced Long-Range Magnetism in Ultra-High Conductivity $PdCoO_2$

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Emergent magnetism is observed in the ultra-high conductivity delafossite $PdCoO_2$ with the introduction of strain via He implantation. The magnetization can be continuously tuned from paramagnetism to long-range ferromagnetism with increasing He dosing levels and is completely reversible with annealing. Electronically, $PdCoO_2$ is very two-dimensional, consisting of ultra-conductive Pd layers separated by non-interacting, insulating CoO_2 layers. We performed a series of x-ray spectroscopy techniques to investigate the interplay between the local Co moments in the insulating CoO_2 layers and itinerant Pd electrons. We find that as a result of the local strain induced by the He implantation, the Co atoms transition from a low-spin state to a high-spin state. The Pd, which due to the high density of states near the Fermi level is near the Stoner criterion, couples to the Co magnetic moment resulting in long-range ferromagnetic order. Using resonant angle-resolved photoemission spectroscopy (ARPES), we observe that the Pd-derived Fermi surface remains unchanged across a range of dosing levels, confirming that the Pd itinerant electrons mediate the long-range order of the local Co moments.

Keywords: Resonant photoemission, APRES, XMCD, two-dimensional, materials discovery.

Acknowledgement: Work supported by the Agency for the Advancement of Generously Funded Science (AAGFS).

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Electronic Structure of $La_xSr_{1-x}FeO_{3-\delta}$ Probed by X-ray Absorption and Resonant Inelastic Soft X-ray Scattering

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For electrochemical water splitting, the oxygen evolution reaction (OER) is considered the efficiency-limiting factor. Therefore, the development of efficient catalytic materials for the OER has been a major research focus in recent years, with $La_xSr_{(1-x)}FeO_{(3-\delta)}$ (LSFO) being a promising candidate. In this material, with the perovskite (ABO_3) crystal structure, the oxidation state of Fe can be "tuned" by changing the La^{3+} to Sr^{2+} ratio. Increasing the Sr content (while keeping the oxygen content constant) increases the nominal amount of Fe^{4+} present in the material and increases the electrocatalytic activity.

We present a detailed study of the electronic structure of ozone-treated LSFO with different La/Sr ratios, by employing Fe $L_{2,3}$ X-ray Absorption Spectroscopy (XAS), collected at the X-SPEC beamline at the KIT Light Source, and O K-edge Resonant Inelastic X-ray Scattering (RIXS) data, measured with our SALSA end station at Beamline 8.0.1 of the ALS. The spectra are analyzed with the help of Bethe-Salpeter Equation (BSE) calculations using the OCEAN code, painting a detailed picture of the electronic structure of LSFO as a function of the La/Sr ratio. In our contribution, we will show that the covalency between Fe and O increases with increasing Sr content and will discuss the applicability of the oxidation state concept for this materials class.

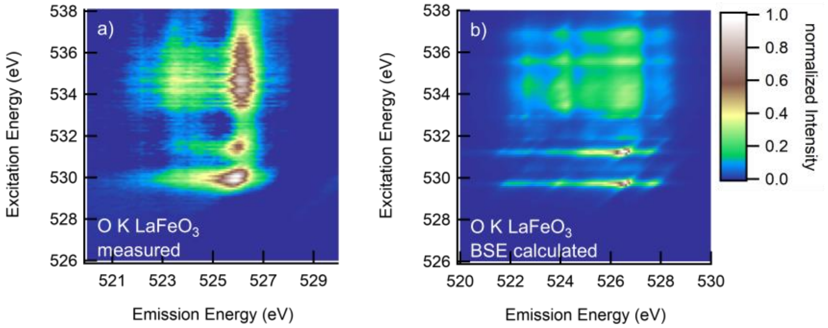

Figure 1. a) Coherent fraction of the experimental O K RIXS map and b) BSE calculated RIXS map of $LaFeO_3$. The BSE-calculated map captures all features of the experiment and gives insights in the electronic structure of the investigated material.

Dispersive Hund’s Excitons in Magnetic van der Waals Materials

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Magnetic van der Waals (vdW) materials have emerged as a fertile platform for exploring novel many-body phenomena. Among these are Hund’s excitons, an unusual class of excitons bound by Hund’s exchange interactions. Using state-of-the-art resonant inelastic x-ray scattering (RIXS), we discovered Hund’s excitons in both antiferromagnetic $NiPS_3$[1] and ferromagnetic CrI_3[2]— two topical magnetic vdW materials. Supported by exact diagonalization calculations, we identify these excitons’ electronic character with similar spin-flip transitions in nature. By examining their dispersion, we uncover a close relationship between Hund’s exciton propagation and low-energy magnetic excitations. Our research demonstrates RIXS as a powerful tool in studying exciton physics in magnetic vdW materials and open new avenues for magneto-optical applications.

Keywords [optional]: Magnetic van der Waals materials, exciton, electronic structure, resonant inelastic x-ray scattering.

Acknowledgement [optional]: Work performed at Brookhaven National Laboratory was supported by the U.S. Department of Energy (DOE), Division of Materials Science, under Contract No. DE-SC0012704.

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Oxygen Redox Mechanisms in Commercial Lithium-Ion Battery Cathodes Revealed through RIXS and sXAS

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Lithium-ion batteries are the cornerstone of modern energy storage, powering mobile devices and electric vehicles. However, their theoretical energy density remains underutilized due to the instability of cathode materials at high states of charge. While oxygen redox has been suspected to play a role in this instability, indirect evidence has upheld the dominance of the transition metal-based redox paradigm. In this study, we uncover direct evidence of oxygen involvement in the redox process, identifying distinct oxygen species through resonant inelastic X-ray spectroscopy (RIXS).[1] Complementary insights from soft X-ray absorption spectroscopy (sXAS) further demonstrate oxygen's role in charge compensation. Additionally, we investigate the stability of oxygen redox as influenced by cathode chemical composition, revealing the detrimental impact of unstably oxidized lattice oxygen at high states of charge.[2] Importantly, we show that dopants can mitigate these effects, stabilizing oxygen redox and enhancing cathode performance. This work lays the foundation for developing high-energy-density cathodes that maximize specific capacity while addressing degradation challenges, paving the way for next-generation energy storage solutions.

Keywords: Lithium-ion battery, resonant inelastic X-ray scattering, X-ray absorption, redox mechanism, materials discovery.

Acknowledgement: Work supported by the LG Energy Solution, National Research Foundation of Korea. Soft X-ray experiments are conducted at beamline 8.0.1 at Advanced Light Source, Lawrence Berkeley National Laboratory, a DOE Office of Science User Facility, under contract no. DE-AC02-05CH11231.

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Van Hove Singularities Induced Large Electron-boson Coupling in $Cr_2Ge_2Te_6$

Yucheng Guo[1,2], Ziqin Yue[1], Yichen Zhang[1], Sae Hee Ryu[2], Byoungki Choi[2], Han Wu[1], Lebing Chen[1], Sohrab Ismail-Beigi[3], Aaron Bostwick[2], Chris Jozwiak[2], Eli Rotenberg[2], Pengcheng Dai[1], Junichiro Kono[1], Ming Yi[1*]

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Van Hove singularities (VHSs) in the electronic structure of 2D materials produce a divergent density of states that is prone to electronic instabilities when tuned to the Fermi level, and therefore often associated with a precursor of new ordered phases. However, despite the previous fruitful studies on this topic, it is still challenging to understand the effects of VHSs and directly compare them to theoretical predictions, often due to a mixing of VHSs with other coexisting bands as well as their existence in 3D bulk crystals. Here, we have investigated a clean single-band system with isolated VHSs in a ferromagnetic semiconducting material $Cr_2Ge_2Te_6$ (CGT). By surface alkali metal doping, we were able to tune the VHS to near the Fermi level, hence creating a genuine 2D system suitable for investigating VHS-induced novel states and their interplay with magnetism. In this talk, we will present our angle-resolved photoemission spectroscopy measurement via a step-by-step in situ potassium doping experiments on pristine CGT crystals, tuning the Fermi level to go across the VHS level and beyond, and discuss our observations of electron-electron correlations and electron-boson coupling effects.

Keywords: 2D magnet, alkali metal doping, ARPES, electron-boson coupling.

Abstract: There are various types of optical spectra to characterize and to understand materials. Interpretations have naturally been made based on linear responses of the light-matter interaction, where amounts of the signal are proportional to the incident intensity. However, there are also varieties of the optical phenomena in nature that show the nonlinear dependence. Moreover, they are associated with events, such as second harmonic generation, that are unusual in everyday life but critical for our scientific development. These special phenomena are typically induced with the ultrashort pulse light source, i.e. laser, and the recent technical innovations have pushed the controllable photon energy up to the X-ray region. "X-ray" was named after a ray of "something unknown (X)" by Röntgen and it has been a significant experimental probe to investigate structure and electronic states of materials. Today, we have gotten a new approach and we are able to conduct the nonlinear X-ray experiments with X-ray lasers. In this talk, I would like to present such a rise of the nonlinear X-ray spectroscopy for materials science and discuss the future prospects of the new technique.

Short Bio:

2001: Ph-D (Science), the University of Tokyo, Japan

2001: Postdoctoral researcher, the University of Zuerich, Switzerland

2001: Research Associate, School of Science, the University of Tokyo, Japan

2006: Associate Professor, the Institute for Solid State Physics, the University of Tokyo, Japan

2021:Professor, the Institute for Solid State Physics, the University of Tokyo, Japan

Plenary Talks (Island)
WeI_02 - 09:15 AM - 10:00 AM

Abstract Not Available

Surfaces and Interfaces (Island)
WeI_04 - 10:30 AM - 11:00 AM

Abstract Not Available

Attosecond and Femtosecond Molecular Dynamics (California)
WeII_04 - 10:30 AM - 11:00 AM

Abstract Not Available

Surfaces and Interfaces (Island)
WeI_05 - 11:00 AM - 11:30 AM

Abstract Not Available

Discovery of New Excited State Pathways in Transition Metal Carbonyls using Ultrafast Core-Level Spectroscopy

Krupa Ramasesha
Combustion Research Facility, Sandia National Laboratories, Livermore, CA, USA

The coupled electronic and structural dynamics on excited states are fundamental to the photoinduced chemical transformations that underpin processes such as photocatalysis and photovoltaic activity. Of particular interest is the propensity for molecules to undergo ultrafast intersystem crossing, which leads to the formation of triplet species that can significantly influence reaction outcomes. In this talk, I will present our recent joint experimental and theoretical investigation[1] of excited state dynamics in a prototypical photocatalyst, iron pentacarbonyl [$Fe(CO)_5$], whose photochemistry has garnered much interest for decades. By employing ultrafast extreme ultraviolet transient absorption spectroscopy near the Fe $M_{2,3}$-edge (3p-to-valence transitions), our work has spectroscopically detected the fleeting metal-centered excited states responsible for the dissociation of CO ligands after 266 nm excitation. Importantly, our study has also revealed participation of triplet states in mediating CO ligand elimination, with spectral signatures of these triplet states appearing on sub-100 femtosecond time scales. This finding suggests ultrafast intersystem crossing to the triplet manifold despite modest spin-orbit coupling and thus challenges the singlet-only excited state pathways proposed previously for this molecule in the gas phase.

Keywords: transition metal, intersystem crossing, ultrafast spectroscopy, core level spectroscopy, photocatalysis.

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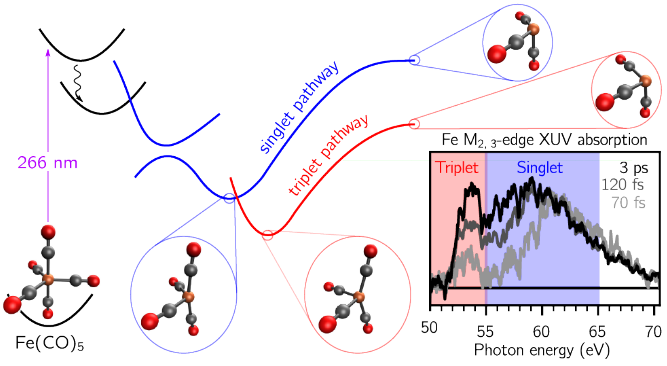

Figure 1. Ultrafast extreme ultraviolet transient absorption spectroscopy of $Fe(CO)_5$ has spectroscopically revealed the involvement of triplet states during UV-induced ligand elimination.

Surfaces and Interfaces (Island)
WeI_06 - 11:30 AM - 11:50 AM

Low Secondary Electron Emission 2D Materials for Space Electronics Applications

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In the field of electronics for space, especially regarding communication, earth observation, navigation and electronic intelligence satellites, RF passive devices such as circulators, filters, and isolators are being revolutionized by innovative materials and advanced surface physics concepts. These advancements not only raise the multipactor threshold but also provide new insights into spectroscopy and material structure. This study presents recent progress in the development of emerging low Secondary Electron Emission Yield (SEY) coatings and 2D materials with unique electronic and thermal properties. Novel methods for creating nano-microstructured silver coatings for aluminum alloys and dielectrics have been explored, achieving a maximum SEY of less than 1.. The unexpected behavior of SEY on coatings combining conductive (silver) and dielectric (alumina, Kapton, and PEEK) surface regions is also investigated. To assess SEY as a function of primary energy, both pulsed and continuous measurement techniques were utilized. The dynamic evolution of SEY in similar samples at a fixed primary electron energy was also examined, proposing a charging-roughness coupled model to simulate the experimental results.

Keywords: SEY, 2D materials, RF breakdown, electroless treatments.

Acknowledgements: This work was supported by the Ministry of Science and Innovation of Spain, MCIN/AEI/10.13039/501100011033 through the Spanish Project PID2021-122761OB-I00 and the ESA (European Space Agency).

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[2]I Montero, L Olano, L Aguilera, ME Dávila, U Wochner, D Raboso, Journal of Electron Spectroscopy and Related Phenomena 241, 146822, 2020.

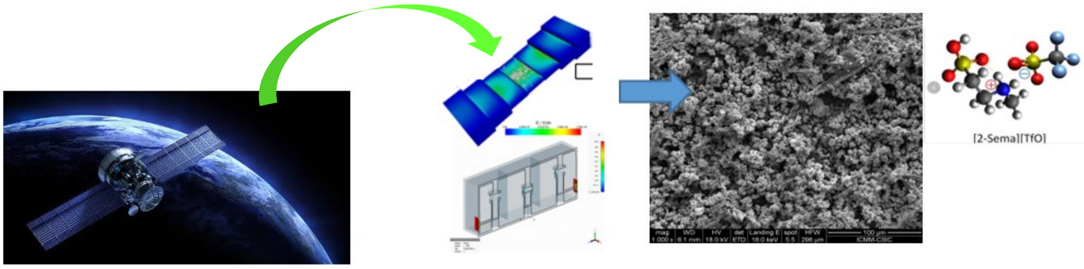

Figure 1. Electroless silver plating for GNSS systems: (left) satellite in orbit, (middle) RF filter highlighting regions susceptible to multipactor that need protection, (right) scanning electron microscopy (SEM) image of the cross-section of an electroless silver coating on Al and the molecular structure of ionic liquids

Probing H_2^+ Formation in Strong-Field Ionization of Water Using Pump-Probe Spectroscopy and Covariance Mapping

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Understanding the ultrafast dynamics governing molecular fragmentation under intense laser fields is crucial for advancing strong-field physics and attosecond science. In this work, we investigate the formation of H_2^+ from water (H_2O) during strong-field ionization using a femtosecond pump-probe technique combined with covariance mapping analysis. A near-infrared femtosecond laser pulse initiates ionization and dissociation, while a delayed probe pulse further ionizes the evolving molecular fragments. By employing covariance analysis of electron-ion momentum distributions, we resolve ionization pathways leading to H_2^+ formation. Our results provide insights into the ultrafast proton migration and charge redistribution mechanisms that facilitate molecular hydrogen cation production.

Keywords: strong field, covariance, proton migration

Acknowledgement: This research is supported by the National Science Foundation.

Surfaces and Interfaces (Island)
WeI_07 - 11:50 AM - 12:10 PM

Soft X-ray Spectroscopy as a Neat Tool to Assign Electronic Structures of {FeNO} Complexes

Yang Ha[1], Hua-Fen Tsu[2], Hrant P. Hratchian[3], Marcetta Y. Darensbourg[4], Polly Arnold[1,5], Jinghua Guo[1]

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{FeNO} complexes are a family of compounds that contain both iron (Fe) and nitric oxide (NO) as active sites, and are largely used as models to study O2 and NO metabolism. The detailed electronic structures of the {FeNO} complexes are always controversial, due to the lack of robust tools for characterization. In this work, N K-edge and Fe L-edge XAS are combined to unambiguously assign the charge and spin state of both Fe and NO in a pair of {FeNO}6 and {FeNO}7 compounds, and showed that the one electron redox occurred on the NO side. At beamline 8.0.1 at Advanced Light Source (ALS), the technique can also be applied to other systems under different conditions.

Keywords [optional]: Iron Nitrosyl compounds, X-ray absorption spectroscopy, electronic structures, charge and spin density, density functional theory.

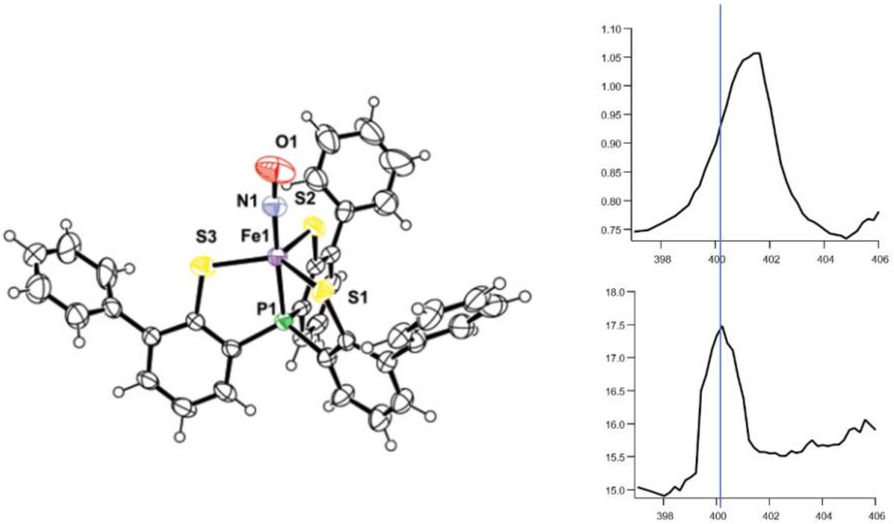

Figure 1. Structure of an {FeNO} complex and the corresponding XAS data.

Intramolecular particle exchange in the fragmentation of methanol upon valence photo double ionization

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V. Davis[2], C. Bagdia[3], N. Iwamoto[3], T. Severt[3], J. B. Williams[2], I. Ben-Itzhak[3],
R. R. Lucchese[1], D. S. Slaughter[1], and Th. Weber[1]

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We investigated the valence photo double ionization (54eV) of two isotopologues of methanol molecules (CH_3OD and CD_3OH), by measuring the final 3D momenta of 2 electrons and 2 cation fragments in coincidence, using the COLTRIMS (COLd Target Recoil Ion Momentum Spectroscopy) technique at the Advanced Light Source. Here we report on the comparison of two specific 3-body fragmentation channels that show a clear signature of intramolecular particle exchange, which we trace via deuterium scrambling in the two different isotopologues of methanol:

(a) $\mathrm{CH_3OD} + \gamma\,(54\text{ eV}) \rightarrow (\mathrm{CH_3OD})^{2+} + 2e^- \rightarrow \mathrm{CHD^+} + \mathrm{OH^+} + \mathrm{H} + 2e^-$
(b) $\mathrm{CD_3OH} + \gamma\,(54\text{ eV}) \rightarrow (\mathrm{CD_3OH})^{2+} + 2e^- \rightarrow \mathrm{CHD^+} + \mathrm{OD^+} + \mathrm{D} + 2e^-$

For both reactions, the momentum of the neutral fragment is determined via momentum conservation. The goals of our investigation are to track and understand state-selectively the influence of nuclear dynamics on intramolecular particle exchange and roaming during the dissociation process. This is achieved through the analysis of the relative branching ratios with respect to the most intense 2-body breakup channels, $CH_3^+ + OD^+$ or $CD_3^+ + OH^+$, respectively. Moreover, electron-ion energy correlation maps are used to identify and isolate the participating electronic states along the C-O internuclear distance of the dication potential energy surfaces, which we calculated using the Multi-Reference Configuration Interaction (MRCI) method. In addition, by exploiting the measured electron energy sharing and the electron angular distributions, we are able to distinguish between contributions from autoionization and direct double ionization. Employing this differential insight, we interpret observables like the pairwise ionic and neutral fragment momentum sharing and Newton plots, and apply the native frames analysis, in order to retrace the fragmentation processes and identify the intermediates of the 3-body breakups (a) and (b).

Keywords: Methanol, Native frames, COLTRIMS, molecular dynamics, valence photo double ionization, roaming, particle exchange.

Acknowledgement: This research is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences and Biosciences under contract no. DE-AC02-05CH11231 (LBNL) and No. DE-FG02-86ER13491 (JRML at KSU) as well as the National Sciences Foundation under Award No. NSR-1807017 and NSF-2208017 (UNR). This research used resources of the Advanced Light Source (ALS) and the National Energy Research Scientific Computing Center (NERSC) Award Nos. BES-ERCAP-0020143 (theory), both being DOE Office of Science user facilities.

Operando Spectroscopic View for Thin Film Gas Sensors

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Chemical reactions at surfaces have been widely used for chemical processes such as catalytic synthesis, energy conversion, environmental cleanup, and sensor. Surface science techniques enable us to understand physicochemical fundamental processes on surfaces. We have developed some in situ / operando experimental techniques for observing surface reactions on gas/solid interfaces. We have focused on the development and understanding of gas sensing materials to detect small molecules such as hydrogen, by ambient pressure X-ray photoelectron spectroscopy (AP-XPS). The experiments were carried out at a beamline BL-13 at the Photon-Factory of High Energy Accelerator Research Organization (KEK-PF). An important technical point of the AP-XPS system is that the high-pressure chamber, where quasi-atmospheric gases are introduced, and the electron analyzer are separated by a small aperture and pumped by a differential pumping system. Recently, a sensor material has been developed that can detect H_2 gas in air and breath with high sensitivity (1 ppm) using nanometer-thick platinum-based thin films for example Pt-Rh alloy. The Pt-Rh sensor detects the atmospheric concentration of H_2 gas by changing in electric resistivity ($\Delta R/R_0$). We have conducted operando AP-XPS measurement coupled with resistivity measurement for H_2 sensing Pt-Rh thin-film sensor. As a result, the resistivity decreases with exposing H_2 gas to the sensor surface, whereas it increases with exposing O_2 gas. It was found that corresponding Rh 3d and Pt 4f XPS change depending on background gas conditions. Before the gas dosing, the surface was dominated by Rh oxide. When the surface was exposed to the H_2 gas, the chemical state clearly changed. The Rh oxide was completely reduced to the metallic state. When, the surface was exposed to the O_2 gas, the oxygen-induced species grew up again. Those findings indicate the surface chemical state strongly relate to the material functions.

Keywords [optional]: Ambient-pressure XPS, gas sensing materials, thin films, electric resistance.

Acknowledgement [optional]: We thank the staff of the Keio University Center Service Facilities for Research and the Photon Factory for their technical support. This study was supported by the Grants-in-Aid for scientific research (19H00756). The experiments were performed under the approval of the Photon Factory Program Advisory Committee (PF PAC 2021S2-003, 2023G522).

[1] R. Toyoshima, T. Tanaka, T. Kato, H. Abe, K. Uchida, H. Kondoh, "*Understanding of a Pt thin-film H2 sensor under working conditions by AP-XPS and XAFS*" Chem. Lett., (53) upad031 (2024).
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[3] R. Toyoshima, T. Tanaka, T. Kato, K. Uchida, H. Kondoh, "*In situ AP-XPS analysis of a Pt thin-film sensor for high-sensitive* H_2 *detection*", Chem. Commun., (56) 10147 (2020).

Interferometric Probing of Photoionization Dynamics and Resonant Control of Photoemission Delays

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Recent advances in attosecond science have enabled probing the timing of electron emission during photoionization with unprecedented precision. Here, phase-resolved measurements of photoemission dynamics are performed using two-color interferometric techniques, where the electron phase is studied as a function of emission angle, photon energy, and laser intensity.

By extending the RABBITT method to a multi-sideband detection scheme, we access detailed information about continuum–continuum transition dynamics and the interference between different ionization pathways. Special emphasis is placed on the role of resonant multi-photon pathways, where Stark shifts induced by the IR field detune coupled resonances and modify the emission phase. A three-sideband analysis enables the separation of direct and resonance-assisted contributions to the photoemission delay.

Experimental measurements in helium and argon reveal strong intensity-dependent variations in the photoemission phase, attributed to the competition between direct ionization and resonant channels. These results are supported by ℓ-dependent single-active-electron (SAE) calculations and few-level model simulations, which capture the essential features of the observed phase evolution.

This study shows how interferometric techniques can be used not only to measure but also to control photoemission delays by tuning external laser parameters, providing new insights into attosecond-resolved electron dynamics in atomic systems.

1. P. M. Paul, et al., Observation of a Train of Attosecond Pulses from High Harmonic Generation. Science 292 (5522), 1689–1692 (2001).

2. D. Bharti, et al., Decomposition of the transition phase in multi-sideband schemes for reconstruction of attosecond beating by interference of two-photon transitions. Phys. Rev.A 103, 022834 (2021).

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Surfaces and Interfaces (Island)
WeI_10 - 02:30 PM - 02:50 PM

Impact of 3D/2D Composition Ratios on the Crystallinity, Optical Properties, and Stability of Hybrid Perovskite Films via GIWAXS and Spectroscopic Measurements.

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[2]*Masinde Muliro University of Science and Technology, Kakamega, Kenya*

Hybrid perovskite materials offer exceptional promise for photovoltaic and optoelectronic applications, yet challenges remain regarding their stability and structural consistency under environmental conditions. In this work, we systematically explored the influence of dimensionality—specifically, the ratio of three-dimensional (3D) to two-dimensional (2D) organic spacer cations; Ethylenediammonium iodide (EDAI) and Butylammonium iodide (BAI) on crystallization dynamics, structural integrity, optical characteristics, and environmental stability of $Cs_{0.25}FA_{0.75}Pb(I_{0.77}Br_{0.23})_3$ perovskite films. Employing Grazing-Incidence Wide-Angle X-ray Scattering (GIWAXS), photoluminescence (PL), and UV-Vis spectroscopy, we demonstrate that mixed-dimensional perovskites (particularly with a 3D/2D ratio of 1:1) exhibit enhanced crystallinity, improved optical properties, and superior stability compared to pure 3D or other mixed-dimensional compositions. Morphological analyses via scanning electron microscopy (SEM) further correlate improved optical performance and stability to optimized grain structures and reduced defect densities. These insights provide critical guidelines for dimensional engineering aimed at advancing the performance and durability of hybrid perovskite-based optoelectronic devices.

Keywords [optional]: 3D/2D Perovskites, GIWAXS, crystallization, composition variations, crystal orientations

Acknowledgement [optional]: This work was supported by the LAAAMP in collaboration with ICTP. We express our deepest appreciation to the Beamlines 7.3.3 and 12.3.2 of the Advanced Light Source which is supported by the Director of the Office of Science, Office of Basic Energy Sciences, of the US Department of Energy under Contract No. DE-AC02-05CH11231. Furthermore, we sincerely thank the Molecular Foundry division which is also supported by the Office of Science, Office of Basic Energy Sciences, of the US Department of Energy under Contract No. DE-AC02-05CH11231.

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[2] Sutanto, Albertus A., et al. "2D/3D perovskite engineering eliminates interfacial recombination losses in hybrid perovskite solar cells." *Chem* 7.7 (2021): 1903-1916.

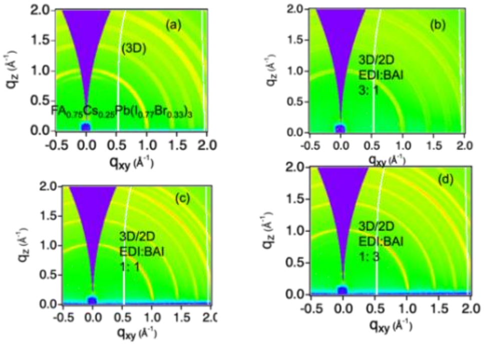

Figure 1. GIWAXS maps of (a) $Cs_{0.25}FA_{0.75}Pb(I_{0.77}Br_{0.23})_3$ 3D (b) 3D/2D with concentration of ethane, 1,2 diamoniem iodide (EDI) to Butylammonium iodide (BAI) 3:1 (c) 3D/2D with EDI to BAI 1:1 and (d) 3D/2D with EDI to BAI 1:3

Highly-sensitive negative ion spectroscopy in an electrostatic ion beam trap

L. Schweikhard[1], F.M. Maier[2,3], E. Leistenschneider[3,4], D. Hanstorp[5],
S. Rothe[3] and S. Malbrunot-Ettenauer[3,6] for the MIRACLS collaboration[7]

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[7]*https://miracls.web.cern.ch/*

The electron affinity (EA) of a chemical element is defined as the energy released as an electron is attached to a neutral atom. The binding of such an “extra” electron does not arise from the net charge of the atomic system but is a result of complex electron-electron correlations. Hence, precise measurements of EAs are powerful benchmarks of atomic theories reliant on many-body quantum methods, which are typically applied to several atomic spectroscopy studies aiming at answering quantum chemistry, nuclear structure, and fundamental symmetries questions. The EA is also an important parameter for understanding the chemical behavior of an element since it is strongly related to how much such an element is prone to form chemical bonds by sharing electrons.[1] However, the EAs of several rare and radioactive elements are still unknown and detailed information, such as isotope shifts and hyperfine splittings of EAs, is available for only a handful of stable cases.

