Joint NSRC Workshop

Big, Deep, and Smart Data Analytics in Materials Imaging

Jointly Organized by the Five DOE Office of Science Nanoscale Science Research Centers and Held at Oak Ridge National Laboratory, Oak Ridge, TN
June 8-10, 2015
(www.cnms.ornl.gov/JointNSRC2015/)

Program
Abstracts
Participant List
Site Information

Program Committee:
Eric Stach, Center for Functional Nanomaterials, Brookhaven National Laboratory
Jim Werner, Center for Integrated Nanotechnologies, Los Alamos National Laboratory
Dean Miller, Center for Nanoscale Materials, Argonne National Laboratory
Sergei Kalinin, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory
Jim Schuck, Molecular Foundry, Lawrence Berkeley National Laboratory

Local Organizing Committee:
Hans Christen, Bobby Sumpter, Amanda Zetans, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory

Joint NSRC Workshop – Big, Deep, and Smart Data Analytics in Materials Imaging

Program

(p): Plenary lecture, 45 min., including questions
(i): Invited talk, 30 min., including questions
(c): Contributed talk, 15 min., including questions
(d): Panel discussion, 30 min.

Monday, June 8:

7:30 – 8:15 Breakfast

8:15 – 8:30 Introduction (M. Buchanan, ORNL Associate Laboratory Director for Physical Sciences)
8:30 – 8:45 Introduction (J. Nichols, ORNL Associate Laboratory Director for Computational Sciences)
8:45 – 9:00 Introduction (H.M. Christen, Director, Center for Nanophase Materials Sciences)

9:00 – 10:30 Morning session 1

- T1 (p): J. Sethian (Berkeley), *CAMERA: The Center for Advanced Mathematics for Energy Research Applications*
- T2 (i): I. McNulty (ANL) *The Argonne Integrated Imaging Initiative*
- T3 (c): T. Potok (ORNL), *Beyond Gutenberg: A Computational Analysis Approach to Scientific Discovery and Innovation*

10:30 – 11:00 Coffee Break

11:00 – 12:30 Morning session 2

- T4 (i): E. Stach (BNL), *Creating a Big Data Ecosystem at the Brookhaven National Laboratory: Successes, challenges and needs*
- T5 (i): J. Ciston (NCEM, Berkeley), *Current Efforts and Future Needs for Recording, Visualizing and Analyzing Multidimensional Datasets in Transmission Electron Microscopy*
- T6 (i): D. Vine (ANL), *Real time phase retrieval in nanobeam ptychography*

12:30- 2:30 Lunch/poster session

2:30 – 4:00 Afternoon session 1

- T7 (i): P. Voyles (U. Wisc), *Extracting Materials Structure Knowledge from STEM Data*
- T8 (i): D. Ushizima (LBL), *Scaling Analytics for Image-based Experimental Data*

- (d) Panel session (B. MacCabe [Chair], J. Ciston, E. Stach, F. DeCarlo), *Big data infrastructure for Imaging – challenges and opportunities*

4:00 – 4:30 Coffee Break

4:30 – 5:45 Afternoon session 2

- T9 (i): Q. Chen (UIUC), *Insights from watching dynamics: from patchy spheres to anisotropic nanocrystals*
- T10 (c): B. Doughty (ORNL), *Reducing the Complexity of Ultrafast Transient Absorption Microscopy Data into Decay Associated Amplitude Maps*
- T11 (c): M. Drouhard (UTK), *Immersion Data Visualization using the Oculus Rift with Applications in Materials Science*
- T12 (c): M.A. Groeber (AFRL) - *DREAM.3D: Thoughts & Lessons Learned While Creating a Common Environment to Store, Share &Work With Digital Microstructure Data*

6:30 Dinner (by reservation only – Riverside Grill, 100 Melton Lake Peninsula, Oak Ridge, TN 37830).
Dinner speaker: A. Tennant, Neutron Sciences Directorate, Oak Ridge National Laboratory

Tuesday, June 9

7:30 – 8:30 Breakfast

8:30 – 10:30 Morning session 1

- T13 (i): S. Sankaranarayanan (ANL), *Imaging and visualizing ultrafast energy transport via molecular simulations*
- T14 (i): D. Skinner (LBL), *Design Patterns in Web-based HPC : Materials Imaging*
- T15 (i): T. Proffen (ORNL), *Neutrons, Materials and Data Challenges*
- (d) Panel session (B. Sumpter [Chair], S. Sankaranarayanan, D. Skinner, P. Kotula) *Linking big data to computing for materials design*

10:30 – 11:00 Coffee

11:00 – 12:30 Morning session 2

- T16 (i): K. Kleese van Dam (PNNL), *Towards streaming analysis of dynamic imaging experiments*
- T17 (c): E. Lingerfelt (ORNL), *A New Computational Infrastructure for the Advanced Analysis of Nanophase Materials Imaging*
- T18 (c): W.C. Yang (NIST), *Automatic Image Analysis of High Resolution Videos Obtained Using Environmental Transmission Electron Microscopy*
- T19 (i): N. Browning (PNNL), *Using Compressive Sensing, Statistics and Automated Tracking Algorithms to Optimize Data Acquisition in High-Resolution In-Situ TEM experiments*

12:30- 2:30 Lunch/poster session

2:30 – 3:45 Afternoon session 1

- T20 (i): M.J. Demkowicz (MIT), *Bringing Bayesian methods into the materials modeling mainstream*
- T21 (c): Rick Archibald (ORNL), *Sparse sampling methods for image processing and data analysis*
- T22 (c): M. Comer (Purdue), *Stochastic Modeling for Materials Images*
- T23 (c): R. Patton (ORNL), *Large-Scale Deep Learning for Scientific Discovery*

3:45 – 4:15 Coffee

4:15 – 5:45 Afternoon session 2

- T24 (i): A. Borisevich (ORNL), *Discovering Materials Physics in Real Space by STEM*
  T25 (i): J. LeBeau (NCSU), *Quantifying the whole STEM image: where we are and where to go from here*
- (d) Panel session (M. Demkowicz [Chair], S. Kalinin, K. Kleese van Dam, J. LeBeau) *Inverse problems and Bayesian inference in image analysis*

6:30 Dinner (on your own)

Wednesday, June 10:

7:30 – 8:30 Breakfast

8:30 –10:30 Morning session 1

- T26 (i): A. Lupini (ORNL), *Electron Ptychography in STEM*
- T27 (c): S. Patala (NCSU), *Identification of Atomic Clusters through Point-Pattern Matching Algorithms*
- T28 (c): D. Frank Ogletree (Berkeley), *Smart Imaging With Scanned Electrons*
- T29 (i): P. Kotula (Sandia), *Acquisition, Analysis and Quantification of Hyperspectral Images*
- T30 (i): D. Gursoy (ANL), *Compressive sampling and its potentials in nanoimaging*

10:30 – 11:00 Coffee

11:00 – 12:30 Morning session 2

- T31 (i): F. DeCarlo (Argonne), *Data intensive science at synchrotron based 3D X-ray imaging facilities: the Argonne National Laboratory experience from data-limited to data-intensive to data-driven science*
- T32 (c): L. Drummy (AFRL), *Data Analytics in Materials Characterization at AFRL*
- T33 (c): S.V. Kalinin (ORNL), *Big data in SPM*
- (d) Panel session (H. Christen [Chair], E. Stach, D. Miller, J. Schuck, S. Kalinin) *Role of NSRCs in integration of big data and imaging: science and user programs*

Posters

Name	Affiliation	Title	Number
Barnard, Rick	Oak Ridge National Laboratory	Sparse Sampling Methods for Image Processing and Data Analysis	**P35**
Bao, Feng	Oak Ridge National Laboratory	A hierarchical Model Reduction Algorithm for Neutron Scattering Optimization Problem	**P21**
Barnard, Edward	Molecular Foundry, LBNL	Multimodal 3D Mapping in Photovoltaics Using Two-Photon Microscopy	**P15**
Belianinov, Alex	Center for Nanophase Materials Sciences, ORNL	Local Crystallography: Phase Recognition and Local Symmetry Analysis	**P3**
Belianinov, Alex	Center for Nanophase Materials Sciences, ORNL	Extraction and Analysis of Complete Information in Dynamic SPM and STEM	**P4**
Belianinov, Alex	Center for Nanophase Materials Sciences, ORNL	Cross Correlation of Multi-Dimensional Data Sets for Studying High Temperature Superconductors	**P5**
Berlijn, Tom	Center for Nanophase Materials Sciences, ORNL	Visualization of Atomic-Scale Phenomena in Superconductors	**P8**
Bilheux, Jean	Oak Ridge National Laboratory	2D/3D Visualization and Analysis of Neutron Imaging Data	**P26**
Chi, Miaofang	Center for Nanophase Materials Sciences, ORNL	4-D Data Acquisition in STEM: A Universal Detector for Sensitive Imaging	**P27**
Dyck, Ondrej	University of Tennessee, Knoxville	Automatic Image Segmentation for 3D FIB/SEM Reconstructions Through The Use of The Trainable WEKA Segmentation Plug-In in Imagej	**P29**
Dycus, Houston	North Carolina State University	Accurate Crystallographic Determination Using Scanning Transmission Electron Microscopy	**P6**
Evans, Barbara	Oak Ridge National Laboratory	Integrated Methods for Imaging of Cellulose Composites	**P41**
Ferragut, Erik	Oak Ridge National Laboratory	Anomaly Detection and Probabilistic Modeling for Image Data	**P37**

Name	Affiliation	Title	Number
Filar, Emre	Ames Laboratory	Correlation Of Structure and Chemical Environment of The Iron In Magnetosomes Residing in Magnetotactic Bacteria	**P44**
Gai, Zhang	Center for Nanophase Materials Sciences, ORNL	Chemically-Induced Jahn-Teller Ordering on Manganite Surfaces	**P7**
Gianfrancesco, Anthony	Center for Nanophase Materials Sciences	Atomic Imaging and Oxide Physics Via Monte-Carlo Methods	**P12**
Gianfrancesco, Anthony	Center for Nanophase Materials Sciences, ORNL	Deep Data Discovery of Atomic Level Structure-Property Relationship in An Iron Superconductor	**P13**
Hagmann, Mark	NewPath Research L.L.C.	Rapid Hyperspectral Measurements and Analysis For 3-D Sub-Nm Resolution Carrier Profiling in Semiconductors	**P14**
Hattrick-Simpers, Jason	University of South Carolina	Use of Bigdata Analytics to Identify High-Temperature Coatings for Nuclear Cladding Materials	**P39**
Henage, Thomas	NewPath Research L.L.C.	Possibility of Modulating a Freequency Comb With 4x10^9 Harmonics For Extremely High Data Rates or Secure Communication	**P31**
Laanait, Nouamane	Center for Nanophase Materials Sciences, ORNL	Driven Reaction Front Dynamics at Mineral-Aqueous Interfaces	**P46**
Maldonis, Jason	University of Wisconsin, Madison	Medium-Range Structure of Zr-Cu-Al Bulk Metallic Glasses from Structural Refinement Based on Fluctuation Electron Microscopy	**P9**
Marksov, Artem	Center for Nanophase Materials Sciences, ORNL	Atomic Resolution Image as A Markov Random Field: Seeing Through Noise	**P30**
Mazumder, Baishakhi	Center for Nanophase Materials Sciences, ORNL	Atom by Atom 3D Chemical Imaging Using Atom Probe Tomography	**P23**
McCullough, Kristin	Grand View University	Approximate Bayesian Computation for Censored Data and its Application to Assess The Reliability of Nanocomponents	**P32**

Name	Affiliation	Title	Number
Ophus, Colin	Molecular Foundry, LBNL	Current Efforts and Future Needs for Recording, Visualizing and Analyzing Multidimensional Datasets in Transmission Electron Microscopy	**P28**
Passian, A	Oak Ridge National Laboratory	Data Noise Reduction in High Spatial Resolution Spectral Imaging of Biomass	**P17**
Pradhan Dhiren	University of Puerto Rico	Humidity and Temperature Effects on Piezoresponse of Pb(Fe0.5Nb0.5)O3	**P47**
Prozorov, Tanya	US DOE Ames Laboratory	Magnetosome Magnetite Biomineralization: Making the Sense of It All	**P45**
Radhakrishnan, Balasubramaniam	Oak Ridge National Laboratory	Challenges in Physically Based Modeling and the Need for Data Driven Approach for Structure-Processing-Property Linkages in Materials	**P34**
Ramanathan, Arvind	Computational Science and Engineering Division, Oak Ridge National Lab	Statistical Inference Techniques for Integrating Neutron Scattering Experiments with Molecular Dynamics Simulations to Characterize Intrinsically Disordered Protein Ensembles	**P24**
Santiago, Mitk'El	Universidad Metropolitana	Crystal Structure Manipulation of Zno Particles Using Organic Ligands: Electrochemical and Photoelectrochemical Characterization and EXAFS Studies for Solar Energy Harvesting Applications	**P43**
Seleson, Pablo	Oak Ridge National Laboratory	Multiscale Material Modeling with Mesoscopic Models	**P33**
Sen, Fatih G.	Argonne National Laboratory	Grain Boundary Structures in Cdte: Atomistic Simulations and Transmission Electron Microscopy Analysis	**P2**

Name	Affiliation	Title	Number
Simpson, Mary Jane	Oak Ridge National Laboratory	Quantitative Separation of Ultrafast Transient Absorption Microscopy Data	**P16**
Sreehari, Suhas	Purdue University	Generations of Spatial Constraints for Electron Tomographic Reconstruction	**P25**
Strelcov, Evgheni	Center for Nanophase Materials Sciences, ORNL	Extracting Physics through Deep Data Analysis	**P18**
Sukumar, Sreenivas	Oak Ridge National Laboratory	Fast and Scalable Image-Analysis Apps on Big Data Architectures	**P38**
Vasudevan, Rama	Center for Nanophase Materials Sciences, ORNL	Big-Data Approaches to Analyzing Piezoresponse Force Microscopy/Spectroscopy: Unearthing New Physics	**P19**
Vasudevan, Rama	Center for Nanophase Materials Sciences, ORNL	Full Information Acquisition and Analysis of Surface Diffraction Data During Oxide Growth	**P20**
Vatsavai, Ranga	North Carolina State University and Oak Ridge National Laboratory	Scalable Machine Learning Approaches for Material Imaging	**P36**
Vorpahl, Sarah	University of Washington	Image Correlation of Scanning Probe, Electron and Fluorescence Microscopy Images for Advanced Materials in Energy	**P42**
Whitelam, Steve	Molecular Foundry, LBNL	Opportunities for Novel Imaging Techniques on Scales From Electrons to Assemblies	**P1**
Zhang, Jiaming	Hewlett Packard Labs	In Situ TEM Study on Crystallization of Nbo2 Thin Film	**P10**
Zhang, Minghao	UCSD	STEM Characterization for Oxygen Vacancies Creation In Li-Rich Layered Oxides for High Energy Li-Ion Batteries	**P11**
Zhou, Youjie	University of South Carolina	Large-Scale Fiber Tracking from Microscopic Composite Images	**P40**

CAMERA: The Center for Advanced Mathematics for Energy Research Applications

J.A. Sethian
Dept. of Mathematics, UC Berkeley and Math Department, LBNL

CAMERA is designed to provide the next generation of computational tools to tackle problems at DOE facilities. It is built around cross-disciplinary teams of applied mathematicians, software engineers, and facility scientists, working together to formulate models, derive appropriate equations, develop algorithms, build and test prototype codes, and deliver useable software. Initial efforts included projects joint with the Advanced Light Source, the Molecular Foundry, and the National Center for Electron Microscopy, and have now grown to include many other partners and facilities. We will discuss how CAMERA started, provide an overview of some of the current projects, and discuss the challenges and opportunities that come with building cross-disciplinary teams.

