Third International Symposium on Advanced Microscopy and Spectroscopy on the Occasion of Celebrating the NSF MRSEC at UCI

University of California, Irvine

November 5-7, 2020

CCAM | Center for Complex and Active Materials | An NSF MRSEC at UCI

UC IRVINE MATERIALS RESEARCH INSTITUTE

Sponsors

To celebrate the establishment of the Center for Complex and Active Materials (CCAM) – a new Materials Research Science and Engineering Center (MRSEC) funded by National Science Foundation (NSF), UCI Irvine Materials Research Institute (IMRI) is delighted to host a special materials research workshop in conjunction with the Third International Symposium on Advanced Microscopy and Spectroscopy (ISAMS-3), on November 5-7, 2020. A workshop centered on CCAM activities will be held on November 5, to kick off the UCI MRSEC, followed by ISAMS-3 on November 6-7, 2020. The ISAMS-3 will bring together the scientific community working on various aspects of transmission electron microscopy to exchange latest research results and ideas, and to address challenges in the development of atomic scale imaging and spectroscopy for advancing materials and biological sciences. Both events will be held virtually via Zoom and are open to the scientific community. For more information about the events, please visit: https://sites.uci.edu/isams3/. To register for the meeting, please visit: https://sites.uci.edu/isams3/#Registration. A Zoom link will be sent to all registered attendees by email.

The National Science Foundation has awarded $18 million to UCI in support of the CCAM – a new NSF MRSEC. The CCAM builds on campus strengths in multidisciplinary science and engineering research, world-class facilities, and commitment to diversity. The primary mission of CCAM is to establish foundational knowledge in the science and engineering of new classes of materials offering unique and broad functionality via an interplay among design, simulation, synthesis, and advanced characterization.

Established in 2015, UCI IMRI is an interdisciplinary special research program under the Office of Research, and a key enabler of the CCAM initiative. It serves as the cross-campus nexus for materials research in Southern California. IMRI operates a wide range of state-of-the-art shared facilities for the analysis and characterization of materials and devices ranging from sub-Å to macroscopic length scales. The facilities are professionally staffed, convenient, and affordable, with user-friendly services.

We are honored to invite you to join us at the special materials research workshop and the ISAMS-3, to share with you our accomplishments and to enable future collaborations amongst attendees.

Xiaoqing Pan

Director, IMRI and CCAM – An NSF MRSEC

Agenda

Workshop on Complex and Active Materials – a Kick-off meeting of the UCI MRSEC

Thursday, November 5, 2020

	Session Chair: Xiaoqing Pan
8:40 – 9:00	Opening Remarks by Prof. Pramod Khargonekar, Vice Chancellor for Research
9:00 – 9:30	Xiaoqing Pan, Overview of UCI MRSEC
9:30 – 10:00	Tim Rupert, IRG 1
10:00 – 10:30	Zhibin Guan, IRG 2
10:30 – 10:45	Regina Regan, Education/Outreach
10:45 – 11:00	Break
11:15 – 11:45	Greg Rohrer, Carnegie Mellon University High Throughput Studies of Metal Oxide Water Splitting Catalysts for the Development of Structure-Property Relations
11:45 – 12:30	Paul Voyles, University of Wisconsin - Madison Relationships Between Stability, Structure, and Dynamics in Glasses and Their Liquids
12:30 – 1:30	**Lunch**
	Session Chair: Ruqian Wu
1:30 – 2:15	Ramamoorthy Ramesh, UC Berkeley & LBNL Observation of Room Temperature Polar Skyrmions
2:15 – 3:00	Wilson Ho, UC Irvine Atomic Scale Inelastic Electron Excitation Spectroscopy and Microscopy
3:30 – 3:45	**Break**
	Session Chair: Zhibin Guan
3:45 – 4:30	Phillip Messersmith, UC Berkeley & LBNL Supramolecular Polymers for Tissue Regeneration
4:30 – 5:15	Takuzo Aida, RIKEN CEMS & University of Tokyo Supramolecular Polymerization: Its Significance and Applications
5:15	Closing remarks (**Ruqian Wu**)

3rd International Symposium on Advanced Microscopy and Spectroscopy

Friday, November 6, 2020

	Session I: Noble Imaging and Spectroscopy of advanced Materials **Session Chair: Xiaoqing Pan**
8:00 – 8:30	Angus Kirkland, University of Oxford Developments in High Speed Structural Imaging of Low Dimensional Materials
8:30 – 9:00	Elizabeth Dickey, North Carolina State University Local Structure Phenomena in Oxide Dielectrics
9:00 – 9:30	Frances Ross, Massachusetts Institute of Technology Opportunities for Understanding Crystal Growth through in situ Electron Microscopy
9:30 – 10:00	David McComb, Ohio State University Monochromated EELS of Organic Functional Materials in the STEM
10:00-10:30	**Break**
	Session II: Liquid Phase Electron Microscopy **Session Chair: Joe Patterson**
10:30 – 11:00	See Wee Chee, Fritz-Haber-Institut der Max-Planck-Gesellschaft Revealing the Dynamics of Electrocatalysts under Reaction Conditions using Liquid Phase Electron Microscopy
11:00 – 11:30	Qian Chen, University of Illinois at Urbana Champaign Liquid-Phase TEM Imaging of Colloidal Crystallization and Protein Transformation
11:30 – 12:00	Joe Patterson, UC Irvine A Close Look at Material Synthesis with Liquid Phase and Cryo Electron Microscopy
12:00 – 1:00	**Lunch Break**
	Session III: Cryo-EM in Materials/Biological sciences **Session Chair: Shane Goenen**
1:00 – 1:30	Hong Zhou, University of California, Los Angels The CryoEM Revolution: From Proteins to Genomes & from *in situ* to in Action
1:30 – 2:00	Brent Nannenga, Arizona State University High-Resolution Structure Determination by Microcrystal Electron Diffraction
2:00 – 2:30	Lena Kourkoutis, Cornell University Progress in Cryogenic STEM for Quantum and Energy Materials
2:30 – 3:00	John Watt, Los Alamos National Laboratory Limiting Damage and Capturing Dynamics of Soft Matter and Beam Sensitive Materials
3:00 – 3:30	**Break**

	Session IV: In-situ S/TEM **Session Chair: Toshiro Aoki**
3:30 – 4:00	Judith Yang, University of Pittsburgh The Surface Dynamics of the Initial Stages of Cu Oxidation
4:00 – 4:30	Wei-Chang D. Yang, National Institute of Standards and Technology Probing Surface-Plasmon-Induced Reactions using a Multimodal Approach for Environmental Scanning Transmission Electron Microscopy
4:30 – 5:00	Ray Unocic, Oak Ridge National Laboratory Atomic Engineering of 2D Materials: Insights from In Situ STEM Experiments, Theory and Functional Properties
5:00 – 5:30	Xuedong Bai, Chinese Academy of Sciences Atomic-Scale Observation of Structural Manipulation in Metal Oxides by in-situ Transmission Electron Microscopy

Saturday, November 7, 2020

	Session III: 4D STEM **Session Chair: Xiaoqing Pan**
8:00 – 8:30	Peter Nellist, University of Oxford Low-dose 2D and 4D STEM Imaging of Beam-sensitive Materials
8:30 – 9:00	Colin Orphus, Lawrence Berkeley National Laboratory Materials Science Applications of 4D-STEM
9:00 – 9:30	Peter Ercius, Lawrence Berkeley National Laboratory The 4D Camera – An Electron Counting Camera for 4D-STEM Experiments
9:30 – 10:00	Xiaoqing Pan, UC Irvine Probing the Local Charge and Phonons of Single Defects by Electron Microscopy and Spectroscopy
10:00-10:30	**Break**
	Session IV: STEM EELS **Session Chair: Huolin Xin**
10:30 – 11:00	Peter Crozier, Arizona State University Exploring Phononic Excitations with Monochromated STEM EELS
11:00 – 11:30	Christoph Koch, Humboldt-Universität zu Berlin Maximizing the Information Extracted from Monochromated EELS and Zero-Loss Filtered 4D-STEM using a Hybrid Pixel Direct Detector with High Dynamic Range.
11:30 – 12:00	Juan-Carlos Idrobo, Oak Ridge National Laboratory Mapping Electron Interactions with a Pocket-Sized Synchrotron (i.e., STEM)
12:00 – 12:30	Huolin Xin, UC Irvine Making Rocket Fuel From CO_2 – from Intermetallics To Non-PGM Biomimetic Catalysts for Small Molecule Activation
12:30	**Closing Remark**

Abstracts

High Throughput Studies of Metal Oxide Water Splitting Catalysts for the Development of Structure-Property Relations

Greg Rohrer

Mingyi Zhang, Wenjia Song, Paul A. Savador, and Gregory S. Rohrer

Department of Materials Science and Engineering, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, USA

The performance of a water splitting catalyst depends on many parameters related to the composition, processing, and structure of the material, as well as the conditions in the reactor. This research uses a newly developed parallel and automated photochemical reactor (PAPCR) that makes it possible to measure the rate of hydrogen yield from up to 108 catalysts in a single experiment, systematically determining the influence of particle shape, particle size, dopants, charged surface domains, protective coatings, co-catalysts, and many other catalyst characteristics on the hydrogen production rate [1]. This talk will focus on the results of studies of $SrTiO_3$, $BaTiO_3$, and heterostructured catalysts consisting of TiO_2 coated $SrTiO_3$ and $BaTiO_3$. We have found that the rate of hydrogen yield from aluminum doped $SrTiO_3$ is a sensitive function of the processing method, the particle size, the particle shape, and the reactor pH. These findings can be understood in terms of the properties of the individual low index surfaces that bound the particles and how they separately promote the reduction and oxidation half-reactions [2]. The prospects for using the PAPCR to develop additional structure-performance relationships for improved water splitting catalysts will be discussed.

References:

[1] W. Song, E.M. Lopato, S. Bernhard, P.A. Salvador, G.S. Rohrer, "High-throughput measurement of the influence of pH on hydrogen production from $BaTiO_3/TiO_2$ core/shell photocatalysts," Applied Catalysis B: Environmental **269** (2020) 118750.

[2] M.Y. Zhang, P.A. Salvador, G.S. Rohrer, "Influence of pH and surface orientation on the photochemical reactivity of $SrTiO_3$" ACS Applied Materials and Interfaces, **12** (2020) 23617-23626

Biosketch

Prof. Gregory S. Rohrer received his Bachelors degree in Physics from Franklin and Marshall College, his Doctoral degree in Materials Science and Engineering from the University of Pennsylvania, and joined the faculty at Carnegie Mellon in 1990, where he is now the W.W. Mullins Professor and Head of the Department of Materials Science and Engineering. Prof. Rohrer has authored or co-authored more than 310 publications. He is a fellow of the American Ceramic Society and his research has been recognized by a number of awards including the Richard M. Fulrath Award, the Robert B. Sosman Award, and the W. David Kingery Award, all of the American Ceramic Society. In 2011, he served as chair of the University Materials Council, from 2016 to 2019 he was a member of the Board of Directors of the American Ceramic Society, and he is currently an editor of Acta Materialia.

