Applied Surface Science
Room 303 - Session AS1+CA+EL-ThM

Emerging Methods for Materials Characterization and Functional Properties

Moderators: Samantha Rosenberg, Kairos Power, **Xiao-Ying Yu**, Oak Ridge National Laboratory, USA

8:00am **AS1+CA+EL-ThM-1 Anisotropic Complex Dielectric Function Response of Mg2SiO4 Single Crystal**, ***Emily Amonette***, University of Toledo; *Darrell Schlom,* Cornell University; *Nikolas Podraza,* University of Toledo

Single crystal Mg_2SiO_4, also called forsterite or peridot in its gem form, has been measured via spectroscopic ellipsometry over a spectral range of 0.78—8.5 eV to determine its anisotropic dielectric response over its a- and c-crystallographic axes. These axes are investigated because the available (010) crystal cut does not allow optical sensitivity to the b-axis. Forsterite has potential to serve as a substrate for epitaxial superconducting RuO_2 film growth and as a bioceramic in bone and dental applications. Single crystal forsterite has an orthorhombic crystal structure belonging to space group Pbnm.Collecting multiple measurements and rotating the sample between each measurement varies the Euler angles and increases sensitivity to anisotropy. Within the measured spectral range, oscillators describing optical absorption are used to model the imaginary part of the dielectric spectra, and Kramers-Kronig integration of these oscillators plus Sellmeier functions model the real part of the dielectric spectra. Absorption onset occurs at 7.5 eV along the a-axis and 6.6 eV along the c-axis.

8:15am **AS1+CA+EL-ThM-2 Spin-Mediated Magnetochromism in Cr3+:β-Ga2O3: From THz Spectroscopy Ellipsometry to Nonreciprocal Thermal Emission Signatures**, ***Sina Khayam***, University of Nebraska-Lincoln, USA; *Viktor Rindert,* Lund University, Sweden; *Ufuk Kilic,* UNIVERSITY OF NEBRASKA-LINCOLN; *Vanya Darakchieva,* Lund University, Sweden; *Mathias Schubert,* University of Nebraska-Lincoln, USA

Magnetic-dipole transitions of paramagnetic spins provide a largely unexplored route for controlling thermal radiation at cryogenic temperatures. We demonstrate this concept using Cr^{3+} centers in β-Ga_2O_3 by combining polarization-resolved millimeter-wave spectroscopy with a spin-Hamiltonian/Bloch electrodynamic model [1–3]. Measurements at T = 15 K, magnetic fields of approximately 6.5–8 T, and 190–230 GHz validate the three field-dependent Cr^{3+} spin transitions through direct comparison of experiment and calculation.

The measured complex permeability is incorporated into a full-wave 4×4 Berreman propagation treatment [4,5] to obtain reflected and transmitted powers and hence spin-resolved absorptivity. Because magnetic bias makes the response nonreciprocal, emissivity is evaluated using the field-reversed adjoint relation rather than the conventional same-state Kirchhoff law [6,7]. With $\mu(-B)=\mu^T(B)$, the circular-polarization channels satisfy $\varepsilon R(+B)=AL(-B)$ and $\varepsilon L(+B)=AR(-B)$, while for unpolarized radiation $\varepsilon unp(+B)=Aunp(-B)$.

Within this framework, magnetic field tunes the spin-resonance frequencies through Zeeman coupling, while temperature redistributes the spin populations and modifies their radiative strength. Planck-weighted calculations show narrow, magnetically tunable thermal-emission channels whose spectral positions are set by the field-dependent Cr^{3+} transitions and whose strengths depend on population differences and overlap with the blackbody spectrum. Maps versus magnetic field and temperature identify operating conditions where one or more resonances contribute most strongly to the emitted thermal power.

The calculations further show that same-state absorption and emission are generally unequal, while the field-reversed adjoint relations are recovered. These results establish paramagnetic spin resonance as an intrinsic, contactless mechanism for spectrally selective thermal-emission control without patterned resonators or cavities, linking experimentally validated magnetic-resonance spectroscopy with nonreciprocal thermal photonics.