The Argonne Integrated Imaging Initiative – Ian McNulty (Argonne National Lab)

Beyond Gutenberg: A Computational Analysis Approach to Scientific Discovery and Innovation

Thomas E. Potok and Robert M. Patton
Oak Ridge National Laboratory

Pursuit of scientific discovery is the central underpinning concept of modern civilization. The immense investment in government-sponsored research in the US has laid the foundation for national scientific, economic, and military security in the 21th century. However, the doubling of scientific publication every nine years, jeopardizes this foundation because it is no longer humanly possible to track relevant research. In order for scientists, to maintain awareness of relevant scientific work and continue advancements in fundamental and applied research requires development of new computational methods that effectively utilize newly emergent computational and machine learning capabilities for accelerating true scientific progress. These machine learning methods can be used to develop recommender systems to help a scientist discover new and relevant publications across the full Web of Science publication base. Compare millions of research papers to help scientists and user facilities to discover potential collaborators working on the same or related materials. By applying these and other novel computational analytic methods to imaging science, we hope to unlocking the human knowledge documented and archived in unstructured text of thousands of image science publication in order to extend scientific discovery in this field and to others.

Creating a Big Data Ecosystem at the Brookhaven National Laboratory: Successes, challenges and needs

This presentation will describe our initial forays into 'Big Data' at the Center for Functional Nanomaterials, and then place them in the broader context of what is happening at BNL as a result of the arrival of the National Synchrotron Light Source —II. At the CFN, we installed a Gatan K2 Direct Electron Detector camera in the fall of 2014: this instrument provides transmission electron microscopy images at an incredible 400 frames per second at 2k x 2k - thus a 3GB/sec data stream. The resulting data stream has created considerable challenges in terms of data transfer, storage and distribution to the User Community, which I will describe and discuss how we have solved. At present, the CFN produces slightly less than 1PB/yr, but with a new instrument coming on line, we expect even more substantial issues. This data rate pales with what is anticipated from NSLS-II (>20PB/yr), and thus I will describe how we seek to provide an integrated Data Ecosystem at BNL. This ecosystem has three primary components: data storage and transfer, quasi-real time analysis and - most importantly - data driven discovery. This presentation will describe how we hope to create this ecosystem, and how we hope to part of a larger effort amongst all the DOE Scientific User Facilities to make data driven discovery in materials science a primary contributor to the national Materials Genome Initiative.

From Picometers to Terabytes: Data-intensive Electron Microscopy

Jim Ciston
Molecular Foundry, Lawrence Berkeley National Laboratory

Electron microscopy has been an indispensible tool for materials science, elucidating the complex local atomic structure of crystals, surfaces, and most critically, defects. Advancements in both hardware and software have extended this resolution deep into the sub-Ångstrom regime and enabled precisions of 1 picometer (below half the electron wavelength). The greatest challenge remains linking the exquisitely precise local measurements to global material properties and performance, a link that transforms mere images to rich data. Larger datasets are required to extend the statistical power of electron microscopy by investigating larger ensembles and fields of view without losing quantitative capabilities.

There is an ongoing revolution in the development of electron detector technology; images are now dramatically clearer and can be obtained at 100 times greater speed, opening new modes of electron microscopy formerly inaccessible. Scanning Transmission Electron Microscopy (STEM) has proven to be an enormously powerful tool to investigate the structure and composition of matter by producing atomically resolved signals of atomic structure, local bonding and valence states, and element identification. However, an enormous amount of information about the sample is discarded in conventional STEM experiments in the form of the converged beam electron diffraction (CBED) patterns produced at every location of the electron probe during scanning. New detectors can acquire over 1000 CBED patters per second over the full 2-dimensional grid of beam positions in an atomic resolution STEM acquisition, producing data at over 20 gigabits per second.

This approach represents a fundamental paradigm shift in the collection, analysis, and methodology of electron microscopy experiments, treating the microscope as a multi-detector electron scattering beamline. This technique supersedes the conventional “one picture at a time” approach and moves toward massive data analytics where one can effectively perform many experiments on a large, multidimensional dataset. Although proof of concept data has been acquired, key algorithm and instrumentation developments are needed to turn this complex methodology into a user-accessible technique that directly outputs materials properties, without burying users in hard drives full of data. With this approach, the complexity and scale of electron microscopy approaches particle physics and astronomy as a data intensive science, and we are only beginning to explore the manifold applications of mining such data.

Real time phase retrieval in nanobeam ptychography – David Vine (Argonne National Lab)

Extracting Materials Structure Knowledge from STEM Data

Paul M. Voyles
Materials Science and Engineering, University of Wisconsin-Madison

Aberration-correction makes it straightforward to obtain atomic-resolution STEM data of various types from most inorganic materials along several crystallographic directions. The next step is to turn that data into knowledge about materials structure. I will present our recent work on extracting structural knowledge in the form of atomic column positions from Z-contrast STEM using non-rigid registration and averaging of a series of short-exposure images. We have demonstrated sub-picometer precision in column positions under extremely high-dose conditions [1], and shown that the same high signal-to-noise ratio imaging method reduces the uncertainty in the number of atoms in each column derived from the standardless atom counting method pioneered by Lebeau [2]. At somewhat lower dose, we can achieve ~2 pm precision [3], but further progress towards high precision at lower dose will require incorporating denoising [4] with registration. Improvement in data quality for extremely noisy STEM EDS spectrum image data using similar approaches will also be discussed.

1. A. B. Yankovich, B. Berkels, W. Dahmen, P. Binev, S. I. Sanchez, S. A. Bradley, S. A. Li, I. Szlufarska, P. M. Voyles, Nat. Commun. **5,** 4155 (2014).
2. J. M. LeBeau, S D. Findlay, L. J. Allen, S. Stemmer, Nano Lett. **10,** 4408 (2010).
3. A. B. Yankovich, B. Berkels, W. Dahmen, P. Binev, P. M. Voyles, Adv. Struct. Chem. Imaging **1,** 2 (2015). DOI: 10.1186/s40679-015-0003-9
4. N. Mevenkamp, P. Binev, W. Dahmen, P. M. Voyles, A. B. Yankovich, B. Berkels, Adv. Struct. Chem. Imaging **1,** 3 (2015). DOI: 10.1186/s40679-015-0004-8

Title: Scaling Analytics for Image-Based Experimental Data

Speaker: Daniela Ushizima

LBNL Data Analytics and Visualization Group

DOE research across science domains is increasingly reliant on image-based data from experiments, demanding construction of new analysis tools that help scientists uncover relevant, but hidden, information in digital images. This data analysis task requires a new *modus operandi* for interpreting experiments conducted at LBNL and other DOE facilities, providing insight to guide and optimize data collection. In collaboration with colleagues at DOE BES and ASCR, and UCB BIDS, we will better exploit the scientific value of a broad array of high resolution, multidimensional datasets. The proposed multi-disciplinary work is designed around a coordinated research effort connecting (1) state-of-the-art data analysis methods with basis on pattern recognition and machine learning; (2) emerging algorithms for dealing with massive data sets; and (3) advances in evolving computer architectures to process the torrent of data. The result will be a set of data science models and new software infrastructure that provides tools that work both “on the factory floor,” as well as workhorse techniques for processing experimental data at ASCR supercomputing centers. These advances will accelerate the analysis of image-based recordings, scaling scientific procedures by reducing time between experiments, increasing efficiency, and opening more opportunities for more users of the imaging facilities.

Talk title:

Insights from watching dynamics: from patchy spheres to anisotropic nanocrystals

Talk abstract:

Rich understanding across fields emerges from visualizing how big model colloids and small nanoscale objects interact and communicate with each other. In our first example, we find the connections between patchy colloids with atoms, and develop new self-assembly strategies. These novel “supracolloidal molecules” are understood using insights from seemingly remote fields, including tessellation and condensed matter physics. For anisotropic nanocrystals, direct imaging of their motions in solution was elusive until the recent development of liquid phase transmission electron microscopy (TEM). We demonstrate a generalizable strategy to utilize their relative trajectories using TEM and measure a spatial map of their interaction potentials. Such nanoscale interactions are crucial for collective behaviors and applications of synthetic nanocrystals and natural biomolecules.

Reducing the Complexity of Ultrafast Transient Absorption Microscopy Data into Decay Associated Amplitude Maps

Benjamin Doughty[1*], Mary Jane Simpson[1], Bin Yang[2], Kai Xiao[2], Ying-Zhong Ma[1]

1. Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN
2. Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN
[*]doughtybl@ornl.gov

Electronic and molecular level dynamics in complex materials take place on a wide range of time and length scales. In order to directly track these dynamical processes, nonlinear optical spectroscopies have been adapted to microscopic platforms to obtain joint spatio-temporal images of excited state relaxation dynamics. The 'snap-shots' of electronic and nuclear motions acquired with femtosecond transient absorption microscopy (TAM) describe the initial mechanistic steps underlying many technologically important fields such as light harvesting and emitting applications, chemical sensors, and photocatalysis to name a few. While this powerful technique allows directly probing various dynamical processes in space and time, the complexity of the multi-dimensional TAM data set often leads to challenges in visualization, analysis, and understanding of the underlying processes. This arises from occurrence of multiple competing processes with distinct signs of transient absorption signals that evolve on timescales ranging from femtoseconds to microseconds. Thus, the methods aimed at analyzing the complex TAM data must be capable of capturing the wide range of dynamical responses and simplifying them into intuitive spatial maps.

This work describes a new data processing scheme intended to reduce the multi-dimensional TAM images into decay associated amplitude maps that represent the spatial distributions of key parameters describing the underlying dynamics. Our approach is based on a piece-meal global lifetime analysis of spatially dependent transient responses from experimentally acquired TAM images. Application of this new method to measured TAM data from a model methyl-ammonium lead iodide ($CH_3NH_3PbI_3$) perovskite thin film allows us to reduce a data set consisting of a 68 time-resolved images into 4 decay associated amplitude maps. These maps offer a simple means to visualize the complex electronic excited-state dynamics in this spatially heterogeneous perovskite film by separating dynamical processes occurring on characteristic timescales. These results provide insight into the ultrafast dynamics associated with excitons and charge carriers, which have recently been found to coexist at spatially distinct locations in this film.

B.D. Research sponsored by the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U. S. Department of Energy. Work by Y.-Z. M. and M.J.S. was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division.

Work by B.Y. and K.X. was conducted at the Center for Nanophase Materials Sciences (CNMS), which is a DOE Office of Science User Facility.

Immersion Data Visualization using the Oculus Rift with Applications in Materials Science

Margaret Drouhard,[1*] Chad Steed[2], Steven Hahn[2], Thomas Proffen[2]

[1]*University of Tennessee, Knoxville, TN*
[2]*Oak Ridge National Lab, Oak Ridge, TN*
[*]*meg.drouhard@utk.edu*

In this work, we introduce an immersive visualization experience for materials science data. Using an Oculus Rift device, we will demonstrate preliminary visualizations of crystal structure and neutron scattering data sets. Currently, researchers have access to powerful three dimensional (3D) visualization tools, including VMD[1], Mantid[2], and ParaView[3], allowing them to interactively explore the data and generate illuminating figures. However, these tools tend to have steep learning curves for standard usage, and navigation of data may be onerous even for advanced users. Moreover, most of the views are limited to two dimensional (2D) projections. The Oculus Rift integrates immersive 3D rendering with predictive head tracking[4] to provide intuitive comprehension and interaction with the data. Our work focuses on providing intuitive interactions that allow users to efficiently navigate and understand large, multi-dimensional data sets in a 3D environment. We anticipate that this tool will assist researchers in rapid data exploration and help them acquire near real-time feedback for ongoing experiments. Figures 1 and 2 show 2D projections of crystal structure data produced using Blender[5], which have been incorporated into an immersive visualization for the Oculus Rift. We will exhibit the initial implementation of our interactive immersion visualization for these structures and other data sets. Workshop participants are invited to interactively explore the data visualizations using the Oculus Rift.

---

[1] Humphrey, W., Dalke, A. and Schulten, K., ``VMD - Visual Molecular Dynamics" *J. Molec. Graphics* 1996, *14.1*, 33-38.

[2] O. Arnold, et al., Mantid—Data analysis and visualization package for neutron scattering and μSR experiments, Nuclear Instruments and Methods in Physics Research Section A, Volume 764, 11 November 2014, Pages 156-166, http://dx.doi.org/10.1016/j.nima.2014.07.029.

[3] Ayachit, Utkarsh, *The ParaView Guide: A Parallel Visualization Application*, Kitware, 2015, ISBN 978-1930934306.

[4] LaValle, S.M.; Yershova, A.; Katsev, M.; Antonov, M., "Head tracking for the Oculus Rift," *2014 IEEE International Conference on Robotics and Automation (ICRA)*, vol., no., pp.187,194, May 31 2014-June 7 2014.

[5] Blender Online Community. *Blender – a 3D Modeling and Rendering Package.* Blender Foundation, Amsterdam, 2015.