Relationships Between Stability, Structure, and Dynamics in Glasses and Their Liquids

Paul Voyles

Department of Materials Science and Engineering, University of Wisconsin – Madison, WI 53706, USA

Glasses are complex materials. They can be synthesized by quenching from a liquid or deposition from a vapor, which results in materials in a variety of metastable states with different enthalpies. They exhibit a wide variety of local atomic arrangements, and those atoms rearrange at different rates in different places, both in the liquid and the solid states. Much of this complexity occurs at the nanoscale, including medium-range structural order and spatially heterogeneous dynamics in the supercooled liquid near T_g. Electron nano diffraction is well-suited to probing this key length scale, in the form of fluctuation electron microscopy (FEM) to probe static atomic structure [1] and electron correlation microscopy (ECM) to provide structural rearrangements [2].

We have used ECM to show directly that dynamics in a metallic glass forming liquid are spatially heterogeneous at the nanometer scale [2]. In the bulk liquid as a function of temperature increasing from the glass transition temperature T_g, the structural relaxation time τ varies from >500 seconds to a few seconds at a characteristic length scale ξ that varies from 1.4 to 0.8 nm. $\xi(\tau)$ over this limited temperature range agrees with all of the major microscopic theories of the glass transition.

The liquid surface exhibits dynamics that are consistently an order of magnitude faster than the bulk, extending ~1 nm in from the surface [2]. The surface remains mobile and liquid-like down to 20 K below the bulk T_g, at which point it exhibits its own glass transition. The fast surface dynamics play a controlling role in synthesis of glass thin films by vapor deposition. For substrate temperatures near the bulk T_g, the fast surface can reach equilibrium while the bulk has insufficient mobility to crystallize. We have used this phenomenon to create ultra-stable metallic glass thin films which exhibit stronger icosahedral structural order than bulk glasses with the same composition. Outside of metals, the same phenomena can be used to synthesize glasses with globally anisotropic structure [3].

References:

[1] M. M. J. Treacy, *et al.* Reports Prog. Phys. **68**, 2899 (2005).
[2] P. Zhang, J. J. Maldonis, Z. Liu, J. Schroers, P. M. Voyles, *Nat. Commun.* **9,** 1129 (2018).
[3] S. S. Dalal, et al., Proc. Natl. Acad. Sci. **112**, 4227 (2015).

Biosketch

Paul Voyles is Professor of Materials Science and Engineering and Harvey D. Spangler Professor of Engineering at the University of Wisconsin-Madison. He earned degrees in physics from Oberlin College and the University of Illinois, Urbana-Champaign, then worked as a post-doctoral member of technical staff at Bell Labs in Murray Hill NJ. He joined the UW-Madison in 2002 as an Assistant Professor. His research specialty is the structure of materials, investigated primarily with electron microscopy, supplemented by simulations and data science. He has worked on metallic and other glasses and on materials for microelectronics, spintronics, and superconductors. He was Chair of the Materials Science and Engineering Department from 2018 to 2018 is currently director of the UW-Madison NSF Materials Research Science and Engineering Center. He has published over 190 journal articles, book chapters, and conference proceedings.

Observation of room temperature polar skyrmions

Ramamoorthy Ramesh

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Complex topological configurations are a fertile playground to explore novel emergent phenomena and exotic phases in condensed-matter physics. For example, the recent discovery of polarization vortices and the associated complex-phase coexistence and response under applied field in superlattices of (PbTiO3)n/(SrTiO3)n suggests the presence of a complex, multi-dimensional system capable of exotic physical responses. I will describe the discovery of polar skyrmions in a lead-titanate layer confined by strontium-titanate layers by atomic-resolution scanning transmission electron microscopy (STEM). Phase-field modeling and second-principles calculations reveal that the polar skyrmions have a skyrmion number of +1 and resonant soft X-ray diffraction experiments show circular dichroism confirming chirality. Such nanometer-scale polar skyrmions exhibit a strong signature of negative permittivity at the surface of the skyrmion, which is furthermore highly tunable with an electric field. They are a new state of matter and electric analogs of magnetic skyrmions and may be envisaged for potential applications in information technologies. I will attempt to describe the exciting observations we have made through many collaborations.

Biosketch:

Ramesh is the Purnendu Chatterjee Chair Professor in the Departments of Physics & Materials Science and Engineering at the University of California, Berkeley. He pursues key materials physics and technological problems in complex multifunctional oxides. Using conducting oxides, he solved the 30-year enigma of polarization fatigue in ferroelectrics. He pioneered research into manganites coining the term, Colossal Magnetoresistive (CMR) Oxides. His work on multiferroics demonstrated electric field control of ferromagnetism, a critical step towards ultralow power memory and logic elements.

His extensive publications on the synthesis and materials physics of complex oxides are highly cited (over 65,000 citations, H-factor =110). He is a fellow of APS, AAAS & MRS and an elected member of the U.S. National Academy of Engineering and a Foreign member of the Royal Society of London. His awards include the Humboldt Senior Scientist Prize, the APS Adler Lectureship and McGroddy New Materials Prize, the TMS Bardeen Prize and the IUPAP Magnetism Prize and Neel Medal. He was recognized as a Thomson-Reuters Citation Laureate in Physics for his work on multiferroics.

He served as the Founding Director of the successful Department of Energy SunShot Initiative in the Obama administration, envisioning and coordinating the R&D funding of the U.S. Solar Program, spearheading the reduction in the cost of Solar Energy. He also served as the Deputy Director of Oak Ridge National Laboratory and the Associate Lab Director at LBNL.

Atomic Scale Inelastic Electron Excitation Spectroscopy and Microscopy

Wilson Ho

Department of Physics and Astronomy, University of California, Irvine, CA 92697, USA

The charge and spin of electrons enable spectroscopy and microscopy of electrostatic and magnetic excitations. Different instruments have been invented based on the energy of the probing electrons, ranging from tens and hundreds of keV in the transmission electron microscope to a fraction and few eV in the scanning tunneling microscope. Inelastic electron excitation informs on the intrinsic states of the system and how they are affected by interactions with its environment. Atomic scale spectroscopy and microscopy probe and provide fundamental knowledge that is difficult to obtain by other techniques.

Biosketch:

Wilson Ho received his B.S. and M.S. degrees in chemistry from the California Institute of Technology in 1975, and his Ph.D. in physics from the University of Pennsylvania in 1979. He spent a year at the AT&T Bell Laboratories and was on the faculty at Cornell University prior to joining the University of California, Irvine in 2000 as Donald Bren Professor of Physics & Astronomy and Chemistry. His research has followed the long tradition of developing new instrumentation and methods for precision measurements of atoms and molecules adsorbed on solid surfaces. Over the last 25 years, he has used homemade cryogenic scanning tunneling microscopes (STM) in ultrahigh vacuum to advance the field of single molecule chemistry, physics, and optics. In 1998, his group discovered spatially resolved inelastic electron tunneling (IET) to enable spectroscopy and microscopy with the STM of quantum excitations at the atomic scale, particularly involving vibration and spin states. In addition, IET drove a wide range of single-molecule motions, revealed previously unknown molecular properties, achieved diffraction unlimited spatial resolution in optical phenomena, and led to the development of the inelastic tunneling probe as new quantum sensors. By combining a femtosecond laser with STM, measurement of coherent phenomena was realized with joint spatial and temporal resolutions at the Å-fs limit.

Supramolecular Polymers for Tissue Regeneration

Jing Cheng[1], Kelsey DeFrates[1], Joakim Engstrom[1], Ellen Heber-Katz[4] and Phillip B. Messersmith[1,2,3]

1. Department of Bioengineering, UC-Berkeley, Berkeley, CA, USA.
2. Department of Materials Science and Engineering, UC-Berkeley, Berkeley, CA, USA.
3. Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA, USA.
4. Lankenau Institute for Medical Research, Wynnewood, PA, USA.

Self-assembly of polymers driven by supramolecular interactions like hydrogen bonding, metal coordination, host-guest, pi-stacking and other noncovalent or dynamic covalent interactions are becoming increasingly explored for use in tissue engineering, drug delivery and regenerative medicine. In this talk I will describe our efforts to exploit supramolecular polymer prodrugs to induce tissue regeneration rather than wound repair by scar, as these represent two distinct processes. Amphibians and primitive organisms have the ability to regenerate tissues with complete restoration of tissue architecture and function (i.e. epimorphic regeneration), whereas most mammals including humans generally heal large wounds by scar. An exception to this pattern is found in Murphy Roths Large (MRL) mouse, an inbred strain that is capable of regenerating tissue in a process reminiscent of epimorphic regeneration. This unusual display of mammalian regeneration is enabled by robust expression of the transcription factor hypoxia-inducible factor 1 alpha (HIF-1α). Inspired by MRL mouse regeneration, we are exploring the concept of therapeutic upregulation of HIF-1α to induce regeneration in otherwise nonhealing mammals. We refer to this concept as *drug-induced regeneration*, a concept that diverges from conventional tissue engineering strategies in two important ways: 1) tissue regeneration is enabled by a therapeutic drug rather than through the use of polymer scaffolds, cells and/or growth factors; 2) in most cases we leave the wound untouched, instead administering the drug by SC, IP, IM injection at a peripheral site. To accomplish this, we have designed polymer-drug conjugates (i.e. polymer prodrugs) that self-assemble into supramolecular nanostructures driven by the hydrophobic nature of a conjugated HIF-1α agonist. The relationship between polymer molecular weight, architecture and supramolecular structures were investigated by SAXS and TEM. The supramolecular polymer prodrugs have high drug loading and are shear-thinning, two attractive features of injectable drug delivery systems. Subcutaneous injection of supramolecular polymer prodrug resulted in tissue regeneration in non-regenerative mice, in a manner that emulates the basic elements of amphibian regeneration.

Biosketch:
Phillip B. Messersmith is the Class of 1941 Professor in the Departments of Bioengineering and Materials Science and Engineering at UC-Berkeley. He earned his B.S. degree in life sciences from the University of Illinois at Urbana, M.S degree in bioengineering from Clemson University, and his Ph.D. degree in materials science and engineering from the University of Illinois at Urbana. Previously, Dr. Messersmith was a postdoctoral fellow at Cornell University (1993-1994), and a faculty member at the University of Illinois at Chicago (1994-1997) and Northwestern University (1997-2014). Dr. Messersmith has published over 200 papers and has 43 patents. His awards and honors include a MERIT award from the National Institutes of Health, the Langmuir Lecture Award from the American Chemical Society, and the 2013 Clemson Award for Basic Research from the Society for Biomaterials. Dr. Messersmith is a fellow of the American Institute for Medical and Biological Engineering, the Royal Society of Chemistry, and the International Union of Societies of Biomaterials Science and Engineering. The Messersmith research group is interested in understanding structure-processing-property relationships of materials in biological systems, and in using this information to inform the design and synthesis of biologically inspired synthetic materials for a variety of practical applications.