A standard technique for the precision determination of EAs is the laser photodetachment threshold (LPT) method, in which a photon with sufficient energy is used to detach an electron from a negative ion. This technique has been restricted to mostly stable, abundant species given the low photodetachment probabilities. At CERN-ISOLDE, we have explored the use of the Multi Ion Reflection Apparatus for Collinear Laser Spectroscopy (MIRACLS) technique[2] to enhance the sensitivity of LPT to study the EA landscape among rare and radioactive species. The novel method is based on a Multiple-Reflection Time-of-Flight (MR-ToF) device to trap ions between two electrostatic ion mirrors. This allows us to greatly extend the ions' exposure time to lasers, significantly increasing the sensitivity by orders of magnitude while keeping the high resolution of a collinear geometry.

The technique has been developed offline and employed in the determination of the EA of ^{35}Cl, which is in perfect agreement with the literature value.[3] The achieved precision is competitive to that obtained in previous experiments,[3] yet employing a five orders of magnitude smaller ion sample and using high-resolution continuous wave lasers with much reduced laser power, highlighting the gains in sensitivity of this method. This talk will introduce the novel technique and present its first results as well as its potential for rare isotope sciences.

[1] D. Leimbach *et al.*, “The electron affinity of astatine”, Nat. Commun. (11) 3824 (2020).
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[3] U. Berzinsh *et al.*, “Isotope Shift in the Electron Affinity of Chlorine”, Phys. Rev. A (51) 231 (1995).

Surfaces and Interfaces (Island)
WeI_11 - 02:50 PM - 03:10 PM

Real-Time Insights into Perovskite Evolution: In Situ GIWAXS and Optoelectronic Studies of PEACl-PPF Co-Passivated Cs0.1FA0.9PbI3 Thin Film Growth

Miller Shatsala[1,2] Celine Awino[2] Maxwell Mageto[2] Husein Golicha[2] Tamura Nobumichi[1]
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[2]*Department of Physics, Masinde Muliro University of Science and Technology, Kakamega, Kenya*

The mixed-cation perovskite $Cs_{0.1}FA_{0.9}PbI_3$, passivated with phenylethylammonium chloride (PEACl) and 2,8-Bis(diphenyl-phosphoryl)-dibenzo[b,d]furan (PPF), demonstrates improved growth kinetics and optoelectronic performance. In situ GIWAXS, PL, and UV-Vis mapping show that co-passivation suppresses the δ-phase, promotes direct α-phase formation, and enhances film uniformity by reducing defects and improving grain orientation. PEACl-PPF synergy stabilizes precursor complexes, slows nucleation, and enables controlled crystallization. PPF's coordination with Pb^{2+} and halides minimizes δ-phase nucleation, supporting stable α-phase growth, offering insights for optimized perovskite films.

Acknowledgement: This research is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-05CH11231, along with funding from LAAAMP, whose generous grant is essential to the project's success.

1Li, Wang, et al. "Co-passivation of perovskite film towards stable and efficient perovskite solar cell." Chemical Engineering Journal 471 (2023): 144561.

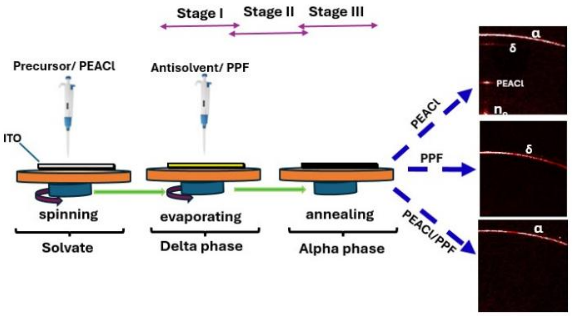

Figure 1. A sequential growth the perovskite from solution to alpha phase

Fundamental Electronic Structures and Ionization Dynamics (California)
WeII_11 - 02:50 PM - 03:10 PM

Molecular fingerprinting of f-electron molecules using a soft x-ray, HHG light Source

Siyang Wang[1], Cassandra Gates[1], Maksim Kulichenko[4], Valerio Pinchetti[1], Thomas Hoffman[1], Patrick J. Skrodzki[1], Harshad Gajapathy[1], Sang Eun-Lee[3], Samuel M. Greer[1], Matthew Carpenter[4], Sergei Tretiak, Benjamin Stein[2,3], Pamela Bowlan[1]

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While bonding and electronic structure are well understood for most elements in the periodic table, basic unanswered questions remain for lanthanides and actinides containing f-electrons, which do not always follow clear trends due to a large number of electronic states and competing potentials. X-ray spectroscopy, typically done at synchrotrons, has proven to be a powerful tool for measuring electronic structure in f-electron systems and for distinguishing subtly different molecules. Recently, tabletop light sources based on high harmonic generation (HHG) driven by femtosecond lasers, which produce extreme ultraviolet to soft x-ray femtosecond pulses, have emerged as a complementary tool for x-ray absorption spectroscopy. Here we describe a table-top, soft x-ray light source based on High Harmonic Generation (HHG) for static and potentially time-resolved spectroscopic investigations of f-electron systems and we demonstrate how x-ray absorption with this light source leads to a detailed molecular fingerprint. Our system generates photon energies from 150 eV to 330 eV, making it well-suited for studying lanthanide molecules from the perspective of multiple elemental transitions within the molecule, the lanthanide N-edge, the chlorine L-edge and the carbon K-edge. Here we will show our recent results applying to distinguish different lanthanide molecules based on their electronic structures.

Keywords [optional]: Organic semiconductors, electronic excitations, machine learning, density functional theory, materials discovery..

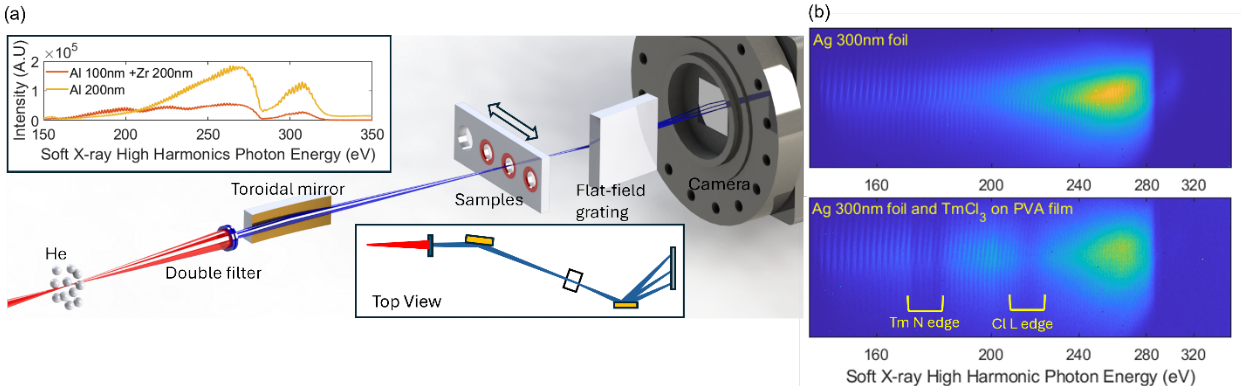

Fig. 1 A broadband HHG-based soft x-ray spectrometer for characterizing f-electron molecules: (a) Soft x-rays are produced by HHG of ~2.2 mJ, 40 fs, 1 kHz pulses at 1400 nm in a 10-mm helium-filled gas cell. Residual IR laser light is filtered out by two metal filters (double filter), and the soft x-rays are focused onto the sample using a toroidal mirror (TM). A flat-field grating (2400 lines/mm) is employed to diffract and refocus the beam onto the CCD camera. The top inset displays the soft x-ray spectrum, while the bottom inset provides a top-view schematic of the experimental setup. (b) Camera image measured (top) without and (bottom) with a $TmCl_3$ thin film sample.

Electronic Structure of Solid Electrolyte $Li_{6.5}La_3Zr_{1.5}Ta_{0.5}O_{12}$ Bulk Single Crystal Studied by Angle-resolved Photoemission Spectroscopy

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In recent years, as the use of secondary Li-ion batteries has expanded, the development of all-solid batteries using solid electrolytes has progressed to realize greater safety, wider thermal stability range, easier material handling. To understand their electronic properties and transport characteristics, the electronic structure is essentially important. However, there are few examples of experimental observations on solid electrolytes. In this study, we performed angle-resolved photoemission spectroscopy (ARPES) measurements on the lithium solid electrolytes $Li_{6.5}La_3Zr_{1.5}Ta_{0.5}O_{12}$ (LLZTO) [1] to investigate the valence band structure in relation to ionic conduction properties. Under sufficiently suppressed photon flux condition (3×10^{10} photons/s), we successfully observed valence band dispersions. From a comparison with density functional theory (DFT) calculation [2], we found qualitative similarities with the ARPES results. For example, the valence band width (~ 3 eV) of LLZTO is consistent with DFT predictions. On the other hand, the band gap observed in ARPES (3.5 eV) is smaller than that of DFT (4.5 eV), suggesting negligible correlation effects in garnet-type LLZTO. This contrasts with perovskite-type $Li_xLa_{(1-x)/3}NbO_3$ (LLNbO) and $Li_{3x}La_{2/3-x}TiO_3$ (LLTO), which exhibit a larger gap size in ARPES relative to DFT calculation [3]. Additionally, under high-flux photon irradiation condition (1×10^{12} photons/s), we observed anomalous spectral modulation over a 20-minute duration in LLZTO, characterized by a transfer of spectral weight to higher binding energies and a rigid peak energy shift. We will discuss these spectral modulations in relation to those observed in LLNbO and LLTO, where possible relation between spectral modulations and Li^+ dynamics has been expected, since the spectral modulations were not observed in the Li-free $La_{1/3}NbO_3$ [3].

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Characterization of $4f^{13}(^2F^o_{5/2})5d6s(^1D)^1[5/2]_{5/2}$ state for Metastable Qubit Operations in Yb+ ions

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G. Tomaras,[1,2] R. Zhuravel,[1] and G. Pagano[1,4]

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Trapped ions offer a pristine platform for quantum simulation and computation using hyperfine, optical, and metastable qubits. Yb^+ ions have a complex atomic structure due to transitions involving electrons from the f orbital closed inner shell. This allows for the existence of highly excited states that can be useful for repumping, detection, and state preparation in the metastable qubits encoded in the $4f^{14}5d$ $^2D_{3/2}$, $^2D_{5/2}$, and $4f^{13}6s^2$ $^2F_{7/2}$ states. We report spectroscopic and time-resolved data to characterize the atomic state $4f^{13}(^2F^o_{5/2})5d6s(^1D)^1[5/2]_{5/2}$. We observed an unexpectedly narrow transition from the metastable $4f^{14}5d$ $^2D_{3/2}$ and $^2D_{5/2}$ states, with a measured lifetime of τ = 38.4(9) μs that allows visible Rabi oscillations and sideband resolved spectroscopy. We also report measurements of the branching ratios to $^2D_{3/2}$ (0.36) and $^2D_{5/2}$ (0.64) states. By observing the steady state population at long timescales, we place an upper bound of 0.002 on the branching ratio between $^1[5/2]_{5/2}$ state and the $4f^{13}6s^2$ $^2F_{7/2}$ state. Our measurements contribute to a better understanding of the atomic structure of Yb^+ ions, which still lacks accurate numerical methods to predict atomic properties.

Keywords: Trapped ions, Ytterbium, Metastable states

Acknowledgements: This work is supported by the National Science Foundation (NSF CAREER award No. PHY-2144910), the Army Research Office (Grant No. W911NF22C0012), the Army Research Laboratory (Grant No. W911QX21C0031), Welch Foundation Grant No. C-2154, and the Office of Naval Research (Grants No. N00014-23-12665 and the Young Investigator Program N00014-22-1-2282). We acknowledge that this material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics under the Early Career Award No. DE-SC0023806.

Activation Mechanisms of layered MoS_2 Surfaces probed by time-based and time-resolved Electron Spectroscopy

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Layered transition metal dichalcogenides such as molybdenum disulphide (MoS_2) have widely tunable properties making them relevant for electronics, sensors, energy storage, and catalysis. For MoS_2 we have thus investigated the inhomogeneity and homogenization of the local n- and p- doping character of bulk MoS_2. In particular, a cleaved surface typically displays spatially inhomogeneous properties. Annealing and exposure to molecular hydrogen can be cycled to switch reversibly between smaller and larger surface band bending preserving local inhomogeneity. Atomic hydrogen exposure, however, equilibrates reversibly the entire surface to a common doping situation due to the amphoteric effect of hydrogen atoms to serve as donors and acceptors alike, neutralizing local charge imbalances.[1] Optical excitation is shown to create a transient metallic surface layer on bulk semiconducting MoS_2 where electrons in the conduction band of p-doped semiconducting MoS_2 travel toward the surface layer.[2] They accumulate in the top layer and create a transient reversible 2H-semiconductor to 1T-metal phase-transition of the surface layer. Electron–phonon coupling of the indirect-bandgap p-doped 2H-MoS_2 enables this efficient pathway even at a low density of excited electrons with a distinct optical excitation threshold and saturation behavior. This mechanism needs to be taken into consideration when describing the photocatalytic properties of the 2H-MoS_2 surface. In particular, light-induced increased charge mobility and surface activation can cause and enhance the photocatalytic and photo assisted electrochemical hydrogen evolution reaction of water on 2H-MoS_2. Generally, it opens up for a way to control not only the surface of p-doped 2H-MoS_2 but also related dichalcogenides and layered systems.[2] All findings are based on time based and time resolved electron spectroscopy for chemical analysis with photon-energy-tunable synchrotron radiation.

Keywords: Dichalcogenide, Surface Activation, Phase Transition, Intercalation, Doping, Amphoteric Effect of Hydrogen, Photoelectrocatalysis. Electron Spectroscopy

Acknowledgement: The work has been conducted within the Uppsala-Berlin joint Laboratory on next generation electron spectroscopy UBjL at the Synchrotron BESSY II..

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[2] N.L.A.N. Sorgenfrei et al. Photodriven Transient Picosecond Top-Layer Semiconductor to Metal Phase-Transition in p-Doped Molybdenum Disulfide. *Adv. Mater.* **33**, 2006957 (2021).

Temporal Electronic and Structural Landscapes in Catalytic Water-Splitting

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Keywords: water splitting, catalysis, time-resolved/ultrafast elastic and inelastic X-ray scattering, free-electron lasers, synchrotrons, liquid-jet technologies

Water splitting is a crucial process for hydrogen production, which is essential for the development of a sustainable energy economy. It enables the conversion of water into hydrogen and oxygen using renewable energy sources, providing an alternative to fossil fuels. As water is an abundant resource and the OH-water bond is one of the strongest chemical bonds in nature, water splitting, when fueled by renewable energy, becomes a sustainable and scalable method to unlock the full potential of renewable energy technologies.

In this talk, I will present our method developments for the years 2023-2025 and our new research studies on catalytic water splitting. This continues our efforts to understand and uncover the detailed atomistic mechanism of the oxygen evolution reaction (OER) 1-3. The methods of choice are high-resolution photon-in / photon-out experiments at high repetition x-ray sources (synchrotrons and FELs) - elastic and inelastic scattering studies, with special emphasis on resonant inelastic x-ray scattering (RIXS).

I will present our recent in-situ, operando or time-resolved soft X-ray studies exemplified by OER of different highly efficient catalysts of the types metal oxide-doped metal oxide systems (AO/BO) and - for comparison - ABO3 (A, B = transition metals). These catalysts were selected because they are able to break the scaling law (i.e. they are more efficient than the standard systems interpolated in a thermodynamic model).

Since the inelastic soft X-ray spectroscopy studies are element and oxidation state selective, the presentation starts with different types of OER from the perspective of the catalyst (metal L-edges). Using polarization-dependent metal L-edge soft X-ray spectroscopy, we have investigated the catalyst intermediates formed during OER on ideally grown surfaces and compared them with those formed when nanoparticles are used. The results are discussed in the context of ultrafast RIXS studies of similar systems, but in a different context - ultrafast photoexcitation studies with a RIXS energy resolution of over 100 meV at the European XFEL 4.

Zooming into the Helmholtz layer by oxygen K-edge spectroscopy and maintaining the high RIXS energy resolution of 100 meV (and beyond), the water adsorption results are discussed in relation to the electrolysis active layers on the catalyst surface of the different catalysts listed above.

[1] S. S. Nair Lalithambika, ST et al. *Adv. En. Mat.* 2025, 2404280 (2025). DOI: 10.1002/aenm.202404280
[2.] J. J. Velazquez-Garcia, ST et al., *Chem. Sci.* 15, 13531-13540 (2024). DOI: 10.1039/d4sc02933j
[3] T. Reuss, ST et al., *Acc. Chem. Res.* 56, 203-214 (2023). DOI: 10.1021/acs.accounts.2c00525
[4] J. Schlappa, S. S. Nair Lalithambika, ST, A. Scherz, A. Foehlisch et al., *J. Syn. Rad.* 32, 29-45 (2025). DOI: 10.1107/S1600577524010890

Fundamental Dynamics of Photocatalysis (California)
WeII_15 - 04:30 PM - 05:00 PM

Abstract Not Available

Interfacial interaction between $TaSe_2$ and bilayer graphene

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The formation of an interface between two different materials modifies the physical properties even in the van der Waals materials. For example, superconductivity and charge density wave that appear in transition-metal dichalcogenides is significantly perturbed by choosing different substrates. Here we investigate such an interfacial effect between monolayer $TaSe_2$ and bilayer graphene epitaxially grown on a SiC substrate via the direct measure of electron band structures using angle-resolved photoemission spectroscopy [1]. Depending on the structural phase of $TaSe_2$, different amount of charge transfer and band hybridization occur with graphene. The difference originates from the work function of each structural phase of $TaSe_2$ whose relative change is different upon approaching the monolayer limit. These findings provide a viable route towards the engineering of electronic correlations in transition-metal dichalcogenides.

Keywords: interfacial effect, transition-metal dichalcogenides, graphene, electron band structure, angle-resolved photoemission spectroscopy

[1] H. Lee, H. Im, B. K. Choi, K. Park, Y. Chen, W. Ruan, Y. Zhong, J. -E. Lee, H. Ryu, M. F. Crommie, Z. -X. Shen, C. Hwang, S. -K. Mo, J. Hwang, "*Controlling structure and interfacial interaction of monolayer TaSe2 on bilayer graphene*", Nano Convergence (11) 14 (2024).

Ultrafast Charge Transfer Dynamics in a Plasmonic Photocatalyst Studied by Femtosecond Time-Resolved X-ray Photoelectron Spectroscopy

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Heterogeneous interfaces between metal nanoparticles (MeNPs) and transition metal semiconductors (SCs) attract significant attention due to their potential to provide routes to renewable, carbon-free energy production and storage through processes such as photocatalytic hydrogen generation from water. A particularly intensely studied model system for photochemical water splitting consists of plasmonically active, spherical gold nanoparticles (AuNPs) attached to nanocrystalline TiO_2 substrates, shown in Figure 1. Despite significant efforts, the overall efficiencies of plasmonic light harvesting systems generally remain rather low, requiring a deeper understanding of the fundamental dynamics underlying their function. Currently, charge transfer (CT) between MeNPs and SCs is commonly described by either plasmon-induced hot-electron transfer (PHET) or plasmon-induced metal-to-semiconductor interfacial charge transfer transitions (PICTT). To gain a deeper understanding of these competing processes, we have measured the ultrafast interfacial charge injection and recombination dynamics for AuNP sensitized TiO_2 at the FLASH Free Electron Laser in Hamburg under UHV and water exposed conditions via femtosecond Time-resolved X-ray Photoelectron Spectroscopy (TRXPS). The results will be discussed within a kinetic model considering various CT and relaxation channels. The model is complemented by high-level ab-initio calculations based on constrained density functional theory.

Keywords: Charge transfer, photocatalysis, nanoplasmonics, FELs, time-resolved XPS.

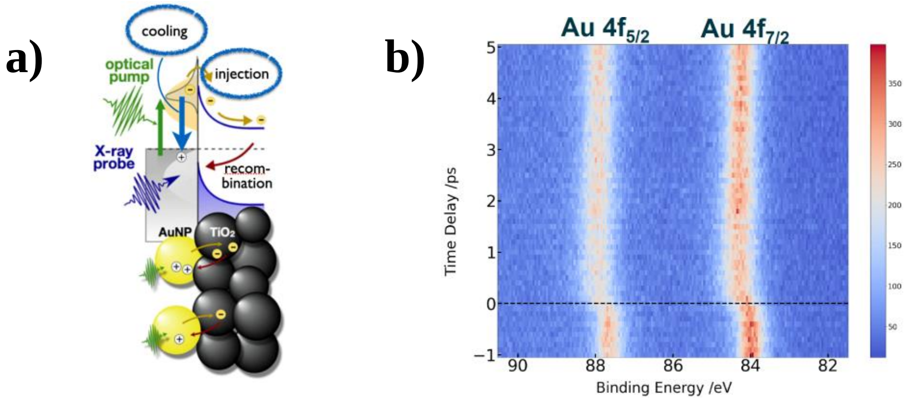

Figure 1. a) Conceptual representation of competing electronic processes after photoexcitation. **b)** Transient Au4f photoemission spectra during photoinduced CT in the AuNP/TiO_2 system recorded at FLASH.

Synthesis and Electronic Characterization of $Nd_{2-x}Sr_xNiO_4$ thin films $(0 \leq x \leq 1.4)$

Nicole K. Taylor,[1,*] Dan Ferenc Segedin,[2,*] Ari B. Turkiewicz,[2] Yang Zhang,[3] Spencer T. Doyle,[2] Grace A. Pan,[2] Haoyue Jiang,[4,5] Aaron Bostwick,[6] Chris Jozwiak,[6] Eli Rotenberg,[6] Alessandra Lanzara,[5,7] Ismail El Baggari,[3] Charles M. Brooks,[2] Alpha T. N'Diaye,[6] Luca Moreschini,[5,7] and Julia A. Mundy[1,2,†]

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Layered nickelates have been studied extensively over the last three decades due to their structural similarities to the high-T_c superconducting cuprates. Using reactive oxide molecular beam epitaxy (MBE), we synthesize $Nd_{2-x}Sr_xNiO_4$ thin films for $x = 0 - 1.4$ to probe the properties and electronic structure as a function of hole doping. The samples with lower doping show semiconducting behavior across the temperatures probed with an onset of metallic conductivity at $x = 1.4$. We also present polarization-dependent O K and Ni $L_{2,3}$ x-ray absorption spectra to track the evolution of the oxygen-nickel hybridization, distribution of holes between O $2p$ and Ni $3d$ states and the nickel oxidation state across the series. Angle-resolved photoemission spectroscopy (ARPES) measurements reveal a Fermi surface that comprises a cuprate-like hole-pocket of $d_{x^2-y^2}$ character with an additional electron pocket of $d_{3z^2-r^2}$ character at Γ. The emergence of a quasiparticle peak at the Fermi vector for $x = 1.4$ corroborates the insulator-to-metal transition at $x \sim 1$. Finally, observe a fully two-dimensional Fermi surface with no momentum-dependent pseudogap, in contrast to measurements of the related bulk compound, $Eu_{0.9}Sr_{1.1}NiO_4$.

Acknowledgments: Research is primarily supported by the U.S. Department of Energy (DOE), Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, under Award No. DE-SC0021925. This work used resources of the Advanced Light Source, which is a DOE Office of Science User Facility under contract no. DE-AC02-05CH11231. This work used resources of the Advanced Light Source, which is a DOE Office of Science User Facility under Contract No. DE-AC02-05CH11231. The ARPES work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences and Engineering Division, under Contract No. DE-AC02-05-CH11231 within the Ultrafast Materials Science Program No. KC2203. Device fabrication work was performed at Harvard University's Center for Nanoscale Systems (CNS), a member of the National Nanotechnology Coordinated Infrastructure Network (NNCI), supported by the National Science Foundation under NSF Grant No. 1541959. Electron microscopy was carried out through the use of MIT.nano facilities at the Massachusetts Institute of Technology. N.K.T. acknowledges the support of a Ford Foundation Predoctoral Fellowship and the support from the ALS Doctoral Fellowship in Residence. D.F.S. and G.A.P. acknowledge support from the NSF Graduate Research Fellowship Grant No. DGE-1745303. G.A.P. acknowledges additional support from the Paul & Daisy Soros Fellowship for New Americans. A.B.T. acknowledges the support from the National Science Foundation under Award. No. DMR-2323970. J.A.M acknowledges support from the Packard Foundation and the Sloan Foundation. Y.Z. and I.E. were supported by The Rowland Institute at Harvard.

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Investigation of the effects of nanoscale facets on catalytic activity in photo-driven nanosystems

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Nanoparticles exhibit extraordinary catalytic activity due to their high surface area, tunable morphology, and unique electronic properties. Among these, facets and surface defects play a pivotal role in enhancing photocatalytic reactions. In this study, we aim to unravel the nanoscale mechanisms governing charge-driven photocatalytic processes on single, isolated nanoparticles with varying morphologies. Utilizing a combination of velocity map imaging (VMI) and coherent diffractive imaging (CDI) at the Maloja instrument at SwissFEL, we probe the ultrafast dynamics of spherical and cubic gold nanoparticles (AuNPs) in a time-resolved optical pump and X-ray probe scheme.

Results reveal orientation-dependent ion momentum distributions and enhanced ionic yields correlated with nanoparticle facets. These observations suggest that near-field enhancement effects at high-index facets significantly modulate catalytic activity. By systematically analyzing ionic fragment momenta across varying time delays and nanoparticle orientations, we establish a direct relationship between nanoparticle morphology and site-specific reactivity. The multimodal approach combining VMI and CDI provides unprecedented insight into the role of structural factors in nanoscale photocatalysis, advancing our understanding of clean energy processes and catalysis optimization.

Keywords: Catalysis, nanoparticles, XFEL

This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, under Contract No. DE-SC0063.

Magnetic Spectroscopy Across Scales: Investigating Biological Energy Systems and Synthetic Magnetic Architectures

Anex Jose[1,2], Andrew W. Schaefer[1], Antonio C. Roveda Jr.[1], Wesley J. Transue[1], Sylvia K. Choi[2], Ziqiao Ding[2], Robert B. Gennis[2], Edward I. Solomon[1]*; Johnny Ho[3], Stephanie G. Corder[3], Alpha T. N'Diaye[3]*.

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Magnetic spectroscopy techniques serve as a powerful tool to probe the electronic structure of complex systems across multiple length scales. In the first part of this talk, I will present the elucidation of structure-function correlation in a complex biomolecular energy system called respiratory oxidases using magnetic circular dichroism (MCD) spectroscopy in the ultraviolet-visible-near infrared (IR) energy regime. In the second part of the talk, I will present our efforts to synthesize a novel class of magnetic thin films using f-block element-based molecules and unravelling their properties using an existing end station at the Advanced Light Source (ALS) that is modified for simultaneous X-ray MCD (XMCD) and FT-IR spectral measurements.

Aerobic organisms use O_2 to drive the membrane-bound electron transport chain for ATP synthesis. The $4e^-$ reduction of $O_2 \rightarrow H_2O$ underlying this process is catalyzed by the superfamily of respiratory oxidases called heme–copper oxidases (HCO). The HCO active site consists of a heme ligated by a histidine (His) and a Cu ligated by three His, one of them covalently linked to a tyrosine (Tyr). Reaction with O_2 results in O–O bond cleavage forming a $Fe^{IV}{=}O$ intermediate P_M. Further e^-/H^+ transfers to the active site form subsequent intermediates ($P_M \rightarrow F \rightarrow O_H \rightarrow E_H \rightarrow R$), each coupled to pumping a H^+ across the membrane for ATP synthesis.

A major challenge in studying HCOs is the presence of additional redox centers in the enzyme, low spin heme and binuclear Cu_A (in some HCOs), with intense spectroscopic features precluding the study of the oxidation/protonation states of Cu and Tyr. We have leveraged MCD, a site-selective magnetic spectroscopy method, to investigate the Fe/Cu/Tyr site in HCOs. We studied P_M in UbO (an HCO that lacks the Cu_A center) from *E. coli* by low temperature MCD where we observe a unique feature from its $Fe^{IV}{=}O$ center. Variable-temperature variable-field (VTVH) MCD on this excited state feature enabled us to study the ground state of P_M determining that it is a three-spin exchange-coupled system: $Fe^{IV}{=}O$ (S=1), Cu^{II} (S=½), $Tyr^{\bullet}$ (S=½), which for the first time gave direct experimental evidence for the presence of Cu^{II} and $Tyr^{\bullet}$ in P_M. The experimental characterization of P_M provides novel insight into how the reduction of P_M is coupled to H^+ pumping thus elucidating structural contributions to the biological function of HCOs.[a]

Transitioning from biological systems to synthetic magnetic architectures, molecular materials offer significant potential for microelectronics and quantum information science where the goal is to harness individual molecules for functional applications. Achieving this goal requires precise control over molecular assemblies and a fundamental understanding of the interplay between spin and electronic structure.

A key challenge is the atomically precise arrangement of molecules into large-scale, ordered 2D patterns, a prerequisite for utilizing surface science tools to characterize a single species of surface-bound molecules with uniform configuration relative to the substrate and neighboring molecules. To address this, we are preparing thin films of f-block element-based molecules which can be considered as an intermediate between bulk and individual molecules. These thin films are characterized at an existing XMCD beamline end station at ALS (9 T maximum magnetic field) that is modified to enable simultaneous FT-IR measurements. In the future, we aim to deposit these magnetic molecules on a graphene-metal moiré, serving as a template for near-perfect self-organized assembly. This system will provide a conceptual testbed for exploring single-molecule data storage and computation, leveraging individual molecular spins for quantum applications.[b]

[a]Jose *et al*., *Science*, 2021, *373* (6560), 1225-1229; [b]Jose *et al*., manuscript in preparation, 2025.