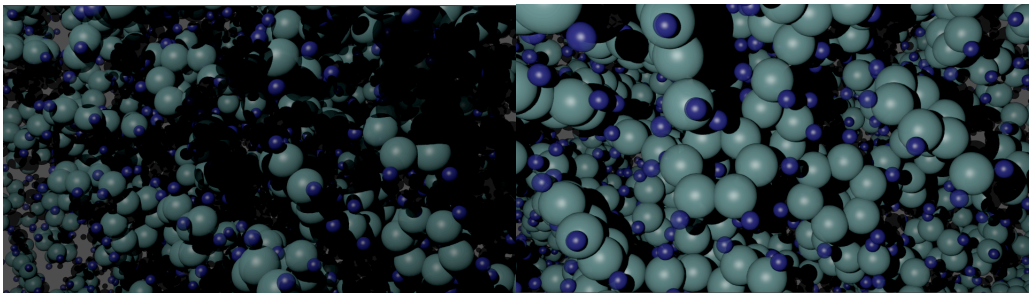


Figure 1: 2D projections of views from within crystal structure.

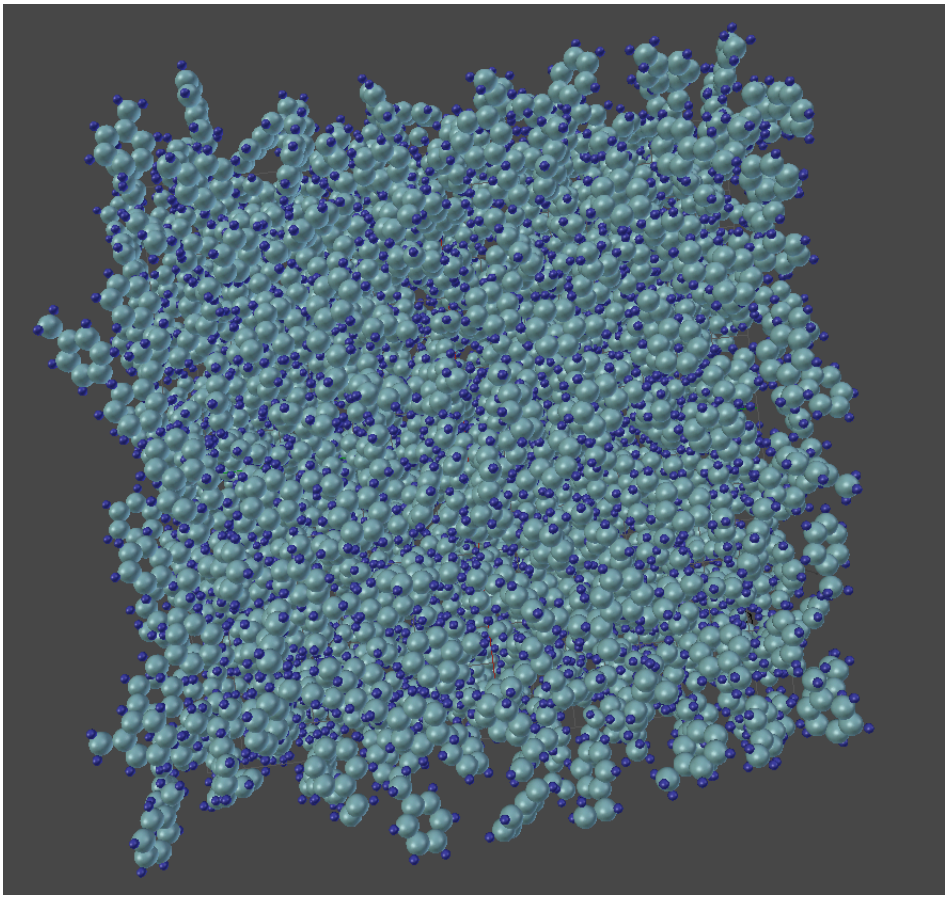


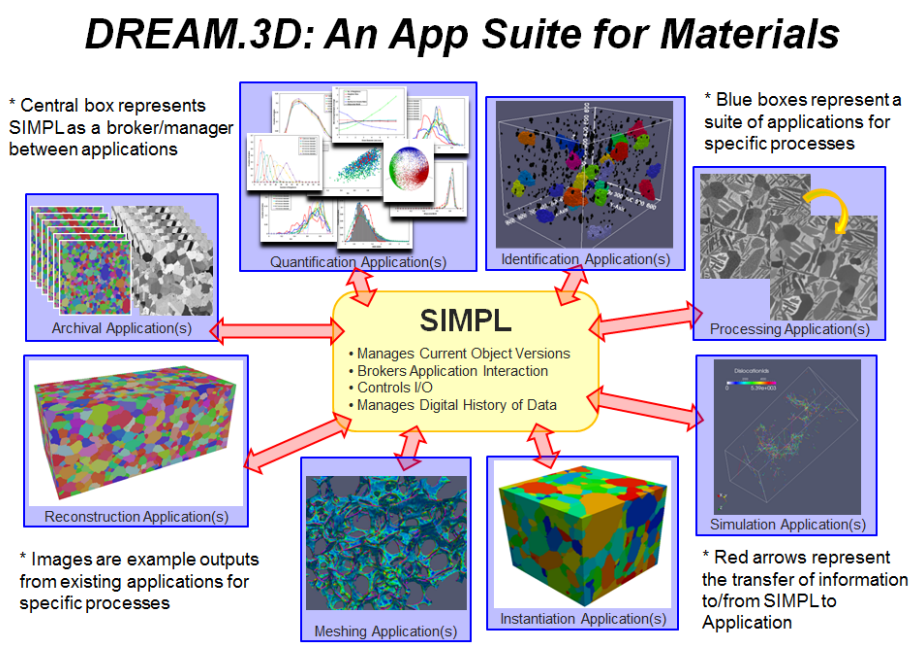
Figure 2: 2D projection of complete crystal structure.

DREAM.3D: Thoughts & Lessons Learned While Creating a Common Environment to Store, Share & Work With Digital Microstructure Data

Michael A. Groeber[1], Michael A. Jackson[2], Sean Donegan[2] and Dennis Dimiduk[2]

[1]*Air Force Research Laboratory*
[2]*BlueQuartz Software*
[*]*michael.groeber@us.af.mil*

This talk will discuss current efforts to develop a software environment that is both open and scalable to address the growing needs for quantitative, digital analysis of microstructural data. The ultimate goal of the DREAM.3D project is to fill the gap in the Integrated Computational Materials Engineering (ICME) chain with respect to `easy-to-use' microstructure quantification and representation tools across all material classes and length scales. Another important goal of the work is to standardize the format of material microstructure data, so that the increasing demand for access to scientific research data can be met. In attempting to accomplish these goals, the authors have encountered a number of issues with respect to data storage, representation scheme and code transferability. This talk will discuss these issues and propose a general/abstracted data structure for working with digital microstructure data across space and time, through SIMPL – a protocol library for managing spatial information. Specific examples will be given of fusing data sets from different imaging modalities, tracking features through time and analyzing data collected across several orders of magnitude in length scale. Finally, the collaborative framework associated with the DREAM.3D project will be presented, highlighting the extent of its current use (AFRL, NRL, ARL, LANL, Sandia, ORNL, APS, INL and many academic institutions).



Imaging and visualizing ultrafast energy transport via molecular simulations

Computing capability in atomistic simulations is growing exponentially at leadership computing facilities. In particular, atomistic simulations such as molecular dynamics are emerging as an effective tool to visualize atomistic phenomena, which occur in the picosecond to nanosecond time scales and over nanometer length scales. Likewise, new analysis and visualization techniques for 4D and higher-dimensional data are being researched. Recently, experimentalists have begun to conduct imaging in time-evolving lattice dynamics. In this talk, I will first highlight a couple of examples where atomistic simulations played a key role in elucidating energy transport and dissipation mechanism by complementing state-of-the-art characterization techniques. These examples also highlight the need to perform accurate calculations and develop work flow tools for computational materials design especially those that involve capture key bond-formation and bond breakage phenomena at reactive interfaces. In this context, I will discuss two of our ongoing efforts towards (a) bridging the atomistic and electronic scales by developing a stream-lined approach to force field development using first principles density functional theory training data and machine learning algorithms (b) integrating ultrafast time-resolved imaging with large-scale molecular dynamics modeling and in situ data analysis and visualization in order to design, conduct, and understand spatiotemporal measurements relevant to energy applications.

Design Patterns in Web-based HPC : Materials Imaging

David Skinner

This talk will review some of the ways teams of scientists leverage web APIs and UIs to access HPC resources. The NERSC Science Gateways effort has focused on building infrastructure to support the basic operations common to many projects engaged in building web interfaces to big iron computing and big data capabilities. Drawing on examples from high energy physics, climate, biology, and materials science we derive some of the design patterns relevant to materials imaging.

Neutrons, Materials and Data Challenges

Thomas Proffen
Director Neutron Data Analysis and Visualization, Neutron Sciences, ORNL

Neutron scattering enables simultaneous measurement of structural and dynamic properties of materials from the atomic scale (0.1 nm, 0.1ps) to the meso scale (1um, 1us). These ranges are remarkable complementary to current capabilities of computational modeling and the simplicity of the scattering cross section allows the prediction of neutron scattering data straight forwardly from computer modeling results.
Oak Ridge National Laboratory is home to two world leading neutron facilities, the Spallation Neutron Source (SNS) and the High Flux Isotope Reactor (HFIR) as well as the Oak Ridge Leadership Computing Facility. Turning these ORNL's unique strengths into the highest impact science is not without challenges. In this talk I will discuss challenges and opportunities and present recent advances at the SNS towards linking materials simulation and neutron scattering and towards real time feedback between experiment and simulation.

Towards streaming analysis of dynamic imaging experiments

Kerstin Kleese van Dam,[1*] Mathew Thomas,[1] Jeremy Teuton[1]

[1]*Pacific Northwest National Laboratory*
[*]*Kerstin.KleesevanDam@pnnl.gov*

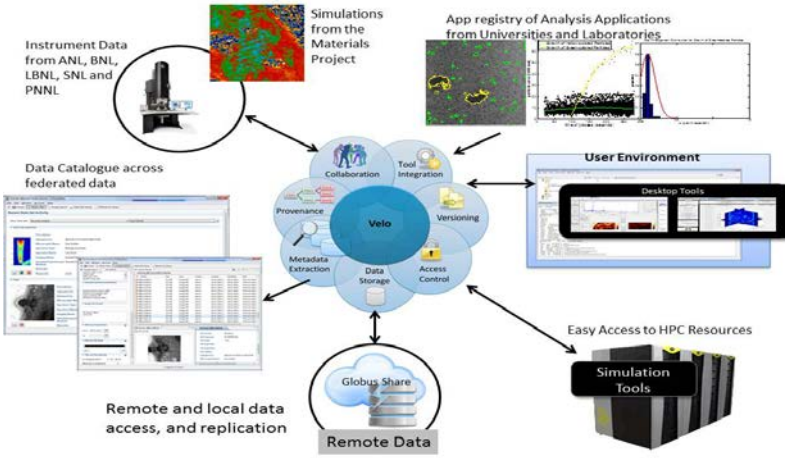
Experimental science is currently undergoing a technology driven transformation. Where in the past experimental methods did not possess the spatial or temporal resolution to capture physical, chemical and biological processes in sufficient details, new experimental in-situ and in-operando modalities promise to enable scientist to capture the evolving processes . Crucially this development in combination with in-situ analysis and interpretation could provide the necessary capabilities for experimental steering and ultimately control of transformations, leading in turn not only to faster scientific progress and better experimental results, but opens up the possibility of completely new scientific discoveries.

The PNNL Chemical Imaging Initiative (http://imaging.pnnl.gov/) started in 2010 to investigate real time analysis methods. It's first target use cases where Nano DESI Mass Spectrometry Imaging (http://www.pnnl.gov/science/highlights/highlight.asp?id=1233) and X-Ray Tomography of Biofilms. Initially the team developed workflows and graphical user interfaces to provide automated analysis capabilities, it became however quickly apparent that this static analysis approach was no sufficient to support the experimental process adequately. In evolving the initial analysis products MSIQuickView [1] and BiofilmViewer [2] we observed a number of key lessons:

- Researchers need to be able to adapt the analysis process during the experiment based on the evolving phenomena and their scientific priorities
- While real time analysis can provide critical assistance, the correct interpretation of these results still relies solely on the expertize of the scientists carrying out the experiment
- With exponentially rising data volumes new computational approaches for real time analysis need to be found, that provide the required speed and flexibility
- A two class system is emerging amongst experimental scientists, those that have advanced data analysis tools and are creating more papers faster, and those that do not and are scientifically less productive.

In response PNNL started to interlinked activities, the Premier Portal for Microscopy a collaborative environment for data and analysis that enables researchers to leverage common data analysis methods, and the Analysis in Motion Initiative focused on developing adaptive streaming data analysis and interpretation approaches in high velocity data environments.

The Premier Portal (http://www.premiernetwork.org/participants.stm) has been operational since mid-2014, serving around 30 collaborating sites. It was successfully demonstrated at SC14 in collaboration with LBNL, BNL, ANL and FSU as part of a series of data infrastructure demonstrations organized by DOE ASCR (http://www.pnnl.gov/science/highlights/highlight.asp?groupid=853&id=3917) . The Premier Portal (see Architecture Diagram in Figure 1) offers the ability to upload experimental results and carry out large scale image analysis utilizing tools contributed by collaborators such as FSU, Baylor and PNNL. Next to the data itself users can upload other supplemental information such as papers, presentations, graphics and videos. Once work is completed users can publish their data sets so it can be referenced in their subsequent publications. The Premier Portal has an agreement with the new Springer journal on Advanced and Chemical Imaging that unlimited supplemental information can be deposited in the Premier Portal and published as part of the normal publication process. Both data and paper are linked at publication and are each citable. Current work is focused on expanding the data management and publication service to be fully compliant with DOE and NSF requirements (using the Premier Portal would enable projects to easily meet their data management plan requirements). Furthermore we are expanding the capabilities offered via the data set 'landing page', in addition to the traditional data set information and download we are adding capabilities to visualize, browse and analyze the published data sets.



The Analysis in Motion Initiative (AIM – http://*aim.pnnl.gov*) started in January 1014 and is focused on research in four core areas:

- Streaming characterization methods that can identify and tag features of importance in high-rate, large-volume data streams
- Continuously evolving models that can interpret identified features as early indicators for phenomena of interest and relate these features and phenomena into explanatory hypothesis that can assist humans and machines in interpretation of results and optimization of future data taking
- Capturing human background knowledge through a new interaction paradigm that will not only support evaluation of candidate hypothesis, but introduce new knowledge into the analysis process
- A streaming analysis integration and execution environment that will ensure the timely analysis, interpretation, and steering required.