Supramolecular Polymerization: Its Significance and Applications

Takuzo Aida

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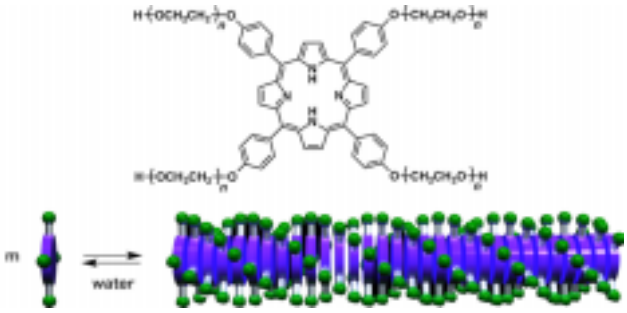

About a century ago, Dr. Hermann Staudinger substantiated the existence of ultralong molecules and won the long term debate against the colloidal theory to establish polymer science. Needless to say, polymer science has made tremendous contributions to the progress of human society, although it coincidentally brought about a critical environmental issue to tackle. In this lecture, I would like to present the significance and applications of supramolecular polymerization, a modernized version of the colloidal approach to polymeric materials. Supramolecular polymers attract attention not only because they are 100% recyclable but also they can be designed to be environmentally friendly, self-healable, responsive, and/or adaptive [1–4]. In 1988, we reported the first prototype of supramolecular polymerization, featuring the formation of a 1D polymeric assembly using an amphiphilic porphyrin with water-soluble oligoether side chains as the monomer and have made fundamental contributions to this field. Representative examples include (1) nanotubular supramolecular polymerization, (2) chain-growth supramolecular polymerization, (3) supramolecular block copolymerization, (4) stereoselective supramolecular polymerization, and (5) thermally bisignate supramolecular polymerization. These contributions are integral elements of conventional polymer science, filling in the critical gap between supramolecular and conventional polymerizations. Furthermore, we have expanded the basic concept of supramolecular polymerization into the noncovalent design of innovative soft materials. Successful examples include the developments of (i) bucky gels, (ii) aquamaterials, (iii) mechanically robust self-healable materials, (iv) supramolecular polymers of biomolecular machines, (v) ferroelectric columnar liquid crystals, and (vi) reorganizable and adaptive core-shell columnar liquid crystals. I will highlight some of these examples to show the significance of supramolecular polymerization for the realization of sustainable society.

References

[1] Aida, Meijer, Stupp, *Science* **2012**, *335*, 813–817. [2] Aida, Meijer, *Israel J. Chem.* **2020**, *60*, 1–16. [3] Aida, *Adv. Matter.* Essay, **2020**, 1908140. [4] Hashim, Bergueiro, Meijer, Aida, *Prog. Polym. Sci.* **2020**, 101250.

Biosketch:

BS: Faculty of Engineering, Yokohama National University (1979)
MS: School of Engineering, The University of Tokyo (1981)
PhD: School of Engineering, The University of Tokyo (1984)

1984–1989: Assistant Professor, The University of Tokyo
1989–1991: Lecturer, The University of Tokyo
1991–1996: Associate Professor, The University of Tokyo
1996–Now: Professor, The University of Tokyo

1996–1999: Researcher, Japan Science & Technology Agency, PRESTO Project
2000–2005: Director, Japan Science & Technology Agency, ERATO Nanospace Project
2005–2010: Director, Japan Science & Technology Agency, EARTO–SORST Project on Electronic Nanospace
2008–2012: Director, RIKEN Advanced Science Institute
2013–2013 Deputy Director, Riken Center for Emergent Matter Science
2004–2006: Associate Editor, *Journal of Materials Chemistry* (RSC)
2009– Board of Reviewing Editors, *Science Magazine* (AAAS) 2014–

Recent Awards: American Chemical Society Award in Polymer Chemistry (**2009**) / Chemical Society of Japan Award (**2009**) / Purple Ribbon (**2010**) / Alexander von Humboldt Research Award (**2011**) / Fujiwara Prize (**2011**) / Leo Esaki Prize (**2015**) / Dean Award, U. Tokyo (**2016**), Chirality Medal (**2017**), Japan Academy Prize (**2018**), The Ichimura Prize in Science for Excellent Achievement (**2020**), Ryoji Noyori ACES Award (**2020**), Member of the Royal Netherlands Academy of Arts and Science (**2020**).

Selected Recent Publications:

(1) Nematic-to-Columnar Mesophase Transition by in situ Supramolecular Polymerization, *Science* **2019**, *363*, 161–165.
(2) Self-Assembly of Lattices with High Structural Complexity from a Geometrically Simple Molecule, *Science* **2018**, *361*, 1242–1246.
(3) Mechanically Robust, Readily Reparable Polymers via Tailored Noncovalent Crosslinking, *Science* **2018**, *359*, 72–76.
(4) Thermally Bisignate Supramolecular Polymerization, *Nature Chem.* **2017**, *9*, 1133–1139.
(5) An Autonomous Actuator Driven by Fluctuations in Ambient Humidity, *Nature Mat*. **2016**, *14*, 1084–1089.
(6) Sub-Nanoscale Hydrophobic Modulation of Salt Bridges in Aqueous Media, *Science* **2015**, *348*, 555–559.
(7) Selective-Assemblies of Giant Tetrahedra via Precisely Controlled Positional Interactions, *Science* **2015**, *348*, 424–428.
(8) A Rational Strategy for the Realization of 'Chain-Growth' Supramolecular Polymerization, *Science* **2015**, *347*, 646–651.
(9) Ultrahigh-throughput Exfoliation of Graphite into Pristine 'Single-Layer' Graphene Using Microwaves and Molecularly Engineered Ionic Liquids, *Nature Chem.* **2015**, *7*, 730–736.
(10) Anisotropic Hydrogel with Embedded Electrostatic Repulsion among Cofacially Oriented 2D Electrolytes, *Nature* **2015**, *517*, 68–72.

Developments in High Speed Structural Imaging of Low Dimensional Materials

A I Kirkland[1,2,3]

[1]Department of Materials, Parks Road, Oxford, OX13PH, UK.
[2] Electron Physical Sciences Imaging Centre (ePSIC), Diamond Light Source, Oxford, UK.
[3]The Rosalind Franklin Institute, Harwell Campus, Didcot, OX11 0FA, UK

I will describe recent work using high speed direct electron detectors and artificial intelligence / machine learning to automatically map defect transitions in graphene. I will also discuss the use of similar detectors in electron ptychography, in particular under extremely low dose conditions using binary counting.

The development of high speed direct electron detectors suitable for use at intermediate electron energies has led to significant progress in imaging, diffraction and spectroscopy[1].

To usefully deploy Graphene and related materials in electronic applications[2-4] it is essential to understand the behavior of defects, which have been the subject of extensive research in silicon devices for decades. Detector advances make it possible to image these defects at primary energies below those that cause significant specimen damage whilst retaining sufficient spatial resolution to resolve local atomic configurations around the defect site[5].

However, the extremely large datasets (typically 10^6 images or greater) that can be acquired makes conventional manual image processing intractable. I will describe how this can be overcome using a deep learning neural network[6] for atomic model abstraction from low dose high framerate graphene images (Fig. 1). Using this approach, it is possible to identify many instances of defect transitions and to map the lifetimes of defect states. In turn these can be used as input to density functional theory to model the potential energy landscape for the transitions.

Finally, I will highlight the use of fast detectors for electron Ptychography at low dose. At low dose the sampling of the diffraction pattern in the far field is sparse and a counting direct electron detector can be operated in a binary mode to provide an effective speed increase.

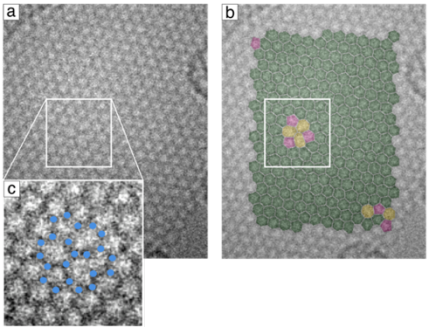

Figure 1. (a) Low-dose image of a graphene sheet recorded with a 1ms exposure at 80KV containing defects; (b) Automatically generated annotation with 5-membered rings (pink), 6-membered rings (green), and 7-membered rings (yellow) overlaid on top of the experimental image; (c) enlarged defect area with carbon atom positions marked by blue circles.

References

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5. Robertson, A.W. *et al.* (2012) Nature Comm. 3, 1144.
6. LeCun, Y. *et. Al*. (2015) Nature 521, 436

Biosketch:

Professor Angus Kirkland was awarded his MA and PhD from the University of Cambridge and has held the posts of Professor of Materials at Oxford since 2005 and JEOL Professor of Electron Microscopy since 2013. In 2016 he was appointed as Director of the National Physical Sciences Imaging Centre at Diamond Lightsource and is also a Science Director at the recently established Rosalind Franklin Institute.

He was awarded the MSA prize in 2005, the Rose prize in 2015, the Quadrennial prize of the European Microscopy Society in 2016 and the Agar Medal for Electron Microscopy in 2017.

He served as General Secretary of the International Federation of Societies for Microscopy in from 2014 -2018 and was elected President in 2018.
He has also served as Editor in Chief of Ultramicroscopy since 2010

Local Structure Phenomena in Oxide Dielectrics

Elizabeth C. Dickey[1], Stephen Funni[1], Nikki Creange[1], Abinash Kumar[2], James M. LeBeau[2]

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The quantification of local symmetry and structure is important to the fundamental understanding of dielectric and ferroic properties of many complex oxides. ABO_3 perovskite and related materials exhibit local structure that is lower in symmetry than the global structure, which is sometimes referred to as *pseudosymmetry*. Local symmetry variants that persist from the ground state into the higher-temperature phases are important attributes of order-disorder phase transformations. Furthermore, local symmetry breaking can be induced by compositional inhomogeneity, where the local bonding environment is spatially inhomogeneous. While numerous scattering and spectroscopy techniques can be used to quantify local structure, modern advancements in aberration-corrected scanning transmission electron microscopy are enabling new insights into local structure phenomena and improving our holistic understanding of the relationship between short- and long-range order in complex oxides. Herein we apply these techniques to study local structure in several functional oxides having varying degrees of lattice disorder.

Biosketch

Elizabeth Dickey is a Distinguished Professor and Associate Head of the Department of Materials Science and Engineering at North Carolina State University. A primary focus of her research is in developing processing-structure-property relationships for ceramic and electronic materials. She has over 150 peer-reviewed journal publications in these areas, which have been cited over twenty-thousand times. Professor Dickey is a fellow and president-elect of the American Ceramic Society, and she is a fellow of the Microscopy Society of America. Professor Dickey serves as an Editor of the Journal of the American Ceramic Society and Editor-In-Chief of Cambridge University Press' Elements in Microscopy and Microanalysis. In January 2021 Professor Dickey will move to Carnegie Mellon University where she will become the head of the Department of Materials Science and Engineering

Opportunities for understanding crystal growth through in situ electron microscopy

Frances M. Ross

Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts, USA

Building functional nanostructures with atomic level precision requires a detailed understanding of the chemistry and physics of crystal growth at the nanoscale. *In situ* experiments in the transmission electron microscope can help by providing unique measurements of individual nanostructures while they grow. I will show how movies recorded in the microscope help to explore the formation mechanisms of nanomaterials, focusing in particular on nanoscale islands of metals and semiconductors grown on two-dimensional materials such as graphene and MoS_2. The difference in the nature of the bonding at the 3D nanocrystal / 2D material interface, compared to the situation for conventional epitaxy, is expected to generate interesting and potentially useful new growth modes and interfacial structures. I will discuss how *in situ* and post-growth measurements using different microscopy techniques can combine to provide insights into the formation and control of these interfacial structures. The rapid pace of advances in electron microscopy instrumentation promise exciting future opportunities as well as new challenges in understanding materials growth and reactions using *in situ* techniques.