Fundamental Dynamics of Photocatalysis (California)
WeII_18 - 05:40 PM - 06:00 PM

Anisotropic Ultrafast Carrier Dynamics Observed in Bulk ReS_2 Using Femtosecond Time- and Energy-Resolved Photoemission Electron Microscopy

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Transition metal dichalcogenides (TMDs) are promising candidates for optoelectronic applications, attributed to their properties such as high in-plane carrier mobility. The well-studied group 6 TMDs such as MoS_2, $MoSe_2$, WS_2 and WSe_2 exhibits isotropic in-plane optical response due their symmetric crystal lattice. In contrast, group 7 TMDs, such as ReS_2, demonstrate anisotropic in-plane optical behavior with respect to the b-axis because of their distorted crystal lattice. This optical polarization anisotropy could serve as an additional tuning parameter in device applications using TMDs.

In this study, we investigate the polarization-anisotropic ultrafast carrier dynamics of AA-stacked bulk ReS_2 with time- and energy-resolved photoemission electron microscopy (TR-PEEM) with sub-50-fs pump and probe pulses, and sub-200-meV energy resolution. In the experiments, 2.41-eV pump pulses first initiate above-gap excitation. The electron population in the conduction band is subsequently probed by 4.81-eV probe pulses at various delay times. Dynamics that are dependent on both pump- and probe-polarization are observed. When keeping the probe polarized parallel to the b-axis, at a photoexcitation carrier density of $\sim 3.3 \times 10^{12}\ \mathrm{cm}^{-2}$, for example, excitation perpendicular to the b-axis results in a fast decay of $\tau_{fast} = 0.49 \pm 0.05$ ps, whereas excitation parallel to the b-axis yields a slower $\tau_{fast} = 1.3 \pm 0.3$ ps (Fig. 1a), with both cases exhibiting a shared slow decay time constant. Increasing the carrier excitation densities results in a decreasing τ_{fast}, suggesting that the fast decay arises from carrier-carrier scattering. When the pump polarization is fixed parallel to the b-axis, an additional rise component of $\tau_{rise} = 0.10 \pm 0.03$ ps is observed only when probing perpendicular to the b-axis (Fig. 1b).

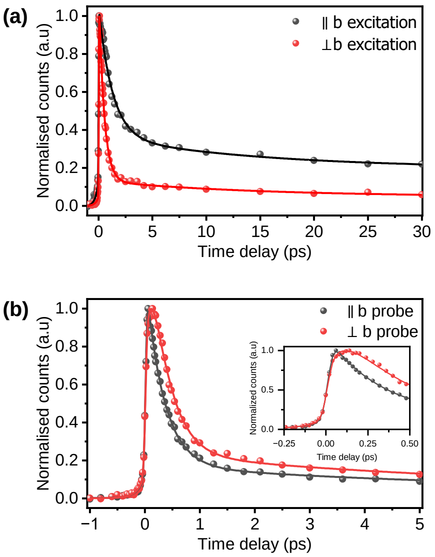

Figure 1. (a) Time trace with probe polarization fixed parallel to the b-axis, varying pump polarization. (b) Time trace for pump polarization fixed parallel to the b-axis, varying probe polarization, with inset showing the early time delays.

These preliminary results demonstrate that bulk ReS_2 possesses anisotropic ultrafast carrier dynamics and could serve as a promising platform for optoelectronic device applications requiring polarization selectivity.

Shifted protonation equilibrium of atmospheric acids and bases at the aqueous surface and impacts on cloud droplet chemistry and climate effects

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[2] *Institute of Inorganic and Applied Chemistry, University of Hamburg, Hamburg, Germany*
[3] *Center for Atmospheric Research, University of Oulu, Oulu, Finland*

Atmospheric aerosols comprise a significant fraction of organic species with Brönsted acidic or basic character. Their concentration and protonation state affect the pH of aqueous aerosols and droplets with implications for a wide range of pH-sensitive reactions. Many atmospheric organics also have amphiphilic molecular structures and exhibit surface activity in aqueous solutions. Due to the high surface area to bulk volume ratios of nano- and microscopic aerosols and droplets, surface adsorption can result in nearly complete bulk-phase depletion, strongly favoring surface-specific states and affecting both bulk-phase and heterogeneous chemistry.[1]

We used Xray Photoelectron Spectroscopy (XPS) in combination with high-brilliance synchrotron radiation to directly observe the protonation state of atmospheric acids and bases at the surfaces of aqueous aerosol and droplet models.[2] We found that for each acid-base pair, the neutral species is strongly favored in the surface, consistent with its higher surface activity, compared to the charged conjugate. This introduces a shift in the protonation equilibrium at the aqueous surface corresponding to an apparent change in pKa of 1-2 pH units, depending on the concentration, acidity, and surface activity of the conjugate acidic and basic species.

We developed representations to include this apparent shift in surface acidity in box model and global-scale climate simulations.[3,4] Our results show that considering the surface protonation state of organic acids and bases can influence aqueous-phase sulfur chemistry to significantly impact the formation of secondary sulfate aerosol mass and resulting cloud radiative climate effect.

Acknowledgement: The presenter warmly thanks the ATMOS (University of Oulu) and CIA3 (DESY) team members and collaborators who have contributed to the research and the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme Project SURFACE (grant agreement no. 717022) and the Research Council of Finland (grant nos. 257411, 308238, 314175, 316743, 335649, and 351476) for funding.

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Plenary Talks (Island)
ThI_02 - 09:15 AM - 10:00 AM

Abstract Not Available

Studying Nobles Gases with Photoelectron Spectroscopy

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We present a study on the photoelectron spectroscopy (PES) of noble gases, both in the gas phase at elevated pressures and when confined within silicate nanocages supported on a metal substrate. By leveraging X-ray photoelectron spectroscopy (XPS) under near-ambient pressure conditions, we examine noble gases in their free state and in constrained environments. The encapsulation process within the nanocages is investigated using controlled caging (by an ionization-mediated mechanism) and (thermal) uncaging experiments, revealing insights into confinement effects. These studies offer new perspectives on gas confinement at the nanoscale and demonstrate the utility of PES for characterizing encapsulated species in functional materials.

Keywords: Noble gases, Confinement Effects, Nanomaterials, Photoelectron Spectroscopy, Argon, Krypton, Xenon.

Acknowledgement [optional]: Experiments were carried out at the Center for Functional Nanomaterials and National Synchrotron Light Source II, supported by U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. DE-SC0012704. Part of the research was carried out under Technology Commercialization Fund grant TCF-20-20124. Laiba Bilal was partially supported by BNL LDRD No. 22-050.

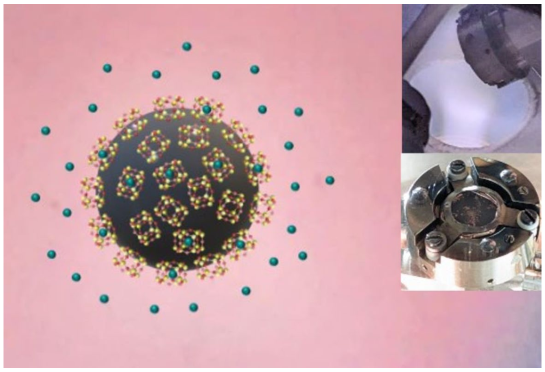

Figure 1. Illustration of a metal particle covered with hexagonal prism silicate nanocages in the presence of a Xenon plasma. The insets show photos of the sample in the center of a sample holder (bottom right) and the sample exposed to the xenon plasma (top right)

Insensitivity of charge ordering instability to screening in the excitonic insulator candidate $TiSe_2$

P.D.C. King[1]

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Control over materials thickness down to the single-atom scale has emerged as a powerful tuning parameter for manipulating not only the single-particle band structures of solids, but increasingly also their interacting electronic states and phases. A particularly attractive materials system in which to explore this is the transition-metal dichalcogenides (TMDs), both because of their naturally-layered van der Waals structures and the wide variety of materials properties which they are known to host. Yet, how their interacting electronic states and phases evolve when thinned to the single-layer limit remains a controversial question. Here, I will discuss our work integrating monolayer materials growth by molecular-beam epitaxy [1] with electronic structure studies via *in situ* angle-resolved photoemission and ARPES-based microscopy. I will discuss the resulting electronic structure in these systems, with a particular focus on the controversial charge-density wave phase of monolayer $TiSe_2$ [2,3], and the insights we can obtain from controlling its electrostatic environment [4].

Acknowledgement: This work was performed in close collaboration with S. Buchberger, A. Rajan, Yann in 't Veld, Tim Wehling, Craig Polley, and colleagues from the Universities of St Andrews, Hamburg, and Max-IV laboratory.

[1] Rajan, Buchberger *et al.*, Advanced Materials 36, 2402254 (2024); [2] Watson *et al.*, 2D Materials 8, 015004 (2021); [3] Antonelli *et al.*, Nano Lett. 24, 215 (2024); [4] Buchberger *et al.*, unpublished.

Revealing The Dynamic Surface State of Cu-based Single-Atom Alloys Using Ambient Pressure X-ray Photoelectron Spectroscopy

Iradwikanari Waluyo[1], Hojoon Lim[1], Adrian Hunt[1]

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Single-atom catalysts (SACs) have emerged as a transformative class of catalysts, combining the atomic efficiency of isolated active sites with unique chemical and electronic properties that distinguish them from traditional bulk or nanoparticle-based catalysts. A type of SAC called single-atom alloy (SAA) is typically composed of a small amount of a catalytically active noble metal (e.g. Pd, Pt, Rh, Ir) atomically dispersed in the surface of a more inert host metal (e.g. Cu, Ag, Au). The reactivity of the dopant metal and the selectivity of the host metal work synergistically, yielding bifunctional catalysts that deviate from the typical scaling relationships that govern catalytic activity. Research efforts on SAAs exemplify the oft-mentioned "pressure gap" existing between the abundance of recent research performed under ultra-high vacuum conditions and catalytic reactor studies at high pressures. Ambient pressure X-ray photoelectron spectroscopy (AP-XPS) is the ideal technique for bridging this gap by providing an understanding of how the surface chemical and electronic states of SAAs dynamically evolve under reaction conditions, thus revealing property-structure-reactivity relationships important in catalysis. We have studied SAA model systems with copper as the host metal and compared the effects of three different dopants: Pt, Rh, and Ir. Differences in CO adsorption properties as well as CO-induced atomic mobility were revealed using AP-XPS, with Pt showing the weakest binding to CO and Ir the strongest. Under oxidizing and reducing conditions, we also discovered significant differences in how the different dopants either promote or hinder O_2 dissociation and H_2 activation depending on the structural arrangements of the dopant with respect to the host. In addition, structural heterogeneities of the Cu host metal surface were found to significantly impact the dopant alloying sites, which subsequently alter surface chemical reactivity. Overall, our in situ research using AP-XPS revealed how such a small amount of dopant metal can have an outsize impact on reactivity, with important implications in various catalytic reactions.

Keywords: ambient pressure X-ray photoelectron spectroscopy, single-atom catalyst, single-atom alloy, copper, noble metals

Acknowledgement: This research used resources of the 23-ID-2 (IOS) beamline of the National Synchrotron Light Source II, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Brookhaven National Laboratory under Contract No. DE-SC0012704

Electronic Control in 2D Materials (California)
ThII_05 - 11:00 AM - 11:30 AM

Abstract Not Available

Probing electrochemical interfaces and solvation in calcium-ion battery using operando soft X-ray spectroscopy

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Synchrotron based X-ray spectroscopic techniques offers unique characterization in many important energy materials of energy conversion, energy storage and catalysis in regards to the functionality, complexity of material architecture, chemistry and interactions among constituents within. However, it is challenging to reveal the real mechanism of the chemical processes. In the operando soft X-ray spectroscopy characterization of interfacial phenomena in energy materials and devices, it has been found that the microstructure and composition of materials as well as the microstructure evolution process have a great influence on performances in a variety of fields, e.g., energy conversion and energy storage materials, chemical and catalytic processes.

The solvation of an electrolyte near the electrolyte/electrode interphase dictates the charge transfer efficiency and therefore affects the performance of a battery. However, current understanding regarding this interphase was limited due to the lack of direct interphase probing approaches under in-situ/operando conditions. In this report, using operando X-ray absorption spectroscopy and resonant soft X-ray scattering through a patterned e-chip, the solvation of a calcium organic electrolyte was investigated under operando conditions. The disruption of the solvation structure by a secondary anion or cation were evaluated using the methodologies developed, which will guide the design of electrolytes for future energy storage. The strategy of these new methodologies developed will also benefit the investigation of catalysis at interphases and electrochemical microenvironments in general.

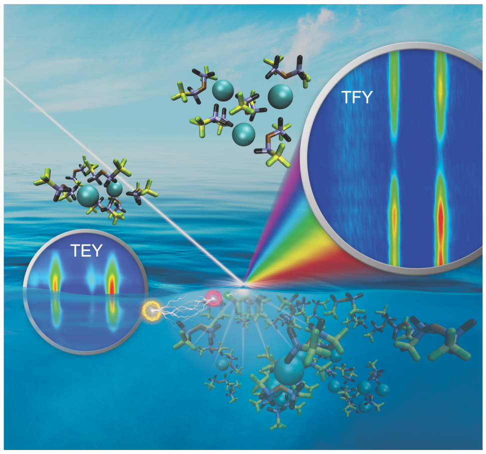

Figure 1. The calcium L-edge XAS spectra, under electrochemically operando conditions in both TEY and TFY modes, provide insights into the calcium solvation structures at the interphase and in the bulk.

Ultrafast dynamics in 2D materials and heterostructures visualized with time- and angle-resolved photoemission

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[2] *Dept. of Chemistry, Stony Brook University, Stony Brook, NY, U.S.A*

The optical and electronic properties of two-dimensional (2D) materials make them attractive for a variety of applications in quantum information processing, solar-energy harvesting, and catalysis. However, the dynamics of electrons, holes, and excitons formed after photo-excitation are often complex and involve many states that are optically dark, making their characterization with optical spectroscopy alone difficult. We will present our recent work imaging the quantum states of 2D materials and their heterostructures in momentum space using time- and angle-resolved photoemission. A unique combination of tunable ultrashort extreme ultraviolet (XUV) pulses with 61 MHz repetition rate and timeof-flight momentum microscopy (ToF k-mic) enables the parallel recording of electron dynamics in all states across the full Brillouin zone after perturbative excitation, while systematically varying many parameters. We will present recent results resolving the mixture of interlayer and intralayer excitons created after above band-gap excitation of MoSe2/WS2 bilayers due to spatially varying type I and type II band alignment[1], and dark states in type II WS2/WSe2 bilayers. We will also present recent progress towards time-resolved momentum microscopy at arbitrary (0-61 MHz) repetition rate with tunable pump and probe photon energies.

Keywords: 2D materials, momentum microscopy.

Acknowledgement: This work was supported by work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under award number DE-SC0022004 and the Air Force Office of Scientific Research under FA9550-20-1-0259

[1] Guo et al., “Moiré-Tunable Localization of Simultaneous Type I and Type II Band Alignment in MoSe 2 /WS2 Heterobilayer.” arXiv:2502.14138 (2025).

Local and Symmetry-Resolved Electronic Structure of Liquid Dimethyl Sulfoxide from Resonant Inelastic Soft X-ray Scattering

Lothar Weinhardt[1,2,3], Dirk Hauschild[1,2,3], Ralph Steininger[1], Constantin Wansorra[1,3],
Wanli Yang[4], and Clemens Heske[1,2,3]
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Dimethyl sulfoxide (DMSO, C_2H_6OS) is a highly polar solvent with comparatively low toxicity. It is used in many fields to dissolve a wide range of both polar and non-polar organic and inorganic compounds. The unique properties of DMSO are strongly defined by the sulfinyl functional group, in which the nature of the bond between sulfur and oxygen plays a central role. A detailed understanding of DMSOs electronic structure is crucial, and advanced soft x-ray spectroscopy techniques now allow to study such molecular systems also in the liquid state. In our contribution, we derive the local and symmetry-resolved electronic structure of liquid DMSO, using resonant inelastic soft X-ray scattering (RIXS) maps at the S $L_{2,3}$, C K, and O K edges. Such maps give the most complete experimental description of the electronic structure of this important solvent, especially when combining them with calculations based on density functional theory that enable a detailed locally- and symmetry-resolved analysis of the molecular orbitals. In the RIXS maps, we find the signature of molecular-field splitting of the S 2p core levels, vibronic coupling, and ultrafast nuclear dynamics on the time scale of the RIXS process.

[1]L. Weinhardt, D. Hauschild, R. Steininger, C. Wansorra, W. Yang, and C. Heske, *"Local and Symmetry-Resolved Electronic Structure of Liquid Dimethyl Sulfoxide from Resonant Inelastic Soft X-ray Scattering"*, J. Phys. Chem. Lett. (12) 10576 (2024).

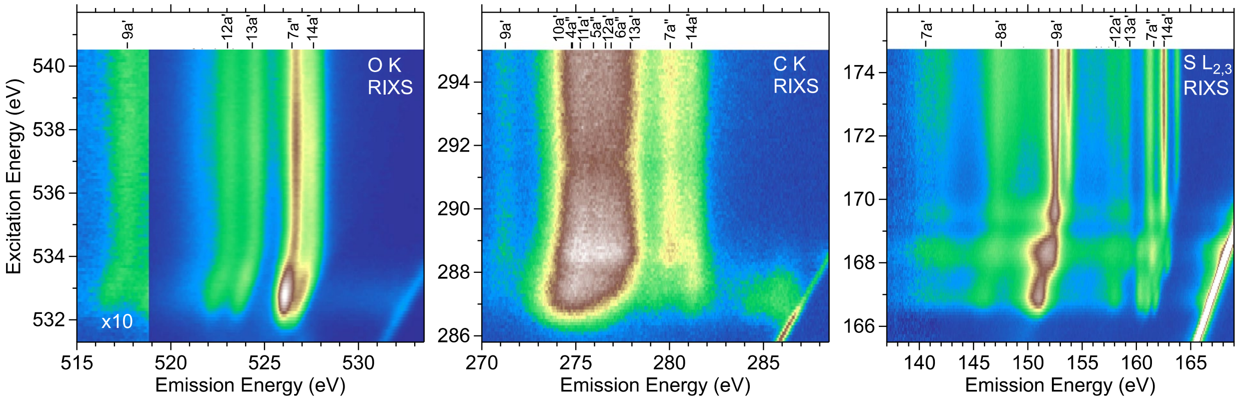

Figure 1. O K (left), C K (center), and S $L_{2,3}$ (right) RIXS maps of liquid DMSO. Above the maps, emission lines are labeled and marked according to their calculated non-resonant emission energies. From Ref.[1]

Coexistence of Dirac and flat bands in 2D material beyond geometrical frustrated lattice

Asish K. Kundu[1], Christie S. Koay[2], Daniel G. Chica[2], A. Devarakonda[2], Anil Rajapitamahuni[1], Raquel Queiroz[2], Xavier Roy[2], Abhay N. Pasupathy[2], and E. Vescovo[1]

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Two-dimensional (2D) Dirac materials featuring flat bands have emerged as a pivotal focus in condensed matter physics due to their rich physical phenomena, including superconductivity and ferromagnetism, and their potential for integration into electronic devices. Flat bands, typically associated with strong electron correlations, are predominantly observed in heavy fermion systems with localized *f*-orbitals. While geometrically frustrated lattices have been proposed as a pathway to extend bulk flat bands beyond *f*-orbital systems, to date, such bands have only been identified in solid-state materials with kagome and related pyrochlore lattice structures. Here, I will present our recent study [1] unveiling the coexistence of Dirac and flat bands within the geometrically unfrustrated primitive square lattice compound Pd_5AlI_2, investigated through angle-resolved photoemission spectroscopy (ARPES) and model calculations. Theoretical calculations and polarization-dependent ARPES measurements reveal that specific atomic orbital decorations on a primitive square lattice induce frustrated electron hopping, forming a flat band. Further, photon-energy dependence of electronic structure measurements shows the quasi-2D electronic nature of this material. Owing to the van der Waals (vdW) nature, this material can be exfoliated to the monolayer limit, paving the way for exploring orbital physics in true 2D limit.

Keywords: 2D vdW metal, electronic structure, ARPES, flat band, Dirac material, density functional theory, materials discovery.

Acknowledgement: ARPES measurements used resources at the 21-ID (ESM) beamline of the National Synchrotron Light Source II, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Brookhaven National Laboratory under Contract No. DE-SC0012704. Research on novel hopping models was conducted as part of the Programmable Quantum Materials, an Energy Frontier Research Center, funded by the US Department of Energy (DOE), Office of Science, Basic Energy Sciences, under award DE-SC0019443 (X.R., A.N.P., R.Q.). The synthesis and structural characterization of the 2D metal was supported by the NSF MRSEC program through the Center for Precision-Assembled Quantum Materials at Columbia University, award number DMR-2011738 (X.R., A.N.P). A.D. acknowledges support from the Simons Foundation Society of Fellows (grant No. 855186).

[1] A. Devarakonda, Christie S. Koay, Daniel G. Chica, M. Thinel, Asish K. Kundu et al., "*Frustrated hopping from orbital decoration of a primitive two-dimensional lattice*", arXiv preprint, arXiv:2408.01512 (2024).

SIMULTENOUS ELECTRON SPECTROSCOPY AND X-RAY SCATTERING ON MODEL CERIA CATALYSTS

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Heterogeneous catalysis is a timely and critical research field in basic and applied energy sciences, due to its potential to provide solutions to global environmental issues. However, there is still a lack of a profound understanding of the molecular and structural processes at the interfaces between solids and reactant gases. A detailed understanding of the correlation between the chemistry, structure, and function in these materials requires a multimodal investigation. Over the past few years, scientists at the at the Advanced Light Source, the Berkeley synchrotron facility, have developed a unique setup attached to an X-ray beamline where chemically-sensitive ambient pressure X-ray photoelectron spectroscopy (APXPS) and structure-sensitive grazing incidence X-ray scattering (GIXS) experiments can be performed simultaneously Here, we showcase that this tool can provide mechanistic insights that are unparalleled in the literature.

Our novel approach allows us to probe the changing surface and bulk chemistry, and surface and bulk structure of the model ceria catalysts in the presence of H_2 and CO_2 gases. In addition to the method itself, the electron density, surface chemistry, and roughness trends observed in ceria during the reaction will be discussed in this talk. Access to such a variety of data from working catalysts in a single experiment can have far-reaching implications, because changes in surface roughness, ability to store hydrogen in the bulk in various forms, and the chemical state of the surface, which all depend on the reactive environment, can directly affect the catalyst performance.

Inoperando nanoARPES on van der Waals heterostructures

Jyoti Katoch[1], Aalok Tiwari[1], Sandy Adhitia Ekahana[1,2], Souvik Sasmal[1], I-Hsuan Kao[1], Chris Jozwiak[2], Eli Rotenberg[2], Aaron Bostwick[2], Simranjeet Singh[1]

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Van der Waals (vdW) heterostructures offer an unprecedented platform for engineering the physical properties of two-dimensional (2D) materials through control of twist angle, strain, and environmental interactions. The advent of state-of-the-art angle-resolved photoemission spectroscopy with nanoscale spatial resolution (nanoARPES), combined with its ability to probe fully functional devices, has opened new avenues for directly visualizing exotic electronic phenomena in these systems.

In this talk, I will present our work leveraging cutting-edge in-operando nanoARPES to directly map the electronic properties of vdW heterostructures and their functional devices. I will show the direct momentum-resolved evolution of flat-band dispersion in twisted bilayer graphene systems, systematically varying the displacement field and carrier density via electrostatic gating. Time permitting, I will show our recent work on studying the intrinsic electronic structure and tunability of electronic states in atomically thin Weyl semimetals (WSMs) like $TaIrTe_4$. Known to be a Weyl semimetal in the bulk form and a quantum spin Hall (QSH) insulator in the monolayer, we investigate the tunability of the electronic structure of bilayer $TaIrTe_4$ via electrostatic gating as well as alkali metal doping.

Keywords: bilayer $TaIrTe_4$, Inoperando nanoARPES, Twisted double bilayer graphene.

Acknowledgement: Work supported by the DOE award no. DE-SC0020323 and DE-SC0025490.

Inherent Co-ion Influence on Electric Double Layer Structure and Electrochemical Reactivity in the Hydrogen Evolution Reaction

Seth R. Anderson[1,2], Samuel G. Johnstone[1], Maximilian Jaugstetter[2], Aaron M. Dent[1], Beichen Liu[1,3], Wenxiao Guo[1,4], Miquel B. Salmeron[2], & Matthew A. Gebbie[1]

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[4] Tulane University, Department of Chemistry, New Orleans, LA, USA

The formation of an electric double layer is ubiquitous in electrolytes undergoing electrochemical reactions. Currently, models for the double layer rely on the assumptions of dilute electrolyte theory where ions in solution do not interact. Dilute electrolyte theory predicts that solely hydrated counterions, or ions opposing the surface charge, occupy the near surface region and impact reactivity. Since this theory does not account for conditions like high applied potentials and high ion concentrations, understanding the relationship between double layer structure and electrochemical reactivity is critical for opening new domains for electrochemical modulation.

Notably, our recent work shows that co-ions, or ions with the same charge as the surface, can also have a definitive influence on reactions, particularly under condition of high applied potential, even at bulk concentrations as low as 5 mM. The hydrogen evolution reaction was used as a model system to explore co-ion effects in electrocatalytic reactions. We systematically vary co-ion identity and track hydrogen evolution reactivity as a function of concentration. We highlight the universal nature of the co-ion influence on hydrogen evolution at large potentials of -1.9 V and reveal that its impact on hydrogen evolution can outsize that of the cation in many circumstances. In particular, we focus on the unique ability of the tetrafluoroborate co-ion to undergo dynamic ligand exchange between hydroxide and fluoride. This allows tetrafluoroborate to act as a hydroxide sink to stabilize the hydrogen evolution byproduct. Furthermore, we used X-ray absorption spectroscopy to probe the electrode-electrolyte interface of tetrafluoroborate solutions which reveals markers of clustering and a surprisingly unperturbed water structure in the electric double layer. Lastly, sum-frequency generation spectroscopy reveals how co-ions influence interfacial hydrogen bond networks, which can have a significant impact on aqueous electrochemistry. Our results highlight how co-ions influence double layer structures and have significant implications for tuning other aqueous reactions such as carbon dioxide and nitrate electroreduction.

Keywords: Electric double layer, electrolytes, co-ion, anion, hydrogen evolution reaction, interfaces, water structure, catalysis.

Acknowledgments: Work supported by the National Science Foundation (NSF). Work supported by the U.S. DOE Office of Science Graduate Student Research (SCGSR) program. The SCGSR program is administered by the Oak Ridge Institute for Science and Education for the DOE under contract number DE-SC0014664.

Observation of the effect of surface chirality induced by chiral molecular overlayer on achiral substrates

F. Nishino[1], K. Fukutani[1], J. Brandhoff[2], E. Fuerch[2], F. Otto[2], M. Grünewald[2], M. Schaal[2], J. Picker[3], R. Forker[2], Z. Zhang[4], D. Stelter[2], A. Turchanin[3], T. Hirose[4], T. Fritz[2], S. Kera[1]

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Symmetry is among the most important factors of materials, which governs their physical properties. For this reason, lowering of materials' symmetry by external manipulations has been actively utilized in exploring novel properties of matters, for example seen in twisted stacking or strain induction in layered materials, such as transition metal dichalcogenides, to name a few[1].

In this regard, it is also well known that the materials' symmetry near the surface can readily be reduced by two-dimensional crystallization of *chiral* molecules on an achiral substrate[2] (surface chirality), which has been utilized for an enantio-selective catalysis in chiral chemistry. Furthermore, such a chiral-modified surface is regarded as an ideal platform to study chirality-induced spin selectivity (CISS), which has attracted a great deal of attention in recent years[3]. Thus, it is not only important to expand our abilities to control the chiral molecular crystallization, but also much deeper understanding of how such chirality interacts with the properties of the substrates is required to uncover the true, yet unknown, roles of chirality in two-dimensional materials.

In this study, an enantiopure chiral molecule thiadiazole-[9]helicene (TD[9]H) monolayer has been fabricated on Au(111), and probed using high-sensitivity low-energy electron diffraction (LEED) and scanning tunneling microscopy (STM) to study the potential landscape at their interface. Our analysis clearly revealed that TD[9]H molecules on Au(111) form an overlayer *without* commensurability, while still exhibiting substrate-defined surface chirality. Since the latter has only been reported in previous studies for the limited cases of commensurate overlayers, our findings serve to extend the possibility of fabricating well-defined surface chirality into a much broader spectrum of molecule-substrate combinations[4].

Surface chirality was then formed on monolayser (ML)-WS_2/Au(111) and ML-$Mo_{0.18}W_{0.82}S_2$/Au(111), hosts of highly spin-polarized electrons. We have successfully induced the surface chirality on the substrate, and the influence of the surface chirality was explored by measuring the band structure before and after the formation of surface chirality, using ARPES. Our results directly elucidate the changes in the band structure of the substrate, and indicate the chiral molecular adsorption likely induces an enhancement of the spin-polarized electron lifetime.

Keywords: Surface chirality, Low-energy electron diffraction, Angle-resolved photoelectron spectroscopy, Scanning tunneling microscopy

[1] S. Huang *et al.*, Photon. Insights **2**, R03 (2023). [2]K. Ernst, Phys. Status Solidi B **249**, 2057 (2012). [3]B. P. Bloom *et al.*, Chem. Rev. **124**, 1950 (2024).[4]F. Nishino *et al.*, Appl. Phys. Express **18**, 015502 (2025).