The initiative utilizes a high rate event messaging service (we achieved 600,000 messages / sec with our first version) to direct the flow of experimental data, analysis results and user input. Within the

infrastructure AIM uses a number of analytical models that work concurrently or collaboratively with the other models to characterize, analyze and interpret the experimental results (see Figure 2). The user has the ability to steer both the data acquisition and analysis process in response to the developing experimental process.

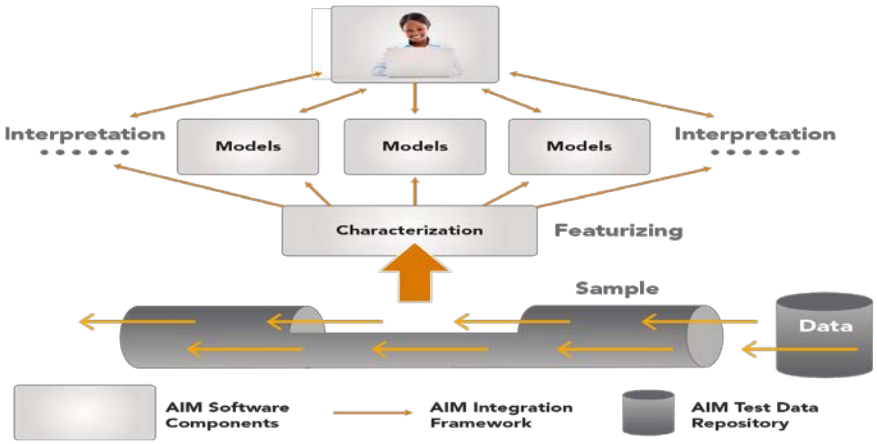


Figure 2 Analysis in Motion Infrastructure

The initial use case focused on the automated analysis of Nuclear Magnetic Resonance (NMR) experiments, aiming to reduce the number of scans needed for successful interpretation of the results and increase the automation level of the analysis process. Initial results with synthetic data showed that all major compounds could be identified without user intervention using 25% - 50% less scans than is standard for these type of experiments. Moving forward our use cases will focus on the analysis and interpretation of compressive sensing microscopy results and the steering of multiple interlinked bioreactors based on multi-modal content analysis.

The talk will briefly introduce the findings of the evaluation of the Chemical Imaging Initiatives real time analysis solutions after 3 years of operational use and describe the capabilities of the Premier Portal. The main focus of the presentation will be the work of the Analysis in Motion initiative.

References

[1] Lanekoff I, Burnum-Johnson K, Thomas M, et al. High-Speed MS/MS in Situ Imaging by Nanospray Desorption Electrospray Ionization Mass Spectrometry. *Analytical chemistry*. 2013;85(20):10.1021/ac401760s. doi:10.1021/ac401760s.
[2] Thomas, M.; Marshall, M.J.; Miller, E.A.; Kuprat, A.P.; Dam, K.K.-V.; Carson, J.P., "3D imaging of microbial biofilms: Integration of synchrotron imaging and an interactive visualization interface," Engineering in Medicine and Biology Society (EMBC), 2014 36th Annual International Conference of the IEEE , vol., no., pp.3304,3307, 26-30 Aug. 2014 doi: 10.1109/EMBC.2014.6944329

A New Computational Infrastructure for the Advanced Analysis of Nanophase Materials Imaging

E. J. Lingerfelt [1,*], A. Belianinov [2], O. Ovchinnikov [3], M. B. Okatan [2], E. Endeve [1], S. Jesse [2], C. T. Symons [4], M. Shankar [4], R. K. Archibald [1]

[1] *ORNL Computer Science and Mathematics Division*
[2] *ORNL Center for Nanophase Materials Sciences*
[3] *Vanderbilt University*
[4] *ORNL Computational Sciences and Engineering Division*
[*] *lingerfeltej@ornl.gov*

We present a new computational infrastructure for the advanced analysis of nanophase materials imaging. The Bellerophon Environment for Analysis of Materials (BEAM) software system will, for the first time, enable instrument scientists at the Center for Nanophase Materials Sciences (CNMS) to leverage the computational power of ORNL's Compute And Data Environment for Science (CADES) platform to perform near real-time data analysis of experimental data in parallel using a web-deliverable, cross-platform Java application. The BEAM system also offers long-term data management services utilizing the CADES large-scale storage system by providing users with a mechanism to easily manipulate remote directories in their private data storage area and transmit data files through the Java client over the network directly to CADES. At the core of this new system is a web and data server enabling multiple, concurrent users to securely access uploaded data, execute materials science workflows, and interactively engage analysis artifacts. In addition, BEAM's n-tier architecture facilitates user workflow needs by enabling integration of custom data analysis routines (serial *and* parallel) into the backend framework. We will discuss the current state of the system and our plans for rapid expansion of its capabilities over the coming year.

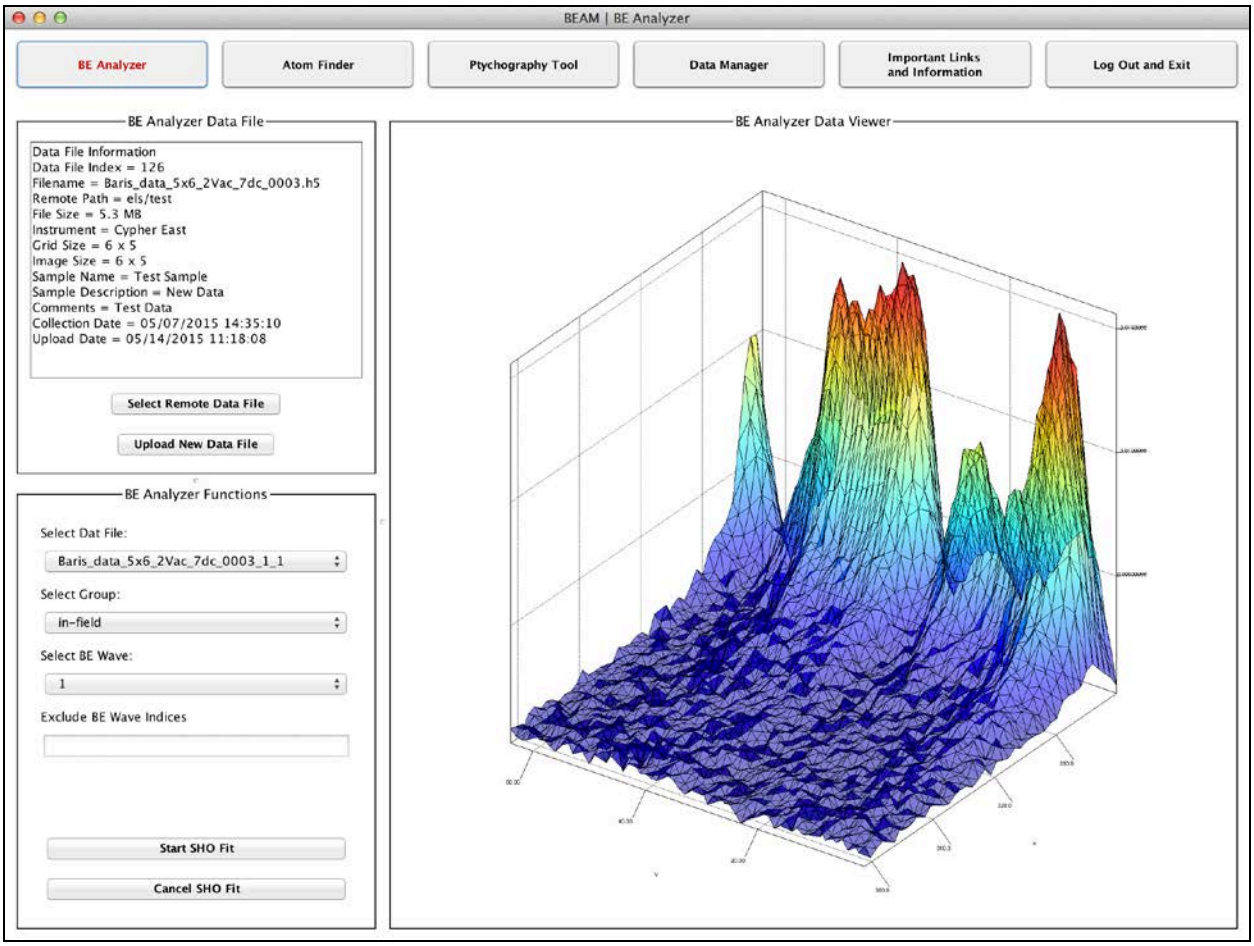


Figure 1. A screenshot of BEAM's BE Analyzer software tool displaying an interactive plot of spectrogram averaged CNMS experimental data.

Automatic Image Analysis of High Resolution Videos Obtained Using Environmental Transmission Electron Microscopy

Zahra Hussaini[1], Pin Ann Lin[1,2], Wei-Chang Yang[1,2], Renu Sharma[1]

[1] Center for Nanoscience and Technology, National Institute of Standards and Technology, Gaithersburg, Maryland, USA

[2] Maryland NanoCenter, University of Maryland, College Park, Maryland, USA

State-of-the-art environmental transmission electron microscopy (ETEM) enables *in situ* measurements of the dynamic changes occurring during gas-solid interaction. These changes usually take place rapidly at the nanometer scale. In order to record them in real time, large, high-speed data sets (≈ GB/sec) of videos may be generated because of the high image resolution and frame rates required. It is laborious to analyze such videos, frame by frame, so that the events of interest can be extracted at the required time resolution. There are two major inherent problems for automatic structural analysis of thousands of images extracted from a video sequence; (a) individual frames are noisy due to shot noise arising from the limited dose per frame and (b) sample drifts during recording periods that may last several seconds or even minutes. In order to overcome these problems we have developed an automatic method to obtain structural information from the images extracted from videos using a combination of algorithms publically available and developed at NIST. Our method has been tested to capture the crystal structure changed in a catalyst nanoparticle during growth of single-walled carbon nanotube (SWCNT) from the videos recorded using two different cameras. Details of the method and its application will be presented.

Using Compressive Sensing, Statistics and Automated Tracking Algorithms to Optimize Data Acquisition in High-Resolution In-Situ TEM experiments

Nigel D. Browning[1], Patricia Abellan[1], Lawrence Carin[2], James E. Evans[3], Richard L. Griswold[4], Eric Jensen[1], Kerstin Kleese Van Dam[1], Libor Kovarik[3], B. Layla Mehdi[1], Chiwoo Park[5], .Andrew Stevens[4], Jeremy R.Teuton[1]

[1]*Fundamental and Computational Science Directorate, PNNL, Richland, WA 99352, USA*
[2]*Electrical and Computational Engineering, Duke University, Durham, NC 27708, USA*
[3]*Environmental Molecular Sciences Laboratory, PNNL,Richland, WA, 99352, USA*
[4]*National Security Directorate, PNNL, Richland, WA, 99352, USA*
[5]*Industrial and Manufacturing Engineering, FSU, Tallahassee, FL 32306, USA*
(Nigel.Browning@pnnl.gov)

The development of aberration correctors for (scanning) transmission electron microscopes ((S)TEM) has increased both the quality and the quantity of image data that can in principle be generated experimentally. However, while it is possible to get direct images of atomic structures with high precision in electron beam tolerant materials, the increased current densities caused by the correctors have made beam damage more prevalent and the limitation to imaging in many cases is now the sample rather than microscope. Similar constraints are implicit during in-situ or operando TEM experiments, where the goal of the experiment is to observe a transient phenomenon without the beam altering the process. For both of these cases the goal now is to more efficiently use the dose that is supplied to the sample to extract the most information from each image. While one approach involves methods to deal with the data once it is acquired, here we discuss ways to optimize the acquisition of images during the experiment. Optimizing the dose/data content involves two main strategies to achieve dose fractionation – reducing the number of pixels being sampled in STEM mode, or increasing the speed of the images in TEM mode (thereby lowering the dose in each image). For the case of the STEM, compressive sensing methods can allow a dose reduction of an order of magnitude in the acquisition, in principle allowing data to be automatically recorded in a compressed form and only decompressed during instrument operation for the user to see the image being acquired. For the TEM mode of operation, the increase in speed increases the number of images and means that automated methods of tracking changes in the structure need to be developed so that only the important changes in the images need to recorded. The application of both of these methods will be discussed along with potential future improvements for quantitative materials observations.

The research described in this paper is part of the Chemical Imaging, Signature Discovery and Analysis in Motion Laboratory Directed Research and Development (LDRD) Initiatives at Pacific Northwest National Laboratory. The in-situ TEM work was primarily supported by Joint Center for Energy Storage Research (JCESR), an Energy Innovation Hub funded by the Department of Energy, Office of Science, Basic Energy Sciences. PNNL is a multi-program national laboratory operated by Battelle for the U.S. Department of Energy (DOE) under Contract DE-AC05-76RL01830. A portion of the research was performed using EMSL, a national scientific user facility sponsored by the Department of Energy's Office of Biological and Environmental Research and located at PNNL.

Bringing Bayesian methods into the materials modeling mainstream

[1]M. J. Demkowicz, [2]Y. M. Marzouk, [3]R. Aggarwal

[1]Dept. of Materials Science and Engineering, MIT, Cambridge, MA 02139
[2]Dept. of Aeronautics and Astronautics, MIT, Cambridge, MA 02139
[3]Dept. of Mechanical Engineering, MIT, Cambridge, MA 02139

Bayesian analysis is widely used in many areas of engineering, but is only now beginning to enter the mainstream of materials modeling. We present example applications of Bayesian analysis to inference and experimental design in two materials-related problems: films deposited on heterogeneous substrates and precipitation at solid-state interfaces. These examples illustrate opportunities for further uses of Bayesian methods in materials science, especially in settings where conventional modes of data analysis are inadequate. In particular, our work highlights the crucial role of reduced order, surrogate models in data-driven uncertainty quantification, inference, experimental design, and model discrimination.

Title:

Sparse sampling methods for image processing and data analysis

Abstract:

This talk will focus on sparse sampling methods and fast optimization developed specifically for image processing and data analysis. Sparse sampling has the ability to provide accurate reconstructions of data and images when only partial information is available for measurement. Sparse sampling methods have demonstrated to be robust to measurement error. These methods have the potential to scale to large computational machines and analysis large volumes of data.