Biosketch

Frances M. Ross received her B.A. in Physics and Ph.D. in Materials Science from Cambridge University, UK. Her postdoc was at A.T.&T. Bell Laboratories, using *in situ* electron microscopy to visualize silicon oxidation and dislocation dynamics, after which she joined the National Center for Electron Microscopy, Lawrence Berkeley National Laboratory, investigating other processes *in situ* including anodic etching of Si. She then moved to the IBM T. J. Watson Research Center where she imaged the growth of nanoscale materials using a microscope with deposition and focused ion beam capabilities, developed liquid cell microscopy for visualizing electrochemical processes, and measured growth and transport properties in a combined focused ion beam-scanning tunneling microscope system. She joined the MIT Department of Materials Science and Engineering in 2018 where her research continues to center on nanostructure self-assembly, liquid cell microscopy, epitaxy and electrochemical processes.

Monochromated EELS of organic functional materials in the STEM

Amanda Trout[1,2], Brittany Ford[1,2], Nuria Bagués[1,2], Robert Colby[3], Robert E.A. Williams[1] and David W. McComb[1,2]

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The ability to probe in the scanning transmission electron microscopes (STEM) chemistry, bonding and electronic properties of materials using monochromated electron energy-loss spectroscopy (EELS) is providing important insights in inorganic functional and structural materials. However, applying these methods to materials that are entirely or predominantly made up of low atomic number elements presents some significant challenges. In particular, in the study of organic materials electron beam damage is always a major consideration. In organic-inorganic composites, or interfaces, in addition to differential damage rates, the challenges of samples preparation may cause significant thickness variations. Materials comprised of low atomic number elements can be more susceptible to electronic charging effects in some cases, and in many cases these materials are sensitive to the environment requiring specialized holders or transfer systems.

When studying such materials, it is essential to understand the "electron budget" for each experiment, *i.e.* the electron dose that will result in changes in the property being measured. In some cases, the budget makes use of some methods prohibitive due to current instrumentation limitations, while in others, more indirect signals of chemical bonding must be explored. In this contribution, I will review and discuss the results of recent investigations into several different types of low Z material systems using mono-STEM-EELS including organic photovoltaics, molecular magnets, 2D materials and polymers. I will show how mono-EELS is providing new insights into structure-property relationships. I will review the impact of recent developments in hardware and software on such studies, in particular for the study ultra-sensitive organic systems, and the implications for spatial resolved mapping of organic functional groups in polymers, composites and biomaterials applications.

Biosketch
David William McComb is an Ohio Research Scholar and Professor of Materials Science and Engineering at The Ohio State University. A chemistry graduate from the University of Glasgow, David did his PhD in Physics at the University of Cambridge. David is an expert in the development and application of STEM and EELS as a sub-nanometre scale probe of chemistry, structure and bonding. He has extensive experience in the application of EELS to the study of problems in solid-state chemistry and materials science. He is a fellow of the Microscopy Society of America, the Royal Society of Chemistry and the Institute of Materials, and former co-director of the London Centre for Nanotechnology as well as Director of Research and Deputy Head of the Department of Materials at Imperial College London. In October 2011 he joined The Ohio State University as the founding director of the Center for Electron Microscopy and Analysis (CEMAS). This state-of-the-art facility houses over $37M worth of electron and ion microscopes performing at or beyond manufacturer specifications.

Revealing the Dynamics of Electrocatalysts under Reaction Conditions using Liquid Phase Electron Microscopy

See Wee Chee[1], Thomas Lunkenbein[2] and Beatriz Roldan Cuenya[1]

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Catalysts play key roles in various energy conversion technologies that needed for sustainable development of our modern society by facilitating the renewable energy-powered generation of chemicals and fuels. However, the rational design of efficient catalysts has been hampered by our lack of insight into the structures that exist during reaction and their associated properties. If we are able to capture the behavior of nanoparticles during catalysis, what we will likely find is a dynamic and inhomogeneous system where the catalysts evolve differently, in different places, and at different times. However, most studies compare catalyst morphology before and after reaction, leaving us blind to these complex dynamics and their impact on catalytic properties.

In situ/operando electron microscopy allows us to visualize the dynamical structures that exist during reaction with its high spatial resolution, relatively high temporal resolution, and provide complementary capabilities for chemical analysis. Here, I will first present an overview of the research efforts at the Fritz Haber Institute looking at the behavior of catalysts under gas and liquid environments that are also realistic for thermal and electro-catalytic reactions. It will be followed by a more in-depth discussion about the research in my research group investigating electrocatalysts for the electrochemical reduction of CO_2 using liquid phase transmission electron microscopy and scanning electron microscopy. Lastly, I will discuss the technical challenges that still prevent us from extrapolating these insights and achieving structure-property correlation in catalytic materials.

Biosketch

Dr. See Wee Chee is currently a Group Leader in the Department of Interface Science at the Fritz Haber Institute of the Max Planck Society. He obtained his PhD in Materials Science and Engineering from the University of Urbana-Champaign and did his postdoctoral training at Arizona State University and Rensselaer Polytechnic Institute. His research group focuses on using liquid phase electron microscopy to interrogate the structure and composition of nanoscale electrocatalysts under reaction conditions and derive the critical structure-property relationships that control their performance.

Liquid-Phase TEM Imaging of Colloidal Crystallization and Protein Transformation

Qian Chen

Department of Materials Science and Engineering, University of Illinois, Urbana, Illinois 61801, USA

I will discuss my group's recent progress on applying low-dose liquid-phase TEM to synthetic and biological colloidal systems. In the first system, we directly image the otherwise elusive crystallization pathways of nanosized colloids into superlattices, where the discreteness and multi-scale coupling effects complicate the free energy landscape and the application forms of the final superlattices. We find that there exist similarities to the prevalent model system of micron-sized colloids, such as a non-classical two-step crystallization pathway, and an agreement with the capillary wave theory. But there are also differences, in particular, a universal layer-by-layer growth mode that we observe consistently for diverse nanoparticle shapes. Single particle tracking, trajectory analysis, and simulations combined unravel the energetic and kinetic features rendering this crystal growth mode possible and universal at the unexplored nanoscale, enabling advanced crystal engineering. In the second system, we sandwich and capture moving membrane proteins in their native lipid and liquid environment at the nm resolution. The proteins exhibit real-time "fingering" fluctuations, which we attribute to dynamic rearrangement of lipid molecules wrapping the proteins. The conformational coordinates of protein transformation obtained from the real-space movies are used as inputs in our molecular dynamics simulations, to verify the driving force underpinning the function-relevant fluctuation dynamics. This platform invites an emergent theme of structural biophysics as we foresee.

Biosketch

Qian Chen has been an assistant professor in the Department of Materials Science and Engineering at the University of Illinois at Urbana-Champaign since 2015. She received her PhD degree from the same department in 2012. Her research focuses on electron microscopy-based imaging, understanding, and engineering of soft materials, such as nanoparticle and colloidal self-assembly, protein transformation, battery materials, and energy-efficient filtration. Her awards include the American Chemistry Society' (ACS) Victor K. LaMer Award in 2015, the Air Force Office of Scientific Research Young Investigator Program (AFOSR YIP) Award in 2017, the National Science Foundation CAREER Award in 2018, an Alfred P. Sloan Research Fellowship in 2018, and the ACS Unilever Award (2018). She also was recognized on the Forbes 30 Under 30 Science List in 2016

A close look at material synthesis with liquid phase and cryo electron microscopy

Joe Patterson

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Chemistry, and its manifestation as self-assembly, provides an elegant strategy to create functional, highly complex, and hybrid materials for a myriad of applications. Through evolution, living systems, have achieved exquisite control over the deposition of both organic and inorganic building blocks to create hierarchical, composite materials with exceptional properties. This is achieved under ambient conditions by utilizing compartmentalization and confinement of chemical environments to control the pathway of formation, realizing structures and shapes that are not readily achievable in synthetic systems. If materials chemists are ever able to have this level of control, it will come from a deep understanding of general mechanisms and pathways that govern the self-assembly of hierarchical and hybrid structures in complex solution environments.[1] Considering that materials synthesis in liquids, and deposition from liquids, pervades the vast majority of polymer science and soft matter research, imaging techniques that provide direct observations of structure and chemistry in solution with nanoscale resolution should be the leading analytical tools for driving self-assembly theory and experimental design. Techniques such as Liquid Phase Electron Microscopy (LPEM),[2, 3] and Cryogenic Electron Microscopy (CryoEM),[1, 4, 5] provided an unprecedented insight into nanoscale reaction mechanisms, however their application is still extremely challenging. The primary challenge for the field of LP-EM is understanding the role that the electron beam plays in the observed mechanism. CryoEM is limited in its ability to resolve complex heterogeneous processes due to the inherent "freezing" of the samples which prevents knowledge of the future or history of any particle under inspection. Consequently, although LPEM and cryoEM enables us to take a close look at materials synthesis we are often left with the question, what does this mean? Our ability to overcome these challenges will be key in translating the insights from LPEM and cryoEM into new theories about the chemistry and self-assembly of materials and will ultimately dictate the future of these techniques for driving innovation in science and engineering.

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Biosketch

Joe Patterson is an assistant professor in the Chemistry Department at the University of California Irvine. He completed his PhD in polymer chemistry and self-assembly at the University of Warwick, working under the supervision of Professor Rachel O'Reilly. He performed postdoctoral research at the University of California San Diego with Professor's Nathan Gianneschi and Kimberly Prather. In 2016 he joined the Eindhoven University of Technology, working in the Laboratory of Materials and Interface Chemistry with Professor Nico Sommerdijk. His interests involve the development of new materials through a deep understanding of their structural dynamics. To address challenges in this area he develops liquid phase and cryogenic electron microscopy for the visualization, quantification and discovery of dynamic nanoscale processes in solution.

The cryoEM revolution: from proteins to genomes, & from in situ to in action

Hong Zhou

California NanoSystems Institute (CNSI) and Department of Microbiology, Immunology and Molecular Genetics, University of California, Los Angeles (UCLA), USA

The technique of cryo electron microscopy (cryoEM) uses electrons to image biological samples that are flash-frozen to preserve their native, water-containing functional states of life processes. Recent advances in cryoEM have revolutionized protein structure determination in the field of structural biology (1-4), sidelining traditional technologies such as x-ray crystallography and nuclear magnetic resonance (NMR). More significantly, direct electron-counting cryoEM combined with advanced computational processing now offers new opportunities to determine structures of genomes and genome replication/transcription in action. Towards this end, viruses are the subject of choice as they are the simplest replicating machines that package their DNA or RNA genomes with a minimal set of proteins to sustain survival and spread. In this talk, I will present the latest breakthroughs in the cryoEM field and how these technology advances have enabled visualization of in situ genome structures in both quiescent, primed and active transcribing states in both RNA and DNA viruses, leading to novel insights into the mechanism of genome compaction and transcription, the fundamental processes of life (5,6).

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Biosketch

Z. Hong Zhou is a Professor of Microbiology, Immunology and Molecular Genetics and the Director of the Electron Imaging Center for Nanomachines at University of California, Los Angeles (UCLA). He received his early education in physics at the University of Science and Technology of China before earning his PhD (in 1995) in biochemistry at the Baylor College of Medicine, in Houston, Texas, USA, under the supervision of cryo electron microscopy (cryoEM) pioneer Wah Chiu. From 1995-1999, he carried out his postdoctoral training in computer science and applied mathematics under Prof. L. Ridgway Scott and developed high-performance image processing software for cryoEM reconstruction.
Zhou has published over 230 research articles and book chapters, among then over 20 in premier journals Nature, Science and Cell. He uses cryoEM to determine three-dimensional structures of molecular complexes near their native functional states at molecular to atomic resolutions. In 2008, his group was the first to demonstrate near atomic resolution cryoEM by single-particle analysis in a milestone Nature paper documenting the first atomic model of the cytoplasmic polyhedrosis virus. Broadly, his research addresses fundamental questions, such as how proteins and nucleic acids interact to store and release energy, to transduce signals, and to perform tasks of chemistry underlying reproduction.