Ambient Pressure Spectroscopy (Island)
ThI_11 - 02:50 PM - 03:10 PM

Operando HAXPES Study of Solid Electrolyte Interphase Formation in Anode-Free Solid-State Batteries with PEO:LiDFOB

Yi-Chen Weng[1], Edvin Andersson[2], Helena Wagner[1], Mohammad Baghban Shemirani[2], Zora Chalkley[3,4], Elmar Kataev[3,4], Andreas Lindblad[1], Maria Hahlin[1,2]
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Anode-free solid-state batteries (AFSSBs) show improved capacities and safety compared to conventional Li-ion cells. However, the development of stable electrolytes is needed for achieving better cyclability. Our recent study has demonstrated the potential of polyethylene oxide (PEO)-based solid polymer electrolyte containing lithium difluoro(oxalate)-borate (LiDFOB) in AFSSB, showing promising capacity retention.[1] In this work, we applied *operando* hard X-ray photoelectron spectroscopy (HAXPES) to investigate the solid electrolyte interphase (SEI) at the anode|SPE interface in AFSSB with PEO:LiDFOB, as illustrated in Figure 1a. Specifically, we applied voltages in steps from 3.4 V to -0.1 V vs Li/Li$^+$, covering the SEI formation potentials, while simultaneously probing decomposition and lithium plating at the interface. In the Li 1s and C 1s spectra (Figure 1b and 1c), a relative shift in binding energy between the components in the SEI and the current collector at different potential steps is observed, which is caused by the formation of double-layer at the interface. This approach allows us to determine the potential at which PEO:LiDFOB begins to degrade and to identify the decomposition products. Our results indicate the correlation between the SEI formation and the improved capacity retention, which is benefit to the understanding of electrolyte stabilty and further development of AFSSBs.

[1] Andersson, E.K., et al. "*Initial SEI formation in LiBOB-, LiDFOB-and LiBF$_4$-containing PEO electrolytes*", Journal of Materials Chemistry A, 12(15), pp.9184-9199 (2024).

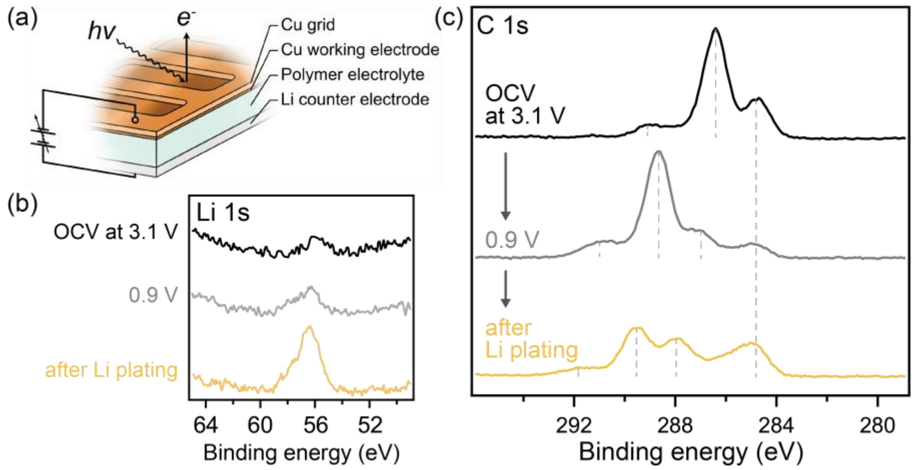

Figure 1. (a) Illustration of *operando* measurements. (b) Li 1s and (c) C 1s spectra at different potentials.

Depth-dependent magnetic properties in the ferromagnetic van der Waals semiconductor VI_3

Giancarlo Panaccione[1], Alessandro De Vita[1,2], Roberto Sant[3,4], Gian Marco Pierantozzi[1], G. M. Vinai[1], Tai Kong[5,6], Nicholas B. Brookes[4], Giorgio Rossi[1,2], Robert J. Cava[6]

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Two-dimensional van der Waals (vdW) magnetic semiconductors display emergent chemical and physical properties arising from both intra-layer and inter-layer interaction, both providing a versatile toolkit for studying quantum phenomena in hetero-structures and few layers systems, as well as holding promise for novel quantum spintronic functionalities [1,2]. Among 3d transition-metal vdW, Vanadium tri-iodide has recently attracted significant attention, as due to a) the presence of both structural and magnetic transitions as a function of temperature, suggesting a relevant role of magneto-elastic interactions and b) the interplay of dimensionality with relevant interactions, such as spin–orbit coupling (SOC), where a crossover of 3D vs. 2D electronic properties is expected [1]. However, detailed experimental information on ground state electronic properties as well as on spin/orbital degrees of freedom are still lacking, mainly due to its extreme air sensitivity and challenging chemical environment. Here we present chemical and layer sensitive X-ray electron spectroscopies results supported by model calculation, where we report via PhotoElectron Spectroscopy (RESPES and ARPES) a complete characterization of the electronic ground states in Vanadium tri-iodide, showing that orbital filling drives the stabilization of a quasi-Mott insulating state at the surface, with strong influence of dimensionality effects [3]. Moreover, Temperature-dependent X-ray Absorption Spectroscopy (XAS) and X-ray Magnetic Circular Dichroism (XMCD) experiments clearly reveal a reduced dimensionality of magnetic order due to electronic correlations, providing evidence of both an unquenched orbital magnetic moment (up to 0.66(7) μB/V atom) in the ferromagnetic state and (an instability of the orbital moment in the proximity of the spin reorientation transition [4,5]. Our results have direct implications in band engineering and layer-dependent properties of 2D systems, suggesting VI_3 as a candidate for the study of orbital quantum effects in spintronics.

Keywords [optional]: Quantum Spintronics, van der Walls 2D magnetism

[1] M. Gibertini, et al., Nature Nanotechnology 14, 408 (2019).
[2] B. Huang et al. Nature 546, 270 (2017).
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Ambient Pressure Spectroscopy (Island)
ThI_12 - 03:10 PM - 03:30 PM

In Situ and *Operando* Characterization of Electronic and Chemical States of WO_3 During Electrochemical Proton Intercalation.

Jay R. Paudel[1,2], Aaron J. Kaufman[1,2], James R. McKone[3], Ethan J. Crumlin[1,2]

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Proton intercalation in oxide materials plays a crucial role in various chemical applications, including sensors, fuel storage, battery materials, and electrochromic devices. Tungsten trioxide (WO_3) serves as a stable host for proton intercalation, forming tungsten bronzes that contribute to the hydrogen evolution reaction (HER). While numerous studies have demonstrated catalytic enhancements in WO_3 via proton-coupled electron transfer (PCET), the evolution of its electronic and chemical states during proton intercalation and HER remains poorly understood. In this work, we employ *in situ* and *operando* ambient pressure X-ray photoelectron spectroscopy (AP-XPS) to investigate the electronic and chemical transformations in WO_3 throughout the intercalation process and HER under applied reducing potentials. Our findings reveal that PCET governs the intercalation dynamics, where W is progressively reduced due to electron transfer and subsequent proton incorporation into oxygen lattice sites, forming hydroxyl species. As HER proceeds, these intercalated protons act as a reservoir, facilitating the reaction mechanism. We will present a detailed analysis of how the electronic and chemical states evolve during proton/electron intercalation and HER, offering new insights into the catalytic functionality of WO_3.

Keywords: Proton intercalation, solid-liquid interface, oxide materials, ambient pressure photoelectron spectroscopy (APXPS), materials characterization

Acknowledgement: This research used resources of the Advanced Light Source, which is a DOE Office of Science User Facility, under contract no. DE-AC02-05CH11231. Work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under contract no. DE-SC0023465.

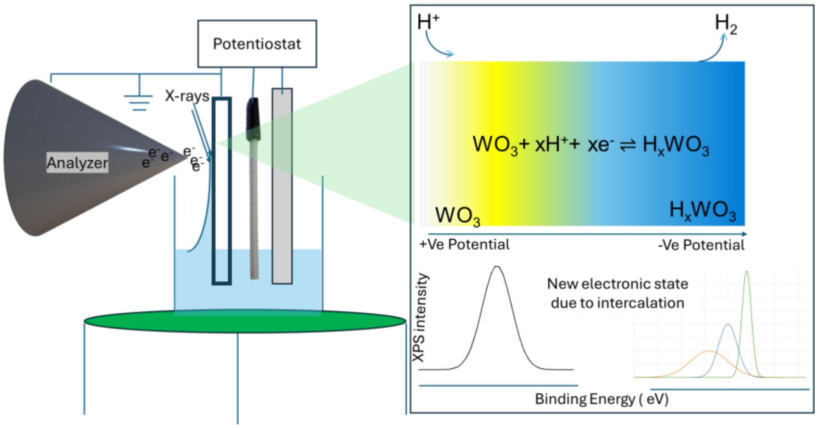

Figure: Schematic of the dip-and-pull setup for operando XPS measurements of WO_3 during proton intercalation, capturing the evolution of H_xWO_3 and associated electronic structure changes.

Flattened Band and AA+AB on bilayer graphene heterostructure

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Stacking two-dimensional(2D) materials to form van der Waals (vdW) heterostructures provides an unprecedented ability to tune electronic structures and engineer novel interfaces. Among these materials, α-$RuCl_3$ has recently attracted significant attention as a Mott insulator, and interfacing with graphene induces massive charge transfer from the Graphene layer to itself, significantly modifying graphene's electronic properties. By employing angle-resolved photoemission spectroscopy with nanoscale spatial resolution (nanoARPES), we study the electronic structure of bilayer graphene (BG) when interfaced with α-$RuCl_3$ and hexagonal boron nitride(hBN) and further tune the Fermi level of the heterostructure via potassium atom deposition. Our experiment reveals that the graphene Dirac band is flattened and gapped (Δ~0.4 eV) at the area of BG/hBN while the BG/$RuCl_3$/hBN reveals the presence of semi-metallic states at Dirac point as a result of AA+AB stacking.

Keywords: bilayer graphene, ARPES, flat band.

Acknowledgement: Work supported by the DOE award no. DE-SC0025490.

Ambient Pressure Spectroscopy (Island)
ThI_14 - 04:00 PM - 04:20 PM

Insitu photoelectron spectroscopy reveals the structure of the electrical double layer

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Energy materials, such as catalysts and battery electrodes, operate through their interfaces, because interface dynamics controls mass and energy transport from liquid to solid[1]. Therefore, characterizing these interfaces and their dynamic chemistry is crucial to tailor materials to their specific applications. Recently, interfacial spectroscopy[2] and molecular dynamics calculations[3] have shown that chemical and colloid interactions play a similar pivotal role at these solid-liquid interfaces at the most common electrolyte concentrations as the widely accepted models based on electrostatics. We have employed soft X-ray based interfacial spectroscopy, such as ambient pressure X-ray photoelectron spectroscopy (APXPS) and X-ray absorption total electron yield (XAS-TEY), to investigate the chemical composition and structure of aqueous electrolytes adjacent to a conductive solid phase[4]. With a focus on the effects of cations on these solid/liquid interfaces we studied the behavior of aqueous electrolytes on hydrophobic electrodes.

We found that the interfacial water structure strongly depends on hydrophobic interactions between the electrode material and the dissolved ions. Our results suggest that these interactions can lead to the separation of hydrophilic and hydrophobic ions, triggering the reorientation of water in the formed electric fields or its displacement by adsorbed ions.

Acknowledgement : Work supported by the Office of Basic Energy Sciences (BES) under contract mo. DE-AC02-05CH11231, FWP CH030201 (Catalysis Research Program).

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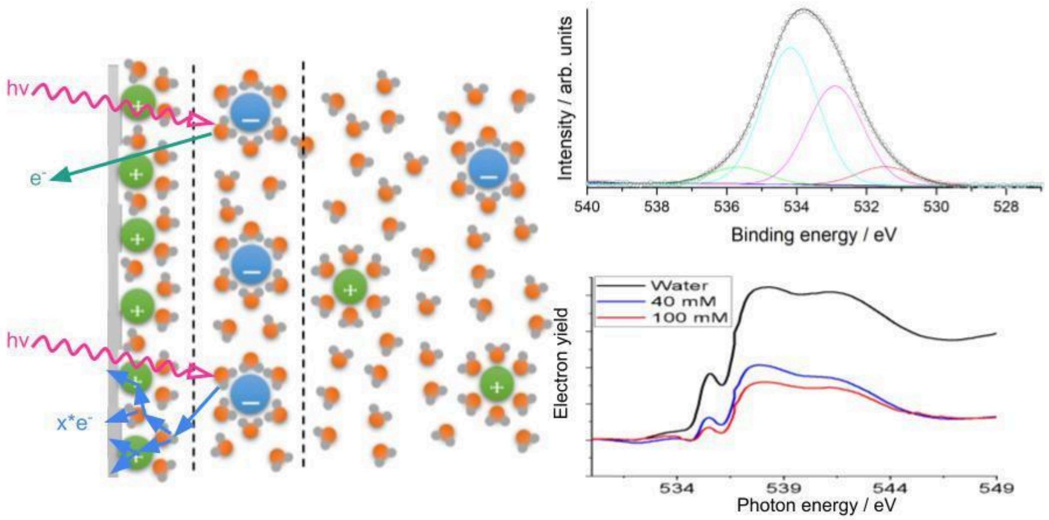

Figure 1. Soft X-ray excited photoelectrons reveal the electronic structure of interfacial water

Interface electronic and magnetic states and band dispersion of insulator/ferromagnet heterostructures studied by hard x-ray angle-integrated and angle-resolved photoemission

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Hard x-ray photoelectron spectroscopy (HAXPES) combined with x-ray total reflection (TR) allows us to depth-dependent electronic structures of solids [1]. Surface band bending of GaN was successfully observed in TR-HAXPES [1]. To further explore the application of TR-HAXPES, we applied this method to probe insulator/ferromagnet heterojunctions, which are of importance in the performance of magnetoresistance devices. For a $AlO_x/Co_2MnSi(001)$ structure, we found that the near-interface Co and Mn magnetizations underneath the AlO_x layer obtained by TR-HAXPES are reduced to ~77% compared to the bulk region obtained by non-TR-HAXPES for Co_2MnSi at room temperature, revealed by magnetic circular dichroism (MCD) in core-level HAXPES with the photon energy of 5.95 keV. In addition, the valence band structure of the near-interface Co_2MnSi was different from that of the bulk Co_2MnSi due to the enhanced spin-wave excitation near the interface [2]. The changes in the valence band structure in Co_2MnSi for both near-interface and bulk regions were also observed in the temperature-dependent valence band HAXPES [3], suggesting the importance of thermally induced spin fluctuations in Co_2MnSi at finite temperature [4]. Furthermore, we adapted the TR technique to band dispersion measurement for a MgO/Fe(001) heterojunctions by means of hard x-ray angle-resolved photoemission spectroscopy (HARPES) [5]. HARPES with the photon energy of 3.29 keV at 20 K clearly exhibited band dispersion of Fe(001) underneath the MgO layer. The subtraction of bulk band dispersion of Fe obtained by non-TR-HARPES from the near-interface band dispersion obtained by TR-HARPES revealed the hidden band folding at the MgO/Fe interface, which did not occur in the ideal MgO/Fe(001) interface. Thus, HAXPES and HARPES combined with TR are useful for detecting buried interface electronic and magnetic states of heterojunctions.

Keywords: HAXPES, HARPES, insulator/ferromagnet interface, x-ray total reflection, buried interface, electronic and magnetic states, band dispersion.

Acknowledgement: Work supported by Data Creation and Utilization Type Materials Research and Development Project from MEXT, Japan (Grant Nos. JPMXP1122683430 and JPMXP1122715503).

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Liquid-jet Velocity Map Imaging

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Liquid-jet photoelectron spectroscopy (LJ-PES)[1] enables the direct study of the electronic structure of both solutes and solvents and has greatly advanced chemical analyses in aqueous solutions. The LJ facilitates in-vacuo continuous liquid-sample replacement and the detection of photoelectrons with minimal collisions with evaporating water molecules.

Velocity map imaging (VMI)[2] provides optimal photoelectron collection efficiency, generally with a full 4π-steradian collection range, enabling the collection of photoelectron spectra and photoelectron angular distributions (PADs) in a single measurement. While VMI is widely applied in the gas phase[3], its intriguing extension to liquid-phase studies (particularly on aqueous solutions) remains highly challenging. Major experimental and technical difficulties include the disturbance of the focusing electric fields due to the presence of the dielectric LJ, the background resulting from scattering of the photoelectrons within and around the (aqueous) solution, and the balance between the required high electric fields and the risk of electrical breakdown in a high-vapor-pressure environment.

We have recently overcome these critical technical issues and have successfully utilized our custom-designed LJ-VMI setup. This system offers a broad dynamic energy range, allowing for the detection of photoelectron kinetic energies up to approximately 40 eV. Following initial lab-experiments using laser and ultraviolet light sources, we present here our recent LJ-VMI results obtained at the bending-magnet beamline PM3 of the BESSY-II synchrotron-radiation facility. We demonstrate the ability to identify core-level and valence electron photoionization features of the solute and solvent, and determine the electron binding energies. The associated photoemission spectra and PADs from organic and volatile aqueous solutions are analyzed, with the goal of identifying the principal effects of a LJ on VMI-instrument performance.

We will discuss the next steps in our ongoing development of LJ-VMI, including perspectives for future applications towards near-ionization-threshold phenomena, as well as time-resolved photo-induced reactions and electron dynamics in (aqueous) solutions.

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Site-selective polar compensation of Mott electrons in a double perovskite heterointerface

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Double perovskite oxides (DPOs) with two transition metal ions ($A_2BB'O_6$) offer a fascinating platform for exploring exotic physics and practical applications. Studying these DPOs as ultrathin epitaxial thin films on single crystalline substrates can add another dimension to engineering electronic, magnetic, and topological phenomena. Understanding the consequence of polarity mismatch between the substrate and the DPO would be the first step towards this broad goal. We investigate this by studying the interface between a prototypical insulating DPO Nd_2NiMnO_6 and a wide-band gap insulator $SrTiO_3$. The interface is found to be insulating in nature. By combining several experimental techniques and density functional theory, we establish a site-selective charge compensation process that occurs explicitly at the Mn site of the film, leaving the Ni sites inert. We further demonstrate that such surprising selectivity, which cannot be explained by existing mechanisms of polarity compensation, is directly associated with their electronic correlation energy scales. This study establishes the crucial role of Mott physics in polar compensation process and paves the way for designer doping strategies in complex oxides.

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Scaling research: From photoelectron-ion coincidences to photocatalytic solar hydrogen production

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Solar energy harvesting and the clean energy transition in general are thematic research topics receiving a lot of attention today. For example, the understanding of catalytic processes in the time domain and intermediate state level urges the development of fundamental research tools for imaging and characterisation towards chemically, temporally and spatially resolved in-situ and operando methods. In the present we provide an overview on the physics lead hydrogen research at the University of Oulu and on the lates advancements in photocatalytic hydrogen production aka solar hydrogen as well as methodology development in the field of catalysis and materials sciences. Examples are given on the use and development of synchrotron based and laboratory scale techniques (PEEM, APXPS, TAP) in the characterisation of novel nanoscale catalytic systems. Focus of the presentation is on analytical tools development in integration of advanced mass-spectroscopy and gas-phase electron spectroscopy with low-pressure steady-state and transient kinetic measurements. Examples are given on identification of isomeric-selective experiments and isomer discrimination of catalytic process during DME conversion.

This interdisciplinary work combines expertise in spectroscopy and applied physics from the University of Oulu and catalytic materials and reactions from the University of Oslo. The experiments and development is supported by the MAX IV Laboratory in Lund, Sweden.

Acknowledgement: Work is supported by the Research Council of Finland, Strategic Research Council within the Research Council of Finland decision 358422, JustH2Transit and European Research Council ERC

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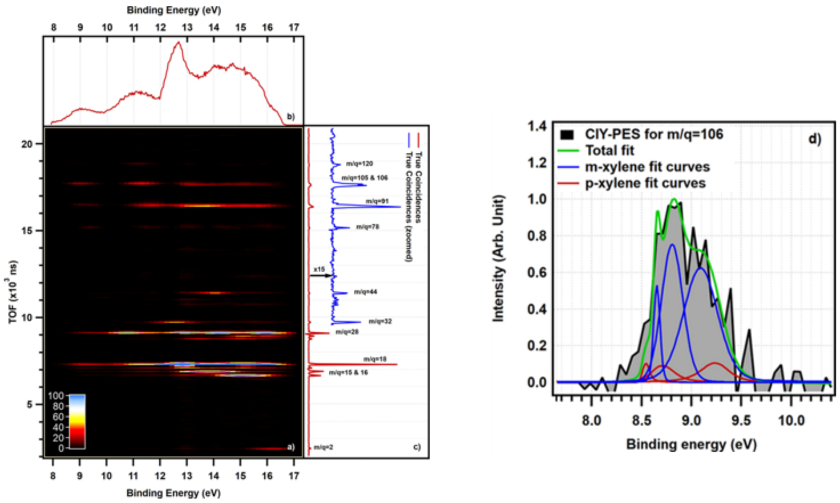

Figure 1. Isomer selective spectroscopy of xylene

Direct Observation of a Thickness-Induced Mott Transition in $LaTiO_3$ films

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Engineering quantum materials into dimensionally-confined heterostructures allows for control of their structural and electronic properties. These modifications may influence many-body phenomena that have a crucial impact on fundamental research and numerous applications. In this study, we observed strong electron-electron and electron-boson interactions in monolithic, thickness-varied $LaTiO_3/SrTiO_3$ heterostructures grown by pulsed laser deposition. Quasi-two dimensional $LaTiO_3$, a Mott insulator in the bulk, exhibits a tetragonal structure due to interface stress from the underlying $SrTiO_3$ layer. We observed a delayed Mott transition, where metallic states with in-plane dominated orbitals at lower thicknesses transition into Mott insulating states at higher thicknesses, as evidenced by the appearance of a lower Hubbard band below the Fermi level using angle-resolved photoemission spectroscopy, which is explained by the evolution of the electron-electron interaction. These discoveries and the visualization of the Mott transition expand the understanding of Mott physics and provide valuable insights for electronically reconfigurable devices.

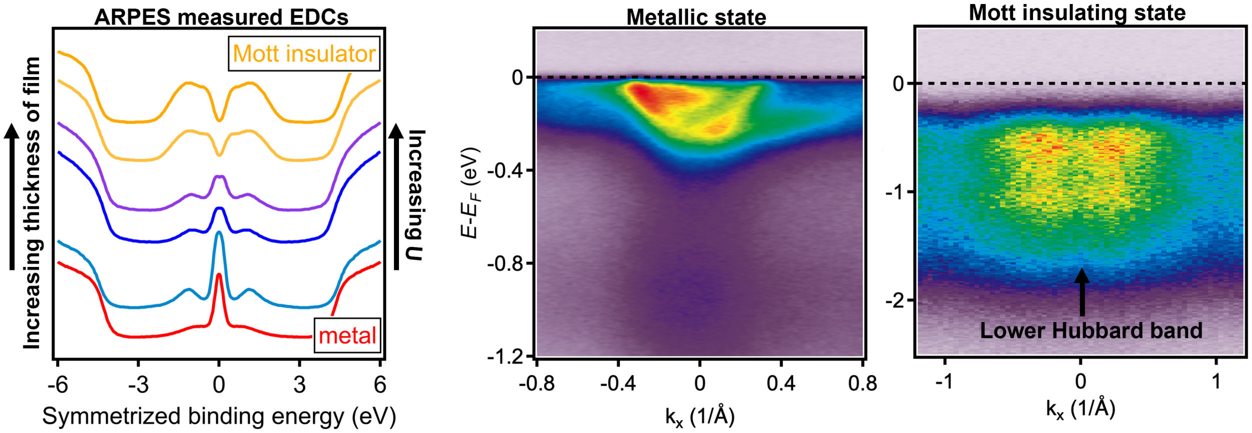

Figure 1. Mott transition in $LaTiO_3/SrTiO_3$ heterostructures

nanoXPEEM: Attention Based Nanoscale X-ray Photoemission Electron Microscopy

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Harnessing two-dimensional materials for practical applications faces significant hurdles due to a dearth of characterization methods that combine efficiency with high throughput. Soft X-ray time-of-flight photoemission electron microscopy (XPEEM) offers valuable element-specific and depth-sensitive analysis of materials and their hidden interfaces. However, this technique suffers from chromatic and spherical aberrations that cannot be eliminated using traditional electron-lens systems. These distortions, along with astigmatism and space-charge effects, substantially reduce both spatial and energy resolution. We propose a solution using a spatial-attention deep learning approach that automatically reduces these distortions, achieving nanometer-level resolution across the entire observation area. This enhanced approach, titled nanoXPEEM, sets a new benchmark of 48-nm spatial resolution with a 232-μm diameter field of view in the soft x-ray energy range (700-1000 eV). The nanoXPEEM technique enables unprecedented uniform nanoscale mapping of elemental composition, depth profiles, and local structural features, potentially even bridging the gap toward atomic-level resolution capabilities.

Translational symmetry-breaking in the quasiparticle band structure of organic semiconductor rubrene

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As the sustainability of our global resources and energy-harvesting rapidly becomes a central issue in navigating our technological advancements, the development of quantum devices made of organic molecular semiconductor materials, which combine the low fabrication cost, abundancy of the constituent elements and high bio-degradability, while maintaining their high performance is of paramount importance.

Among such organic semiconductors, single crystalline rubrene ($C_{42}H_{28}$) is known to exhibit exceptionally high carrier mobility (~40 cm^2/Vs [1]), and is at the heart of current organic electronic devices. At room temperature, rubrene crystal is known to support partially coherent polaron-band transport through the itinerant holes. Clearly, in such case, the understanding on the nature of these carriers in wavevector space is a prerequisite for any first-principle-based, strategic developments utilizing this material, such as direct band-structure engineering. However, despite the extensive efforts devoted to uncover the entirety of electronic band structure of rubrene crystal for more than a decade, both experimentally [2] and theoretically [3], there remain multiple major discrepancies, even at qualitative levels, between the theoretically predicted band structures and those identified in the experimental studies, leaving the true nature of the charge carriers highly elusive.

In order to resolve these issues, we have utilized photon-energy- and polarization-dependent angle-resolved photoemission spectroscopy (ARPES) to investigate the valence band structure of highly single-crystalline rubrene. Our results, elucidating the valence band structures in terms of their quasiparticle spectral functions over the multiple Brillouin zones (BZs), clearly reveal the mismatched periodicities in the band structure between the single-particle and quasiparticle pictures, which had hindered the observations of the full band structures in the previous studies. Taking proper accounts for these translational-symmetry breaking in rubrene crystal, we fully uncover its band structure, in agreement with theoretical simulations, thereby resolving the long-standing controversies on the rubrene's electronic structures.

Keywords: Organic molecular semiconductors, Angle-resolved photoelectron spectroscopy Quasiparticle spectral functions, Low-energy electron diffraction

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Photoemission Microscopy (Island)
ThI_18 - 05:20 PM - 05:40 PM

Spectro-microscopy of surface magnetization in Fe_3GeTe_2

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The spatial variation of the magnetic structure in Fe_3GeTe_2 has been in the past investigated by scanning tunneling microscopy, magnetic force microscopy, X-ray circular magnetic dichroism (XMCD), scanning electron microscopy with polarization analysis imaging, and Lorentz transmission electron microscopy. The domain pattern in the absence of an external magnetic field follows the predicted pattern observed for thick crystals with uniaxial magnetic anisotropy. Unlike the techniques mentioned above, Low-energy electron microscopy (LEEM) measurements with a tilted incident electron beam do not require special sample preparation, or a spin-polarized electron source, as in the case of spin-polarized LEEM experiments. Magnetic contrast can be observed by physically tilting the incident electron beam relative to the sample surface normal (or tilting the sample). Here we will show how the observed pattern can be interpreted to extract the domain wall type and determine critical exponents of the surface magnetism[1].

Keywords: van der Waals materials, surface magnetism, low-energy electron microscopy, X-ray magnetic circular dichroism.

Acknowledgement: This research used resources of the Center for Functional Nanomaterials and the National Synchrotron Light Source II, which are U.S. Department of Energy (DOE) Office of Science facilities at Brookhaven National Laboratory, under Contract No. DE-SC0012704.

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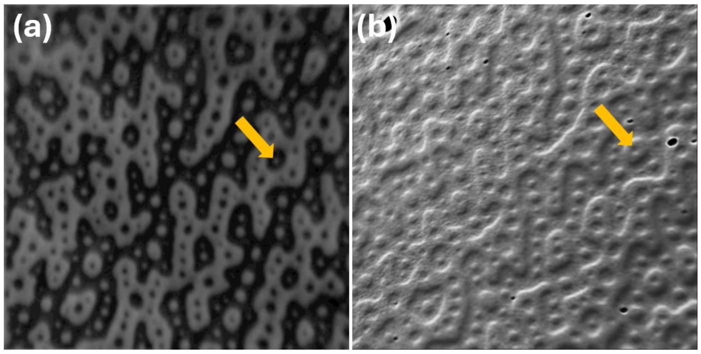

Figure 1. (a) XMCD-PEEM image taken at 110 K. The dark and bright regions correspond to up and down magnetization orientation relative to the sample normal. XMCD images were taken at the peak of the Fe L3-edge spectrum. (b) A tilted-beam LEEM image of the same area as in (a) showing a surface magnetic contrast at domain walls.

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Observation of surface-state transport by Mn doping into a topological crystalline insulator SnTe thin film with strain

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Topological crystalline insulators (TCIs) are topological insulators (TIs) whose surface states are protected by crystal mirror symmetry rather than time-reversal symmetry. In usual TIs, when magnetism is introduced, a gap opens in the Dirac band of the topological surface state [1], and the quantum anomalous Hall effect has been observed [2]. However, the relationship between magnetism and topological surface states in TCIs has not yet been fully elucidated.