Stochastic Modeling for Materials Images

Mary Comer[1,*] , Huixi Zhao[1], Dae Woo Kim[1], and Shruthi Kubatur[1]

[1]*School of Electrical and Computer Engineering, Purdue University, W. Lafayette, IN*
[*]*comerm@purdue.edu*

Materials scientists have long relied on statistics to help characterize properties of materials. The use of statistics is essential, since materials systems are fundamentally random in nature. However, some of the most powerful tools from probability theory, such as conditional probability, Bayes Theorem, and the total probability law, are not commonly used in materials science. These tools are of critical importance in the modeling of systems having anywhere from two to millions (or even billions) of random variables. Stochastic modeling of the joint behavior of these random variables requires a major step forward in the use of probability theory by materials scientists. We will present an overview of the work we have been doing to address this problem, in collaboration with materials scientists and engineers, for most of the past decade.

We will first discuss the general problem of stochastic modeling of complex systems, describing the use of tools from probability theory in understanding the joint behavior of multiple random variables. We will then provide an example of the application of some of these concepts for a specific materials problem, namely the joint modeling of characteristics at the microscale and the mesoscale, based on image data collected at the microscale [1]. This modeling allows us to investigate links between the geometry of a material at the mesoscale and local (pixel-wise) interactions at interfaces at the microscale. The mesoscale modeling is based on prior geometric information about a given material, while the microscale modeling is based on the Ising or Potts model [2].

The final topic we will discuss is rare event modeling in materials systems. The mathematical theory that underlies rare event modeling is large deviations. We will provide a high-level overview of large deviations theory and discuss our current plans to use this theory to model abnormal grain growth in polycrystalline materials.

It should be noted that the presentation will not assume any particular expertise in probability theory among the audience.

[1] Huixi Zhao, Mary Comer, Marc De Graef, “A unified Markov random field/marked point process image model and its application to computational materials”, IEEE International Conference on Image Processing (ICIP), Oct. 2014, pp.6101-6105.

[2] J. Simmons, C. Przybyla, S. Bricker, D. W. Kim, and M. Comer, “Physics of MRF regularization for segmentation of materials microstructure images,” IEEE International Conference on Image Processing (ICIP), Oct. 2014, pp. 4882–4886.

This research is supported by the Air Force Office of Scientific Research MURI contract # FA9550-12-1-0458 and by the Air Force Office of Scientific Research contract # FA9550-14-1-0313

.

Large-Scale Deep Learning for Scientific Discovery

Robert Patton, Thomas Karnowski, Seung-Hwan Lim, Derek Rose, and Steven Young

[1]*Oak Ridge National Laboratory*
pattonrm@ornl.gov

Machine learning has been extensively used in recent years to facilitate knowledge discovery and exploration in "Big Data" data sets, including scientific data sets through various 'grand challenges' comprised of teams of machine learning experts. A specific subset of machine learning, deep learning (DL), has recently enjoyed success and attention in the media and would be an ideal tool to perform scientific discovery. Deep Learning has been applied to computer vision data sets for classification and general object recognition to great success. In practice, however, deep networks have been they can be difficult to utilize for a variety of reasons. One reason is the process of defining the neural network topology and hyper parameters requires significant expertise. We have started development of tool called Multi-node Evolutionary Neural Networks for Deep Learning (MENNDL), primarily using ORNL's Titan supercomputer, which searches for both the optimal hyper parameters of a deep learning network and the optimal topology of the network. As a result of an evolutionary approach to searching, interactions between hyper parameters and their corresponding values can be easily studied. Initial results show clear correlations between hyper-parameter values of the neural network and the classification performance of the network, thereby providing deeper insight into the application of the neural network to a specific data. MENNDL will provide a scalable deep learning framework that will enable the key capability to adapt and apply DL technology to scientific data sets that are currently too large to be analyzed; potentially leading to new breakthroughs.

Discovering Materials Physics in Real Space by STEM

Qian He,[1] Jae Hyuck Jang,[1] Young-Min Kim,[2,1] Alex Belianinov,[3] Oleg Ovchinnikov,[4] Rick Archibald,[5,6] Sokrates T. Pantelides,[4] Sergei V. Kalinin[3,6], Albina Y. Borisevich[1,6*]

[1]*Materials Science and Technology Division, ORNL, Oak Ridge, TN, USA*
[2]*Korea Basic Science Institute, Deajeon, South Korea*
[3]*Center for Nanophase Materials Sciences, ORNL, Oak Ridge, TN, USA*
[4]*Department of Physics and Astronomy, Vanderbilt University, Nashville, TN, USA*
[5]*Computer Science and Mathematics Division, ORNL, Oak Ridge, TN, USA*
[6]*Institute for Functional Imaging of Materials, ORNL, Oak Ridge, TN, USA*
[*]*albinab@ornl.gov*

Atomic-scale characterization of materials is necessary to understand their properties and build predictive theoretical models. Aberration corrected scanning transmission electron microscopy (STEM) can provide direct structural and chemical information at the unit cell level with picometer precision. These capabilities enable local crystallography, real-space quantitative structural characterization approach based on STEM images. Images of single crystals and epitaxial films can be interpreted using the familiar unit cell language, by stretching overall lattice onto an image. In that framework, atomic positions can be used to map the quantities such as strain and polar displacements at interfaces and surfaces. Peak shape analysis can reveal finer details, such as 3D symmetry. For images where multiple phases or areas of the same phases with different orientations are present, local configuration analysis – purely real space approach to structure description - can be used, augmented by automated classification algorithms such as k-means clustering.[1]

These developments set the stage for images becoming a treasure trove of data, rather than illustrations. Simultaneously, the importance of automating the associated data processing approaches increases greatly. Automated analysis will enable us to evaluate all of the images collected, as opposed to select few as is the current practice. It will be invaluable for multi-frame sequences of images showing structural evolution in time or under other changing external parameters and is the only way to make full use of fast detectors capable of acquiring up to ~1000 frames per second. Scaling up local crystallography and local configuration approaches for analysis of multiple images and multi-frame sequences brings additional challenges. Ultimately, the atomic configurations retrieved in the analysis can be used for theoretical calculations, enabling theory to experiment comparison devoid of human bias. This can be further facilitated by establishing a continuous data pipeline from the microscope to supercomputer

This research is sponsored by the Materials Sciences and Engineering Division, Office of Basic Energy Sciences of the U.S. DOE. Part of this research was conducted at ORNL's Center for Nanophase Materials Sciences (CNMS), which is sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. DOE. Computational and automation work supported by ORNL's Laboratory Directed R&D Fund.

Quantifying the whole STEM image: where we are and where to go from here

James M. LeBeau*

Department of Materials Science & Engineering, North Carolina State University, Raleigh, NC 27695-7907

**jmlebeau@ncsu.edu*

Electron microscopy has been revolutionized by the advent of the aberration corrector. We can now acquire structural and chemical information well into the sub-ångstrom regime, unlocking information about material properties. In particular, atomic resolution scanning transmission electron microscopy (STEM), provides the flexibility to capture images that are directly interpretable and whose intensities depend sensitively upon the atomic species and number present. And with development of methods to place intensity and distance measurements on an absolute scale, quantitative STEM imaging is leading to new materials insights directly at the atomic scale.

In this talk, I will discuss the current state-of-the-art in quantitative STEM imaging along with thoughts on future directions for this important topic. I will present the experimental methods that place STEM images on an absolute intensity scale for truly quantitative imaging that providing unambiguous direct comparisons with theory. Equipped with these absolute intensity images, I will highlight applications such as absolute atom counting and composition determination from theory alone.

While these recent advances in quantitative intensity measurements have enabled new possibilities for atomic scale characterization, measurement of atomic displacements and distances from STEM images have typically been hampered by the presence of scan distortion. I will discuss new image analysis algorithms that can remove this distortion and show that sub-picometer precision and accuracy below 0.1% are now possible. Multiple case studies will be presented to demonstrate the power of these techniques; for example, I will show how picometer level precision enables the capability to quantify chemically correlated atomic displacements within a complex oxide solid solution.

Finally, we will end by looking towards the future where these results open a new world of atomic scale exploration that was previously just beyond our reach. I will discuss how concepts used in analyzing total diffraction data, such as the pair distribution function, can be applied to STEM data for direct real-space analysis of structure on an atom-column by atom-column basis. From this perspective, I will emphasize that we should aim to quantify all aspects of our electron microscopy data and that there remains much to be explored.

Electron Ptychography in STEM

Andrew R. Lupini,[1,3,*] Miaofang Chi,[2] Sergei V. Kalinin,[2,3] Albina Y. Borisevich,[1,3]

Juan Carlos Idrobo,[2] Stephen Jesse.[2,3]

[1] Materials Science & Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN

[2] Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN

[3] Institute for Functional Imaging of Materials, Oak Ridge National Laboratory, Oak Ridge, TN

*9az@ornl.gov

Recent advances in detector technology have enabled a variety of powerful new imaging modes in the scanning transmission electron microscope (STEM). It is possible to acquire the scattering distribution as a function of angle (the Ronchigram) at every probe position in a 2 dimensional (2D) scanned image to give a 4D dataset that we call a ptychogram [1]. Working with this 4D dataset provides a variety of advantages over traditional STEM imaging where a range of scattering angles are integrated to give a single value. For example, common STEM imaging modes can be generated numerically from this dataset (Fig. 1). Thus detector positions and angles can be optimized in post-processing rather than during the experiment, and detectors that could not be directly constructed, such as numerical filters that have sensitivities varying according to spatial frequency can be synthesized. This allows novel techniques such as extremely dose efficient imaging [2] or rapid measurement of optical aberrations. [3]

References:
[1] J. M. Rodenburg, B. C. McCallum and P. D. Nellist, Ultramicroscopy, **48** (1993), p. 304.
[2] T.J. Pennycook, *et al*, Ultramicroscopy (2015), *in press*.
[3] Research supported by Division of Materials Sciences and Engineering Division, Office of Basic Energy Sciences, U.S. DOE (ARL, AYB) and by Oak Ridge National Laboratory's Center for Nanophase Materials Sciences (CNMS), which is sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. Department of Energy (MC, SVK, JCI, SJ).

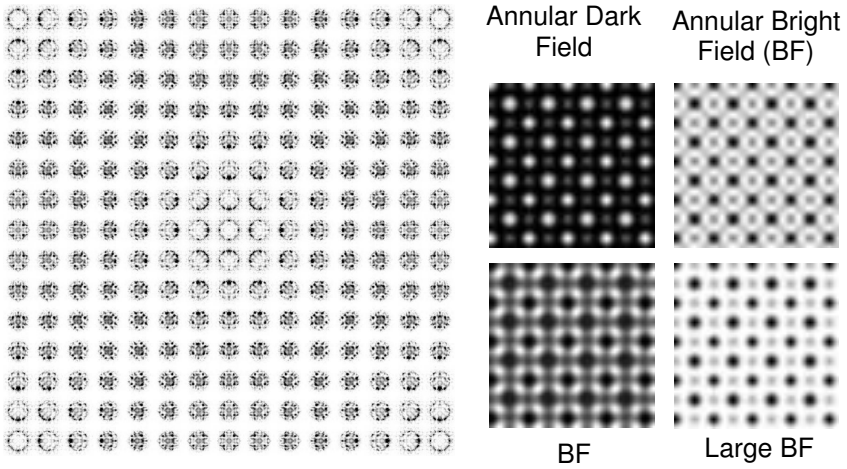


Figure 1. (Left) A simulation showing a series of Ronchigrams from different probe positions on $SrTiO_3$. (Right) this dataset can be integrated over different angular ranges to give traditional STEM images.

Identification of Atomic Clusters through Point-Pattern Matching Algorithms

Arash D Banadaki,[1] Sirkanth Patala[1,*]

[1]*North Carolina State University*
[*]*spatala@ncsu.edu*

I will present a novel application of pattern recognition algorithms for identifying atomic structures in defects. Atoms are the building blocks in all material systems and the diversity in their spatial arrangement gives rise to interesting material phenomena. For example, the periodic arrangement of atoms without an inversion center gives rise to the piezoelectric effect in ceramics. On the other hand, the lack of long range order in the arrangement of atoms gives glasses their unique viscoelastic properties. The materials science community has developed numerous techniques to describe atomic structures at these two extremes, i.e. the infinitely periodic crystals and the completely disordered glasses. However, there do not exist any general quantitative techniques for the identification and the description of patterns formed by atoms in defects, such as dislocations, surfaces and interfaces. In these defective regions, the crystal symmetry is broken but the arrangement of atoms is not entirely disordered. The properties of defects, which control the macroscopic material phenomena, is directly related to the spatial arrangement of atoms in these regions.

In this talk, the arrangement of atoms in the grain boundaries (GBs) of Aluminum will be used as a template to show how point pattern matching algorithms may be used to describe complex atomic arrangements. GBs exhibit atomic structures ranging from ordered (e.g. (111) twin boundary) to highly disordered (high-angle general GBs). These structures are obtained through atomistic simulations but the methods presented here are applicable for the analysis of images obtained both from computer simulations or microscopy techniques. We adopted the graph theory based point pattern matching algorithms to successfully identify polyhedral structural units in a variety of twist and tilt GBs (see Figure 1). The structure of general GBs may then be described as a stacking of these polyhedral units. This technique provides an exciting opportunity to *identify* complex structural units not only in defects but also in complex material systems such as distinguishing different types of short-range order in metallic glasses. Machine learning techniques may also be used to extend the types of atomic clusters that may be identified and improve the accuracy of the graph theory based matching algorithms.

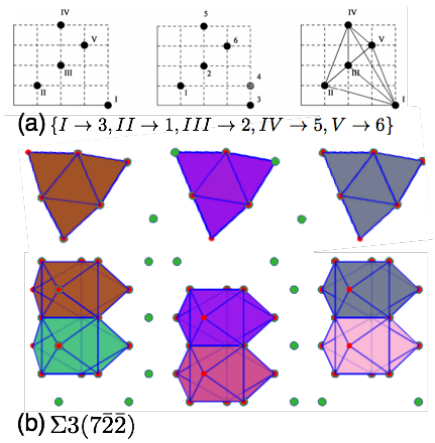


Figure 1. (a) Illustration of the point pattern matching algorithm between the two sets of points [1]. (b) Snub-Disphenoid deltahedrons identified in Σ3(722) general grain boundary.

[1] **Learning graph matching**, TS Caetano, JJ McAuley, L Cheng, LV Quoc and AJ Smola, *Pattern Analysis and Machine Intelligence*, IEEE Transactions, 31, 6, 1048 – 1058, 2009

Smart Imaging With Scanned Electrons

D. Frank Ogletree[1,*], Ed Barnard[1], Hao Wu[1,2], Shaul Aloni[1]

[1]*Molecular Foundry, Imaging Facility, Lawrence Berkeley National Lab, Berkeley CA*
[2]*University of California, Berkeley CA*
[*]*DFOgletree@LBL.gov*

In the "classical" paradigm for scientific imaging, commercial vendors first develop imaging hardware, followed by the creation of usually-proprietary software to operate the instrument in predefined modes. Image data is acquired interactively by researchers, alone or in collaboration with microscopists, guided by qualitative on-line interpretation of images. Quantitative, physics-based analysis of multichannel images, if done at all, is performed off-line and after the experiments, using a variety of commercial, open-source or user-developed software.