Zhou was a Pew Scholar in Biological Sciences and a Basil O'Connor Scholar of the March of Dimes Foundation. He is a recipient of a Burton Award and K. H. Kuo Distinguished Scientist Award.

High-resolution structure determination by Microcrystal Electron Diffraction (MicroED)

Brent L. Nannenga

Chemical Engineering, School for Engineering of Matter, Transport and Energy, Arizona State University, Tempe, AZ 85287 USA
Center for Applied Structural Discovery, The Biodesign Institute, Arizona State University, Tempe, AZ, 85287 USA

A common barrier to high-resolution structure determination is the growth of large well-ordered crystals. Electron diffraction is capable of producing diffraction data from crystals that are orders of magnitude smaller than those needed for conventional X-ray crystallographic experiments. In this presentation, the technique of microcrystal electron diffraction, or MicroED, will be described, which allows the collection of high-resolution diffraction data from extremely small nano and microcrystals. MicroED methods will be described along with representative applications where MicroED was used for structure determination. Additionally, current work in our lab, which is focused on improving MicroED methodology and extending this technique to new samples will be presented.

Biosketch

Brent Nannenga is currently an Assistant Professor of Chemical Engineering in the School for Engineering Matter, Transport and Energy at Arizona State University and a member of The Biodesign Institute's Center for Applied Structural Discovery. He received his Ph.D. in Chemical Engineering from the University of Washington in 2011 under the supervision of François Baneyx. Following his graduate studies, he conducted postdoctoral research at Janelia Research Campus (Howard Hughes Medical Institute) in the lab of Tamir Gonen, where he focused on the development and application of cryo-electron microscopy (cryo-EM), specifically the development of microcrystal electron diffraction (MicroED).

Since joining Arizona State University in 2015, his research has focused on developing and using methods for high-resolution structure determination, and employing structural insights gained from these methods in order to engineer biomolecules and materials with novel and unique properties. His work has been recognized by awards including an NSF CAREER Award, Air Force Office of Scientific Research (AFOSR) Young Investigator Award, and the Burton Medal from the Microscopy Society of America.

Progress in Cryogenic STEM for Quantum and Energy Materials

Lena F. Kourkoutis[1,2], Ismail El Baggari[3], Berit H. Goodge[1], Elisabeth F. Bianco[2], Noah Schnitzer[4], Michael J. Zachman[1], David J. Baek[5]

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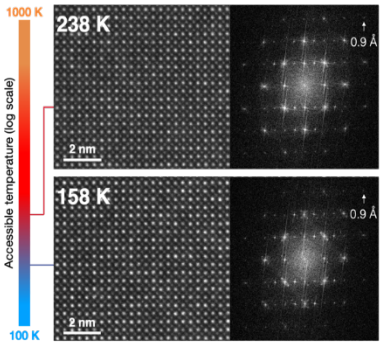
Electron microscopy has enabled imaging of our natural word with exceptional detail. Today, the three-dimensional structure of biomolecules can be studied down to the atomic scale and single defects present in atomically thin materials can not only be identified but also spectroscopically probed. The developments that have enabled these successes in the life sciences and the physical sciences have been recognized by the 2017 Nobel Prize in Chemistry for cryogenic electron microscopy of biomolecules and the 2020 Kavli Prize in Nanoscience for sub-Ångstrom imaging enabled by aberration correction.



Despite these breakthroughs in imaging there are entire classes of materials and devices that have not been able to be explored at the relevant microscopic length scales [1-2]. In this talk, we will discuss recent advances in cryogenic scanning transmission electron microscopy (cryo-STEM) for the physical sciences [3-7] which have opened a new window to probing phenomena in quantum [4, 7, 8] and energy materials [9-11] that have not been accessible before.

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Biosketch

Lena F. Kourkoutis is an Associate Professor of Applied and Engineering Physics at Cornell University and the co-Director of the Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials (PARADIM). Dr. Kourkoutis received her undergraduate degree in Physics from the University of Rostock, Germany in 2003, and completed her Ph.D. at Cornell in 2009. As a Humboldt Research Fellow, she spent 2011-2012 exploring cryo-electron microscopy in the Molecular Structural Biology Group at the Max Planck Institute of Biochemistry in Martinsried, Germany. She returned to Cornell as a Postdoctoral Associate in 2012 and joined the Applied and Engineering Physics Faculty in 2013. The Kourkoutis electron microscopy group focuses on understanding and controlling nanostructured materials, from biomaterials to materials for energy to quantum materials. They have developed new cryogenic scanning transmission electron microscopy techniques to gain access to low temperature electronic states, to study processes at liquid/solid interfaces in energy devices and to image thick biological specimens. Prof. Kourkoutis is recipient of a Packard Fellowship for Science and Engineering, a Presidential Early Career Awards for Scientists and Engineers (PECASE), an NSF CAREER award, the Burton Metal awarded by the Microscopy Society of America, the Kurt Heinrich Award from the Microanalysis Society, and she is a Kavli Fellow of the National Academy of Sciences.

Limiting Damage and Capturing Dynamics of Soft Matter and Beam Sensitive Materials

John Watt[1], Katherine L. Jungjohann[2]

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[2.] Center for Integrated Nanotechnologies, Sandia National Laboratories, Albuquerque, USA.

Soft matter and its organic-inorganic interfaces have historically been unlikely candidates for investigation by electron microscopy techniques due to damage by the electron beam, as well as inherent instability under a high vacuum environment. The recent development of cryogenic transmission electron microscopy (cryo-TEM) and low-dose, high-speed direct detection cameras provides the soft materials science community with an exciting opportunity to probe the structure of these materials in real space. Cryo-TEM reduces beam damage and allows for the characterization of materials in a native, frozen-hydrated (or solvated) state, providing a direct visual representation of soft structure. Direct detection can be used to capture dynamic transformations of nanomaterials at high speed, capturing crucial intermediate structures. Additionally, the two techniques can be paired to provide optimal imaging conditions for beam-sensitive materials; including low-Z metals, metal-organic frameworks, and sensitive hydrated interfaces. In this talk I will present some of the recent research performed at the Center for Integrated Nanotechnologies applying low dose and high speed imaging techniques in the imaging of sensitive materials.

References

This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Los Alamos National Laboratory, an affirmative action equal opportunity employer, is managed by Triad National Security, LLC for the U.S. Department of Energy's NNSA, under contract 89233218CNA000001.

Biosketch

Dr. John Watt is a Scientist and electron microscopist at the Center for Integrated Nanotechnologies (CINT) at Los Alamos National Laboratory. He received his PhD from Victoria University of Wellington, New Zealand and held postdoctoral positions at both VUW and CINT. His research interests include the synthesis and characterization of soft matter, inorganic materials, and their unique interfaces.

The Surface Dynamics of the Initial Stages of Cu Oxidation

Judith C. Yang[1,2], Meng Li[1], Matthew T. Curnan[1], Richard Burke Garza[3], Michael A. Gresh-Sill[1], Stephen D. House[1,2], Wissam A. Saidi[1,4]

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Fundamental understanding of the oxidation of metals and alloys is critical to corrosion protection, catalyst design, and nano manufacturing. Much is known about oxygen interaction with metal surfaces and about the macroscopic growth of thermodynamically stable oxides. However, the transient stages of oxidation - from nucleation of the metal oxide to formation of the thermodynamically stable oxide - represent a scientifically challenging and technologically important terra incognito. Classical oxidation theories, such as Cabrera-Mott and Wagner, make the simplifying assumption of uniform oxide scale growth as previous instruments were not capable of visualizing microstructural changes during oxidation at the nanoscale and below. With the development of in situ transmission electron microscopy (TEM), we are now able to visualize the relevant oxidation processes at the atomic scale. We have demonstrated via in situ TEM that the formation of epitaxial Cu_2O islands during the transient oxidation of Cu films bear a striking resemblance to heteroepitaxy, where the initial stages of growth are dominated by oxygen surface diffusion and strain impacts the evolution of the oxide morphologies. We are presently using correlated in situ environmental high-resolution TEM (ETEM) and atomistic simulations (Figure 1) to gain deeper fundamental understanding.

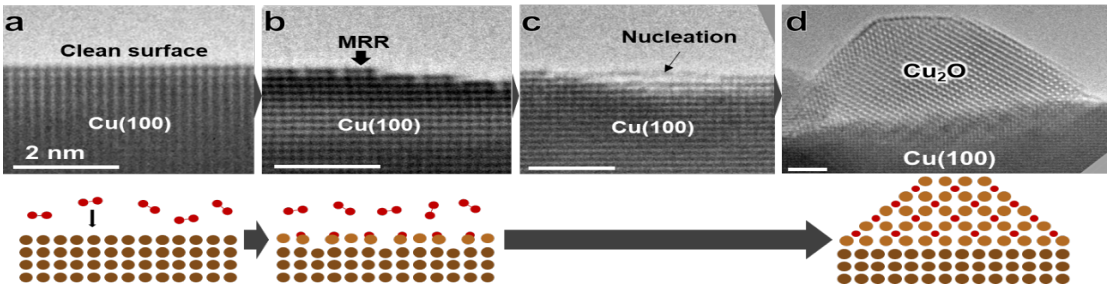

Figure 1. In situ ETEM observation of oxidation on pure Cu(100) surfaces at 350 C under O_2. (a) Clean Cu(100). (b) Missing Row Cu-O Surface Reconstruction (MRR) under 0.1 Pa O_2. (c) Cu_2O nucleation and (d) Cu_2O island growth under 0.3 Pa O_2. Scale bar = 2 nm.

Biosketch

William Kepler Whiteford Professor Judith C Yang received her PhD in physics from Cornell University in 1993. She was a post-doc at the Max-Planck-Institute of Metallforschung, Stuttgart, Germany and then a post-doc and visiting lecturer at U. Illinois at Urbana-Champaign. In 1999, she joined the engineering faculty at U. Pittsburgh. She is an American Physical Society fellow (2017) and Microscopy Society of America fellow (2018).

Probing surface-plasmon-induced reactions using a multimodal approach for environmental scanning transmission electron microscopy

Wei-Chang David Yang[1], Canhui Wang[1,2], David M. Raciti[3], Amit Agrawal[1], Henri J. Lezec[1] and Renu Sharma[1]

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Optically-excited localized surface plasmon (LSP) resonances of metallic nanostructures (Au, Ag, and Al) were found to induce H_2 or O_2 dissociation at reduced temperatures, mimicking photocatalysis. Energy harnessed by plasmonic nanostructures is theorized to overcome reaction barriers by compensating for the thermal energy required otherwise. Resolving the reaction mechanism at the sub-nanoparticle scale, including (1) reactant adsorption, (2) LSP resonance mode coupling, and (3) product desorption, could elucidate the design rule for plasmonic catalysts. However, such sub-nanoparticle information has not been obtained under reactive environments by variants of optical spectroscopy due to the diffraction-limited spatial resolution. In this talk, we show that the reaction details, gained from in-situ electron energy-loss spectroscopy (EELS), cathodoluminescence (CL), and gas chromatography-mass spectrometry (GC-MS) connected to a gas-cell holder, provides insight into the reaction mechanism of plasmonic catalysis.