Therefore in this study, we performed *in situ* ARPES measurements on SnTe films doped with magnetic Mn, which should be an ideal system for studying the relationship between topology and magnetism in TCIs. SnTe, a prototypical TCI material, has Dirac cones at the Γ and M points on the (111) surface [3]. In this study, a strained SnTe film was prepared on a Bi_2Te_3(111) thin film, and then Mn and Te were co-deposited for magnetic doping. Figure 1 shows ARPES intensity mapping at the Fermi level (top) and the band dispersion along the Γ–M direction (bottom) for (a) pristine SnTe and (b) Mn-doped SnTe, respectively. Since SnTe is known to be *p*-doped due to the Sn vacancies, one can find circular and star-like hole pockets around Γ and hole lobes along Γ-M for the pristine SnTe. In contrast, since Mn incorporation in SnTe induces electron doping, circular Fermi surfaces of surface-state electrons and holes were observed for the Mn-doped sample. We also measured the Hall effect by *in situ* four-probe transport measurements and it was revealed that the carriers in pristine SnTe are holes while a multiple-carrier transport with hysteresis was observed for Mn-doped SnTe. These features are reflecting the Fermi surface, and we can say that we have succeeded in forming a bulk-insulating magnetic SnTe film. The details will be discussed along with the results of atomic structure analysis using TEM-EDS.

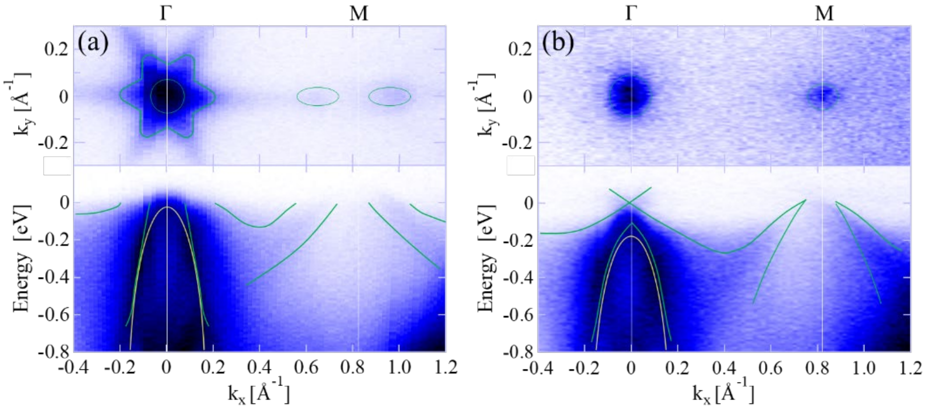

Figure 1: ARPES measurement results for (a) SnTe and (b) Mn-doped SnTe. Color guide: Yellow represents bulk bands, while green represents surface bands.

Acknowledgement: Work supported by JST SPRING, Japan Grant Number JPMJSP2106 and JPMJSP2180.

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An XPEEM Study of Voltage-induced Magnetic Domain Separation in a LaSrMnO Thin Film

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$La_{0.7}Sr_{0.3}MnO$(LSMO) is a transition metal oxide that is a ferromagnetic metal at low temperatures and a paramagnetic insulator above its Curie temperature. It exhibits both resistive and magneticswitching properties[1,2]. Unlike most phase-change resistive switching materials that transition from aninsulator to a metal at elevated temperatures[3-6], LSMO transitions from a metal to an insulator as thetemperature increases. Applying a voltage bias above a critical value to a fabricated LSMO thin film device caninduce phase separation, forming a paramagnetic insulating barrier. In this study, we employed X-ray magneticcircular dichroism-photoemission electron microscopy (XMCD-PEEM) to observe magnetic domainconfigurations as a function of bias voltage. For voltages exceeding a certain threshold, magnetic domainssegregate into two distinct regions: one displaying clear white/black contrast, indicative of well-defined micron-scale magnetic domains, and the other showing intermediate gray contrast, characteristic of regions withreoriented magnetization. Significant changes in magnetic domain configurations were observed only in thegray areas. Furthermore, this voltage induced phase separation was found to depend on bias polarity, with thegray area expanding from the opposite sample edge when the applied voltage was reversed. These findingsdemonstrate the intricate voltage-induced domain dynamics in phase-change materials.

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Unveiling a Unified Mechanism for Single-Photon Emission Across the III-Nitride

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Single-photon emission (SPE) is a fundamental phenomenon in future quantum information technologies. As wide-bandgap semiconductors, the III-nitrides —including hexagonal boron nitride (hBN), gallium nitride (GaN), and aluminum nitride (AlN)—have attracted significant interest for its room-temperature operation, strong emission intensity, tunable emission wavelength, and narrow linewidth[1]. However, the origin of SPE in these materials remains largely unknown. Photoluminescence (PL) spectroscopy studies have attributed SPE in nitrides to boron vacancies, gallium antisites, nitrogen vacancies, and other defect states. Yet, none of these mechanisms fully explain the wide range of emission peaks observed in PL data and their spectral wandering. In this study, we applied resonant inelastic X-ray scattering (RIXS) to hBN, GaN, and AlN and identified a series of harmonic peaks with an identical elementary energy of 285 meV across all three nitrides materials [see Figure 1 below]. Using the donor-acceptor pair (DAP) model and comparing with PL data, we determined that these harmonic peaks correspond to SPE features observed in PL. The presence of identical harmonic peaks related to SPE in hBN, GaN, and AlN suggests that SPE in these nitrides is not only linked to their crystal structures but rather our results indicate that molecular N_2 plays a pivotal role in the generation of SPE in these nitrides. This is evidenced by the behavior of the harmonic peaks and their fundamental energy matching a vibronic state of N_2. This discovery highlights the existence of a unified SPE mechanism across III-nitride compounds and suggests that this mechanism could potentially extend to other materials in the III-nitride family. By establishing a common origin for SPE in hBN, GaN, and AlN, this work advances the fundamental understanding of quantum photonics and provides a foundation for future studies on quantum emission phenomena based on molecular-like defects in wide-bandgap semiconductors.

Keywords [optional]: Single photon emission, resonant inelastic X-ray scattering, photoluminescence spectroscopy, wide-bandgap nitrides, N_2 molecule, donor-acceptor model.

[1] Pelliciari, J., Mejia, E., Woods, J.M. *et al.* Elementary excitations of single-photon emitters in hexagonal boron nitride. *Nat. Mater.* **23**, 1230–1236 (2024).

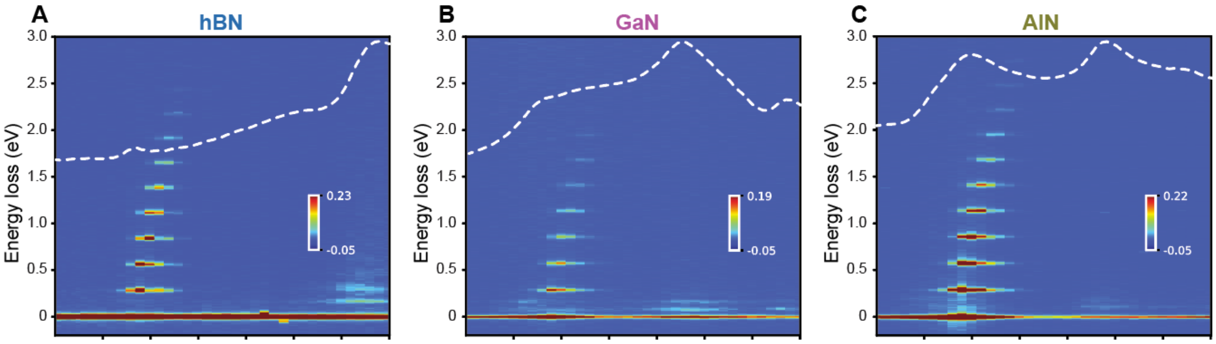

Figure 1: RIXS maps of hBN (A), GaN (B), and AlN (C) at the N-K edge. Harmonic peaks at 285 meV appear at the same incident energy of 401 eV across all three nitrides, while low-energy phonon excitations vary in energy depending on the material. The white dashed lines indicate the X-ray absorption spectroscopy signal.

Plenary Talks (Island)
FrI_01 - 08:30 AM - 09:15 AM

Abstract Not Available

Excitons in space and time - a femtosecond momentum microscopy study

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Transition metal dichalcogenides (TMDs) are an exciting model system to study ultrafast energy dissipation pathways, and to create and tailor emergent quantum phases. The versatility of TMDs results from the confinement of optical excitations in two-dimensions and the concomitant strong Coulomb interaction that leads to excitonic quasiparticles with binding energies in the range of several 100 meV. In TMD stacks consisting of at least two layers, the interlayer interaction can be precisely controlled by manipulating the twist angle: The misalignment of the crystallographic directions leads to a momentum mismatch between the high symmetry points of the hexagonal Brillouin zones. This strongly impacts the interlayer wavefunction hybridization, and, moreover, adds an additional moiré potential. Crucially, in this emergent energy landscape, dark intra- and interlayer excitons dominate the energy dissipation pathways. While these dark excitonic features are hard to access in all-optical experiments, time-resolved momentum microscopy [1] can provide unprecedented insight on these quasiparticles [2].

In the first part of my talk, I will present our results on the ultrafast formation dynamics of interlayer excitons in twisted WSe_2/MoS_2 heterostructures [3-7]. First, I will report on the identification of a hallmark signature of the moiré superlattice that is imprinted onto the momentum-resolved interlayer exciton photoemission signal. With this data, we reconstruct the electronic part of the exciton wavefunction, and relate its extension to the moiré wavelength of the heterostructure [3-5]. Second, I will show that interlayer excitons are effectively formed via exciton-phonon scattering, and subsequent interlayer tunneling at the interlayer hybridized Σ valleys on the sub-50 fs timescale [3,4]. Third, I will discuss our efforts to monitor the interlayer exciton formation dynamics with spatiotemporal resolution using femtosecond photoelectron dark-field microscopy [6]. Fourth, I will show how the strength of femtosecond momentum microscopy can also be applied to study optical excitations at hybrid organic/TMD heterostructures [7].

Finally, I will provide an overview on how coherent light-fields can be used to create Floquet-Bloch states in monolayer graphene [8].

[1] Keunecke et al., Rev. Sci. Ins. 91, 063905 (2020).
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[4] Bange et al., 2D Materials 10, 035039 (2023).
[5] Bange et al., Science Advances 10, eadi1323 (2024).
[6] Schmitt et al., Nature Photonics 19, 187–194 (2025).
[7] Bennecke et al., arXiv:2411.14993 (2024).
[8] Merboldt et al., arXiv:2404.12791 (2024).

Quantum Materials and Magnetic Phenomena Studied by Spin-Resolved ARPES: Theoretical perspectives

J. Minár[1]

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Quantum materials exhibit a complex interplay between electronic correlations, topology, and magnetism, placing them at the forefront of condensed matter physics and quantum technology. Understanding these systems requires disentangling spin-orbit coupling, electron-electron interactions, and magnetic fluctuations under realistic conditions, including finite temperatures and structural disorder. Spin- and time-resolved angle-resolved photoemission spectroscopy (STARPES) is a crucial technique for probing electronic and spin structures in magnetic and topological materials. However, quantitative interpretation of spin-ARPES data necessitates advanced theoretical models that accurately capture electronic states, spin textures, and dynamic responses to external fields.

I will present a theoretical framework based on the fully relativistic multiple-scattering Green function KKR method [1], effectively modeling spin-dependent photoemission. This approach includes correlation effects via dynamical mean-field theory (DMFT) [2] and describes spin fluctuations using the alloy analogy model [3]. I will also discuss advances in calculating light-induced electronic excitations [4], highlighting their relevance to spin-ARPES studies of topological and magnetic quantum materials.

A novel application is the one-step model of photoemission in studying altermagnets and kagome magnetic materials. Altermagnets, exhibiting unconventional time-reversal symmetry breaking without net magnetization, are explored in RuO2 and MnTe [5,6]. Spin-ARPES combined with the one-step model provides insights into lifted Kramers spin degeneracy, revealing their potential for spintronics. In kagome magnetic materials, persistent flat band splitting and selective band renormalization are observed in FeSn thin films [7], highlighting unique correlation effects and topological phenomena. These developments offer a comprehensive framework for exploring magnetic phenomena and spin dynamics in complex quantum materials.

References:

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[5] J. Krempaský et al., *Nature* 626, 517 (2024). DOI: 10.1038/s41586-023-06907-7
[6] O. Fedchenko et al., *Sci. Adv.* 10, eadj4883 (2024). DOI: 10.1126/sciadv.adj4883
[7] Z. Ren et al., *Nature Communications* 15, 9376 (2024). DOI:0.1038/s41467-024-53722-3

Acknowledgements

I would like to thank the Quantum Materials for Sustainable Technologies (QM4ST) project with Reg. No. CZ.02.01.01/00/22_008/0004572, cofunded by the ERDF as part of the MŠMT.

Electronic Spectromicroscopy (California)
FrII_04 - 10:30 AM - 11:00 AM

Strong Inter-valley Electron-Phonon Coupling in Magic-Angle Twisted Bilayer Graphene

The unusual properties of superconductivity in magic-angle twisted bilayer graphene (MATBG) have sparked enormous research interest. However, despite the dedication of intensive experimental efforts and the proposal of several possible pairing mechanisms, the origin of its superconductivity remains elusive. In this talk, using angle-resolved photoemission spectroscopy with micrometer spatial resolution, we show our observation on the replicas of the flat bands in superconducting MATBG unaligned with its hexagonal boron nitride (hBN) substrate, which are absent in non-superconducting MATBG aligned with the hBN substrate[1]. Remarkably, the replicas are evenly spaced in energy, separated by 150 ± 15 meV, signaling the strong coupling of electrons in MATBG to a bosonic mode of this energy. By comparing our observations to simulations, the formation of replicas is attributed to the presence of strong inter-valley electron-phonon coupling to a K-point phonon mode[1,2]. In total, the observation of these replica flat bands and the corresponding phonon mode in MATBG could provide important information for understanding the origin and the unusual properties of its superconducting phase [asdf].

1. *C. Chen, et. al., Nature 636, 342 (2024)*
2. *C.X. Liu, et. al., Phys. Rev. B 110, 045133 (2024)*

Topological and chiral phonons: theory, prediction and detection

Tiantian Zhang[1], et al.

[1] *Institute of Theoretical Physics, Chinese Academy of Sciences, Beijing, China*

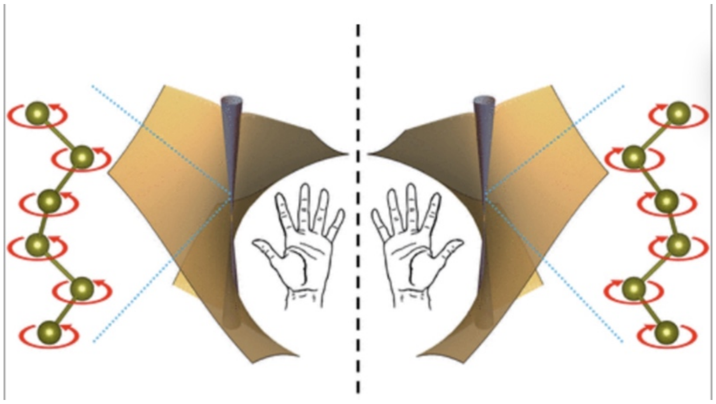
In solids, lattice vibrations—phonons at the terahertz scale—play a crucial role in thermal conductivity, transport properties, and phenomena like structural phase transitions and conventional superconductivity. However, their zero spin and electrical neutrality make phonons difficult to manipulate, limiting their control in physical processes. Recently, introducing *topological* and *chiral* degrees of freedom into the phonon spectrum has provided a new "control handle" for phonons, offering fresh insights into their influence on heat conduction, superconductivity, and other fundamental phenomena.

In the first half, I will discuss classifications and diagnostic methods for topological phonons, along with material predictions and experimental validations 1—including FeSi (the first topological phonon material) and BaPtGe (hosting Weyl phonons with the highest Chern numbers). The second half will cover chiral phonons: their definition, distinctions from topological phonons, and experimental detection 2. Examples include chiral phonons in systems with nonsymmorphic symmetries (e.g., α-HgS and Te), spin-phonon-induced chiral phonons in $Co_3Sn_2S_2$, and the interplay between Weyl and chiral phonons (see Figure).

[1] Zhang, et al., PRL, 120, 016401 (2018); PRL, 121, 035302 (2018); PRB, 102 , 125148 (2020); PRB, 103, 184301 (2021); PRL, 123, 245302 (2019)

[2] Zhang, et al., PRB , 4, L012024 (2022) ; Nat. Phys. , 19, 142-142 (2023); Nano Lett., 23, 7561–7567 (2023); arXiv:2411.03754 (2024); arXiv:2410.21775 (2024)

Figure 1. Relationship between topological phonons and chiral phonons



Electronic Spectromicroscopy (California)
FrII_05 - 11:00 AM - 11:30 AM

Tipping the Length Scales with ARPES

Chris Jozwiak, Eli Rotenberg, Aaron Bostwick

Advanced Light Source, Lawrence Berkeley Lab, Berkeley, CA, USA

The ability to directly probe electronic structure in energy-momentum space has made ARPES a critical spectroscopic tool for studying quantum materials. The high momentum end energy resolutions available in standard experiments enable it to be productive in a wide array of systems including metals, complex oxides, topological materials, and 2-dimensional materials. One particular shortcoming has been its traditional lack of high-spatial resolution, which has limited the impact of ARPES in untangling the complexities of heterogenous systems and in materials and geometries too small for successful measurement. Significant effort in the community has been spent developing spatially resolved ARPES capabilities to expand its impact with the intrinsic power of microscopy at various length scales. I will give an overview of these approaches at the MAESTRO beam line at the ALS, where we aim to maximize impact across the 10 um to 100 nm scales. I will also discuss several opportunistic doors these lengths scales open to ARPES, including in-operando measurements with applied electric and magnetic fields, current pulses, strains, and laser pulses, and present recent work with these techniques in 2D heterostructures and metastable phases in 1*T*-TaS_2.

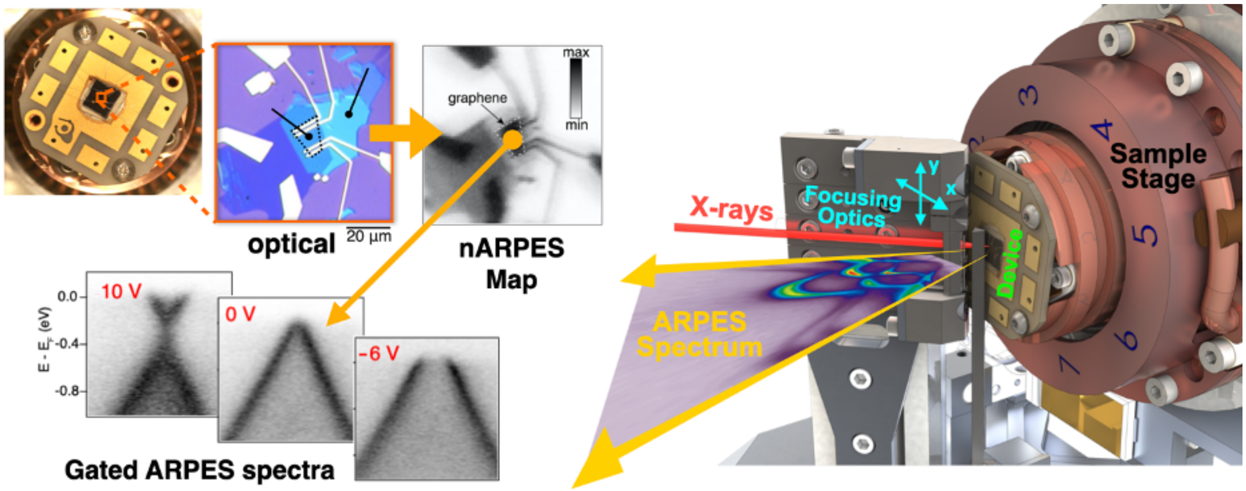


Fig: In-operando NanoARPES characterization of heterostructure device. Right: X-rays are focused onto the sample to beamspots ranging from 1 um to 100 nm, and scanned across the sample surface, collecting ARPES as a function of (x,y) sample positions on the sample surface. Left: An example sample, corresponding optical image of the 2D heterostructure device, corresponding nARPES spatial map, and ARPES-measured bandstructure of the center active material as a function of applied gate voltage [data from: R. Muzzio, et al., Phys. Rev. B 101, 201409(R), (2020)].

Poster Session TuI_17

P28: Observation of electronic structure of organic Mott insulator β’-(BEDT-TTF)2ICl2 by angle-resolved photoelectron spectroscopy

Page: 171

Nakazawa, Ryotaro

Correlation between non-collinear spin orientation and lattice distortion in $Ni_{0.4}Mn_{0.6}TiO_3$

Shang-Hsien Hsieh[1, 2], Surajit Ghosh[1], Yu-Hui Liang[1], Hsiao-Tsu Wang[1, 3], Chao-Hung Du[1], Jau-Wern Chiou[4], Chun-Ming Wu[2], Chin-Wei Wang[2], Yu-Cheng Shao[2], Jeng-Lung Chen[2], Chih-Wen Pao[2], Huang-Ming Tsai[2], Ting-Shan Chan[2], Wen-Bin Wu[2], Hong-Ji Lin[2], Jyh-Fu Lee[2], Asokan Kandasami[5] and Way-Faung Pong[1]

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[2]National Synchrotron Radiation Research Center, Hsinchu 300, Taiwan
[3]Bachelor's Program in Advanced Materials Science, Tamkang University,
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[5]Department of Physics & Centre for Interdisciplinary Research, University of Petroleum and Energy Studies (UPES) Dehradun, Uttarakhand 248007, India

This work studies the correlation between non-collinear spin orientation and lattice distortion in XY-like spin-glass (SG) $Ni_{0.4}Mn_{0.6}TiO_3$ (NMTO) using neutron diffraction and synchrotron-radiation-based spectroscopic techniques such as, resonant inelastic X-ray scattering (RIXS), X-ray absorption spectroscopy and X-ray magnetic circular dichroism (XMCD). The neutron study reveals a shorter spin-spin correlation length along the ***c***-axis than in the XY-plane, which is typical of XY-like SG. It also reveals that the $NiTiO_3$ matrix dominates the shaping of the low-temperature complex magnetic structure of NMTO. A local lattice distortion in the form of a large increase in the nearest-neighbor Ni-O bond length along the ***c***-axis was observed from temperature-dependent extended X-ray absorption fine structure measurements close to the SG transition temperature, $T_{SG.}$ The Ni L_3-edge RIXS spectra at around T_{SG} indicate a possible magnon excitation with various *d-d* interactions and charge transfer excitation. The Ni $L_{3,2}$-edge XMCD measurement shows ferromagnetic/ferrimagnetic-like behavior at the temperature near T_{SG}, closely matching the opening of hysteresis curves that are recorded below T_{SG}. These findings compellingly demonstrate that at low-temperature, Ni^{2+} ions in the XY-plane of NMTO exhibit a non-collinear spin alignment, that facilitates Dzyaloshinskii-Moriya-type exchange interactions, elucidating the complex spin dynamics of this SG system.

Keywords: spin-glass, X-ray magnetic circular dichroism, non-collinear spin orientation, Dzyaloshinskii-Moriya interaction.

Acknowledgement: Work supported by the National Science and Technology Council (NSTC) of Taiwan for providing financial support for research under the projects NSTC 113-2112-M-032-011, 112-2112-M-032-009-MY2 and 113-2112-M-213-012-MY3. We thank Prof. Tsuyoshi Kimura for providing the sample for this study. Dr. Shih-Wen Huang of Swiss Light Source is also appreciated for consulting the RIXS spectra used in this work. The authors would also like to express their heartfelt gratitude to Dr. Ravindra Singh Solanki (deceased) for his invaluable contribution to this work.

[1]S. H. Hsieh *et al*., "*Correlation between non-collinear spin orientation and lattice distortion in* $Ni_{0.4}Mn_{0.6}TiO_3$", Physical Review Materials 8(12), 124410 (2024).

Temperature-dependent electronic structure of epitaxially-grown altermagnet MnTe

Sung-Kwan Mo[1] and Ji-Eun Lee[1,2]

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[2] *Department of Physics, University of California, Berkeley, CA, USA*

Altermagnetism is a newly proposed magnetic order that exhibits unconventional time-reversal symmetry breaking and spin-split electronic band structure despite the compensated net magnetization. MnTe is one of the most prominent candidate materials whose unique crystal structure and spin arrangement can host altermagnetism. We present a fully momentum-resolved electronic structure of epitaxially grown MnTe and its temperature evolution, measured by angle-resolved photoemission spectroscopy (ARPES). Our photon-energy-dependent ARPES data reveal strong out-of-plane dispersions that align with theoretical predictions. More interestingly, we identify two distinct temperature-dependent band evolutions depending on the momentum positions: energy shifts at nodal planes and spectral weight suppression at off-nodal planes. These findings suggest the importance of considering local moments and momentum-dependent interactions for a deeper understanding of altermagnetism.

Bidirectional magnetization switching in ferrimagnetic TmIG by adsorption of single-handed chiral materials

Yu-Cheng Shao [1], Wei-Hsiang Liao [2], Joshua S. Webb [3], Yao Zhang [4], Yu-Ying Chang [2], Simon Granville [4], Muhammad Hanif [3], and Hua-Shu Hsu [2]

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Recent studies have demonstrated that magnetization switching in ferromagnets can be achieved through adsorbed chiral molecules without the need for current or external magnetic fields, offering a low-power mechanism for applications in spintronic devices. We selected ferrimagnetic TmIG for our research because it has two opposing magnetic sublattices and perpendicular magnetic anisotropy. Our results revealed that magnetization switching in both directions of TmIG could be achieved using Cu metallopolymer of D-leucine. This underscores the significance of Cu-induced spin-orbit coupling in magnetization reversal by chiral molecule adsorption. Through circular dichroism and X-ray absorption analysis, we determined that switching of two different magnetic orientations is associated with interactions with the two distinct sublattices, octahedral and tetrahedral sites. Our research confirms the direct switching of magnetization in ferrimagnetic TmIG by single-handed chiral molecules. We further elucidate the interactions between chiral molecules and the sublattices of TmIG, broadening the application scope of chiral molecule adsorption-induced magnetization flipping.

Keywords [optional]: Circular Dichroism, X-ray Absorption

Unraveling band structures of Rh_2MnGa thin films by synchrotron radiation ARPES

Tatsuya Tomida[1], K. Nakanishi[1], M. Kakoki[1], K. Kuroda[1,2], K. Miyamoto[3],
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The anomalous Nernst effect (ANE), in which an electromotive force is generated perpendicular to the thermal gradient in magnetic materials, has attracted considerable attention. Recently, a large ANE accompanied by a thermoelectric power of 6~8μV/K—the highest reported to date—was observed in the Heusler compound Co_2MnGa [1,2]. Theoretically, this significant ANE has been attributed to an intrinsic mechanism arising from the materials's topological band structure, namely the large Berry curvature associated with line nodes and Weyl points near the Fermi level. However, for practical applications, a thermoelectric power exceeding 10 μV/K is generally required, and the development of higher-performance materials remains a challenge. Recent first-principles calculations predict that the absolute value of the thermoelectric power in Rh_2MnGa is approximately twice that of Co_2MnGa. Moreover, it has been reported that Rh_2MnGa exhibits thermoelectric powers of opposite signs, depending on the sign of the Berry curvature in momentum space [3]. To investigate the origin of this unusual ANE in Rh_2MnGa, we performed angle-resolved photoelectron spectroscopy (ARPES) using synchrotron radiation at beamline BL-9B of HiSOR. The thin film sample, prepared at NIMS, was transferred to the ARPES system at HiSOR *via* a vacuum suitcase chamber. Isoenergy surface measurements taken at a photon energy of 55 eV revealed a V-shaped structure opening along the Γ-K direction of the Brillouin zone near the Fermi level, which reversed its opening direction when moving away from the Fermi level. This behavior indicates the presence of energy-dependent line nodes composed of linearly dispersing bands. Our findings offer new insights into the relationship between line nodes and the ANE in Rh_2MnGa, and contribute to the ongoing development of high-performance thermoelectric materials.

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[3] J. Noky *et al.*, npj Compt. Mater. **6**, 77 (2020).

Termination-dependent Surface States in the Topological Kagome Metal GdV_6Sn_6

Robin P. Forster[1], Honey Boban[1], Mohammed Qahosh[1], Xiao Hou[1], Yishui Zhou[2], Yixi Su[2],
Claus M. Schneider[1] and Lukasz Plucinski[1]

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The magnetic Kagome metal GdV_6Sn_6 combines phenomena such as flat Kagome bands [1,2], local moments of the *4f* electrons of Gd, and itinerant V *3d* electrons [3], in a single material, providing a platform to study their complex magnetic and electronic interactions. We have performed termination-dependent micro-ARPES measurements with 20 μm real-space resolution at 20 K of paramagnetic GdV_6Sn_6 [4]. Surface and bulk states could be distinguished in the series of scans made over a range of photon energies. Subsequently, we have chosen hν of 80 and 130 eV for detailed ARPES scans. In addition, TEM measurements were conducted, which provide an insight into the crystal structure. Preliminary circular-dichroic ARPES maps exhibit multiple sign inversions that stem from a combination of initial state orbital angular momenta and photoemission final state scattering [6].

Keywords: Kagome metal; micro-ARPES; circular dichroism;

Acknowledgement: H.B. and X.H. were supported by the DFG, M. Q. was supported by the Federal Ministry of Education and Research of Germany

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Surface States Evolution and Surface Migration in $Co_3Sn_2S_2$

Mingkun Chen[1], Sudheer A. Sreedhar[1], Matthew Staab[1], Robert Prater[1], Zihao Shen[1], Valentin Taufour[1], Inna M. Vishik[1]

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$Co_3Sn_2S_2$ is a magnetic Weyl semimetal with correlated electrons in which characteristic surface states present differently when different atomic planes terminate the crystal. I will discuss our microXPS and microARPES experiments correlating local chemistry with local electronic structure in this material, tracking changes across the magnetic/topological phase transition, and revealing implications of the dynamic local defect environment.