References
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[2] V. Rindert et al., Phys. Rev. Lett. 134, 086703 (2025).
[3] V. Rindert et al., Appl. Phys. Lett. 126, 082105 (2025).
[4] D. W. Berreman, Opt. Soc. Am. 62, 502 (1972).
[5] M. Schubert, Phys. Rev. B 53, 4265 (1996).
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[7] C. Khandekar et al., New J. Phys. 22, 123005 (2020).

8:30am **AS1+CA+EL-ThM-3 Near-Infrared to VUV Characterization of a Si-doped α-(AlxGa1-x)2O3 Transistor Structure with a 7 eV Bandgap using General Spectroscopic Ellipsometry**, ***Preston Sorensen***[1], University of Nebraska - Lincoln; *Jacob Steele, Debaditya Bhattacharya, Kazuki Nomoto, Naomi Pieczulewski,* Cornell University; *Viet-Anh Ha, Nick Pant,* University of Texas at Austin; *Ihit Shukla, Shaon Das,* University of Buffalo; *Ufuk Kilic,* University of Nebraska - Lincoln; *Madhav Ramesh,* Cornell University; *Feliciano Giustino,* University of Texas at Austin; *Baishakhi Mazumder,* University of Buffalo; *Matara Kankanamge Indika Senevirathna, Michael Williams,* Clark Atlanta University; *Mathias Schubert,* University of Nebraska - Lincoln; *David Muller, Huili Xing, Debdeep Jena, Darrell Schlom,* Cornell University

Demand in high-power switching electronics has driven the need for wider and wider bandgap semiconductors. Recent developments have moved from Gallium Oxide towards solid solutions of Aluminum-Gallium Oxide. Alpha-phase Aluminum-Gallium Oxide is a colossal bandgap semiconductor with a rhombohedral crystal structure. Multilayer heterostructures and a MESFET with layers of varying Aluminum concentration were grown by Suboxide Molecular-Beam Epitaxy on commercial sapphire substrates. General Spectroscopic Ellipsometry was performed on the samples and the template in the spectral range of 0.7 to 9.2 eV. The anisotropic dielectric functions were modeled using the direct band-to-band transition critical-point model developed by Hilfiker *et al.*. The channel layer of the MESFET was found to have an optical bandgap of 6.71 eV. Thick, relaxed epitaxial layers were found to have bandgaps up to 7 eV.

8:45am **AS1+CA+EL-ThM-4 XPS Spectrometer Transmission Correction using the Consistent Retardation Assumption**, *Neal Fairley,* Casa Software, UK; ***Alexander Shard***, National Physical Laboratory, U.K.; *Pascal Bargiela,* Universite de Pau, France; *Jonas Baltrusaitis,* Lehigh University; *Miklós Németh,* HUN-REN Centre for Energy Research, Hungary; *Shaoliang Guan,* University of Cambridge, UK; *David Morgan,* Cardiff University, UK; *Aurelien Renard,* Universite de Lorraine, France; *Timothy Livingston Large,* Sandia National Laboratories; *Vincent Fernandez,* Universite de Nantes, France; *Yusuf Koçak,* Bilkent University, Turkey; *Huanyong Li,* Jinan University, China; *Anna Kallistova,* University of California; *Robert Hengstebeck,* Pennsylvania State University; *Olivier Heintz,* Ecole Centrale de Lyon - LTDS, France; *Solene Bechu,* Universite de Versailles, France; *Guillame Sauthier,* Catalan Institute of Nanoscience and Nanotechnology, Spain; *Isidoro López,* Universite de Bourgogne, France; *Hannah Levene,* University of Edinburgh, UK; *Ryan Thorpe,* Lehigh University