Electron-beam-excited LSP resonances on Au and Al nanoparticles are exploited to drive chemical reactions in an environmental scanning transmission electron microscope (ESTEM) equipped with a monochromated electron gun. Coupling efficiency maps of LSP resonances on Au and Al nanoparticles, obtained by applying the non-negative matrix factorization (NMF) to EELS hyperspectral images, illustrate the optimized electron beam position to excite specific resonance modes of which resonance energies are theoretically matching the selected reaction barriers. For triangular Au nanoprisms on TiO_2 support in CO environment, gas adsorption maps are attained for the first time using in-situ core-loss EELS, indicating that CO is adsorbed on the selective Au edges but not on the entire surfaces. We track the deposition of carbon, through the room-temperature Boudouard reaction: $2CO_{(g)} \square CO_{2(g)} + C_{(s)}$, driven by the near-field enhancement of the dipole mode on Au nanoprisms. Based on the location of carbon on Au edges, we find the reaction active sites is where preferential adsorption sites and locations of strong plasmonic field superimpose. Energy transfer through nonradiative plasmon decay during the reaction is also identified using correlative CL and EELS. We further show that the endothermic reverse reaction can be driven in CO_2 environment by the ultraviolet (UV) surface plasmons on Al nanoparticles instead of the visible-light surface plasmons on Au, resulting in carbon etching and CO formation. We also demonstrate that the small amount of CO from the local reverse reaction is measurable using GC-MS coupled to a gas-cell holder and increases in proportion to the volume of etched carbon. Our multimodal approach provides unprecedented information to explicate plasmonic catalysis at the nanometer scale and shed light on the design principles for new hybrid plasmonic catalysts that enable low-temperature chemical processes

Biosketch

Wei-Chang David Yang is a materials research engineer in the Physical Measurement Laboratory at the National Institute of Standards and Technology (NIST), leading the environmental transmission electron microscopy project. He received his bachelor's and master's degrees from the National Tsing Hua University, Taiwan, and his Ph.D. in Materials Engineering from Purdue University. Prior to his current appointment, he conducted his postdoctoral research, advised by Renu Sharma, developing in situ and operando methods for TEM to measure dynamic processes at NIST. His research presently focuses on the discovery of nanoscale light-matter interactions in materials correlated to their structures and processing conditions that potentially have an impact on photovoltaics, photocatalysis, and nanoelectronics. He was awarded the Early Career Award from the Nanometer-scale Science and Technology Division of the American Vacuum Society in 2019 for his work that elucidates the key structural features in plasmonic nanoparticles resulting in photochemistry that induces chemical reactions without the need of additional heat. He was also awarded the Microscopy Society of America's Postdoctoral Scholar Award in 2019

Atomic Engineering of 2D Materials: Insights from In situ STEM Experiments, Theory and Functional Properties

Raymond R. Unocic[1], Xiahan Sang[2], Xufan Li[3], Wen Zhao[4], Jicheng Dong[4], Guoxiang Hu[1], Victor Fung[1], Panchapakesan Ganesh[1], Christopher M. Rouleau[1], David B. Geohegan[1], Feng Ding[4,5], Kai Xiao[1]

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Designing new materials for functional applications (i.e. nanoelectronics, catalysis, membranes, energy storage and conversion, etc.) depends upon our ability to understand and correlate a materials structure and chemistry to functional material properties. This is even more important for two-dimensional (2D) materials where thicknesses are on the order of single atoms to a few-atomic layers; therefore, any structural or chemical modification at these length scales can have a profound effect on modifying the electronic, magnetic, optical, and catalytic properties Therefore, it is important to understand the mechanisms by which the atomic structure can be manipulated in order to tune specific functional properties. In this talk, we use *in situ* scanning transmission electron microscopy (STEM), to directly observe and manipulate the edge structure of 2D W-doped $MoSe_2$ as a function of temperature and controlled electron beam irradiation. From time-resolved, atomic resolution HAADF STEM images we track the formation of a variety of edge configurations that structurally transform into theoretically predicted atomic configurations by thermal and chemical driving forces. *Ab initio* molecular dynamics (AIMD) simulation confirms that the observed atomic structure evolution is attributed to changes in the Mo chemical potential during *in situ* heating. As the local Mo chemical potential increases, the edge structure evolves from Se-terminated zigzag edges to nanowire-terminated zigzag edges [1]. As determined by density functional theory (DFT), different edge structures exhibit very different electronic properties. While Se-terminated edges are generally semiconducting, nanowire (NW) -terminated edges are conductive and can also be ferromagnetic due to the specific atomic rearrangement of NW-terminated zigzag Se edges. Moreover, the non-stoichiometric engineered edges can be catalytically active for the hydrogen evolution reaction [2] and a variety of edge configurations have been predicted from theory [3]. The work presented here helps pave the way to engineer the atomic edge structure of 2D materials for targeted functional applications [4].

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Biosketch

Raymond Unocic is a Senior R&D Staff Scientist, Microanalysis of Materials Group Leader and Directed Nanoscale Transformation theme science leader at the Center for Nanophase Materials Sciences at Oak Ridge National Laboratory (ORNL). He received his Ph.D. in Materials Science and Engineering from The Ohio State University in 2008. He started his career at ORNL under the Alvin M. Weinberg early career distinguished fellowship program from 2009-2011 then transitioned to R&D Staff Scientist. His research focuses on the development and application of novel *in situ* and *operando* electron microscopy techniques to probe the functionality of nanomaterials used in energy storage, catalysis and electronic applications. He holds an adjunct professor position in the School of Materials Science and Engineering at Georgia Institute of Technology and is a joint faculty member in the Bredesen Center for Interdisciplinary Research and Graduate Education at the University of Tennessee, Knoxville and ORNL. He is active in professional societies such as the Microscopy Society of America (MSA), Materials Research Society (MRS), the Electrochemical Society (ECS) and was the past leader for the focused interest group on *in situ* electron microscopy in liquids and gases for MSA. He has co-authored over 140 peer-reviewed publications and 3 book chapters. He was awarded 4 U.S. Patents and received several notable awards such as 2 R&D 100 Awards, the Microanalysis Society Birks Award and the National Collegiate Inventors Competition Award.

Atomic-Scale Observation of Structural Manipulation in Metal Oxides by in-situ TEM

Xuedong Bai

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The In-situ transmission electron microscopy (TEM) method is powerful in a way that it can directly correlate the atomic-scale structure with physical and chemical properties. In this presentation, we will report on the construction and applications of the in-situ TEM setup including mechanical, electrical and optical holders, which were built by scanning probe microscopy technique. So the manipulation and physical measurement have been realized inside TEM, where the atomic-scale imaging of electrically and/or mechanically driven structural evolution at atomic scale has been carried out by homemade in-situ TEM setup.

Exploring the lattice degree of freedom of metal oxides in transition between different structural phases may provide a route to enable new functionality in oxide materials with potential applications. Here we report on the oxygen diffusion in CeO_2 (1), and the experimental finding of strain-inhibited structural transition from perovskite to brownmillerite during the oxygen vacancy electromigration in epitaxial $LaCoO_3$ thin films (2), as well as the electrical and mechanical manipulation of topological polar configurations in $PbTiO_3/SrTiO_3$ superlattice (3, 4) will be included. This is a fundamental research for the future nanoelectronics such as memories and also valuable insight into lattice–charge interactions at nanoscale.

Thank Profs. Peng Gao, Zhi Xu, Congbing Tan, Jie Wang, Xiangli Zhong and Drs. Lifen Wang, Liang Zhu, Pan Chen et al. for their contribution to this talk.

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Biosketch

Xuedong Bai is a professor at Institute of Physics, Chinese Academy of Sciences (CAS). He obtained his PhD in materials science and engineering from Institute of Metal Research, CAS (1999). He ever worked at National Institute of Material Science (NIMS), Japan (2004-2005) as a visiting professor and at Georgia Institute of Technology (2001-2003) as a post-doctoral fellow. Prof. Bai's research interest is focused on in-situ TEM characterization method and its applications on nanoscience and materials physics. He is the co-author of over 200 papers in peer-reviewed journals, such as Nature and its sister journals, PNAS, PRL and JACS etc.. He delivered over 80 invited talks including MRS and AVS conference. Xuedong Bai was named a number of academic awards, including Hu Gang-Fu Physics Award (Chinese Physics Society, 2015), the 2nd Place of the National Science and Technology Award (2011), and Outstanding Young Research Foundation of China (NSFC, 2007) etc.

Low-dose 2D and 4D STEM imaging of beam-sensitive materials

Peter D. Nellist[1], Mathias Uller Rothmann[2], Hui Luo[1], Judy S. Kim[13], Juliane Borchert[2], Kilian Lohmann[2], Colum M. O'Leary[1], Alex Sheader[1], Laura Clark[1], Hazel E. Assender[1], Henry J. Snaith[2], Michael B. Johnston[2] Laura M. Herz[2],

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Scanning transmission electron microscopy (STEM) has not been widely regarded as an instrument suitable for low-dose imaging work, yet there is mounting evidence that a focused probe scanning rapidly over a sample can lead to reduced damage compared to wide-field illumination for the same total electron fluence. Many materials of current interest, including hybrid perovskites for photovoltaic applications, Li-ion battery materials and polymers, are highly beam sensitive. Here we demonstrate the application of both 2D and 4D STEM methods for low-dose structure determination. In Fig. 1A below, low-angle annular dark-field (LAADF) imaging reveals previously unobserved defect structures in the hybrid perovskite formamidinium lead iodide ($FAPbI_3$). In Fig. 1B below, electron ptychography was used to measure the small phase shifts that arise during electron transmission through light elements. Here the atomic structure of a crystalline domain in the polyester, polyethylene naphthalate (PEN). There is disagreement with the previous X-ray diffraction determined model, in particular the contrast between the strong (002) layers.

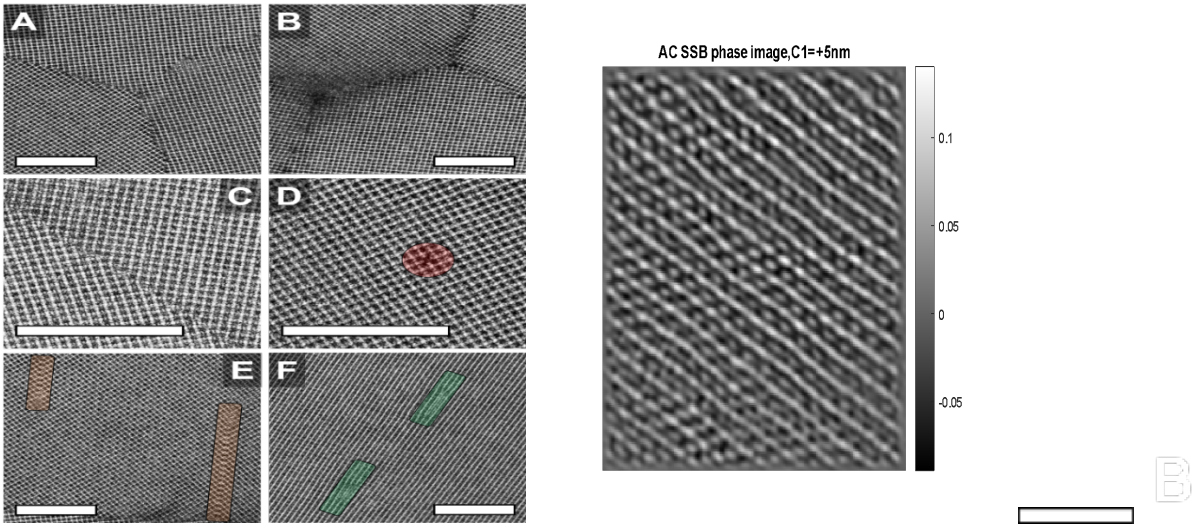

Figure 1 A: Grain boundaries in a $FAPbI_3$ thin film viewed using LAADF STEM. The sharness of the grain boundaries are apparent and the dislocations forming the low-angle boundary in the top right can be clearly seen. Scale bar 10 nm. B: The structure of PEN viewed using the phase image of a ptychographic reconstruction in STEM. Scale bar 1 nm.