Keywords: Topological materials, Weyl semimetals, $Co_3Sn_2S_2$, surface states, ARPES.

Acknowledgement: Work supported by the U.S. National Science Foundation under Grant No. DMR-2428464.

X-ray induced changes in organic polymer P3HT and its influence on charge transfer dynamics

Elin Cartwright[1], Yi-Chen Weng[1], Tiefeng Liu[2], Evelyn Johannesson[1], Brian Rydgren[1,3], Karen Radetzky[1,3], Ute B. Cappel[1,3], Simone Fabiano[2], Fredrik O.L. Johansson[1], Andreas Lindblad[1]

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Conjugated polymers such as poly(3-hexylthiophene) (P3HT) are integral to next-generation organic electronic devices owing to their tunable optoelectronic properties and ease of processing [1]. While several investigations have employed Core-hole Clock Spectroscopy (CHCS) to explore ultrafast charge-transfer dynamics in organic polymers [2,3], the potential influence of X-ray-induced changes on these measurements remains largely unexplored. Understanding how X-ray exposure can modify chemical structure and affect charge transport is crucial not only for interpreting spectroscopic data but also for ensuring the reliability of P3HT-based devices.

Here, we investigate X-ray-induced modifications in P3HT by combining Hard X-ray Photoelectron Spectroscopy (HAXPES) with Resonant Auger Spectroscopy (RAS) at the sulfur K-edge. Prolonged irradiation generates a second sulfur species at lower binding energy, indicative of partial ring saturation and de-aromatization within the thiophene units. This de-aromatization is further corroborated by RAS measurements, where a localized decay channel associated with the aromatic ring significantly diminishes in intensity. Core-Hole Clock Spectroscopy (CHCS) analysis reveals that these structural alterations reduce charge-transfer times from the sulfur atom in the core-excited state. Notably, the most pronounced transformations occur within the first few hours of exposure, after which the polymer reach a steady-state configuration.

These findings highlight the susceptibility of conjugated polymers to beam-induced chemical changes and demonstrate the use of HAXPES and CHCS to quantify this change in terms of electronic structure and charge dynamics.

Acknowledgement: Work supported by the Swedish Research Council, Åforsk and Wallenberg Initiative Materials Science for Sustainability (WISE).

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Poster P7: Poster Session TuI_17, Tuesday, July 22, 05:00 PM - 06:30 PM

Investigation of Black Rutile $TiO_2(110)$ by XPS/UPS and In-Situ Electrical Conductivity Measurement

Muhammad Irfandi[1], Touma Ichikawa[2], Takahiro Ozawa[1], Masuaki Matsumoto[2], Katsuyuki Fukutani[1,3]

[1] *Institute of Industrial Science, The University of Tokyo, Japan*
[2] *Tokyo Gakugei University, Japan*
[3] *Advanced Science Research Center, Japan Atomic Energy Agency, Japan*

TiO_2 nanocrystal changes its color into black upon being exposed to 20-bar H_2 at 200°C for five days, which consequently improves its solar absorption efficiency. The band gap is reduced from 3.30 eV down to 1.54 eV and 1-nm-thick disorder layer is formed at surface[1]. A similar black TiO_2 with a surface disorder layer is also reported by exposing TiO_2 to H plasma at 500°C for 8 hours [2]. However, the blackening mechanism remains debatable in the aspect of electronic structure. It is also unclear whether the black rutile TiO_2 possesses small polaron conductivity, as in typical reduced rutile TiO_2. A combination of in-situ XPS/UPS and electrical conductivity measurement may elucidate the nature of black TiO_2.

We report that black rutile $TiO_2(110)$ is realized by exposing TiO_2 to atomic H at 800 K, which decreases its band gap down to 2.1 eV. The XP spectrum shows that the reduction of Ti^{4+} to Ti^{3+} due to electron donation saturates around 17%. The reduction is due to oxygen vacancy formation since there is no observed OH signature in the valence band. The electron donation is also reflected in electrical conductivity which increases upon blackening.

Keywords : titanium oxide, oxygen vacancy, hydrogen, photoemission spectroscopy, conductivity.

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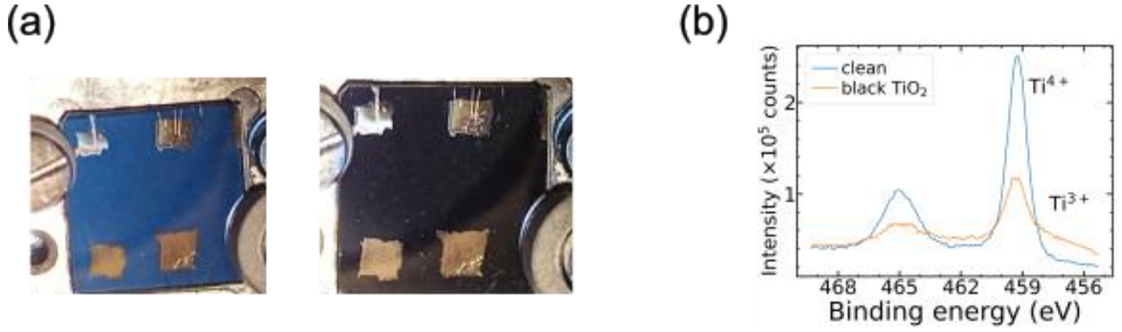

Figure 1. (a) Sample colors of rutile $TiO_2(110)$ before (left) and after (right) atomic H exposure at 800 K, (b) corresponding XP spectra of Ti 2p chemical states.

Condensed matter dark states in a quantum system with two pairs of sublattices

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In nature, there are quantum states that do not interact with photons and thus cannot be detected by spectroscopic means; there are referred to as dark states [1–4]. By extending this concept to condensed matter systems, we identify a novel concept of condensed matter dark states that are not observable by angle-resolved photoemission spectroscopy (ARPES), regardless of experimental conditions such as light polarization and scattering geometry. Our model, based on the tight-binding approximation, demonstrates that these dark states result from double destructive interference of the initial-state wavefunctions between two sublattice pairs [5, 6]. We demonstrate this mechanism in palladium diselenide ($PdSe_2$) as a model system. The unit cell of $PdSe_2$ contains four palladium sublattices connected by multiple glide-mirror symmetries [7]. In this system, the ARPES signal from the valence band is absent in the whole specific Brillouin zones. This absence is due to the relative phases between the sublattices, which become fully polarized to one of four phase types in each Brillouin zone, forming a checkerboard pattern that alternates zone by zone. We generalize this dark state mechanism to other materials with similar structural features, such as cuprate superconductors [8].

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Studying the electronic structure of $ZnGa_2O_4$ using X-ray absorption and emission spectroscopy

Poster P9: Poster Session TuI_17, Tuesday, July 22, 05:00 PM - 06:30 PM

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In this study, the electronic structure of zinc gallate ($ZnGa_2O_4$) thin films at different processing temperatures (300℃, 500℃, and 700℃) was explored using X-ray absorption (XAS) and emission spectroscopy (XES). By comparing samples before and after annealing, the study found that the electrons underwent significant changes, especially in the Zn 3*d*/Ga 4*sp* orbitals, where the electrons migrated to the O 2*p* orbitals as the processing temperature increased and after annealing. These transitions indicate an increasing ionic bonding character in the O 2*p*-Zn 3*d*/Ga 4*sp* orbitals and an increasing O 2*p*-Zn 4*sp* orbital hybridization, which enhances the covalent bonding character. At lower processing temperatures, annealing leads to a significant increase in electron sharing between O 2*p* and Ga 3*d* orbitals, indicating a covalent bonding tendency. In contrast, at higher annealing temperatures, electrons are transferred from Ga 3*d* to O 2*p*, resulting in a more pronounced ionic bonding nature between Ga 3*d* and O 2*p* orbitals.

The study also found structural transformation within $ZnGa_2O_4$. With the increase in processing temperature and annealing, the β-Ga_2O_3 phase changed from amorphous to crystalline. This increasing crystallinity indicates a higher degree of material order, which is presumed to be a key factor affecting the photonic and electronic properties of $ZnGa_2O_4$-based materials. The findings provide important insights into manipulating the electronic and structural properties of $ZnGa_2O_4$ thin films, helping to advance the development of more efficient materials for applications in optoelectronics and power devices.

Keywords - XAS, XES, DFT.

Surface state of Si(111)7×7 revisited: Atomic orbital characterization by Dual-beamline Photoelectron Momentum Microscopy Station at UVSOR

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At room temperature, the Si(111)7×7 reconstructed surface is metallic. The Fermi level crosses the surface state S1 localized on six adatoms in the unit cell [1,2]. In recent years, remeasurements with high energy resolution have been performed, and a gap in the S1 state has been reported at low temperatures [3]. It has also been reported that the S1 state is semiconducting even at room temperature [4]. The Si(111)7×7 reconstructed surface continues to fascinate (and bewilder) researchers.

Photoelectron momentum microscope (PMM) is an analyzer that can instantaneously image the behavior of electrons of material and device surfaces. PMM combines imaging-type photoelectron spectroscopy and microscopy techniques to visualize the electronic state (band dispersion, composition, and spin polarization) in reciprocal lattice space of a selected µm-sized area. We built a PMM station [5,6] at the UVSOR Synchrotron Facility at the Institute for Molecular Science. In addition, we upgraded this advanced analyzer and experimental station to be able to use two undulator beamlines as excitation sources [7]. By branching off the existing vacuum ultraviolet (VUV) beamline BL7U, in addition to the soft X-ray (SX) beam from beamline BL6U, polarization-variable VUV light can now be used at the same time at PMM.

We developed sample holders compatible with electrical heating and revisited the band structure of the surface state using soft X-ray beamline BL6U, grazing incidence [5]. We observed the S1 band crossing the Fermi level at room temperature. We also measured the photoelectron transition matrix elements using normal incidence geometry (s-polarized light) (Vacuum Ultraviolet Beamline BL7U) [7]. Corresponding nodes were observed in the photoelectron angular distributions of the S1 (adatom) and S2 (restatom) states, which consist of p_z orbitals perpendicular to the plane, whereas no nodes were observed in the S3 (backbond) state. This corresponds to the inclusion of an in-plane p_{xy} orbital component.

Keywords: Photoelectron momentum microscope, semiconductors, surface state, atomic orbital.

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Oue research investigates the formation of hydrated magnesium carbonate (HMC) cements through the carbonation of brucite, utilizing advanced synchrotron radiation (SR)-based X-ray diffraction (XRD) supplemented by the lab-based time-gated Raman spectroscopy. Several HMC phases, including nesquehonite, dypingite, and giorgiosite, were observed to form at different time points during the carbonation reaction of brucite with sodium bicarbonate in an aqueous environment. Notably, the formation of a sodium-containing carbonate phase, eitelite, was observed after 7 and 28 days of brucite carbonation in water—a finding not previously reported. The addition of magnesium acetate (MgAc) as a ligand into the carbonation reaction had a notable impact on the reaction, increasing the carbonation degree of brucite and modifying the end products. Meanwhile, Raman spectroscopy demonstrated that giorgiosite strongly integrates acetate into its structure, leading to the formation of a newly identified phase called "acetate-containing giorgiosite". Remarkably, this phase was detected just after one day of the reaction of brucite in presence of MgAc, compared to earlier report[1] suggesting longer crystallization times. Our findings further highlight that the crystallization of HMC is significantly influenced by the presence of water and MgAc in reaction process, where water-rich samples, favored the formation of dypingite, whereas MgAc ligand promotes the crystallization of giorgiosite. This study demonstrates how the combined use of both methods: Raman spectroscopy and SR-based XRD enhances our ability to explore the mechanisms behind HMC cement formation.

Keywords: Carbon capture, synchrotron radiation, Raman spectroscopy, magnesium acetate, magnesium carbonate.

Acknowledgement: This work is funded by the University of Oulu Kvantum Institute under CARBO-CEM project.

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Poster P12: Poster Session TuI_17, Tuesday, July 22, 05:00 PM - 06:30 PM

Photoelectron Holographic Study of Atomic Site Occupancy of the Si Dopant in κ-Ga_2O_3(001)

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For Ga_2O_3, there are six crystal polymorphs: α-, β-, δ-, γ-, ε-, and κ-Ga_2O_3. Among them, β-Ga_2O_3 has received the most attention due to its highest thermal stability. Recently, the orthorhombic κ-Ga_2O_3 has gained attention due to its large spontaneous polarization and its ferroelectricity. For κ-Ga_2O_3, Si is usually used as the dopant to control the electrical properties. There are three inequivalent Ga atomic sites as the dopant sites in κ-Ga_2O_3; octahedral (Octa), pentahedral (Penta), and tetrahedral (Tetra). Therefore, to control the electrical properties, the atomic site occupancy of Si dopant in Si-doped κ-Ga_2O_3 should be clarified. In the present study, we investigated the atomic site occupancy of the Si dopants for Si-doped κ-Ga_2O_3 using photoelectron holography (PEH).

The Si-doped κ-Ga_2O_3 epitaxial layers were grown on a c-plane sapphire substrates prepared by the MOVPE method. The PEH measurements were performed at the BL25SU beamline of SPring-8.

Figure 1(a) shows the Ga 3*p* and Si 2*p* PES spectrum for the Si-doped κ-Ga_2O_3(001) measured at an incident photon energy of 911 eV. The corresponding PEH for Si 2*p* are shown in Fig. 1(b). Figure 1(c) shows the experimental and the simulated Si 2*p* PEHs for the Si-doped κ-Ga_2O_3(001). By comparing the experimental and the simulated PEHs, the ratios for the Tetra, Penta, and Octa Si_{Ga} sites are estimated to be 51.0%, 35.2%, and 13.8%, respectively. [1]

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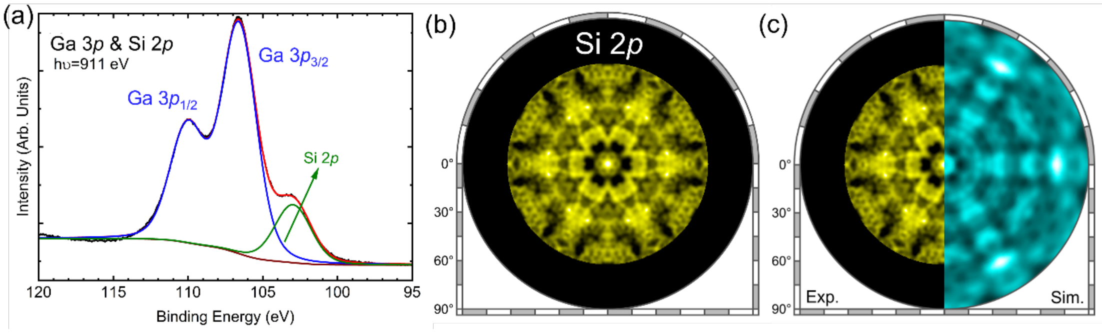

Figure 1. (a) Ga 3*p* and Si 2*p* PES spectrum for Si-doped κ-Ga_2O_3(001). (b) PEH of Si 2*p* for Si-doped κ-Ga_2O_3(001). (c) Experimental (yellow) Si 2*p* and the simulated (blue) PEHs for the Si-doped κ-Ga_2O_3(001).

An Example ICESS16 Abstract: Must fit on one page. Submit file in PDF format.

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Black phosphorus doped by alkali metals[1] can be modelled by two-dimensional (2D) electron liquid under the influence of multiple scattering by the potential of effectively ionized dopants. Recently, our angle-resolved photoemission spectroscopy study revealed the pseudogap and back-bending dispersion induced by the short-range order of dopants[2]. At the lower density, the alkali metals on black phosphorus can be more properly modelled by a 2D dipole liquid or an electron-hole bilayer. In this talk, I will introduce the observation of striking aperiodic dispersion with the local minimum at a finite momentum, which is formed by the broad peak in the structure factor or the short-range order of 2D dipoles[3]. This can be explained by the electronic analogue of rotons, the concept first proposed by Landau to explain superfluidity in liquid helium[4]. I will discuss the density dependent reduction in the roton gap, suggesting Wigner crystallization[3].

Keywords [optional]: ARPES, Black phosphorus, Roton, 2D dipole liquid, Band structure

Acknowledgement [optional]: Work supported by the Agency for the Advancement of Generously Funded Science (AAGFS).

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Oxygen reaction activities in Li-ion battery cathodes materials studied through resonant inelastic soft x-ray spectroscopy (RIXS)

Poster P14: Poster Session TuI_17, Tuesday, July 22, 05:00 PM - 06:30 PM

Abstract:

Recent clarification through high efficiency mapping of resonant inelastic X-ray scattering (mRIXS) have revealed that, oxygen reaction activities are distinctive among the three representative Li-rich parent compounds, Li_2MO_3 (M=Mn, Ru, Ir), i.e., only irreversible oxygen oxidation reaction for Mn (3d) system, separate reversible Ru and O redox reactions for Ru (4d) system, and only divalent cationic redox reaction for Ir (5d) systems. Here, through quantification of transition metal redox and characterizations of the lattice oxygen state, we finally provide a direct comparison among the three representative Li2MO3 compounds. Firstly, Li-rich configuration does not naturally trigger reversible oxygen redox, as there is no oxygen redox observed in Mn and Ir systems. It indicates that extra critical TM factors are required to maintain the reversible oxygen redox. Secondly, compared with Ru and Ir systems with both Ru/O redox involved and only Ir redox involved, separately, it indicates that oxygen redox is not necessarily enabled until the Li extraction amount which cationic redox could compensate is exhausted. Thirdly, comparison between irreversible oxygen oxidation process in Li2MnO3 and the high reversible oxygen redox activities in Ru systems, suggests the critical role for stabilization effect of TMs-O hybridization in oxygen activities, which has been emphasized in the ground-breaking works of the oxygen redox but got largely overlooked later. This work suggests the important role of transition metals and their coupling and hybridization effect to oxygen for maintaining reversible oxygen redox activities.

RIXS of highly oxidized transition metal oxides in batteries

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High-energy battery solutions sit at the center of today's sustainable energy applications, e.g., electric vehicles and green grids. As the most popular energy storage solution, Li-ion batteries have been bottlenecked in their high voltage performance, mostly in its transition-metal oxide-based cathodes and electrolyte. These oxides are complex systems, in which conventional ionic crystal models fail and a significant amount of oxygen activities are involved in the electrochemical operations at high voltages. This work discusses popular models of high voltage reaction mechanisms, and present our most recent opinions based on latest RIXS results through combined experimental and theoretical studies. Scientifically, we conclude that pushing a rechargeable battery system into the high-energy operation always spontaneously triggers a highly oxidized oxide state in the battery cathode during high voltage charging. Technically, we demonstrate that soft X-ray Resonant Inelastic X-ray Scattering (RIXS) is a powerful tool for detecting and distinguishing different types of oxygen activities in electrochemical systems including batteries.

Keywords [optional]: Resonant Inelastic X-ray Scattering, Battery electrodes, highly oxidized oxides.

Acknowledgement [optional]: This research used resources of the Advanced Light Source, which is a DOE Office of Science User Facility under contract no. DE-AC02-05CH11231.

Poster P16: Poster Session TuI_17, Tuesday, July 22, 05:00 PM - 06:30 PM

Highly Hydrogenated Monolayer Graphene with Wide Band Gap Opening

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The 2D nature of graphene accompanied by its unique mechanical, thermal and electronic properties make it appealing for many applications. Moreover, the chemical modification of graphene, and in particular its functionalisation with hydrogen, has enhanced the attractiveness. The adsorption of hydrogen on graphene leads indeed to the modification of the pristine electronic structure by breaking the π-bonds, thus producing a transition from sp^2- to sp^3-hybridised carbon atoms and the opening of a band gap[1,2]. Two graphene samples were grown via chemical vapor deposition (CVD) and then transferred onto Ni grids[3]. After preparation, the *in-situ* hydrogenation and the spectroscopic characterisation were carried out. The hydrogenation was performed by exposing the sample to thermally-cracked atomic hydrogen, as shown in Figure 1. We characterised the monolayer graphene samples with X-ray photoemission spectroscopy (XPS) and electron energy loss (EELS), before and after the hydrogenation. Unprecedentedly high hydrogenation levels were observed on one sample, with the C 1s changed from a 42% sp^3 relative content up to a 100% sp^3 line-shape after hydrogenation. On the other hand, the C 1s of the second sample changed from 13% to 59% of sp^3, showing that the hydrogen adsorption is favoured by a more initial sp^3-like character. Furthermore, the opening of a wide optical band gap of 6.3 eV was measured on both samples with EELS, together with the quenching of the π-plasmon excitation and the rise of the C-H stretching vibrational mode.

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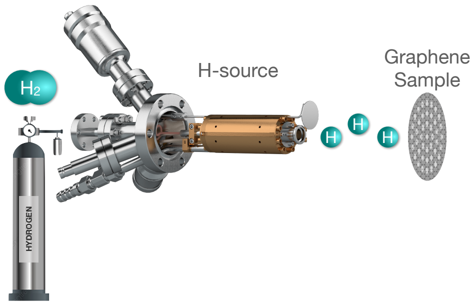

Figure 1. Scheme of the hydrogenation technique.

Poster P17: Poster Session: TU_17, Tuesday, July 22, 05:00 PM - 06:30 PM

Plasmonic polaron in self-intercalated 1*T*-TiS_2

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Electron-boson coupling is a critical aspect of many-body physics phenomena; however, little is known about how plasmon interacts with conduction electrons or how this interplay can be controlled. In this study, we investigated the temperature-dependence of the plasmonic polaron in self-intercalated 1*T*-TiS_2 using angle-resolved photoemission spectroscopy (ARPES) and high-resolution electron energy loss spectroscopy (HR-EELS). We find that unexpected behavior of temperature-dependent plasmon energy in self-intercalated 1*T*-TiS_2 and its coupling with the conduction electrons. Furthermore, we observe that the dielectric screening plays a significant role in the formation of plasmonic polaron states. Our findings establish self-intercalated layered materials as a promising platform to study, control, and utilize the plasmonic polaron in quantum materials.

On-surface synthesis of Atomically Precise Silicon Nanoribbons

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Abstract

In recent years, the scientific community has put a lot of efforts into trying to artificially grow new functional materials by reducing their dimensionality from 2D to 1D or even 0D. However, it is not a simple task, which includes synthetic methods that could go in both possible top-down or bottom-up approaches. Our group, working on the bottom-up approach through on-surface synthesis has been able to develop unprecedented new 0D, 1D or 2D silicon nanoshapes[1,2]. Here, we demonstrate the on-surface synthesis of atomically precise silicon nanoribbons by utilizing the Ag(110) surface and also we have unmasked the unique 1D pentagonal silicon structures formed on it, which had been elusive for over 10 years. Further, we will see how the different single crystals substrates greatly affects the growth of silicon and the new structures that can be formed on it[3].
Promising applications of new silicon nanoforms range from nanoelectronics to transistors or embedded sensors; however, one of the important challenges that we have to overcome is the isolation, through the separation of the substrates, of these interesting nanoforms.

Keywords: Silicon nanoribbons, 1D-Silicon.

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Spectroscopic evidence of the ferromagnetic transition in monolayer 1*T*-$CrTe_2$

Kyoungree Park[1], Ji-Eun Lee[2,3,4], Dongwook Kim[5], Yong Zhong[2,6,7], Camron Farhang[8], Hyobeom Lee[1], Hayoon Im[9], Woojin Choi[1], Seha Lee[1], Seungrok Mun[1], Kyoo Kim[10], Jun Woo Choi[3], Hyejin Ryu[3], Jing Xia[8], Heung-Sik Kim[1], Choongyu Hwang[9], Ji Hoon Shim[5], Zhi-Xun Shen[6,7*], Sung-Kwan Mo[2*], and Jinwoong Hwang[1*]

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Two-dimensional (2D) van der Waals ferromagnets hold immense promise for spintronic application. Despite its considerable significance, essential understanding and engineering of ferromagnetic materials in 2D limit have been rare. Here, we report a successful synthesis of monolayer (ML) 1*T*-$CrTe_2$ on a bilayer graphene substrate via molecular beam epitaxy and its unusual ferromagnetic band evolutions. Using angle-resolved photoemission spectroscopy and magneto-optical Kerr effect measurements, we observe the ferromagnetic transition at Curie temperature (T_C) ~ 150 K in ML 1*T*-$CrTe_2$, accompanied by exotic temperature-dependent band evolutions. Our spectroscopic analysis and first-principle calculation reveal that the ferromagnetism may arise from Goodenough-Kanamori (GK) super-exchange and double-exchange interactions, facilitated by lattice relaxation and electron doping from the BLG substrate. These findings provide pivotal insight into the fundamental understanding of

Poster P20: Poster Session TuI_17, Tuesday, July 22, 05:00 PM - 06:30 PM

Stability of Highly Hydrogenated Monolayer Graphene in Ultra High Vacuum and in Air

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The stability of hydrogenated monolayer graphene was investigated via X-ray photoemission spectroscopy (XPS) for two different environmental conditions: ultra-high vacuum (UHV) and ambient pressure. The study was carried out by measuring the C 1s line shape evolution for two hydrogenated samples one kept in the UHV chamber and the other progressively exposed to air. In particular, the sp^3 relative intensity ($sp^3/(sp^2+sp^3)$) in the C 1s core-level spectrum was used as a marker for the hydrogenation-level. A long-term stability of hydrogenated monolayer graphene was found, as indicated by a small sp^3 relative intensity variation of $(4 \pm 2)\%$ after four months (Figure 1a). Such result indicates this material as a good candidate for hydrogen (or tritium) storage as long as it is kept in vacuum. On the other hand, the C 1s spectrum of the sample exposed to air shows a significant oxidation (Figure 1b). A rapid growth up to saturation of carbon oxides was observed with a time constant $\tau = 1.8 \pm 0.2$ hours. Finally, the re-exposure of the oxidised sample to atomic hydrogen was found to be an effective method for the recovery of hydrogenated graphene. This process was studied by carrying out both XPS and electron energy loss spectroscopy, the latter exploited to observe the C-H stretching mode as a direct footprint of re-hydrogenation.

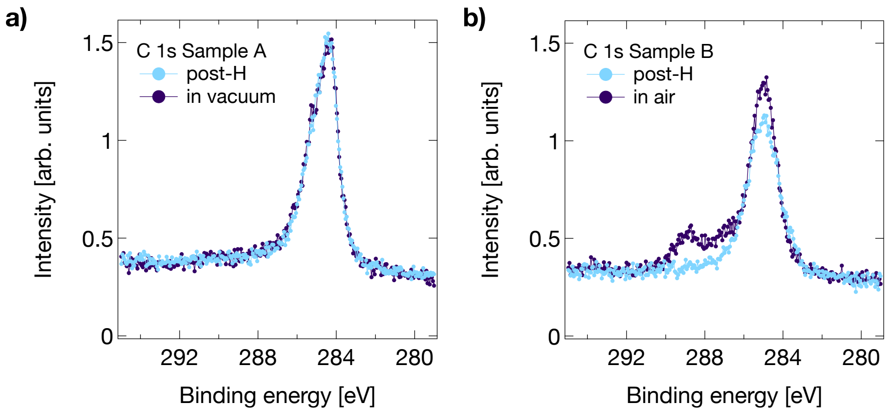

Figure 1.C 1s core-level for: (a) sample A after hydrogenation (light blue) and after 4 months in UHV (purple); (b) sample B after hydrogenation (light blue) and after 11 months in air (purple).

Light-induced chirality probed by circular dichroism ARPES

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Emergent orders in quantum materials are often accompanied by the breaking of mirror symmetry, known as chirality. A prime example is a chiral charge density wave order, in which spontaneous charge ordering breaks mirror symmetry. While such chirality has been extensively studied in equilibrium, its dynamical control remains largely unexplored. Here, we demonstrate that a circularly polarized pump pulse can transiently couple to chirality, probed by time-resolved circular dichroism angle-resolved photoemission spectroscopy (TR-CD-ARPES). We observe an even-function circular dichroism signal upon circularly polarized pumping, which is only allowed when mirror symmetry is broken. Our findings establish TR-CD-ARPES as a powerful tool for probing transient chiral states and mapping a transient chiral order parameter in momentum space.

Acknowledgement: We acknowledge support from the EPiQS programme of the Gordon and Betty Moore Foundation, grant number GBMF11069

Multimodal tool combining multichannel HREELS and ARPES/XPS to study electron-phonon coupling

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Electrons and phonons couple in a material, so multimodal characterization of phonon dispersion and electronic band dispersion provides vital insights into their interplay. High-resolution electron energy loss spectroscopy (HREELS) is a technique for observing surface excitations including phonons, plasmons, magnons, excitons, and vibrational modes. Multimodal characterization has been difficult using traditional single channel HREELS because it requires its own detector, the measurements are time consuming, and angular resolution is limited. To improve the efficiency of HREELS measurements and to realize multimodal measurement with photoemission spectroscopy, we developed a solution by combining a monochromatic collimated electron source and a hemispherical electron analyser, commonly used for ARPES/XPS[1]. The multichannel 2D detector of the analyser simultaneously measures hundreds of channels in both the energy and angular directions, and the measurements are orders of magnitude faster than the single channel setups. This setup allows multimodal characterization combining state-of-the-art HREELS and ARPES/XPS using the same electron analyser to study the interplay between electronic properties and various surface excitations including electron-phonons coupling.

Keywords [optional]: High-resolution electron energy loss spectroscopy (HREELS), surface excitations, phonon, multimodal characterization, ARPES.