Accurate and comparable XPS data relies upon the correct calibration of the intensity scale. A method, called here the “Consistent Retardation Assumption” (CRA) procedure and used by some manufacturers to compute the energy-dependent transmission response of an X-ray photoelectron spectroscopy (XPS) spectrometer, is evaluated for modern XPS instruments. The CRA computes a transmission curve which depends only upon the retardation ratio of the analyzer. A step-by-step guide using an exemplary instrument is given, and the method was applied to more than 20 instruments in this study. The internal consistency of CRA calibrations was assessed from the scatter between corrected spectra taken using different pass energies. The accuracy of CRA calibrations was assessed by comparing the calibrated gold spectra to the appropriate gold reference spectra from the National Physical Laboratory, UK. The exemplary instrument produces an excellent CRA calibration in its main operational mode. The calibrated spectra are consistent with each other and agree closely to the NPL reference spectrum. In different modes of operation, specifically small area analysis modes, the CRA procedure works poorly for this instrument. Other XPS instruments were calibrated using the CRA method with various degrees of success. Approximately one third of XPS instruments in the study could be accurately calibrated to within 5 % of the NPL reference spectrum. We recommend that, before using the approach, the suitability of the CRA method first be assessed and methods to do this are outlined.

9:00am **AS1+CA+EL-ThM-5 Autonomous ToF-SIMS Studies with AI-Agent Quality Control**, ***Anton Ievlev***[2], Oak Ridge National Laboratory

Time-of-flight secondary ion mass spectrometry (ToF-SIMS) is a workhorse of local chemical characterization across a broad range of materials and systems, with more than 5,000 publications reporting its use since 2016. Yet the technique requires constant supervision by an expert to watch data acquisition, adjust sputter and analysis parameters, and decide when

[1] **JVST Highlighted Talk**
[2] **JVST Highlighted Talk**

something has gone wrong. Here, we present PACE-SIMS (Pause–Assess–Correct–Execute for SIMS), an autonomous workflow on an unmodified commercial TOF.SIMS 5 instrument in which those judgments are delegated to an artificial-intelligence (AI) agent under human-approved limits. The researcher specifies the study in plain language; the agent builds the measurement plan and, after a go/no-go review, executes it, pausing at checkpoints after every acquisition to assess data quality the way an operator would (layer thickness, signal levels, crater depth consistency, detector saturation) and to accept the measurement, correct parameters and remeasure, retry, or escalate to the human. We validated the approach in a randomized, blind, two-polarity study of ^{18}O-enriched WO_x films with a buried tracer layer: 35 measurements in 8.1 hours from about 40 minutes of total human attention. The agent recalibrated per-sample sputter settings that differed by 44% across the films, held isotope ratios steady through a 32% swing in absolute yields, and correctly attributed a sustained 30% intensity drop to a primary-beam emission fault rather than the sample, escalating once for source realignment before resuming without loss of the sample budget. Beyond demonstrating autonomous operation, the experiment also provided insight into the dynamic properties of the WO_x films, whose oxygen stoichiometry controls the resistive-switching and electrochromic behavior that motivates these materials. The proposed architecture is instrument-portable and can be applied to any analytical technique, especially destructive ones.

The research and developments were supported by the Center for Nanophase Materials Sciences (CNMS), which is a US Department of Energy, Office of Science User Facility at Oak Ridge National Laboratory.

9:15am **AS1+CA+EL-ThM-6 in Situ Molecular Imaging of Ion Clusters Reveals the Acid Gas Capture Capacity and Mechanism of Water-Lean Ionic Liquids**, ***Xiao-Ying Yu***, *Difan Zhang,* Oak Ridge National Laboratory*; Zihua Zhu,* Pacific Northwest National Laboratory*; Gabriel Parker, Roger Rousseau,* Oak Ridge National Laboratory