Biosketch

Pete Nellist is a Professor and Co-Head of the Department of Materials, and a Tutorial Fellow at Corpus Christi College, University of Oxford. He leads a research group that focuses on the applications and development of high-resolution electron microscope techniques, in particular scanning transmission electron microscopy (STEM), including atomic resolution Z-contrast imaging, ptychography, electron energy-loss and energy-dispersive X-ray spectroscopy and applications of spherical aberration correctors. Pete gained his PhD from the Cavendish Laboratory, University of Cambridge. Since then he has worked in academia and in the commercial world in the UK, USA and Republic of Ireland. In 2007 he was awarded the Burton Medal by the Microscopy Society of America for exceptional contributions to microscopy, in 2013 the Ernst Ruska Prize of the German Microscopy Society, the 2013 Birks Award of the Microbeam Analysis Society and is the senior author on the publication awarded the 2015 Outstanding Paper Award in Materials Science from the European Microscopy Society. He is a former President of the Royal Microscopical Society he is a Board Member of the European Microscopy Society. He was elected to Fellowship of the Royal Society in 2020.

Materials Science Applications of Four Dimensional–Scanning Transmission Electron Microscopy (4D-STEM)

C. Ophus[1], B. H. Savitzky[1], S. E. Zeltmann[2], T. C. Pekin[3], K. Bustillo[1], and A. M. Minor[1,2]

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Traditional scanning transmission electron microscopy (STEM) detectors are large, monolithic regions that integrate a subset of the transmitted electron beam signal scattered from each electron probe position. With the introduction of extremely high speed direct electron detectors, we can now record a full image (2D data) of the diffracted electron probe scanned over the sample (2D grid), producing a four-dimensional dataset we refer to as a 4D-STEM experiment. Figure 1 demonstrates the rich atomic-scale information contained in these diffraction images, such as the sample structure, orientation, composition, defect structure and more. The spacing between adjacent STEM probes can be varied from below one Angstrom to hundreds of micrometers, allowing us to probe the functional length scale of materials and devices.

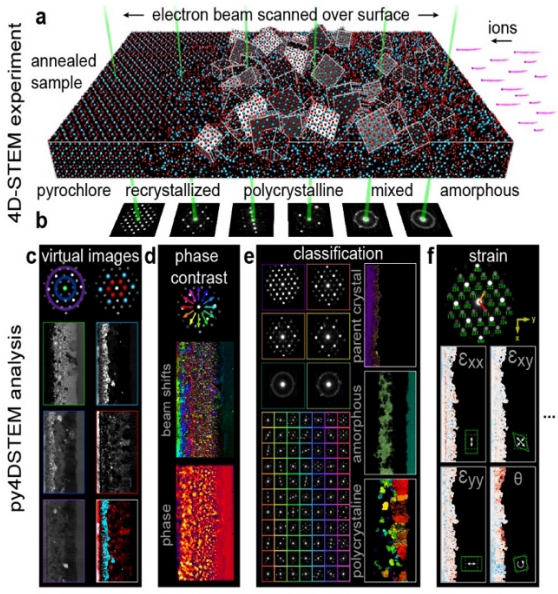

Figure 1 – **(a)** A 4D-STEM experiment performed on a gadolinium titantate sample, which has been amorphized via ion bombardment, and subsequently annealed. **(b)** Some diffraction images from a scan size of approximately 0.5 x 1 μm, showing characteristic patterns from single crystal, polycrystalline and amorphous sample areas. **(c)** Images formed by applying virtual detectors.**(d)** Differential phase contrast reconstruction. **(e)** Classification of the various sample regions. **(f)** Strain maps of the single crystal patent structure.
All analysis performed with **py4DSTEM**, figure adapted from BH Savitzky et al., **arXiv:2003.09523**.

In this talk, I will discuss several 4D-STEM applications in materials science. I will show several examples of nanobeam electron diffraction used to measure sample structure, orientation and strain, for samples ranging from metallurgical alloys to conductive polymers. I will also describe phase contrast imaging methods such as differential phase contrast, ptychography and STEM holography. Finally, I will also show how the scattering matrix (S-matrix) formalism can be used both to numerically invert sample structure under conditions of multiple scattering of the electron beam, and also for very fast simulation of large 4D-STEM datasets. I will show how these simulations can be performed using the newly developed PRISM algorithm and the open source code Prismatic, as well as describing future extensions such as double-channeling STEM electron energy loss spectroscopy (STEM-EELS) simulation.

Biosketch

Colin Ophus is currently a Staff Scientist at the National Center for Electron Microscopy (NCEM), part of the Molecular Foundry user facility, located at Lawrence Berkeley National Laboratory in the USA. He holds a BSc in Engineering Physics and a PhD in Materials Engineering, both from from the University of Alberta in Canada. After graduation, he was awarded a Postdoctoral Research Fellowship from NSERC Canada, for a joint position between the National Center for Electron Microscopy (NCEM) at Berkeley Lab and the MSE Department at the University of California Berkeley. He joined NCEM full time in 2012, and in 2018, Colin was awarded a Department of Energy Early Career Research Award to study the application of Quantum Metrology to Electron Microscopy.

Colin currently runs a user program that provides computational support to facility users for quantitative analysis and simulation of electron microscopy experiments. He also runs an independent research program that focuses on developing methods, algorithms and codes for simulation, analysis and instrument design for high resolution and scanning transmission electron microscopy (HRTEM and STEM). He has published over 110 peer-reviewed publications, and given over 30 invited lectures around the world. He is the project leader for 2 open source microscopy software packages; the open source Prismatic STEM simulation code (https://prism-em.com/) and the py4DSTEM python analysis toolkit for 4D-STEM (https://github.com/py4dstem/py4DSTEM).

The 4D Camera – An Electron Counting Camera for 4D-STEM Experiments

Peter Ercius[1], Ian J. Johnson[2], Hamish Brown[1], Phillip Pelz[1], Shang-Lin Hsu[5], Brent R. Draney[3], Erin Fong[2], Azriel Goldschmidt[2], John M. Joseph[2], Jason R. Lee[3], Jim Ciston[1], Colin Ophus[1], Mary C Scott[1], Ashwin Selvarajan[3], David Paul[3], David E. Skinner[3], Marcus Hanwell[4], Chris Harris[4], Patrick Avery[4], Thorsten Stezelberger[2], Craig S. Tindall[2], Ramamoorthy Ramesh[5], Andrew M. Minor[1,5] and Peter Denes[6]

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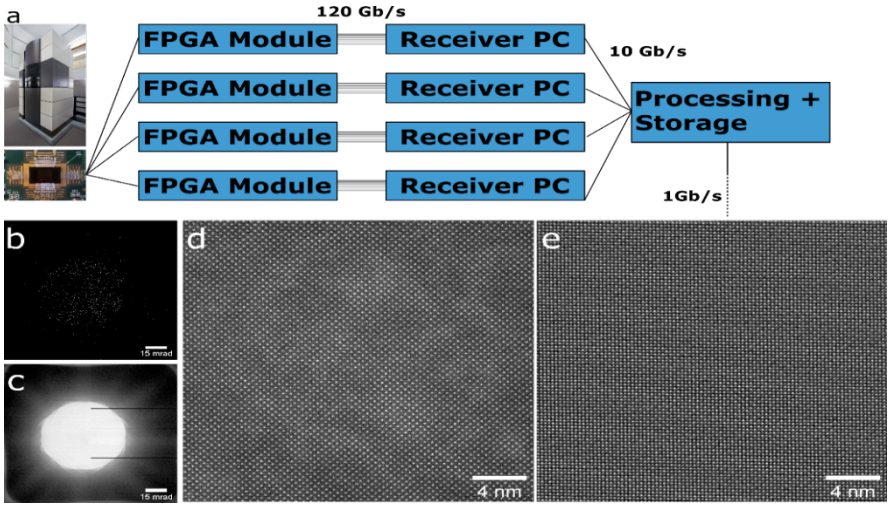
Direct electron detectors have provided electron microscopy with much more sensitivity and speed improving current capabilities and enabling entirely new imaging modalities. In scanning transmission electron microscopy (STEM) the ability to capture the full 2D diffraction pattern at every scan position provides a four-dimensional dataset which can be used to create STEM images with any detector geometry during post processing as well as new imaging techniques such as center-of-mass, ptychography, strain mapping and STEM holography. Essentially, the microscopist acquires all of the forward scattering data and then determines the ideal image contrast during post-processing. The 4D Camera was designed to operate at 87,000 Hz comparable to traditional microsecond scanning speeds producing a ~400 Gbit data stream and minimizing scanning artifacts like drift. Electron strike counting makes it possible to reduce this data stream 10-100x keeping only the electron scattering information and suppressing detector noise. I will show details of the detector and applications for STEM imaging as well as the open-source processing ecosystem being developed to handle large counted datasets.
Biosketch



a) Edge compute system for detector acquisition, storage and analysis at the microscope. The detector streams raw data to 4 receiver PCs which saves the data on a 5th PC for analysis and storage. b) A single detector frame with single electron counts. c) Summed frames showing a CBED pattern c) ADF-STEM and d) DPC-STEM images constructed from the electron counted data set of $PbTiO_3$.

Biosketch

Peter Ercius graduated from Cornell University with a B.S. in applied and engineering physics in 2003. He remained at Cornell and completed a Ph.D. in applied and engineering physics with Professor David A. Muller in 2009. His dissertation project focused on three-dimensional (3D) electron tomography of semiconductor devices using scanning transmission electron microscopy (STEM). He then joined the NCEM facility as a collaborative postdoctoral researcher for 2 years before being hired as a permanent Staff Scientist of the Molecular Foundry. Peter is currently in charge of the electron tomography program at NCEM and the dual aberration-corrected TEAM 0.5. Dr. Ercius is a leading expert in electron tomography and collaborates with users of the Molecular Foundry on a wide range of projects including S/TEM atomic resolution imaging, electron tomography, 4D-STEM scanning diffraction, in situ liquid TEM, and electron energy loss spectroscopy (EELS).