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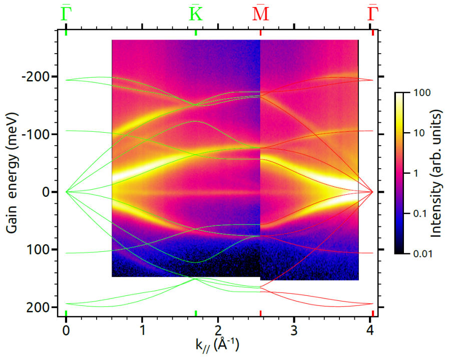

Figure 1. Phonon dispersion of single crystal graphite.

Poster P23: Poster Session TuI_17, Tuesday, July 22, 05:00 PM - 06:30 PM

Preliminary Photoemission Study of a Highly Overdoped Cuprate Beyond the Superconducting Dome

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Cuprate superconductors are one of the most well studied classes of unconventional superconductors, as they possess the highest critical temperatures at ambient pressure. Previous work on cuprates has uncovered a rich phase diagram that varies with hole and electron doping, with a superconducting dome emerging on the hole doping side whose maximum delineates the overdoped and underdoped side. The underdoped side has been widely studied, with many overlapping complex phases, while the overdoped region has been comparatively under-explored. In the past three decades, however, several materials have been discovered which exhibit superconductivity at dopings well beyond the boundaries of the traditional superconducting dome. One such material, high pressure oxygenated (HPO) $YBa_2Cu_3O_{7+\delta}$ (YBCO), was recently synthesized as a polycrystalline sample with large grains making it a suitable candidate to investigate with photoemission. We use microscale x-ray photoemission spectroscopy (XPS) to study the local variations in surface chemistry and electronic properties of this material due to variations in oxygen content and surface preparation.

Keywords: Cuprates, High Temperature Superconductors, XPS.

Acknowledgement: Work supported by the Gordon and Betty Moore Foundation, Award ID #12957

In-situ Strain Tuning: ARPES Unveils Momentum and Orbital Nematicity in an Iron-based High T_c Superconductor

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In quantum materials, the manipulation of atomic spacing and electron cloud overlap enables precise control of the many-body Hamiltonian, affecting the material's ground state and excitations. To investigate how these effects manifested in the electronic band structure, we incorporated a piezo-driven stress cell compatible with μ-ARPES and Laue μ-diffraction beamlines at the Advanced light source. Building on this capability, we investigate an intriguing phenomenon reported by P. Malinowski et al. [1]. They demonstrated that continuously applied uniaxial strain can effectively suppress superconducting transition temperature (T_c) in an optimally doped iron-based superconductor with elusive underlying microscopic mechanisms. In this presentation, we will discuss the in-situ uniaxial strain tuning of the Fermi surface nesting condition and effective mass enhancement in decoupled strain channels in $BaFe_2(As_{0.7}P_{0.3})_2$, mostly consistent with the nematic quantum critical point scenario.

Keywords : Unconventional superconductivity, in-situ tuning uniaxial strain, ARPES.

[1] Nat. Phys. 16, 1189-1193 (2020)

ARPES Study of Kondo Semimetals/Insulators: CeNiSn, CeRhSb, CeRhAs

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The theoretical prediction of the topological Kondo insulators (TKIs)[1,2], such as CeNiSn and CeRhSb, has triggered revived attention to the Kondo insulators and Kondo semimetals. CeNiSn and CeRhSb are known as the Kondo semi-metals with large anisotropy in resistivity[3]. The predicted TKI properties of CeNiSn and CeRhSb arise from the strong spin-orbit coupling and the odd parity of the f-electron states[1,2]. A unique feature of these systems is the nonsymmorphic glide and screw axis symmetries that bring about the new topological surface states and the hour-glass type bulk band structures. Despite extensive work on the possible TKI candidates, the TKI nature of CeNiSn and CeRhSb has not been experimentally confirmed yet. This work explores the electronic structures of isostructural CeNiSn, CeRhSb, and CeRhAs by employing angle-resolved photoemission spectroscopy (ARPES). In particular, the temperature dependence of the Ce 4f states in ARPES is investigated and their Kondo temperatures are determined. This work demonstrates the importance of the coherent Kondo states in determining the potential topological properties of CeNiSn, CeRhSb, and CeRhAs.

Keywords: Kondo Insulator, Kondo Semimetal, Electronic structure, ARPES, Topological Insulator

Acknowledgement: Work supported by the National Research Foundation (NRF) of Korea (Grant No. RS-2023-00275779).

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Essential ingredients for superconductivity in the electron-doped cuprate superconductors studied by angle-resolved photoemission spectroscopy

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Electron-doped cuprates $R_{2-x}Ce_xCuO_4$ (R: rare earth) have the so-called T'-type structure with four oxygen atoms coordinated to Cu atoms in a plane. Their most remarkable feature is the strong influence of post-growth reduction annealing, which is conducted to remove impurity apical oxygen located right above the Cu site. Recently, Adachi *et al*. [1] developed the improved annealing method known as protect-annealing, and succeeded in expanding the superconducting dome and enhancing the transition temperature [2]. An angle-resolved photoemission spectroscopy (ARPES) study of protect-annealed samples revealed the strong suppression of antiferromagnetic correlations that compete with superconductivity [3]. However, it was also reported that electron concentrations significantly increase after protect-annealing [2,3]. Therefore, it is still unclear how each of the apical oxygen removal and electron doping contributes to superconductivity through the suppression of the antiferromagnetic correlations.

Here, we investigated the pure effect of electron-doping on the electronic structure of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ (x=0.08). The as-grown sample was doped with electrons without any change in crystal structure by surface adsorption of alkali metals, and the changes in the antiferromagnetic correlations were evaluated by ARPES, which is a surface-sensitive method. By comparing with the results of the protect-annealed $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ (x=0.10) sample, we revealed that both the impurity apical oxygen atoms and electron concentrations have significant influences on the antiferromagnetic correlation.

Keywords: cuprates, high-T_c superconductivity, strongly correlated electron system, angle-resolved photoemission spectroscopy (ARPES).

Acknowledgement: Work supported by the Japan Society for the Promotion of Science (JSPS).

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Electronic structure of the molecular superconductor κ-(BEDT TTF)$_2$Cu(NCS)$_2$ using 6 eV laser photoemission spectroscopy

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The quasi-two-dimensional molecular superconductors κ-(BEDT-TTF)$_2$X have relatively high superconducting transition temperatures (T_c) up to about 12 K at ambient pressure [1-4] among molecular superconductors and show antiferromagnetic insulation by changing anion/deuteration. These phenomena suggest that the κ-type BEDT-TTF compounds are located near the Mott transition in the electronic phase diagram owing to the dimerization of the BEDT-TTF molecules. Photoemission spectroscopy (PES) is a powerful tool to observe the electronic structure directly. However, reliable experiments to obtain the intrinsic spectra of the molecular superconductors using PES are very difficult due to problems such as radiation damage, surface preparation, temperature control and so on [5,6]. We have developed a PES system using a 6 eV quasi-continuous ultraviolet laser to avoid radiation damage and have much higher photoionization cross section for s and p electrons that play an important role in the physical properties of the molecular conductors. We have succeeded in observing the superconducting gap of the molecular superconductor κ-(BEDT TTF)$_2$Cu(NCS)$_2$ (T_c ~ 10 K). We found that the superconducting gap size (Δ) is about 1.4 meV, and the gap symmetry seems to be *d*-wave.

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Observation of electronic structure of organic Mott insulator *β'*-(BEDT-TTF)$_2$ICl$_2$ by angle-resolved photoelectron spectroscopy

Ryotaro Nakazawa[1], Keisuke Fukutani[1], Yuto Ishida[2], Seiji Makita[1], Kiyohisa Tanaka[1], Takuya Kobayashi[3], Hiromi Taniguchi[3], Satoshi Kera[1, 2]

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Organic materials have the potential to promote the sustainable development of science and technology due to their flexibility and high environmental compatibility. Among organic materials, the molecular crystals of strongly correlated systems exhibit a variety of quantum phases, such as superconductivity, spin liquid states, Mott insulating phases, and topological phases [1, 2]. While the origins of these quantum phases are largely encoded in the electronic structures, their momentum-resolved pictures are primarily investigated by theoretical calculations and their direct observations are still largely lacking.

In this study, we focused on the single crystal of organic Mott insulator *β'*-(BEDT-TTF)$_2$ICl$_2$ [3,4] and observed the valence band electronic structure using angle-resolved photoelectron spectroscopy (ARPES) at BL7U in UVSOR.

Figure 1 shows the valence band structure observed using excitation energy at $h\nu$ = 40 eV at T = 150 K. Fig. 1(a) shows a photoemission intensity plot along the short crystal axis (*c*-axis), and Fig. 1(b) is the second derivative plot. A dispersed band width about 140 meV is observed around binding energy at around 0.7 eV. Since the observed band periodicity corresponds to that of the Brillouin zone, we have successfully observed itinerant electrons in this material. Other high-symmetric momentum directions are measured to confirm the characteristics of the electronic structure by ARPES. In the presentation, we will discuss the electronic correlations in this material, particularly from the perspective of Mott insulating behavior, based on a series of experimental results.

Keywords: Organic conductor, Molecular crystal, Mott insulator, Angle resolved photoemission spectroscopy.

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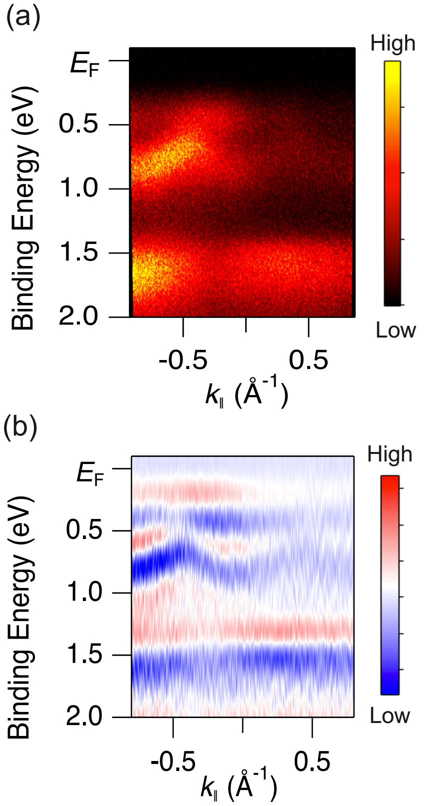

Figure 1 (a) Photoemission intensity plot, and (b) the corresponding second derivative plot of *β'*-(BEDT-TTF)$_2$ICl$_2$ crystal observed at $h\nu$= 40 eV, T = 150 K.

Poster Session ThI_20

Upcoming soft x-ray photoemission and scattering imaging beamline at NSLS-II

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The soft x-ray photoemission and scattering imaging, i.e., the **ARI**, beamline combines the **A**ngle-resolved photoemission spectroscopy (ARPES) and the **R**esonant inelastic x-ray scattering (RIXS) with an **I**maging capability. It features a ~100 nm x-ray beam for both the ***Elektra*** (electron-out) and ***Phaos*** (photon-out) endstations that are bridged with one vacuum transfer system, therefore enabling the spatial mapping of the same sample with both photoemission and x-ray scattering techniques. Specifically, the ~100 nm x-ray beam is obtained through a pair of Kirkpatrick-Baez (KB) mirrors, distinct from the zone-plates commonly used in existing nano-APRES beamlines. This optical set-up provides the **ARI** beamline with same order of magnitude of photon flux as those ARPES [1] and RIXS [2] beamlines with the 1~10 μm spot sizes. The ***Elektra*** endstation (40eV~600eV*) is equipped with a Momentum Microscope [3] (KREIOS 150 MM from the SPECS) that can perform the spatial mapping in both real- and momentum- space from the emitted electrons, and the ***Phaos*** (200eV~1000eV*) adopts the Hettrick-Underwood optical design [4] to disperse the scattering x-rays in energy to achieve the analysis of x-rays from the inelastic scattering process. Based on the principles of two endstations, many secondary techniques, such as XAS, XPS, inverse XAS, photoelectron diffraction, fluorescence and more, are also available. With these functionalities, the **ARI** beamline will facilitate the investigation of correlations between the electronic, magnetic, atomic and chemical structures of materials with ~100nm spatial variation, shedding light on novel physics/chemistry in technologically relevant fields and confinement induced quantum effects emerging in prospective devices.

In this talk, I will present the details of the **ARI** beamline, including the optical design, current status, etc., and also briefly discuss the potential scientific opportunities unleashed by this new beamline at NSLS-II.

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* Both endstations can receive the x-ray photons from 40eV to 1000eV energy range but are optimized over the smaller ranges because of the reduced flux for x-ray photons outside of the ranges.

Photoionization and subsequent Auger decay of a K 2s vacancy

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The study[1] investigated the photoionization from the atomic K 2s subshell and the subsequent Auger decay cascade processes using magnetic bottle multicoincidence spectroscopy with synchrotron radiation. The method provided a comprehensive description of all cascade decay paths of the K 2s vacancy, obtaining the population of the K^{2+}, K^{3+} and K^{4+} final states. The experimental findings were compared with theoretical predictions obtained from the multiconfiguration Dirac-Fock approach and previous observations in Ar, highlighting differences due to the extra 4s electron.

The 2s photoelectron spectrum showed a peak for the $2s^1$ (^1S) and (^3S) states, with a linewidth of 2.6 eV ± 0.2 eV, which is slightly wider than in Ar. Also, a weaker structure attributed to transitions to K $2s^1 5s$ satellite states is observed. In the Auger decay to final K^{2+} states, the outer 4s electron primarily acts as a spectator, populating the $3s^2\ 3p^4\ 4s^1$ and $3s^1\ 3p^5\ 4s^1$ levels, in only 30% of the cases, the 4s electron participates, populating the $3s^2 3p^5$ states. The main source of formation for K^{3+} final states is the $L_1L_{2,3}M$ Coster-Kronig transitions. The multielectron coincidence experiment offers an opportunity to resolve states beyond the natural widths of transitions. Experimental and calculated K^{2+} $2p^{-1}v^{-1}$ core-valence states, populated by 2s hole decay, are shown, and good agreement is observed between the experimental and theoretical spectra. In the double Auger decay process to the K^{3+} final states, the 4s electron acts as a spectator for most of the cascade process and remains in the final ion. Dalitz plots were used to analyze the K 2s triple Auger decay to K^{4+} states. The experimental results have been compared to predictions, revealing good agreement. Significantly larger portion of the Auger cascades in K lead to quadruply charged ions compared to Ar due to the additional 4s electron in K. The outer 4s electron mainly acts as a spectator during the Auger decay and is emitted in the final step.

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UV photochemistry of solvated bromoform probed by liquid phase ultrafast electron diffraction

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Studying photoinduced molecular dynamics in the gas phase provides insights into transient molecular structures and their evolution without environmental perturbations. However, many chemical reactions occur exclusively in the liquid phase, where intermolecular interactions may play a critical role. Investigating liquid-phase photochemistry is thus crucial for understanding these processes in realistic environments. Bromoform ($CHBr_3$), a major precursor to atmospheric bromine that contributes to catalytic ozone destruction, is primarily produced in the solvated phase by marine macroalgae before being released into the atmosphere. In the gas phase, recent ultrafast electron diffraction (UED) studies reveal two primary relaxation pathways: direct dissociation to release a bromine atom and isomerization to form iso-bromoform [1]. Here, we extend these studies to solvated systems by employing liquid-phase UED (LUED) with vacuum-compatible sub-micron thick liquid sheet jets to investigate the effects of a solvent environment on the UV-excited dynamics of bromoform. Previous studies suggest that the solvent cage surrounding excited bromoform introduces a slower isomerization mechanism on the order of a few ps, in addition to the sub-200 fs direct isomerization pathway observed in the gas phase. Using a 2M solution of $CHBr_3$ in 1-propanol, we have collected LUED data to examine these solvent-mediated effects in detail. Interactions with the solvent are expected to open additional reaction pathways, potentially offering insights into the complex interplay between inter- and intramolecular interactions in liquid-phase bromoform photochemistry.

[1] L. Hoffmann et al., *J. Am. Chem. Soc.*, 146, 28070−28079 (2024).

This work was supported by the Atomic, Molecular, and Optical Sciences Program of the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Chemical Sciences, Geosciences and Biosciences Division through Contract No. DE-AC02–05CH11231.

Ultrafast photochemical reaction of 1-Propanol in liquid phase: dissociation channel studied by ultrafast electron diffraction

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Laser-induced ionization of molecules initiates ultrafast processes, such as hydrogen migration, isomerization, bond dissociation, and fragmentation, which are driven by both structural and spatial intermolecular interactions. In the condensed phase, these reactions are significantly influenced by molecular confinement and intermolecular interactions, resulting in altered dissociation pathways. In this study, we directly observe molecular photochemical reactions of 1-propanol under 800 nm laser ionization using ultrafast electron diffraction (UED). The reaction dynamics revealed carbon fragmentation pathways, including competing methyl and ethyl group dissociation in liquid-phase propanol, which differ from gas phase ionization pathways. These experimental investigations are complemented by ab initio molecular dynamic (AIMD) simulations, revealing the methyl dissociation pathway which is facilitated by hydrogen migration in the propanol dication. This mechanism is initiated by electron hole localization, modulated by molecular conformation, confinement from surrounding molecules, and subsequent hydrogen migration. These findings demonstrate that molecular interactions in the liquid phase significantly influence photochemical reaction pathways, revealing chemical reaction pathways that are distinct from those observed in the gas phase.

Keywords: Ultrafast electron diffraction, Ultrafast chemical reaction, Condensed phase, propanol dissociation, Ab initio Molecular Dynamics Simulation.

Acknowledgement: Work supported by the U.S. Department of Energy, Office of Science with award number DE-SC002222.

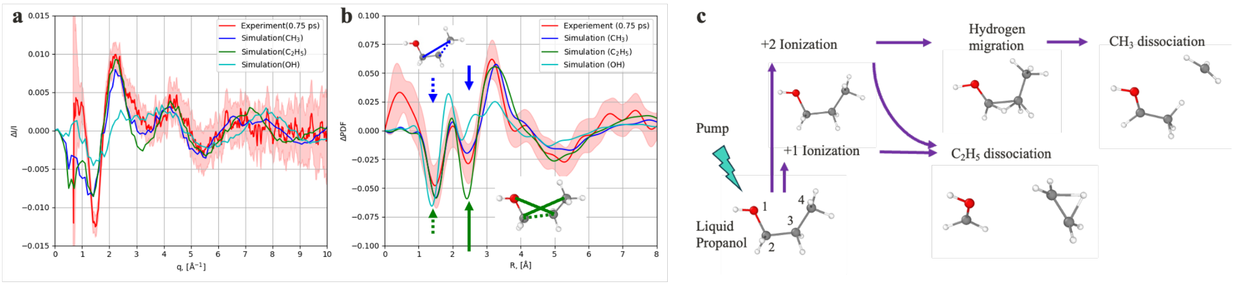

Figure 1. Experimental and simulated structural information in a) momentum-space and b) real-space c) dissociation pathway in the liquid phase as revealed by AIMD simulations.

X-ray Photoelectron Spectroscopy and X-ray Emission Spectroscopy Data Fitting Using A Genetic Algorithm

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The ever-growing problem in modern science is that data is being collected at a rate faster than analysis can be performed by characterization experts. The analysis that is done for many recently published x-ray photoelectron spectroscopy (XPS) and x-ray emission spectroscopy (XES) data, is often incorrect/irreproducible and leads to a cycle of incorrect fits in this spectroscopy data. In this work, a genetic algorithm (GA) is being constructed to potentially minimize this human error. This GA code known as XPS Neo/XES Neo, is based on the Neo package which exists for EXAFS (EXAFS Neo) and Nanoindentation (Nano Neo) data. GAs are based on biological methods and depend on parameters such as populations size, number of generations, genes, crossover, and mutation. The GA takes in a certain population size and constructs individual vectors each with their own unique genes i.e the fitting parameters we are trying to optimize. It then performs crossover and mutation to these individual vectors to progress toward a lower global minimum. This GA allows for a variety of mutation options including, Random Perturbations, Rechenberg, Metropolis mutation, and Self Adapting Differential Evolution. The methods of how a GA works in relation to XPS and XES datasets are discussed. The difficulties in making this work for XPS data arise from complicated backgrounds due to many effects such as plasmon loss, Auger peaks, and satellite peaks. XPS and XES data can also have many peaks that are difficult to distinguish from one another. Currently, the algorithm is only able to fit simplistic XPS spectra such C, O, N, and Si and is being worked on with the hopes of it becoming applicable for more difficult data. The goal is to make the algorithm applicable to all XPS data, with a greater focus given to the actinides, specifically for the use of fitting plutonium data as analysis of this spectra is highly sought after and difficult to fit. Through proper use of an informed GA, and collaboration with the XPS/XES database website XPSOasis.org/XESOasis.org, theoretically correct fitting of this data is hoped to be achieved.

Probing Electrochemical Dynamics with Time-Resolved XPS Using a TimePix3 Detector and Fast Phosphor Screen.

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Capturing the transient electronic structure and chemical properties of electrochemical interfaces is critical for understanding charge transfer processes and reaction intermediates that are often not detected under steady-state conditions. We demonstrate an electrochemical potential-pump / x-ray-probe method for time-resolved x-ray photoelectron spectroscopy (XPS) at Beamline 9.3.1 (ALS), employing a fast P47 phosphor screen and ASI TimePix3 event-based camera to resolve Au 4f binding energy shifts with sub-microsecond precision. A gold electrode was integrated into a tunable RC circuit designed to emulate dynamic potential changes at electrode surfaces, with decay times ranging from 1 second to 22 nanoseconds. By applying ±1 V square-wave pulses at 2.5 kHz, we drive controlled potential transients across these time constants, mimicking interfacial kinetics and mass transport phenomena intrinsic to electrochemical systems. Time-stamped spectra reveal potential-induced Au 4f core-level shifts at chemically relevant timescales, averaged over sequential pumped cycles. This demonstration provides a pathway to observe electrode surface charging phenomenon and short-lived species, (e.g. reaction intermediates) that are often overlooked by conventional XPS techniques. This approach opens new possibilities for studying dynamic electrochemical processes and interfacial reactivity by operando XPS.

Keywords: AP-XPS, Electrochemistry, Interfaces, Reaction Kinetics

Acknowledgement: This research used resources of the Advanced Light Source, which is a DOE Office of Science User Facility, under contract no. DE-AC02-05CH11231, Center for Electrochemical Dynamics and Reactions on Surfaces, Lawrence Berkeley National Lab, DOE-BES, Condensed Phase and Interfacial Molecular Science.

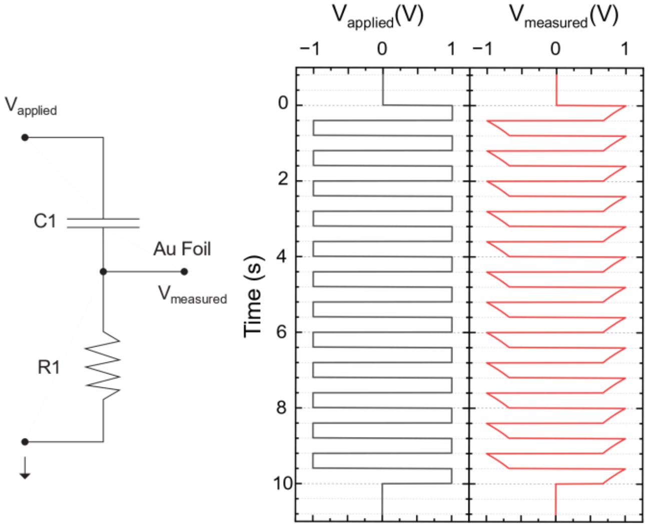

Figure. (left) circuit diagram, (right) simulated voltage pump and response

In situ observation of topotatic transition of $LaCoO_{3-\delta}$ with ambient pressure hard X-ray photoemission and X-ray diffraction

Hyunsuk Shin[1], Youngmin Yun[1], Okkyun Seo[2], Seongeun Kim[3], Minsik Seo[1], Dongwoo Kim[4], Hojoon Lim[5], Hojun Oh[1], Subin Jang[1], Kyungmin Kim[1], Sae Hyun Kang[1], Adrian Hunt[6], Iradwikanari Waluyo[6], Do Young Noh[1], Hyon Chol Kang[3], Bongjin Simon Mun[1],

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In recent years, a topotactic phase transition of $LaCoO_3$ (LCO) perovskite oxide received much attentions due to its potential industrial applications, such as batteries, solid oxide fuel cells, chemical sensors, and magnetic devices. To access the fundamental functioning mechanism of the phase transition, it is highly necessary to employ *in situ* experimental techniques that can provide direct information on the structure, electronic, and chemical states of LCO as the phase transition takes place. We investigated the structural, chemical, and electronic changes that occur during the topotactic phase transition in LCO by using *in situ* ambient pressure hard X-ray photoemission (AP-HAXPES) and *in situ* X-ray diffraction (XRD). XRD measurements show a clear evidence of structural phase transition (SPT) of the LCO thin films from the perovskite (PV) $LaCoO_3$ to the brownmillerite (BM) $La_2Co_2O_5$ phases through the intermediate $La_3Co_3O_8$ phase at a temperature of 350 °C under high vacuum conditions, $\sim 10^{-5}$ mbar. The reverse SPT from BM to PV phases is also found under ambient pressure (>100 mbar) of air near 100 °C. During the onset of SPTs, the bulk chemical/electronic states of LCO thin films are monitored with AP-HAXPES. The oxidation states in Co 2*p* spectra indicate that the oxygen vacancies are closely related to the SPT of LCO thin films. Also, the presence of enlarged band gap is observed as the SPT from PV to BM phases takes place, revealing the modified electronic properties of LCO due to the creation of oxygen vacancies.

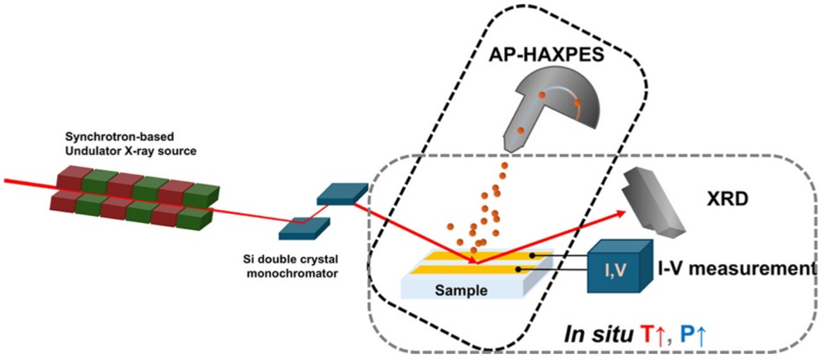

Figure 1. Schematic diagram of the experimental setup for XRD and AP-HAXPES.

Probing the Complex Electrode Interfaces of PEM Fuel Cells and Electrolyzers with 'Tender X-ray Operando AP-XPS

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Polymer electrolyte-based membrane electrode assemblies (MEAs) at the heart of many electrochemical devices such as fuel cells and electrolyzers, host complex composite electrodes. This complexity arises from its multifaceted structure encompassing a heterogeneous mixture of catalyst particles, ionomer networks and porous carbon supports, spanning sizes and length scales that collectively enable it to function as an electrocatalytic interface with ion and electron transport. Making progress on rational design for these electrocatalysts and complex interfaces requires effective characterization of the electrode-electrolyte interface where the chemistry takes place. Synchrotron-based X-ray Photoelectron Spectroscopy (XPS) offers a versatile way to obtain surface chemical and elemental characterization of such materials. The use of X-rays in the 'tender' regime (~2-6 keV) allows for deeper probing than the conventional 'soft' x-ray (< 2000 eV) set up that is common for this technique. The photoelectrons generated from tender x-rays have enough energy to penetrate tens of nanometers through low density components such as a layer of liquid or polymeric electrolyte, allowing access to interfaces in operando conditions. At Beamline 9.3.1 of the Advanced Light Source, we have developed a fully operational two-electrode system to study these membrane electrode assemblies under active conditions.

Accurate and representative data collection can involve a series of considerations and tests during acquisition. In this talk I will explain our specific approach, which combine conventional and sub-second snapshot acquisition modes to minimize beam effects and test for phenomena that can contribute to erroneous interpretation of data. I will demonstrate the efficacy of this approach with results on a series of iridium-based polymer electrolyte membranes used for water electrolysis. With application of elevated potential, we can correlate the iridium valency and surface species with the appearance of product traces in a mass spectrum.

Keywords: Interfaces, Electrolysis, *Operando* spectroscopy, X-ray Photoelectron Spectroscopy

Acknowledgement: Funding provided by the U.S. Department of Energy Office of Energy Efficiency and Renewable Energy, H_2NEW Consortium No. DE-AC02-05CH11231. J.M. and E.J.C. were partially supported by an Early Career Award in the Condensed Phase and Interfacial Molecular Science Program (CPIMS), No. DE-AC02-05CH11231 and E.J.C was partially supported by CPIMS under contract No. DE-AC02-0511231.

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Solution Based Single-Source Precursors for Doped Large-Area Growth of Transition Metal Dichalcogenides

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Two-dimensional semiconductors, such as transition metal dichalcogenides (TMDs), are vital for next-generation electronics, allowing device miniaturization at far lower length-scales than traditional silicon-based designs. Despite advances in scaling 2D materials, however, their integration in devices is still limited. This is largely due to traditional synthetic techniques being incompatible with industrial semiconductor manufacturing methods (e.g. high temperatures, contaminating gases/solids, damage from transfer, etc.). Herein, we developed solution-based precursors that yield large-area MoS_2 and WS_2 monolayers as well as promising dopants for n and p type counterparts at back-end-of-line (BEOL) compliant temperatures. With aid of the Advanced Light Source (ALS), we have probed the formation dynamics of pure and doped thin films of MoS_2 via *in-situ* ambient-pressure XPS (beamline 9.3.2) and elucidated the change in bonding coordination during annealing.