"Smart imaging" implies performing *experiments* in the microscope, not just recording "images", with adaptive control of data acquisition to maximize the "*data-to-damage*" ratio with nanoscale samples, combined with on-line physical analysis that feeds back to guide the experiment and the acquisition of microscopy data. Reaching this goal will require development of open-source software architectures that control the hardware – the experimental environment, the microscope with its detectors and spectroscopic tools – while integrating data visualization and theoretical analysis of real-time data streams.

We will describe one approach to this problem we are taking at the Molecular Foundry in the context of quantitative scanning electron microscopy (SEM) and spectroscopy, including Cathodoluminescence, Auger and reflected electron energy loss imaging. SEM vendor software is historically limited, and generally does not support quantitative multichannel data acquisition, low-dose imaging or spectroscopy. We are developing a framework based on scientific python[1] to control the hardware and visualization aspects of smart SEM. The issues are very similar for other types of scanned imaging, including analytical STEM, STM, AFM, STXM, NSOM and confocal optical spectroscopy.

Understanding SEM image contrast and spectroscopy requires analysis of sample morphology and the electron-sample interactions. [2] Although the physics of SEM image formation has been studied[3, 4], and scientific tools, some of them open-source, are available for data analysis,[4, 5] these methods also must be a part of "smart imaging".

[1] T. E. Oliphant, *Computing in Science & Engineering* **2007**, *9*, 10; C. Severance, *Computer* **2015**, *48*, 7.
[2] W. S. M. Werner, *Journal of Electron Spectroscopy and Related Phenomena* **2010**, *178-179*, 154.
[3] D. C. Joy, *Journal of Physics: Conference Series* **2010**, *241*, 012002 (8 pp.).
[4] J. S. Villarrubia, A. E. Vladar, M. T. Postek, *Surface and Interface Analysis* **2005**, *37*, 951.
[5] W. Smekal, W. S. M. Werner, C. J. Powell, *Surface and Interface Analysis* **2005**, *37*, 1059; H. Demers, N. Poirier-Demers, A. R. Couture, D. Joly, M. Guilmain, N. de Jonge, D. Drouin, *Scanning* **2011**, *33*, 135; C. A. Schneider, W. S. Rasband, K. W. Eliceiri, *Nat Methods* **2012**, *9*, 671.

Acquisition, Analysis and Quantification of Hyperspectral Images

Paul G. Kotula, Sandia National Laboratories
PO Box 5800, MS 0886, Albuquerque, NM, 87185-0886
paul.kotula@sandia.gov

Hyperspectral imaging, where a complete spectrum is acquired from an array of pixels or voxels, was first described in 1979 and has been available commercially since the early 1990s. From the start though, analysis of the large data sets was a challenge for researchers given the large number of observations and spectral channels and limitations of early personal computers. This presentation will look at acquisition and analysis of hyperspectral images in the context of electron microscopy (X-ray energy dispersive spectroscopy and electron energy-loss spectroscopy). The discussion of analysis will focus on data preprocessing and multivariate statistical analysis (MSA) methods, both of which are for the most part automated, followed by adding additional knowledge to make the output quantitative. Examples from a number of materials systems will be used to illustrate the power of MSA to reducing a large amount of raw data to a handful of readily interpreted factors while facilitating subsequent quantification.

Sandia National Laboratories is a multi- program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Company, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Compressive sampling and its potentials in nanoimaging

Doga Gursoy, ANL

Nyquist-Shannon (NS) sampling theorem plays a fundamental role in signal sampling and processing and is of vital importance for many engineering applications ranging from consumer electronics and electromagnetic sensing to modern communication devices. According to the NS theorem, the sampling rate of the signals should be at least as twice of the maximum frequency component of the signal in order to reliably describe it. The compressive sampling (CS) theory, on the other hand, allows recovery of signals from far fewer samples or measurements than the NS theory predicts. In other words, the information rate of a signal may be much smaller than suggested by its bandwidth. This allows us to formulate inverse models for the recovery of model parameters using less number of measurements without sacrificing accuracy of the solutions. Commonly the sparsity of the solutions is governed by a regularization term that penalizes certain features or variations in solutions. In this talk, I will describe some of the basics in CS theory and in which ways this can be applied for nanoimaging applications to reduce the number of measurements required to image certain types of samples. This understanding has the potential to impact design of future experiments, especially for scanning based systems that have multitude degrees of freedom.

The Advanced Photon Source (APS) is the US premier source of hard X-rays. In recent years the very high X-ray photon flux generated by the APS has been matched by a new generation of X-ray area detectors that are able to collect images at unprecedented speed with exposure time ranging from hundreds of milliseconds to hundreds of picoseconds allowing for a full tomographic dataset to be acquired in less than one second. This new capability opens the door for real-time, in situ and dynamic 3D studies, but also presents new challenges for data management, analysis and distribution.

In this talk we will describe how we are handling this transformation and how we are preparing for the even brighter X-ray source promised by the APS upgrade.

Data Analytics in Materials Characterization at AFRL

Lawrence Drummy[1]

[1]*Air Force Research Laboratory*
Materials and Manufacturing Directorate
Wright Patterson AFB, OH 45433
lawrence.drummy.1@us.af.mil

Air Force materials characterization requirements range from long-term fundamental research to rapid response needs for fielded systems, from biomaterials, to nanocomposites, metals, semiconductors and ceramics. Multi-scale, multi-dimensional data is becoming ever more prolific in materials characterization techniques such as electron microscopy and synchrotron X-ray scattering, and the development of new algorithms is necessary to gain meaningful knowledge from the data obtained. Direct coupling of physics-based models with the experimental characterization process has been a focus of Integrated Computational Materials Science and Engineering (ICMSE) at AFRL. This integration of models and materials characterization allows for new possibilities in materials analysis and development, and new challenges in data.

As microscopes and other instruments become more advanced and sensors such as CCDs and direct electron detectors become more sensitive and run at higher frame rates, fundamental physical limitations in data collection still remain, such as Poisson noise inherent to all experimental measurements. Therefore, an improved understanding of the types of noise present in detectors is required, and algorithms such as Model Based Iterative Reconstruction (MBIR) which model noise in tomographic data quantitatively can provide dramatically improved results over existing methods. MBIR uses a forward model to describe the image formation process, and physics-based prior models such as the Markov Random Field to constrain the results to reconstructions which are physically realistic. This talk will describe recent efforts at AFRL to develop algorithms such as MBIR which extract useful knowledge from raw data. Current work in multi-modal data acquisition and reconstruction methods, dynamic sampling, and autonomous experimentation will be highlighted. A new ICMSE digital collaborative environment at AFRL will also be discussed, in the context of providing a unifying framework for data management and software development.

Big data in SPM – Sergei Kalinin (Oak Ridge National Lab)

Opportunities for novel imaging techniques on scales “from electrons to assemblies”

Stephen Whitelam, Molecular Foundry

In the Molecular Foundry Theory Facility we are interested in phenomena that occur on a wide range of scales. These phenomena include electronic excitations and charge transfer, X-ray-matter interactions, and the self-assembly of components into large-scale structures. In this talk I will describe examples of interest to us that call out for new ways of imaging. For instance, I will describe the self-assembly of a multicomponent metal-organic framework [Whitelam, Yaghi, Stoddart, et al. PNAS, 2015] whose novel properties can be fully realized only through the development of imaging techniques that can see with chemical and spatial resolution inside a 3D structure.

Grain boundary structures in CdTe: Atomistic simulations and transmission electron microscopy analysis

Fatih G. Sen[1], Christopher Buurma[2], Tadas Paulauskas[1], Ce Sun[3], Moon Kim[3], Sivalingam Sivananthan[2], Robert F. Klie[2], Maria K.Y. Chan[1]
[1]Argonne National Laboratory, Lemont, IL, 60439, U.S.A.
[2]University of Illinois at Chicago, Chicago, IL, 60607, U.S.A.
[3]University of Texas at Dallas, Dallas, TX, 75080, U.S.A.

The achieved power conversion efficiency of polycrystalline CdTe photovoltaic cells has remain far below its theoretical maximum. It is believed that the presence of defects in poly CdTe is one of the main contributors to low efficiencies in CdTe. Therefore, it is important to understand the role of vacancies, interstitials, dislocations and grain boundaries on the electronic structure of CdTe. Atomistic-level characterization, combining transmission electron microscopy (TEM) and first principles modeling, is crucial in developing such a fundamental understanding. The modeling of the electronic structure of the grain boundaries and dislocation cores requires accurate description of the atomic structure at the grain boundary interfaces. In the present study, to systematically understand the role of grain boundaries on CdTe optical properties, different bi-crystals of CdTe formed between different orientations are fabricated. The bi-crystal interface crystal structures were analyzed with an aberration corrected TEM. We generated atomic structures of interfaces using image analysis methods and also using the crystallographic information. We carried out first principles density functional theory (DFT) calculations for the grain boundaries structures. We analyzed the atomic structures based on thermodynamics of grain boundary formation and discussed the pertinent changes in the electronic structure of CdTe. The electronic density of states of different CdTe grain boundaries will be reported and evaluated based on their photovoltaic performance. With underlying mechanisms revealed, we will discuss new techniques that can be developed to eliminate recombination centers and allow for high-performance CdTe devices.

ACKNOWLEDGEMENT: Use of the Center for Nanoscale Materials was supported by the U. S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357. The submitted abstract has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. The U.S. Government retains for itself, and others acting on its behalf, a paid-up nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government. (no table selected)

Local crystallography: phase recognition and local symmetry analysis

Alex Belianinov,[1,2] Qian He,[3] Oleg Ovchinnikov,[1,2] Artem Maksov[1,2] Rick Archiblald[1,3], Chris Symons[1,4], Stephen Jesse,[1,2] Albina Borisevich,[1,3] Sergei V. Kalinin[1,2]

[1] Institute for Functional Imaging of Materials,

Oak Ridge National Laboratory, Oak Ridge, TN 37831

[2] The Center for Nanophase Materials Sciences,

Oak Ridge National Laboratory, Oak Ridge, TN 37831

[3] Computer Science and Mathematics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

[4] Computational Sciences and Engineering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

5 Materials Sciences and Technology Division,

Oak Ridge National Laboratory, Oak Ridge, TN 37831

Abstract

With advances in high resolution imaging in (scanning) transmission electron microscopy and scanning probe microscopies, precision measurements (10 pm or better) of atomic positions are routinely obtained. This level of fidelity is sufficient to correlate bond length (and hence energy), as well as bond angle, to functional properties of materials. We introduce an approach for local analysis of material structure based on statistical analysis of individualized, local atomic

neighborhoods. Both supervised and unsupervised learning algorithms are used; this allows for a highly flexible framework of specified metrics which in turn are sensitive various lattice aspects. Using the local neighborhood approach we demonstrate phase identification as well as unit cell identification and classification. Multiphase catalytic materials, complex oxides and 2D materials are presented as case studies .Furthermore approaches for building image genomes and structure-property libraries, based on conjoining structural and spectral realms through local atomic behavior are discussed.

Acknowledgement:

Research related to atomic resolution imaging (AB, RA, CS, SJ) was sponsored by Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Department of Energy. The computational portion of this research for (OO, AM, SVK) was conducted at the Center for Nanophase Materials Sciences, which is a DOE Office of Science User Facility.Imaging research portion supported Q.H. A. B. by the U.S. Department of Energy, Basic Energy Sciences, Materials Sciences and Engineering Division.

Extraction and analysis of complete information in dynamic SPM and STEM

Alexei Belianinov, Suhas Somnath, Sergei Kalinin, Stephen Jesse

Institute for Functional Imaging of Materials and the Center for Nanophase Materials Science, Oak Ridge National Laboratory,
Oak Ridge, TN 37831

In the last three decades, Scanning Probe Microscopy (SPM) as well as Scanning (Transmission) Electron Microscopy (STEM) have emerged as primary tools for exploring and controlling the nanoworld. A critical part of SPM measurements is the information transfer from the tip-surface junction to a macroscopic measurement system. This process reduces the many degrees of freedom of a vibrating cantilever to relatively few parameters recorded as images. Similarly, the details of dynamic electron-beam to sample interactions at various time scales and beam energies are averaged out by transitioning to millisecond time scale of pixel acquisition, and pertinent material details are lost due to insufficient sampling of the detector space. Hence, the amount of information available to the external observer is severely limited, and its selection is biased by the chosen data processing method. We report a fundamentally new approach to SPM and STEM imaging using information based analysis of the data stream from the detector. This approach allows full exploration of complex probe-surface interactions, spatial mapping of multidimensional variability of material properties and their mutual interactions, and data collection at the information channel capacity limit. Extension of the technique to spectroscopic and higher dimensional imaging modes will also be discussed. Challenges and opportunities in transitioning to a high performance computing environment, and the outlook for expanding this technique into other microscopy fields will be presented.

Acknowledgements

Research for all authors was conducted at the Center for Nanophase Materials Sciences, which is sponsored at Oak Ridge National Laboratory by the Scientific User Facilities Division, Office of Basic Energy Sciences, U. S. Department of Energy. Research was supported by the US Department of Energy, Basic Energy Sciences, Materials Sciences and Engineering Division.

Cross correlation of multi-dimensional data sets for studying high temperature superconductors

A. Belianinov, A. Gianfrancesco, W. Lin, A. Sefat, B. Sales, S. Jesse, S. Kalinin

We explore atomic level spatial variability of electronic structure in Fe-based superconductor FeTe0.55Se0.45 (Tc = 15 K) using current-imaging tunneling-spectroscopy (CITS). We then use multivariate statistical analysis, as well as more traditional Dyne's fitting techniques[1], to analyze the data and differentiate regions of dissimilar electronic behavior that can be identified with the segregation of chalcogen atoms, as well as boundaries between terminations and near neighbor interactions.

Multivariate and clustering analysis of the 3D (or higher) dimensional data set allows identification of the spatial localization of electronically dissimilar regions. However, the quality of the CITS experiment depends largely on the drift of the tip while the measurement is performed. Ideally, imaged areas and spectroscopy areas would be maximally correlated; e. g. the tip position on the spectroscopy grid can be mapped exactly on the STM image. This allows accurate identification of the source in the electronic signal which allows for chemical discrimination of the STM data including separation of atomic identities, proximity and local configuration effects.