Probing the Local Charge and Phonons of Single Defects by Electron Microscopy

Xiaoqing Pan

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Spherical aberration correction marks a milestone in the development of transmission electron microscopy (TEM) which allows the quantitative determination of 3D structure and composition of nanostructures with atomic resolution. In combination with in-situ techniques, one can follow the phase transformations, chemical reactions, and dynamic behaviors of materials in response to applied fields and/or to the change of environment in real-time. The development of pixelated direct electron detectors and monochromators unlocked a door for a new era of discovery by electron microscopy. Today, beside the structure and composition, many properties of nanostructures and single defects can be determined by using scanning transmission electron microscopy (STEM). In this talk, I will present a novel 4D STEM diffraction imaging technique that we developed for mapping the local electric field and charge density with sub-Å spatial resolution. Using this technique, we are able to directly measure the electrical charge density, dipole moment, valence electron distribution between atoms, and charge distribution at heterointerfaces. Furthermore, recent instrumentation advances in electron energy-loss spectroscopy (EELS) in STEM enables an energy resolution of <5 meV with an ability to detect energy shifts <1 meV at the atomic spatial resolution. We demonstrate that space- and angle-resolved vibrational spectroscopy in a TEM allows the study of the vibrational properties of nanostructures and individual crystal defects. We observed a red shift of several meV and major changes in the intensity of acoustic vibration modes of a single stacking fault in SiC, and demonstrate that the changes are confined to within a few nanometers of the stacking fault. Our work opens the door to studying phonon propagation around defects, and to providing guidance to the engineering of specific thermal properties of materials

Biosketch

Xiaoqing Pan is the Henry Samueli Endowed Chair in Engineering, professor of materials science and engineering, and professor of physics and astronomy. He is also the inaugural director of the Irvine Materials Research Institute (IMRI), and founding director of the Center for Complex Active Materials – an NSF MRSEC. Pan is an internationally recognized materials scientist and electron microscopy expert due to his pioneering development and applications of novel transmission electron microscopy (TEM) methods for probing the atomic scale structure, properties and dynamic behaviors of materials. His work has led to the discoveries of new materials and novel functionalities. Pan has received the NSF CAREER Award and the Chinese NSF's Outstanding Young Investigator Award. He is an elected fellow of the American Ceramic Society, American Physical Society, Microscopy Society of America, and the Materials Research Society. He has published over 400 peer-reviewed scientific papers in high impact journals

Exploring Phononic Excitations with Monochromated STEM EELS

Kartik Venkatraman[1], Barnaby Levin[1], Katia March[2], and Peter A. Crozier[1]

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The enhanced energy resolution of recently developed monochromators now make it possible to use electron energy-loss spectroscopy (EELS) in the scanning transmission electron microscope (STEM) to probe materials systems in new ways. This powerful spectroscopic capability combined with the small focused electron probe opens the door to explore photonic excitations with high spatial resolution. In particular, the ability to probe short wavelength modes rapidly with atomic resolution will help establish a direct connection between vibrational excitations and local atomic level defects and structural heterogeneities such as surfaces and interfaces in materials. Here we discuss the two primary electron interaction processes that yield the vibrational EELS spectrum. Varying the experimental conditions allows long and short wavelength phonon modes to be sampled.

The fast electron can excite vibrational modes in materials either through dipole scattering (similar to IR absorption spectroscopy) or impact scattering (similar to inelastic neutron scattering). The spatial localization of these two scattering mechanisms is very different. The long-range dipole interaction is usually dominant in polar materials and leads to delocalized excitation of phonon modes associated with the center of the Brillouin zone (small scattering angles and momentum transfer). A weaker impact scattering signal typically involves higher momentum transfers yielding a spatially localized spectral signal. In a crystal, high momentum transfer interactions are associated with exciting the short wavelength phonon modes at the Brillouin zone boundaries (BZB). Provided suitable experimental conditions are employed, the weaker impact signal can be enhanced allowing long and short wavelength phonons to be probed simultaneously with on-axis vibrational STEM EELS. Probing long and short wavelength phonons simultaneously with EELS offers vibrational information on materials similar to that available from neutron inelastic scattering spectroscopy. A huge advantage of the EELS approach compared to neutron scattering is that we can probe short wavelength modes rapidly and with high spatial resolution.

Biosketch
Peter A. Crozier is a professor of materials and is chair of the Materials Graduate Program at Arizona State University. He develops and applies advanced transmission electron microscopy techniques to problems related to energy and the environment with special emphasis on electroceramics and catalytic materials. He has published extensively on in situ electron microscopy and is a recognized international leader in developing and applying the technique of aberration corrected transmission electron microscopy to problems in catalytic materials and oxide electrolytes. He also works with monochromated electron energy-loss spectroscopy to determine the optical and vibrational properties of ceramics. He is a member of the American Ceramics Society, Microscopy Society of America, Materials Research Society, the North American Catalysis Society and is a Fellow of the Microscopy Society of America. He is currently President Elect of the Microscopy Society of America.

Maximizing the information extracted from monochromated EELS and zero loss filtered 4D-STEM using a hybrid pixel direct detector with high dynamic range.

C.T. Koch[1], B. Haas[1], A. Eljarrat[1], W. Van den Broek[1], M. Schloz[1], J. Müller[1], H. Kirmse[1], T. Pekin[1], T. Lovejoy[2], O. Krivanek[2,1]

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Recent progress in aberration correction and monochromation in the scanning transmission electron microscope (STEM) has enabled a wide range of new experiments that greatly enhance the depth of information becoming accessible in materials characterization. Direct detection of electron diffraction patterns and spectra became available just at the right time to push the limits of detectability and resolution of these electron optically advanced instruments even further and make experimental data fully quantifiable, to the point, where the match of simulations and experiment is ultimately only limited by counting statistics.

We will report on the recent development of advanced data reconstruction algorithms for recovering the dielectric function from relativistic EELS spectra [1], a versatile ptychography reconstruction algorithm that includes various regularization options [2], and other ways to process 4D-STEM data [3]. We will present results obtained by applying these algorithms to data having been recorded using a monochromated STEM (Nion HERMES microscope operating between 30 … 200 kV, EELS-resolution < 6 meV, spatial resolution < 0.07 nm) [4] equipped with a Dectris ELA hybrid pixel detector [5] that combines true single electron sensitivity, high speed (2250 fps for 514 x 1030 pixels at 16 bit, 4500 full fps at reduced bit depth of 8 bit, and 18,000 fps with 1030 x 130 pixels and 8 bit) and nearly perfect point spread function (1.3 pixels in integration mode) with the capacity for a high dynamic range (> 10^7). Being able to detect spectra and (zero-loss filtered) diffraction patterns without read-out noise greatly increases the level at which these reconstruction algorithms can extract the underlying dielectric function, local crystal structure or maps of the electrostatic potential.

Example applications that will be presented include the extraction of IR-dispersion information from high-resolution EELS of plasmonic nanostructures and a comparison of various data processing algorithms (ptychography, center-of-mass, etc.) applied to (zero-loss filtered) 4D STEM data

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Biosketch

Christoph received his PhD at Arizona State University in 2002 under the supervision of Prof. John Spence. He then went on to a postdoctoral position at the Max Planck Institute for Metals Research in Stuttgart, Germany. In 2011 he accepted a professorship at Ulm University in Germany and in 2015 at full professorship at Humboldt University of Berlin, where he currently heads the structure research and electron microscopy group. His group operates several electron microscopes, and focuses on the development of novel imaging, diffraction, and spectroscopy techniques and their application to relevant problems in materials science

Mapping Electron Interactions with a Pocket-Sized Synchrotron (i.e., STEM)

Juan-Carlos Idrobo

Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, USA

The new generation of monochromated aberration-corrected scanning transmission electron microscopes (MAC-STEMs) are now capable of rival the capabilities of synchrotrons and in many cases supersede them. The later is evident when exploring local emerging phenomena in materials with high spatial resolutions.

In this talk, I will present few examples of how we are utilizing the new MAC-STEMs to study the thermal, optical and electronic properties of materials. In the first example, I will present our efforts in study the optical response of aperiodic nano-fabricated structures that locally (~ 1000 nm) are the same but at larger scales are translational invariant. I will also show how we can reveal the spatial range of electron-optical coupling of moire potentials in twisted graphene multilayers. Finally, I will discuss our efforts in measuring the limits of thermal property measurements using electron energy-loss spectroscopy (EELS) from cryogenic to room temperatures [1].

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[1] This research was supported by the Center for Nanophase Materials Sciences, which is a Department of Energy Office of Science User Facility, and instrumentation within ORNL's Materials Characterization Core provided by UT-Battelle, LLC under Contract No. DE-AC05-00OR22725 with the U.S. Department of Energy.

Biosketch

Juan Carlos Idrobo is a Senior Staff Scientist at Oak Ridge National Laboratory and Group Leader of the Scanning Transmission Electron Microscopy (STEM) Group at the Center for Nanophase Materials Sciences. His research consists in developing and applying analytical techniques in electron spectroscopy within monochromated and aberration-corrected scanning transmission electron microscopy to study the structure, electronic, magnetic, thermal, optical and topological properties of materials. He has published over 160 papers in peer review journals with over 15,000 citations. In 2018 Idrobo was recognized by Clarivate Analytics to be among the top 1% of researchers being cited in Cross-Field in *Web of Science*, between 2006 and 2016. Idrobo holds Physics degrees from Universidad de Los Andes in Colombia (B.Sc., 2000), University of Illinois at Chicago (Master, 2003) and University of California Davis (Ph.D., 2004).

From making disinfectants and rocket fuels to powering heavy-duty vehicles -- intermetallics and non-PGM biomimetic catalysts for small molecule activation

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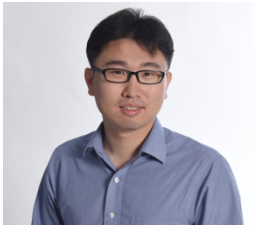
The ammonia you use to clean and disinfect your kitchen floor starts off as nitrogen, a gas that makes up almost 80 percent of Earth's atmosphere. But the conversion requires the breaking of a strong chemical bond in a high-heat, high-pressure industrial process known as the Haber-Bosch process. In nature, however, bacteria convert nitrogen gas to ammonia with a nitrogenase enzyme, whose active center is molybdenum, an abundant, nonprecious metal. By mimicking this biological nitrogen fixable process, my group has recently developed a series of new catalysts that can produce ammonia, rocket fuels, and power heavy-duty vehicles in a more sustainable way. The key is anchoring single metal atoms in a nitrogen/oxygen/carbon-coordinated environment to form a so-called single-atom catalyst (SAC). Unlike the Haber-Bosch process, which consumes massive amounts of energy and emits significant quantities of carbon dioxide to the atmosphere, these cheap, single-atom catalysts can be incorporated into modular and compact electrolyzing cells to produce commodities (ammonia, methane, and formate), as well as high-value-added chemicals like pharmaceuticals, with renewable solar or wind power. This emerging technology will not only make the traditional chemical production process greener but can also pave the way for a decentralized chemical industry.

Biosketch



Huolin Xin graduated from the Physics Department of Cornell University in 2011 and joined the University of California, Irvine in 2018. He received an accelerated tenure promotion to Associate Professor in 2020. Prior to becoming a professor at UCI, he worked at Brookhaven National Laboratory as a scientific staff member and a principal investigator from 2013 to 2018. He received the DOE Early Career Award, the UCI Distinguished Early-Career Faculty Award, MAS Distinguished Scholar Award, Castaing Award, and MSA Presidential Scholar Award. His work has twice been selected as the Top-10 Scientific Achievements in 2014 and 2019 by Brookhaven National Lab. His research has resulted in more than 200 peer-reviewed publications and 1 patent, 35 of which are published in Science/Nature sister journals (corresponding author on 10 of the 35).