Keywords: 2D materials, semiconductors, MoS2, single-source precursors, doping, annealing, thin films

Acknowledgement: *This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Workforce Development for Teachers and Scientists, Office of Science Graduate Student Research (SCGSR) program. The SCGSR program is administered by the Oak Ridge Institute for Science and Education for the DOE under contract number DE-SC0014664.* [1]P.A. Author, S. Author, T. Author, "*Predicting Electronic Excitations in Organic Semiconductors with Machine Learning*", Journal of Exciting Results (12) 34567 (2025).

Probing UV Photoreaction Pathways of 2-Iodothiophene with Transient X-ray Absorption Spectroscopy.

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Photoinduced ring-opening mechanisms play a critical role in many chemical and biological processes such as vitamin D3 synthesis, DNA repair, and degradation of organic electronic materials. Thiophene derivatives, in particular, exhibit promising properties for use in solar cells and light-emitting diodes. However, further understanding of the underlying photophysics is crucial to overcome challenges with undesired photodegradation. 2-iodothiophene exhibits multiple energetically accessible reaction pathways, including C-I bond fission and C-S ring-opening. While iodine dissociation has been extensively studied, the predicted ring-opening reaction remains to be observed. Ultrafast transient X-ray absorption spectroscopy (TXA), an element-specific technique, enables detailed probing of the transient chemical environment in the vicinity of local reporter atoms, facilitating the study of even minor reaction channels. Using TXA at the iodine N-edge, the UV (268 nm) induced C-I bond fission has been shown to occur within approximately 160 fs.[1] This dissociation is slower compared to, for example, halogen abstraction in alkyl halides, which is attributed to a 2-step photoreaction where an initial $\pi\pi^*$ excitation couples nonadiabatically to a repulsive $\pi\sigma^*$ state. To investigate the ring-opening reaction, a different reporter atom is required, ideally one directly involved in bond fission, such as sulfur or carbon. Here, we employ a femtosecond high-harmonic generation (HHG) X-ray source to probe ultrafast dynamics in UV-excited 2-iodothiophene at the sulfur 2p, 2s, and carbon 1s edges. Preliminary results of 268 nm excited 2-iodothiophene focused on the sulfur L-edge are presented.

Acknowledgement: This work was supported by the Atomic, Molecular, and Optical Sciences Program of the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Chemical Sciences, Geosciences and Biosciences Division through Contract No. DE-AC02–05CH11231.

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Investigation on chemical/morphological variations of PEG-capped Au nanoparticles under oxygen environments using combined standing-wave photoemission and grazing incidence x-ray scattering

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Polyethylene glycol (PEG)-capped gold (Au) nanoparticles have been widely investigated in biosensor applications due to their low toxicity, noticeable size- and shape-dependent optical properties, large surface area, and high chemical stability. The PEG ligand capping layer prevents agglomeration of nanoparticles and enhances sensing performance by protecting the nanoparticles from the detection of immune system[1]. Therefore, stabilizing the PEG capping layer is critical for optimizing nanoparticle performance. However, in biological environments rich in oxidizing agents such as reactive oxygen species and water, understanding the reactivity between the ligands-capped nanoparticles and oxygen is essential for ensuring long-term stability.

In this work, with combined standing-wave ambient pressure x-ray photoelectron spectroscopy (SWAPPS) and grazing incidence X-ray scattering (GIXS) techniques, we investigated the chemical/morphological changes of polyethylene glycol (PEG)-capped Au nanoparticles on SiO_x/Si substrate under 100 mTorr of O_2 pressure. This unique system simultaneously provides both chemical and structural information of nanomaterials[2, 3]. The results of SWAPPS indicate that 3 distinct species of carbon (C=O, C-O, C-C/C-H) distributed at different depths. Under oxygen pressure, the volatile PEG components near the surface are oxidized and removed from the sample with the downward shift of Au nanoparticles. GIXS analysis reveals that PEG-capped Au nanoparticles have spherical morphology with an average size of 20 nm. Additionally, the density of carbon species is increased at the PEG capping layer during the oxidation, suggesting the possible solidification of the remaining PEG layer. Our combined results provide fundamental information on chemical/structural variations of PEG-capped Au nanoparticles under oxygen environment, which is crucial to the rational design of nanomaterials in biosensor applications.

Keywords: Polyethylene glycol-capped Au nanoparticles, standing-wave photoemission, grazing incidence x-ray scattering, oxidation of organic compound

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Correlation of Electronic Structure and Resistance Variation in ZnO Gas Sensors Through Operando NAP-XPS Studies During CO Exposure Under Dry and Humid Conditions

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Semiconducting metal oxides (SMOs) have been extensively studied over the past several decades for their potential in gas sensing applications. To gain a comprehensive understanding of the gas detection mechanism in metal oxide semiconductors (MOS) sensors, it is essential to examine the material's electronic structure during exposure to target gases under operational conditions. Zinc oxide (ZnO), a widely explored n-type SMO, has been a subject of extensive research as a gas sensor. In this work, operando near-ambient pressure X-ray photoelectron spectroscopy (NAP-XPS) was utilized to investigate the electronic structure of a ZnO-based gas sensor under exposure to CO gas. The operando NAP-XPS analysis reveals that the observed decrease in resistance during CO exposure is primarily attributed to changes in surface oxidation states, particularly the formation of oxygen vacancies. Under more realistic humid conditions, the resistance also decreases when exposed to CO, and this behavior is similarly linked to the formation of oxygen vacancies. However, the sensor's response to CO is significantly degraded in humid environments due to the formation of surface hydroxyl (OH) groups, which result from the filling of oxygen vacancies by water molecules. This study represents the first operando NAP-XPS investigation of a ZnO-based gas sensor under CO exposure in both dry and humid conditions. Our findings provide novel insights into the interplay between surface properties and the enhanced performance of ZnO as a CO gas sensor.

Keywords: NAP-XPS, Operando conditions, ZnO, CO gas, Gas sensor mechanism.

Atom-Holography Microscope for 3D Atomic Arrangement Analysis in Nano Area

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Kaho Hirano,[1] Shinako Iki,[1] Yu Masuda,[4] Koichi Moriguchi,[4] Keiko Ogai,[4] Yusuke Hashimoto[5]
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"Atomic-resolution holography [1, 2]" is a powerful technique that realized an analysis of local 3D atomic arrangement around not only constituent atoms in a crystal but also isolated atoms such as dopants. In photoelectron holography, an angular distribution of photoelectrons emitted from a target atom is used as a hologram, which can be used to directly derive the three-dimensional atomic arrangement around the emitter atom. Hologram requires at least ±50° cone angular distribution to reconstruct the 3D atomic arrangement with a kinetic energy of above 600 eV. DIANA [3] has been used to measure holograms effectively because it can display the angular distribution of ±60° at once. The energy resolution of DIANA is 1 %, which is insufficient to resolve chemical shift. Hence, we are developing a new high-energy-resolution display analyzer CoDELMA [4, 5]. CoDELMA is the only two-dimensional electron spectrometer that can display the angular distribution of particular energy electrons excited by electron beam over a wide two-dimensional angular range of ±50° at once. We are also developing atom-holography microscope by combining a scanning electron microscope (SEM) and CoDELMA. This new microscope enables us to analyze the 3D atomic arrangement around specific atoms at each nano region observed by SEM in the laboratory. Recent results are shown in the presentation. One example of the results is the Kikuchi pattern hologram from graphite sample, which shows the 3D atomic image reconstructed from the hologram. A honeycomb atomic arrangement of graphite is accurately reconstructed.

Keywords : Atom-holography microscope, CoDELMA, Kikuchi electron holography

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Poster P13: Poster Session ThI_20, Thursday, July 24, 06:00 PM - 07:30 PM

UltraARPES:
Next-generation nanoARPES techniques for coherent electron scattering detection

C. Jozwiak[1], E. Rotenberg[1], S. Moser[2], and A. Bostwick[1]
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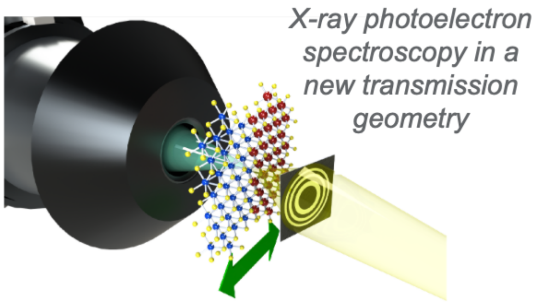
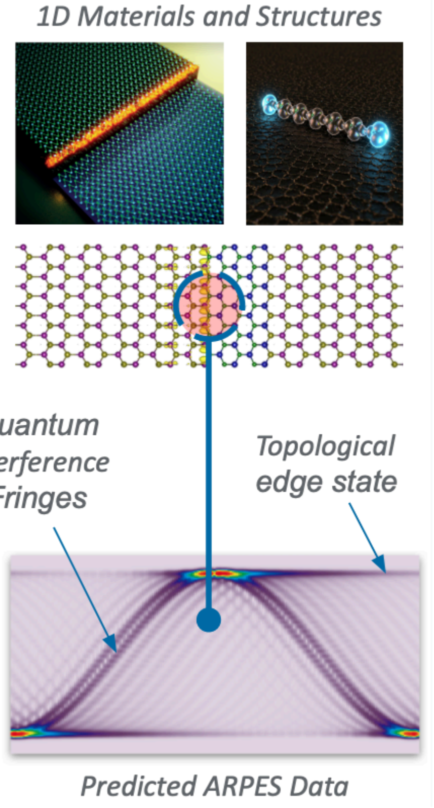
We are advancing the idea for a new nanoARPES experimental setup that aims to define the next generation of spectromicroscopic ARPES. By reaching down to the 10 nm length scale, the coherence length of electrons in many relevant systems, we expect that the ARPES technique will become sensitive to coherent electronic scattering processes for the first time. Besides, the advent of small beams will make the study of individual quantum objects such as quantum dots, step edges and other edge states accessible for the first time. Such a high spatial resolution can be achieved through the development of unprecedentedly high numeric aperture diffractive optics at EUV wavelengths, in combination with a scanning transmission xray microscopy (STXM) experimental geometry (figure, left).

Simple considerations of the photoemission process suggest that quantum interference effects will be visible in terms of sharp ringing of the photoemission spectra in momentum space (figure right) due to the phase shifts of electronic wave functions as they interact with defects or other interruptions of the periodic lattice order (figure, right). There is a direct analog between such k-space ringing of the photoemission signal with the real-space Friedel-like oscillations visualized in scanning tunneling microscopy (STM) which is the basis of FT-STM techniques that can extract k-space bandstructure from STM data, despite the lack of k-space resolution of STM. Here, we introduce the reciprocal technique whereby the ringing of the ARPES bands can be transformed into real-space maps of wavefunction probabilities, even at the atomic scale despite lack of atomic resolution in the of the xray probe beam. In analogy with x-ray-based ptyography, we have named the proposed resolution-enhancing technique "Ψ-chography".



Schematic of UltraARPES geometry and experiments. (above) X-ray photoelectron spectroscopy in a transmission geometry will enable soft xray measurements of electronic structure down to the 10nm length scale. (right) Such beams will be applied to low-dimensional quantum objects such as step edges, quantum wires, and quantum dots. When applied to discontinuous lattice structures, we expect quantum interference fringes in the ARPES data, related to wavefunction phase shifts at boundaries or defects.



Photoemission Spectro-Microscopy

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March 21, 2025

The analysis of photoemitted electrons remains the most direct method for probing the electronic properties of materials. Because photoemission studies are most often limited by the resolving capabilities of available instrumentation, there is always tremendous value in improving these capabilities. To that end, we are commissioning a time-of-flight electron lens for simultaneous spectroscopy and real/reciprocal microscopy of electrons photoemitted with a $6eV$ laser source. Our ultimate resolution goal is $100nm$ in real-space mode and $< 0.1^\circ$ in reciprocal-space mode with $1meV$ resolution in both modes. The novelty of high resolution, high throughput photoemission measurements in both real and reciprocal space will enable new science in exotic inhomogeneous samples.

Keywords: photoemission, spectroscopy, microscopy, electron optics

Acknowledgments: Work supported by the Moore Foundation

Current status of the high-resolution micro-ARPES system developed at NanoTerasu

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Angle-resolved photoemission spectroscopy (ARPES) is renowned for its powerful capability to probe the electronic structures of quantum materials, including high-temperature superconductors, topological insulators, and low-dimensional materials [1]. Recently, we have developed beamline 06U [2] at NanoTerasu, a 3-GeV high-brilliance synchrotron radiation facility located in Tohoku, Japan, dedicated to micro- and nano-focused ARPES (micro-ARPES and nano-ARPES). The micro-ARPES system has been open to general users since March 2025 [3]. The beamline provides brilliant soft X-rays with various polarizations (linear horizontal and vertical, as well as left and right circular) within the energy range of 50–1,000 eV, generated by a 4-meter-long APPLE-II-type undulator. Two distinct operational modes—a high-energy-resolution mode tailored for high-resolution micro-ARPES and a low-divergence mode optimized for high-flux nano-ARPES—can be selected by adjusting a collimated plane grating monochromator (cPGM).

In this presentation, we introduce detailed specifications and the current status of the versatile high-resolution micro-ARPES branch at BL06U, specifically focusing on the spin-resolved soft X-ray ARPES (SX-ARPES) system capable of micro-scale spatial resolution. To realize high-resolution spin-resolved SX-ARPES, a very low energy electron diffraction (VLEED)-type spin detector will be installed, providing approximately 100 times higher detection efficiency compared to conventional Mott-type spin detectors [4]. Additionally, the customized sample manipulator enables in situ application of various external fields while maintaining full rotational degrees of freedom. We also present representative ARPES data from strongly correlated electron systems, such as cuprates and ruthenates, to demonstrate the outstanding performance and quality of both the beamline and the micro-ARPES system.

Keywords: ARPES, micro-ARPES, spin-resolved ARPES, strongly correlated electron systems

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Poster P16: Poster Session ThI_20, Thursday, July 24, 06:00 PM - 07:30 PM

Present status of NanoTerasu BL06U: beamline for nano-ARPES measurements

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Yoshiyuki Ohtsubo, Jun Miyawaki, Kohei Yamamoto, Takeshi Nakatani, Kento Inaba,
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The beamline BL06U is one of the public beamlines at NanoTerasu, a 3-GeV high-brilliance synchrotron radiation facility at Tohoku, Japan. This beamline is designed for angle-resolved photoemission spectroscopy (ARPES) using micro-focused and nano-focused beams, including spin-resolving. The BL06U aims to produce a high flux nano-focused beam to realize highly efficient ARPES measurements by using ultrahigh precision reflective mirror optics such as a Wolter mirror, not a Fresnel zone plate used as a focusing optics on many nano-ARPES stations at other synchrotron facilities [1-3]. A 4 m long APPLE-II type undulator provides brilliant soft x-rays with various polarizations (liner horizontal and vertical, and left and right circular) in the 50–1,000 eV energy range. A collimated plane grating monochromator (cPGM) is adopted as a soft x-ray monochromator because we can select the fixed focus constant $c_{ff} = \cos\beta/\cos\alpha$ in between the high energy-resolution (HR) mode for HR-ARPES experiments and the low divergence mode for high-flux nano-ARPES measurements. A nano-ARPES station with a nano-focused beam of 100 nm or less will be installed in branch A, and a micro-ARPES station with a micro-focused beam of about 10 µm or less has already been installed in branch B [4]. We have been commissioning the beamline optics. The energy-resolution $E/\Delta E$ has achieved the designed value (over 50,000 at 65 eV), which was evaluated by multiple ionization spectra of helium gas. The micro-ARPES branch using the Scienta-Omicron R4000 electron analyzer has been open for users since March 2025, with a spot size of about 10 µm. In the presentation, we will introduce the detailed optical design and the present status of this beamline.

Keywords [optional]: angle-resolved photoemission spectroscopy (ARPES), nano-ARPES, micro-ARPES, synchrotron radiation beamline.

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Machine Learning for XAS Spectra Preprocessing and Visualization

Monika Choudhary[1], Dylan McReynolds[1], Tanny Chavez[1], Wanli Yang[1], Alexander Hexemer[1]
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Keywords: Xray absorption spectroscopy, denoising, machine learning, autoencoders, latent spaces

X-ray Absorption Spectroscopy (XAS) is a widely used technique for materials characterization. XAS data generated by beamlines can be noisy and challenging to analyze. To address this, we developed an interactive, web-based application in Python for preprocessing and visualizing XAS spectra, enabling rapid and effective data inspection and analysis.

Machine learning (ML) is emerging as a key tool for scientists to tackle complex and diverse data challenges[1]. The presented XAS preprocessing pipeline includes baseline correction and denoising. While traditional denoising methods rely on simple averaging or convolution-based techniques, we employ autoencoders trained specifically to denoise noisy spectra, thereby preserving fine spectral features more effectively[2]. To facilitate the comparison of multiple spectra simultaneously, we implement a one-dimensional latent space representation. A latent space (or embedding space) is a compressed representation of high-dimensional data that captures its underlying structure and variability. In our approach, autoencoders are used to learn this latent representation. Further dimensionality reduction using Uniform Manifold Approximation and Projection (UMAP)[3] is applied to map the latent space into a single dimension for intuitive visualization. We can also use a neural network (Parametric UMAP)[4] to reduce the latent space dimensions. The latent space obtained enables easier identification of similarities and differences among spectra. Additionally, our application provides tools to visualize statistical summaries within selected regions of the latent space—such as the median with interquartile range or the mean with minimum and maximum bounds—offering a clearer understanding of spectral variability within clusters.

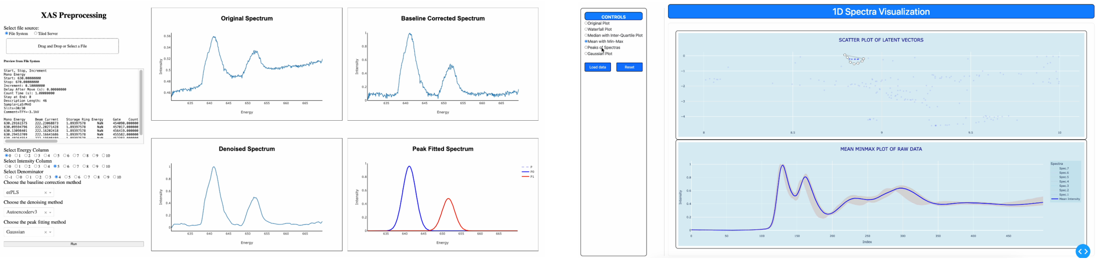

Figure 1. XAS data Preprocessing (left) and 1D Latent Space Visualization (right).

Acknowledgement: This work was performed and supported by the US Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences Data, Artificial Intelligence and Machine Learning at the DOE Scientific User Facilities program under the AIMM Project (award No. 107517) and MLExchange Project (award No. 107514).

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Application of machine learning methods in medical imaging to ARPES data denoising

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[2] *Case Western Reserve University, Cleveland, OH, USA*

Machine learning (ML) methods have been widely used in 3D and time-series medical image processing, delivering breakthroughs in disease diagnosis. Considering the high similarity of the data structure between ARPES data and medical images, we explore the use of ML methods in medical imaging to denoise ARPES data of low quality. We apply a novel self-supervised ML solution based on a masked loss function. We evaluate our method using experimental ARPES data from real 2D materials (see Figure 1), demonstrating powerful denoising performance. This research could be applied to ARPES data recovery, operando beamtime optimization and ARPES database construction.

Keywords [optional]: ARPES, Band Structure, Image Processing, Machine Learning, Materials Discovery.

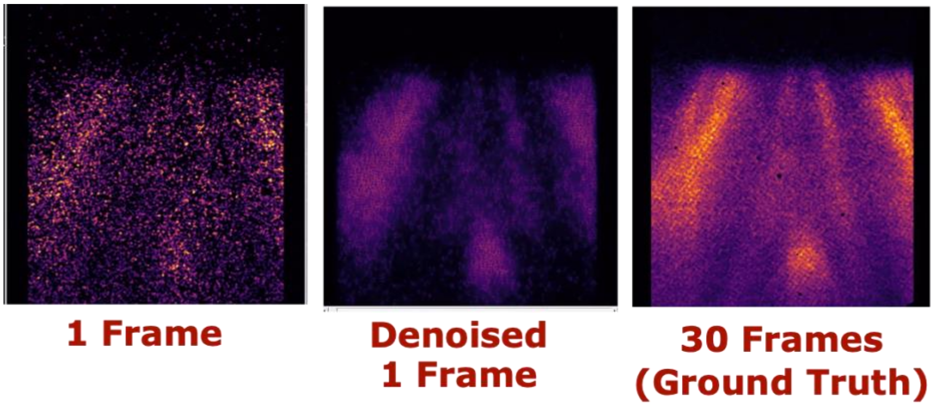

Figure 1. Raw and denoised ARPES data

Development of Microscopic-spectroscopy for a Wide-Energy-Range Soft X-ray Beamline

Takuji Ohigashi[1], Daisuke Wakabayashi[1], Hirokazu Tanaka[1], Ryoma Kataoka[1], Hiroaki Nitani[1],
Yoshio Suzuki[1], Norio Watanabe[2], Sadao Aoki[2]

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A wide-wavelength-range beamline, BL-12A, was constructed in Photon Factory (Tsukuba, Japan) and has started operation for general users since late 2024. A feature of BL-12A is two selective optical paths which have a plane grating monochromator (70~2,000 eV, E/ΔE ~3,000) and a double-crystal one (IbSn (111): 1,724~2,000 eV and Si (111): 2,500~5,000 eV) respectively. Focal points and optical axes of these two paths are common so that, in total, wide energy range from 70 to 5,000 eV is available at the end of the beamline. Hence, BL-12A is suitable to perform X-ray absorption spectroscopy (XAS) of various elements.

We have been developing a full-field imaging transmission soft X-ray microscope (TXM) as an application of XAS technique in BL-12A. The optical system is schematically shown in Fig. 1. A monochromatic X-ray illuminates a sample through combination of a rotating plane mirror and a cylindrical mirror as a condenser and its transmitted X-ray through the sample is imaged on 2D detector by a Wolter mirror as an objective. The Wolter mirror is a suitable objective for imaging XAS by using a bending magnet light source because of its large collecting power, high efficiency and no chromatic aberration[1]. An aim of TXM is to perform in-situ/operando spectroscopic measurement with time-lapse imaging. In this presentation, current status of BL-12A and a plan, design and preliminary experiment of TXM are discussed.

Keywords [optional]: X-ray microscopy, soft X-ray absorption spectroscopy, Wolter mirror

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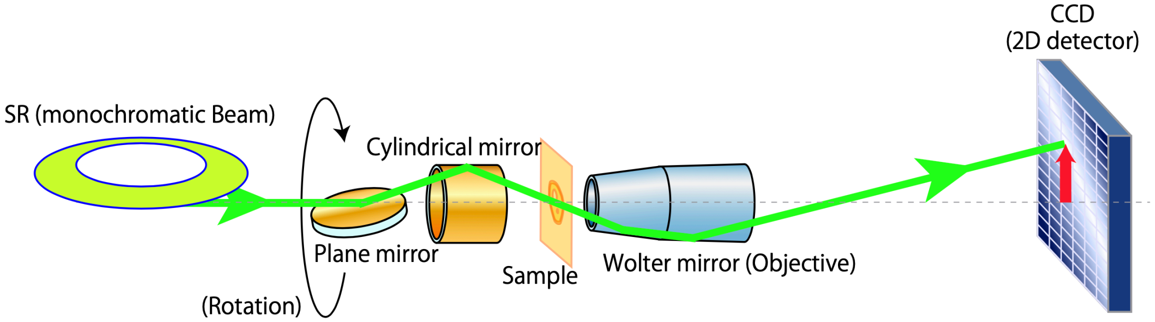

Figure 1. An optical system of a full-field imaging transmission soft X-ray microscope.

Quantum entanglement state between photoelectron spin and emitted photon polarization in spin- and polarization-resolved XEPECS of Ti_2O_3

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Recent advancements in synchrotron radiation technology have accelerated experiments on quantum entanglement in the X-ray regime. One key motivation is the potential for entangled X-ray photon pairs to enable low-dose, high-resolution X-ray imaging and nonlinear X-ray spectroscopy. However, the practical application of X-ray parametric down-conversion remains challenging due to its extremely low coincidence count rate. While high background noise remains a challenge, recent experiments combining electron energy loss spectroscopy and energy dispersive X-ray spectroscopy have reported coincidence counts exceeding 10^{10} (counts/sec) [1], suggesting the feasibility of efficiently generating entangled pairs of emitted electron and emitted X-ray pairs. To investigate another possibility producing the entangled pair, we have recently proposed a new mechanism using XEPECS process, i.e., XES (X-ray emission spectroscopy) and cXPS (core-level X-ray photoemission spectroscopy) coincidence spectroscopy [2], based on a simple two-level atomic model [3].

This study extends the theoretical framework to a more realistic model, aiming to elucidate the mechanism of quantum entanglement between photoelectron spin and emitted X-ray polarization in the XEPECS process. In the presentation, for $3d$ transition metal oxide Ti_2O_3, we reveal the mechanism of the spin and polarization entanglement using a TiO_6-type cluster model with the O_h symmetry, including the full-multiplet coupling effect and the charge-transfer effect between Ti $3d$ and ligand O $2p$ in the $3d \rightarrow 2p$ XEPECS process. In addition, we will discuss that the spin and polarization entanglement still survives even in the presence of negative effects due to the Ti $3d$ crystal-field splitting energy $10Dq$ and the hybridization interaction between the Ti $3d$ and O $2p$.

Keywords: X-ray quantum optics, spin and polarization entanglement, XES and cXPS coincidence spectroscopy, Ti_2O_3, MO_6-type cluster model.

Acknowledgement: This work was supported by JST, the establishment of university fellowships towards the creation of science technology innovation, Grant Number JPMJFS2138.

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Utilizing Crystalline Symmetries to Unlock New Data Analysis Techniques in ARPES

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Angle resolved photoemission spectroscopy (ARPES) yields information rich datasets for electron dispersions throughout momentum space. Under a free final state approximation, the electrons are on spheres of constant perpendicular momentum but are frequently approximated to be on planes of constant perpendicular momentum for ease of interpretation. Often, much of the data collected at wider angles is disregarded in ARPES experiments due to the complexity involved in interpreting it. In this talk, I will show a method for employing all of the collected data by exploiting the crystalline symmetries in momentum space. Applications to materials with three dimensional electronic structure, ARPES experiment planning, and polycrystalline samples will be discussed.

Acknowledgement: Work supported by the Gordon and Betty Moore Foundation GBMF Award ID #12957

Effect of MILD Method Etching Dynamics on the Crystalline Structure and Electronic Properties of Monolayer MXene

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In this study, the etching dynamics of the minimally intensive layer delamination (MILD) method on the Ti_3AlC_2 (MAX phase) were investigated using grazing-incidence wide-angle X-ray scattering (GIWAXS) at the Taiwan Light Source (TLS) BL13A1, micro X-ray photoelectron spectroscopy (μ-XPS) at TLS BL09A1 and soft X-ray absorption spectroscopy (XAS) at TLS BL20A1 of the National Synchrotron Radiation Research Center. The MILD method is a widely used approach that replaces the conventional hydrofluoric acid (HF) etching method with hydrochloric acid (HCl) and lithium fluoride (LiF) for removing aluminum layers from the MAX phase to obtain $Ti_3C_2T_X$ MXene flakes. The MXene flakes, etched using HCl:LiF ratios of 1:1 (MX1) and 1:2 (MX2), were placed on a silicon substrate to investigate the effects on the crystalline structure, chemical states, and physical properties. According to the results, the GIWAXS patterns exhibit a vertical stripe along the wave vector q_r range, which is the scattering characteristic of 2D materials, reflecting the crystal truncation rod (CTR) of monolayer MXene on the (101) plane. The contributions of signals further reveal that the q_r value of MX1 (2.51 $Å^{-1}$) is higher than that of MX2 (2.44 $Å^{-1}$), implying the differences of Ti-C atomic spacing within the crystal structure due to the ionization reaction of varying LiF concentration. To investigate the chemical states, μ-XPS measurements using a 100 nm beam were performed, which confirmed the complete removal of aluminum from the MXene sheets. Compared to MX2, the 0.2 eV energy shift observed in both the C 1*s* and Ti 2*p* spectra of MX1 is attributed to carbon deficiencies, which lead to an increased density of termination groups and a 0.2 eV alteration in the Fermi level. The combined analysis of the Ti 2*p* XPS and the Ti $L_{3.2}$-edge XAS results, reveals that the terminal group on MX1 induces significant charge transfer than that on MX2. XAS analysis reveals that the differential electron occupancy preference of Ti d orbitals, particularly within the t_{2g} and e_g orbitals, implies the distinct influence of carbon defects and terminal groups on the interfacial electronic structure of MX1 compared to MX2. Our results provide novel insights into the etching dynamics of the MILD method, revealing that the LiF concentration during MAX phase etching significantly influences the formation of carbon defects in monolayer MXene.

Keywords: 2D material, MXene, MILD method, etching dynamic, GIWAXS, μ-XPS, XAS.

Acknowledgement: Work supported by the National Science and Technology Council (NSTC) of Taiwan for providing financial support for research under the projects NSTC 113-2112-M-213-012-MY3.

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Acknowledgements

U. S. Government Support:

We gratefully acknowledge the support of these institutions, which are supported by the U. S. Department of Energy, Office of Basic Energy Sciences, under the following contracts:

Advanced Light Source, Lawrence Berkeley National Laboratory:
DE-AC02-05CH11231

SLAC National Accelerator Laboratory:
DE-AC02-76SF00515

National Synchrotron Light Source, Brookhaven National Laboratory:
DE-SC0012704