Acknowledgement:

The computational and experimental portions of this research for (AB, AG, SJ, SVK) was conducted at the Center for Nanophase Materials Sciences, which is a DOE Office of Science User Facility.
Sample growth portion of this research supported AS BS by the U.S. Department of Energy, Basic Energy Sciences, Materials Sciences and Engineering Division.

References:

1. W. Lin, Q. Li, B. C. Sales, S. Jesse, A. S. Sefat, S. V. Kalinin and M. Pan, ACS Nano 7 (3), 2634-2641 (2013).

Accurate crystallographic determination using scanning transmission electron microscopy

J. Houston Dycus[1*], J. Harris[1], X. H. Sang[1], C. M. Fancher[1], A. A. Oni[1], S. D. Findlay[2], E. T. Chen[1], J. L. Jones[1], C. C. Koch[1] , L. J. Allen[3], D. L. Irving[1], and J. M. LeBeau[1]

[1]*Department of Materials Science and Engineering, North Carolina State University, Raleigh NC 27695, USA*
[2]*School of Physics and Astronomy, Monash University, Victoria 3800, Australia*
[3]*School of Physics, University of Melbourne, Parkville, Victoria 3010, Australia*
[*]*jhdycus@ncsu.edu*

Accurate crystallographic determination is important for insight into material behavior. X-ray and neutron based diffraction methods, for example, provide excellent precision and accuracy, but are limited by poor spatial sensitivity. In contrast, scanning transmission electron microscopy (STEM) can directly probe real space crystallography with atomic resolution. Although STEM has advanced significantly in recent years, accurate distance information has remained elusive due to specimen drift and scan system distortion.

We will demonstrate that revolving STEM (RevSTEM) enables direct, accurate and precise length measurements at the unit-cell scale [1]. By measuring distortion from a set of images acquired with rotated scan coordinates, we correct the effects of drift from every frame that are then averaged to provide the representation of the sample. The final image has enhanced signal to noise, which enables precise determination of atom column positions. Using silicon as a calibration standard, we remove residual distortion introduced by the scan system to obtain an accurate and precise distance scale. This calibration is then applied to subsequent imaging of other samples. As a test of accuracy, we will show that lattice parameters of pure Bi2Te3 and Bi2Se3 obtained from STEM match those from X-ray powder diffraction to within 0.1 %.

Further, we will demonstrate that the approach can determine alloy composition by applying Vegard's Law to the measured lattice parameters using a Bi2Te3-Bi2Se3 alloy. We will also discuss the atomic structure employing atomic resolution chemical spectroscopy to determine site preference of the atomic species. We will show that Se resides almost exclusively within the middle layer,Te(2), of the Bi2Te3 quintuple - Te(1)-Bi-Te(2)-Bi-Te(1) - which are each held together by van der Waals forces. We will show that within the alloy unit-cell, bond lengths vary between those of the pure samples, as expected. The van der Waals gap, however, is found to change anomalously by increasing relative to the pure compounds. This behavior is shown to be caused by charge redistribution within upon incorporation of alloying elements. Finally, we will discuss the general applicability of the approach to other materials systems.

[1] Sang, X. and LeBeau, J.M. . Revolving scanning transmission electron microscopy: Correcting sample drift distortion without prior knowledge. **2014** Ultramicroscopy 138:28–35.

Chemically-induced Jahn-Teller ordering on manganite surfaces

Zheng Gai[1], Wenzhi Lin[1], J. D. Burton[2], Evgeny Y. Tsymbal[2], Stephen Jesse[1], Sergei V. Kalinin[1], and Arthur P. Baddorf[1]

[1] Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

[2] Nebraska Center for Materials and Nanoscience and Department of Physics and Astronomy, University of Nebraska Lincoln, Lincoln, NE 68588, USA

Physical and electrochemical phenomena at the surfaces of transition metal oxides and their coupling to local functionality remains one of the enigmas of condensed matter physics. Understanding the emergent physical phenomena at surfaces requires the capability to probe the local composition, map order parameter fields, and establish their coupling to electronic properties. We demonstrate that measuring the sub 30 pm displacements of atoms from high-symmetry positions in the atomically resolved scanning tunneling microscopy (STM) allows the physical order parameter fields to be visualized in real space on the single atom level. Here, this local crystallographic analysis is applied to the in-situ grown manganite surfaces. In particular, using direct bond-angle mapping we report direct observation of structural domains on manganite surfaces, and trace their origin to surface-chemistry-induced stabilization of ordered Jahn-Teller displacements. Density functional calculations provide insight into the intriguing interplay between the various degrees of freedom now resolved on the atomic level.

This research was conducted at the Center for Nanophase Materials Sciences, which is sponsored at Oak Ridge National Laboratory by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. Department of Energy.

Zheng Gai, Wenzhi Lin, JD Burton, K. Fuchigami, T. Z. Ward, P.C. Snijders, J. Shen, Stephen Jesse, Sergei V. Kalinin, and Arthur P. Baddorf, Nature Comm. 5, 4528, (2014)

Visualization of atomic-scale phenomena in superconductors

Presenter: Tom Berlijn

We developed a simple method [1] for calculating inhomogeneous, atomic-scale phenomena in superconductors which makes use of the wave function information traditionally discarded in the construction of tight-binding models used in the Bogoliubov–de Gennes equations. The method uses first-principles Wannier functions to visualize the effects of superconducting pairing on the distribution of electronic states over atoms within a crystal unit cell. Local symmetries lower than the global lattice symmetry can thus be exhibited as well, rendering theoretical comparisons with scanning tunneling spectroscopy data much more useful. An application of the method [2] to Zn impurities in the cuprates puts earlier crude “filter” theories on a microscopic foundation and solves a long standing puzzle.

[1] P. Choubey, T. Berlijn, A. Kreisel, C. Cao, and P. J. Hirschfeld, Phys. Rev. B 90, 134520 (2014)

[2] A. Kreisel, P. Choubey, T. Berlijn, W. Ku, B. M. Andersen and P. J. Hirschfeld, arXiv 1407.1846 (accepted in Phys. Rev. Lett)

Medium-Range Structure of Zr-Cu-Al Bulk Metallic Glasses from Structural Refinement Based on Fluctuation Electron Microscopy

Jason J. Maldonis,[1,*] Pei Zhang,[1] Matt Besser,[2] Matt Kramer,[2] Paul M. Voyles[1]

[1]*Department of Materials Science and Engineering, University of Wisconsin, Madison, WI, USA*
[2]*Ames Laboratory (DOE), Ames, Iowa, USA and Department of Materials Science and Engineering, Iowa State University, Ames, Iowa, USA*
[*]*maldonis@wisc.edu*

Fully determining the structure of complex materials like glasses from straightforward inversion of experimental data is not typically possible. Advanced techniques like fluctuation electron microscopy add more information, but still do not sufficiently constrain the structure. We have developed an approach using hybrid reverse Monte Carlo (HRMC) structural optimization to combine multiple data sources including simulated total system energies and experimental data to generate realistic structural models. By combining FEM data and an empirical interatomic potential using HRMC, we aim to identify important links between structure and glass-forming ability. The FEM data constrain the medium-range order of the structure, and the potential constrains the short-range and chemical order. Analysis of the resulting structural models for two different compositions of Zr-Cu-Al bulk metallic glass using conventional techniques based on Voronoi polyhedra found two different nanoscale structure types, one icosahedral and the other more crystal-like. In $Zr_{50}Cu_{35}Al_{15}$, the poorer glass former, the crystal-like structure is more stable under annealing below the glass transition temperature, T_g, than in $Zr_{50}Cu_{45}Al_5$, a better glass former. We have also developed new model analysis techniques. One is based on visualization and filtering of the computed three-dimensional reciprocal space of the model and the other is based on connectivity graphs and clustering of similar Voronoi polyhedra in real space. Based on structures identified with these techniques, we conclude that achieving better glass forming ability in this alloy system may depend more on destabilizing crystal-like structures than enhancing non-crystalline ones.

This work was supported by the NSF (DMR-1205899 and CMMI-1232731, JJM, PZ, and PMV). The facilities and instrumentation for microscopy were supported by the University of Wisconsin Materials Research Science and Engineering Center (DMR-1121288). The synthesis of alloys was supported by U.S. DOE, Office of Basic Energy Sciences, through the Ames Laboratory (MFB and MJK), and Iowa State University under contract DE-AC02-07CH11358.

In situ TEM study on crystallization of NbO_2 thin film

Jiaming Zhang[1], Kate Norris[1], Katy Samuels[1], Ning Ge[1], Max Zhang[1], Joonsuk Park[2], Robert Sinclair[2], Gary Gibson[1], Zhiyong Li[1], R. Stanley Williams[1]

[1]*Hewlett Packard Labs,* [2]*Stanford University*

[*]*Jiaming.zhang@hp.com*

Metal oxides Resistive Random Access Memory (RRAM) is one of the most viable candidates for next generation nonvolatile memory applications, due to its fast switching, excellent retention, good scalability, low operation voltage and simple device structure. Niobium dioxide (NbO_2) is one of the transition metal oxides that exhibit current-controlled negative differential resistance, also referred as threshold switching, when used in two-terminal devices. This phenomenon is caused by a reversible insulator-to-metal phase transition, which is proposed to induce a conductive channel in the device that bridges the two electrodes.[1, 2] Previous study have shown NbO_2 undergoes a Metal-Insulator-Transition (MIT) at ~1081K and simultaneous structural transition from rutile to a distorted variant with a body-centered tetragonal lattice. By using transmission electron microscopy analysis, microstructural evolution such as crystal structure transition and chemistry and in the sputtered NbO_x film grown from RF magnetron sputtering can be investigated as function of temperature. Both ex situ and in situ annealing experiments were carried out in both NbO_2-based device in nanovia platform and 200nm thick blanked film. Our preliminary results have shown that the annealing behavior of NbO_2 (10nm thin film) in via device is quite different compared to the bulk NbO_2 film (200nm thick).

1. F. A. Chudnovskii, L. L. Odynets, A. L. Pergament and G. B. Stefanovich, Journal of Solid State Chemistry **122** (1), 95-99 (1996).
2. M. D. Pickett, G. Medeiros-Ribeiro and R. S. Williams, Nat Mater **12** (2), 114-117 (2013).

STEM Characterization for Oxygen Activities in Li-rich Layered Oxides for High Energy Li-ion Batteries

Minghao Zhang,[1] Bao Qiu,[2] Danna Qian,[1] Ying Shirley Meng[1,*]

[1] *Department of Nano Engineering, University of California San Diego, La Jolla, CA 92093, USA.*

[2] *Ningbo Institute of Materials Technology and Engineering (NIMTE), Chinese Academy of Sciences, Zhejiang 315201, P. R. China*

[*] *shirleymeng@ucsd.edu*

With the increasing demand of energy and power in electric vehicles (EVs), a high battery capacity together with high-rate and long-life performance must be achieved. In this regard, Li-rich layered oxides, which can deliver discharge capacity over 280 mAh g^{-1}, have been extensively studied as cathode materials. However, the huge initially irreversible capacity loss, poor rate capability and serious voltage decay have prevented Li-rich layered oxides from practical application. We have performed a novel effective and facile modification approach for Li-rich cathodes. STEM-EELS results reveal that oxygen vacancies were created on the particle surface after the modification. Combining with first principles calculations, we are able to demonstrate for the first time that the surface oxygen vacancies are highly correlated to the electrochemical performance of the Li-rich cathodes. The target material after the surface modification is able to deliver discharge capacity as high as 306 mAh g^{-1} with initial coulombic efficiency of 90.6%, and the modified material exhibits no obvious capacity decay and voltage degradation after 100 cycles at 0.1 C-rate.

Atomic Imaging and Oxide Physics via monte-carlo methods

Anthony G. Gianfrancesco,[1,2,3] Rama K. Vasudevan,[1,2] Alexander Tselev,[1,2]
Arthur P. Baddorf[1,2] and Sergei V. Kalinin[1,2,3]

[1]Center for Nanophase Materials Sciences and [2]Institute for Functional Imaging of Materials,
Oak Ridge National Laboratory
[3] UT/ORNL Bredesen Center, University of Tennessee

We have used *in-situ* scanning tunneling microscopy to visualize atomic-scale surface structure of a mixed-valence manganite $La_{5/8}Ca_{3/8}MnO_3$ films grown by pulsed laser deposition. Surface termination and chemical composition were identified *in-situ* with angle-resolved x-ray photoelectron spectroscopy (XPS). The spatial variation of the mixed terminated surface was studied through use of a sliding Fast Fourier Transform (FFT) filter on atomically resolved data, with the resulting dataset investigated by principal and independent component analyses. This method appears to be highly useful in detection of edge features, lattice types and their spatial variation across the surface. In parallel, we observe mound-like structures reflecting surface roughening at a mesoscopic length scales. These results can be interpreted as a strong influence of oxygen on the adatom mobility during growth. Using this information, a lattice-gas model was created to examine the diffusion of adatoms on the exposed surface of the sample. The Ehrlich Schwoebel Barrier was found to have a height of approx. 0.05 - 0.17 eV, depending on temperature. These studies highlight the utility of atomically resolved information, sliding windows and Monte-Carlo methods in elucidating factors relevant to the growth of complex oxides.

Deep Data Discovery of Atomic Level Structure-Property Relationship in an Iron Superconductor

Anthony Giacomo Gianfrancesco,[1,2,3] Alex Belianinov,[1,2] Stephen Jesse,[1,2] Albina Borisevich,[1,4]Sergei V. Kalinin[1,2]

[1] Institute for Functional Imaging of Materials, Oak Ridge National Laboratory, Oak Ridge, TN 37831.
[2] The Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN 37831.
[3] UT/ORNL Bredesen Center, University of Tennessee, Knoxville, TN, 37916.
[4] Materials Sciences and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831.

Understanding the underlying physics of superconductive materials can be greatly facilitated by establishing a relationship between atomic structure and electronic properties at the nanoscale. Here, the structural data is obtained from high-resolution scanning tunneling microscopy (STM) imaging, whereas tunneling spectroscopy mapping provides information on the local electronic properties such as band structure and, in our case, the superconductor gap. We further explore local interactions with statistical methods such as Principle Component Analysis (PCA) and Bayesian Statistics to perform unsupervised classification and cross-correlative analysis of these data to establish the internal data structure and reveal the correlations between structure and functionality. In addition to this, other variables such as nearest neighbor distance, neighboring heights, unit cell area and volume were also compared to test the hypothesis of structural effects on electronic response. Then maps are created of each variable, through interpolation of the information, and compared using a 2-d correlation analysis to examine the largest correlations of structure to electronic response. In this approach, the statistically significant atomic configurations are established and further used as an input into first principle modelling. Thus, the determined electronic structure is then compared to the local tunneling spectra.