Water-lean solvents are a promising technology for capturing acid gases like carbon dioxide (CO_2).In situ liquid time-of-flight secondary ionization mass spectroscopy (ToF-SIMS) is used to study a representative solvent N-(2-ethoxyethyl)-3-morpholinopropan-1-amine (2-EEMPA) with different CO_2 loadings to reveal the complex solvent structure upon CO_2 capture.Characteristic peaks of 2-EEMPA, such as m/z^- 215 $C_{11}H_{23}N_2O_2^-$ (deprotonated 2-EEMPA) and m/z^+ 217 $C_{11}H_{25}N_2O_2^+$ (protonated 2-EEMPA), are detected due to acid gas uptake.Also, solvent molecules and carboxylate ion pairs, such as m/z^- 259 $C_{12}H_{23}N_2O_4^-$ [(deprotonated 2-EEMPA$\cdots CO_2$)] and m/z^+ 261 $C_{12}H_{25}N_2O_4^+$ (protonated 2-EEMPA$\cdots CO_2$), are observed.Interestingly, more than one CO_2 molecule can be captured per each solvent molecule as evidenced in SIMS mass spectra, for example, m/z^- 321 $C_{13}H_{25}N_2O_7$ [(deprotonated 2-EEMPA)$\cdots 2CO_2\cdots H_2O$], m/z^+ 305 $C_{13}H_{25}N_2O_4^+$ [(protonated 2-EEMPA)$\cdots 2CO_2$], m/z^- 389 $C_{17}H_{29}N_2O_8$ [(deprotonated 2-EEMPA)$\cdots 3CO_2\cdots 3CH_2$], and m/z^+ 373 $C_{16}H_{25}N_2O_8^+$ [(protonated 2-EEMPA)$\cdots 3CO_2\cdots 2C$].However, the monomer of 2-EEMPA and CO_2 seems to be most prevalent.Furthermore, solvent clusters are detected in loaded solvents, for instance m/z^+ 433 $C_{22}H_{49}N_4O_4^+$ [(2-EEMPA)$_2\cdots$H] and m/z^+ 646 [(2-EEMPA)$_3$-2H], while capturing CO_2 at different amounts.Relative abundance of cluster ions provides a semi-qualitative venue to assess the free energies gas capture energetics, indicating the relative stability trend within the same solvent system, previously impossible.These observed ion clusters are verified with molecular modeling, where dimer, trimer, and cluster ions are validated for their presence either due to weak molecular interactions or hydrogen bonds.In situ molecular imaging of ionic liquids and molecular modeling reveals that the acid gas capture mechanism by ionic liquids includes both physical adsorption and chemical bonding with multiple reaction pathways, engaging cluster formation and alteration of solvent structures.

9:30am **AS1+CA+EL-ThM-7 Co-Adsorption of Acetic Acid with H2O on Anatase TiO2(101)**, ***Joshua Wagner***, *Zbynek Novotny, Zdenek Dohnalek,* Pacific Northwest National Laboratory

The adsorption geometry of carboxylic acids on metal oxides controls key steps in ketonization. While molecular and deprotonated binding modes of acetic (and formic) acid on anatase TiO_2(101) are known, how co-adsorbed water influences adsorption, proton transfer, and surface mobility on this facet remains unclear. Here, we combine angle- and energy-resolved molecular beam scattering with scanning tunneling microscopy to probe acetic-acid binding motifs in the presence of water on anatase TiO_2(101). To first build a molecularly resolved understanding of the structure of water on a-TiO_2(101), precise surface coverages of water are prepared revealing complex coverage dependent adsorption motifs. Water molecules in the second layer are coordinated to the surface through hydrogen bonding and are shown through timelapse STM to participate in coordinated surface diffusion. The site-specificity and stability of acetic acid co-adsorbed with water is then visualized through STM. Overall, this work provides a molecularly resolved description of the dynamics and structure of water, acetic acid, and acetate intermediates on a-TiO_2(101) with results relevant for ketonization on metal oxides.

Author Index

Bold page numbers indicate presenter

— A —
Amonette, Emily: AS1+CA+EL-ThM-1, **1**
— B —
Baltrusaitis, Jonas: AS1+CA+EL-ThM-4, 1
Bargiela, Pascal: AS1+CA+EL-ThM-4, 1
Bechu, Solene: AS1+CA+EL-ThM-4, 1
Bhattacharya, Debaditya: AS1+CA+EL-ThM-3, 1
— D —
Darakchieva, Vanya: AS1+CA+EL-ThM-2, 1
Das, Shaon: AS1+CA+EL-ThM-3, 1
Dohnalek, Zdenek: AS1+CA+EL-ThM-7, 2
— F —
Fairley, Neal: AS1+CA+EL-ThM-4, 1
Fernandez, Vincent: AS1+CA+EL-ThM-4, 1
— G —
Giustino, Feliciano: AS1+CA+EL-ThM-3, 1
Guan, Shaoliang: AS1+CA+EL-ThM-4, 1
— H —
Ha, Viet-Anh: AS1+CA+EL-ThM-3, 1
Heintz, Olivier: AS1+CA+EL-ThM-4, 1
Hengstebeck, Robert: AS1+CA+EL-ThM-4, 1
— I —
Ievlev, Anton: AS1+CA+EL-ThM-5, **1**
Indika Senevirathna, Matara Kankanamge: AS1+CA+EL-ThM-3, 1
— J —
Jena, Debdeep: AS1+CA+EL-ThM-3, 1
— K —
Kallistova, Anna: AS1+CA+EL-ThM-4, 1
Khayam, Sina: AS1+CA+EL-ThM-2, **1**
Kilic, Ufuk: AS1+CA+EL-ThM-2, 1; AS1+CA+EL-ThM-3, 1
Koçak, Yusuf: AS1+CA+EL-ThM-4, 1
— L —
Levene, Hannah: AS1+CA+EL-ThM-4, 1
Li, Huanyong: AS1+CA+EL-ThM-4, 1
Livingston Large, Timothy: AS1+CA+EL-ThM-4, 1
López, Isidoro: AS1+CA+EL-ThM-4, 1
— M —
Mazumder, Baishakhi: AS1+CA+EL-ThM-3, 1
Morgan, David: AS1+CA+EL-ThM-4, 1
Muller, David: AS1+CA+EL-ThM-3, 1
— N —
Németh, Miklós: AS1+CA+EL-ThM-4, 1
Nomoto, Kazuki: AS1+CA+EL-ThM-3, 1
Novotny, Zbynek: AS1+CA+EL-ThM-7, 2
— P —
Pant, Nick: AS1+CA+EL-ThM-3, 1
Parker, Gabriel: AS1+CA+EL-ThM-6, 2
Pieczulewski, Naomi: AS1+CA+EL-ThM-3, 1
Podraza, Nikolas: AS1+CA+EL-ThM-1, 1
— R —
Ramesh, Madhav: AS1+CA+EL-ThM-3, 1
Renard, Aurelien: AS1+CA+EL-ThM-4, 1
Rindert, Viktor: AS1+CA+EL-ThM-2, 1
Rousseau, Roger: AS1+CA+EL-ThM-6, 2
— S —
Sauthier, Guillame: AS1+CA+EL-ThM-4, 1
Schlom, Darrell: AS1+CA+EL-ThM-1, 1; AS1+CA+EL-ThM-3, 1
Schubert, Mathias: AS1+CA+EL-ThM-2, 1; AS1+CA+EL-ThM-3, 1
Shard, Alexander: AS1+CA+EL-ThM-4, **1**
Shukla, Ihit: AS1+CA+EL-ThM-3, 1
Sorensen, Preston: AS1+CA+EL-ThM-3, **1**
Steele, Jacob: AS1+CA+EL-ThM-3, 1
— T —
Thorpe, Ryan: AS1+CA+EL-ThM-4, 1
— W —
Wagner, Joshua: AS1+CA+EL-ThM-7, **2**
Williams, Michael: AS1+CA+EL-ThM-3, 1
— X —
Xing, Huili: AS1+CA+EL-ThM-3, 1
— Y —
Yu, Xiao-Ying: AS1+CA+EL-ThM-6, **2**
— Z —
Zhang, Difan: AS1+CA+EL-ThM-6, 2
Zhu, Zihua: AS1+CA+EL-ThM-6, 2