Rapid hyperspectral measurements and analysis for 3-D sub-nm resolution carrier profiling in semiconductors

Mark J. Hagmann,[1,*] Thomas E. Henage[1], and Dmitry A. Yarotski[2]

[1]*NewPath Research L.L.C., Salt Lake City, Utah*
[2]*Los Alamos National Laboratory, Los Alamos, New Mexico*
[*]*mhagmann@newpathresearch.com*

Acronyms: MFC-microwave frequency comb, SFCM-scanning frequency comb microscopy, SSRM-scanning spreading resistance microscopy, STM-scanning tunneling microscope

Introduction

The semiconductor industry needs new technology for sub-nm resolution carrier profiling as they progress to sub-10nm lithography nodes for new devices with improved performance [1]. SSRM is now used at these nodes because of its wide dynamic range, high sensitivity, and fine resolution [2]. However, SSRM is a destructive method, and cannot provide resolution finer than 1 nm [3]. Furthermore, SSRM requires cleaving or polishing for 3-D carrier profiling [2].

Our technology

We generate a unique MFC, with hundreds of measurable harmonics setting the present state-of-the-art in narrow linewidth, by focusing a mode-locked ultrafast laser on the tunneling junction of a STM [4]. It appears feasible to achieve sub-nm resolution carrier profiling by advanced data analytics with low-noise hyperspectral measurements of the attenuation of the MFC in the spreading impedance of a semiconductor at the tunneling junction [5].

Each pulse of the laser forms a spot of excess electrons (for p-type) or holes (for n-type) at the surface of the semiconductor, and this spot expands rapidly in a Coulomb explosion followed by scattering processes. This pulse is quickly attenuated and dispersed so the MFC is measured by a probe at the surface of the semiconductor within 10 μm of the tunneling junction.

This method is analogous to SSRM, but has the following advantages:

(1) Much finer resolution down to the 0.1 nm radius of the tunneling junction
(2) Non-destructive continuously-scanned measurements with no contact to the semiconductor
(3) Rapid hyperspectral measurements at high data rates to characterize the semiconductor
(4) Spot-size adjustable by a factor of 2 to change the sampled depth for 3-D profiling
(5) Low-noise measurements due to the state-of-the-art narrow linewidth in the MFC.
(6) Acquisition time reduced with MFC replacing dc tunneling current for feedback in scans.

References:

1. L.H. Vanamurthy, M. Huang, H. Bakhru, T. Furukawa, N. Berliner, J. Herman, Z. Zhu, P. Ronsheim, and B. Doris, J. Vac. Sci. Technol. A 31 (2013) 031403.
2. W. Vandervorst, A. Schulze, A.K. Kambham, J. Mody, M. Gilbert, and P. Eyben, Phys. Status Solidi C 11 (2014) 121-129.
3. K. Arstila, T. Hantschel, C. Demeulemeester, A. Moussa and W. Vandervorst, Microelectron. Eng. 86 (2009) 1222-1225.
4. M.J. Hagmann, F.S. Stenger, and D.A. Yarotski, J. Appl. Phys. 114 (2013) 223107.
5. M.J. Hagmann, P. Andrei, S. Pandey and A. Nahata, J. Vac. Sci. Technol. B 33 (2015) 02B109

Multimodal 3D mapping in photovoltaics using two-photon microscopy

Edward S. Barnard[1,2*], Benedikt Ursprung[1,2], Eric T. Hoke[2], Shaul Aloni[1], Craig H. Peters[2], Brian E. Hardin[2], P. James Schuck[1]

[1]*The Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley CA*
[2]*PLANT PV Inc. Oakland CA*
[*]*esbarnard@lbl.gov*

Traditional optical techniques are inherently surface sensitive due to light-absorption that predominately occurs near the surface, thus making determination of bulk or sub-surface properties of photovoltaic materials difficult. Here we show that two-photon absorption permits sub-surface optical excitation, enabling us to map optoelectronic properties in three-dimensions. For example, carrier lifetimes vary throughout a photovoltaic material due to defects such as grain boundaries, traps and surface defects that act as local recombination centers. Mapping and quantifying the effect of these defects is critical to improving the performance of devices where defects limit efficiencies. We will describe our custom microscope and data acquisition system that allows for the simultaneous acquisition of spectrally- and temporally-resolved photo-luminescence and photocurrent as a function of 3D-localized optical excitation. Additionally we will show how we visualize and model these 3D maps to extract relevant semiconductor properties such as bulk minority carrier lifetime and surface recombination velocity.

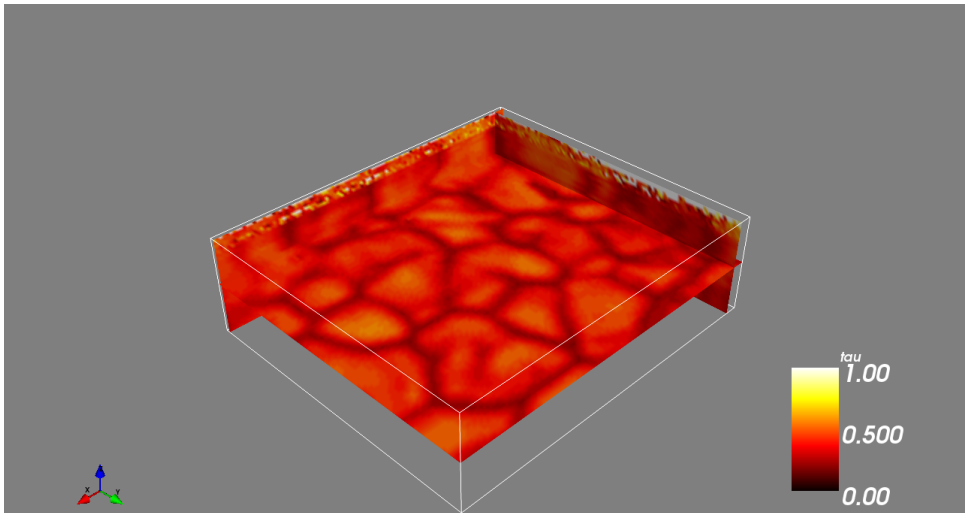


Figure 1. 3D map of carrier lifetime in a 20x30x8µm region of a polycrystalline CdTe photovoltaic thin film. This map shows long lifetime at center of grains and strong reductions in carrier lifetime near grain boundaries due to material defects.

Quantitative Separation of Ultrafast Transient Absorption Microscopy Data

Mary Jane Simpson[1], Benjamin Doughty[1], Bin Yang[2], Kai Xiao[2], Ying-Zhong Ma[1]*

1. Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN
2. Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN
**simpsonm@ornl.gov*

Transient absorption microscopy (TAM) is an emerging microscopy technique that leverages differences in electronic excited state dynamics as a source of contrast. An experimental TAM data set typically consists of a stack of transient absorption (TA) images at various delay times between the pump and probe pulses, and the TA response for each pixel is characteristic of contributing chemical or electronic photoexcitation species. For systems with high spatial heterogeneity due to, for instance, the presence of multiple distinct chemical species or different phases of a same material, the TAM images can be substantially complex and can exhibit vastly different features that each evolve on variable time scales. The challenge, therefore, is to reduce the data set into one or few two-dimensional images where the characteristics of distinct transient absorption responses can be represented by different colors. This would allow direct visualization of the microscopic distributions of chemical or excited state species, which has broad applications throughout materials and biological sciences.

This work focuses on distinguishing the spatial distribution of neutral excitons and free charge carriers in a polycrystalline thin film of methylammonium lead tri-iodide ($CH_3NH_3PbI_3$) interfaced with the commonly used electron acceptor [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) layer. Based on phasor and independent component analysis, we show that it is possible to identify spatial distributions of individual photoexcitation species. A combination of independent component analysis and linear unmixing analysis further allow us to estimate the relative contribution of each species to the TA signal across the entire image area. Our analysis further allows us to compare the strengths and weaknesses of these data analysis approaches for separating distinct photoexcitation species in semiconducting materials.

Work by Y.-Z. M. and M.J.S. was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division. B.D. Research sponsored by the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U. S. Department of Energy. Work by B.Y. and K.X. was conducted at the Center for Nanophase Materials Sciences (CNMS), which is a DOE Office of Science User Facility.

Data noise reduction in high spatial resolution spectral imaging of biomass

A. Passian[1,2,*], R. H. Farahi[1,2], V. V. Zaharov[1], B. H. Davison[1,2]

[1] *Oak Ridge National Laboratory, Oak Ridge, TN 37833, USA*
[2]*Department of Chemical and Biomolecular Engineering, University of Tennessee, Knoxville, TN 37996, USA*
[*]*passianan@ornl.gov*

Nondestructive nanoscale high spectral resolution physical and chemical imaging is a highly active area of material characterization. Ambient imaging however generates image data that is highly contaminated with noise originating from various sources such as the Brownian probe dynamics in atomic force microscopy, thermo-plasmonic response of nanoparticles, and photo-thermal properties of the material domains. We present an imaging modality that is capable of generating morphological and chemical information of plant cells accessible from cross sections of biomass at 10 nm length scales. The approach capitalizes on a form of infrared photo-acoustic spectroscopy in conjunction with nanomechanical frequency mixing to generate the sought data. High levels of Brownian noise are anticipated in the resulting images due to the elevated temperatures associated with the spectral response of the material. Karhunen-Loève transform is introduced to more specifically process the Brownian noise component of the spectral data. It is shown that inclusion of just a limited number of the leading eigenvalues of the covariance matrix allows the reconstruction of the spectral data that displays significantly improved spectral peak determination and can thus be proposed as a contrast enhancement approach.

1- V . V . Zaharov, R. H. Farahi, P . J. Snyder, B. H. Davison, and A. Passian “Karhunen Loeve treatment to remove noise and facilitate data analysis in sensing, spectroscopy and other applications,” Analyst 139(22), 5927-5935 (2014).

Extracting Physics through Deep Data Analysis

Evgheni Strelcov[1], Alexei Belianinov[1], Ying-Hui Hsieh[2], Stephen Jesse[1], Arthur Baddorf[1], Ying-Hao Chu[2,3] and Sergei V. Kalinin[1]*

1 The Institute for Functional Imaging of Materials and the Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, 37831
2 Department of Materials Science and Engineering, National Chiao Tung University, Hsinchu 30010, Taiwan
3 Institute of Physics, Academia Sinica, Taipei 105, Taiwan
*strelcove@ornl.gov

In recent decade the scientific community routinely produces hundreds of petabytes of data per year. The reason for this is that materials and phenomena in the world around us exist in an interweaved, entangled form, which gives rise to the complexity of the Universe and determines the size and complexity of the data that describes it. Decomposition of complex behavior is the key to understanding manifestations of Nature. However, tools to carry out this task are not readily available, and therefore, intricate systems often remain well-characterized experimentally, but still not well understood due to intricacy of the collected data.

The information hidden in the data can be explored using two complementary frameworks: *statistical* and *physical*. The former reveals the variability and abundance of different behaviors and internal correlations within the data set. The latter postulates the physical mechanisms underpinning the observed behaviors, or attempts to infer them based on comparison with similar systems. Multivariate statistical methods used in data mining allow for extraction of the statistical information. In order to go deeper and also extract the physical information, more sophisticated mathematical tools are required, tools that are based on physical models and incorporate physical constraints to the outcome of the statistical analysis.

One such approach – Bayesian linear unmixing (BLU) – has been recently developed by Dobigeon et al. This algorithm treats experimental data as a linear combination of a specified number of behaviors (e.g., spectra) and endeavors to unmix them into pure components with respective abundance maps. BLU features the abundance nonnegativity and sum to one constraint, which ensure that the resultant maps and components are physically meaningful (e.g., intensity of the spectral signal is nonnegative at all energies), something that a number of multivariate methods explicitly lack. In our work BLU was applied to SPM voltage spectroscopy data collected from a two-materials nanocomposite. Analysis showed that both material components and their interface can be identified and that 4 different conductive behaviors corresponding to two conduction mechanisms (Schottky tunneling and Ohmic transport) are responsible for the diversity in the raw data. Unlike the common interpretation of the optical or electron spectroscopy data, which is performed based on *a priori* known spectra of pure chemical elements/compounds, the individual conductive behaviors of the nanocomposite were not known, but found in the process of analysis. The fact that the chemical composition and physical mechanisms became identifiable in these two examples via BLU highlights its usefulness and destines its future applicability. Comparison of conductive behaviors at different oxygen partial pressures (in ambient and in ultra-high vacuum), shed light onto the complexity of their control via local oxygen vacancy concentrations.

'This research was conducted at the Center for Nanophase Materials Sciences, which is sponsored at Oak Ridge National Laboratory by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. Department of Energy.'

Big-Data Approaches to Analyzing Piezoresponse Force Microscopy/Spectroscopy: Unearthing New Physics

Rama K. Vasudevan,[1,2] Shujun Zhang,[3] M. Baris Okatan,[1,2] Stephen Jesse,[1,2]
Nazanin Bassiri-Gharb[4,5] and Sergei V. Kalinin[1,2]

[1]Center for Nanophase Materials Sciences and [2]Institute for Functional Imaging of Materials,
Oak Ridge National Laboratory
[3]Department of Materials Science and Engineering, Pennsylvania State University
[4]School of Materials Science and Engineering and [5]G.W. Woodruff School of Mechanical Engineering,
Georgia Institute of Technology

Piezoresponse force microscopy (PFM) is a well-known, widely used and powerful scanning probe technique that offers unparalleled resolution for exploring electromechanical behavior on the nanoscale in systems including oxides, nitrides, and biological systems. More recently, the proliferation of spectroscopic modes that take advantage of PFM and couple it with studies of piezoresponse as a function of voltage, pulse width, time and cycling history, in conjunction with band-excitation approaches for partial and full spectrum acquisition have resulted in enormous increases in file size, that can be on the order of ~20GB for single spectroscopic experiments (see Fig. 1a below). We discuss the physical requirements that necessitate these high dimensional measurements, and review their developments over the last several years. Yet, the visualization and rigorous analysis of the large, multidimensional PFM datasets remains in its infancy. Here, we highlight an example of a relaxation PFM mode applied to a ferroelectric relaxor system. The relaxation response is measured as a function of voltage amplitude and polarity, yielding 10 distinct relaxation curves at every (x,y) point in the measurement grid. The mean response is shown in Fig. 1b below, and indicates large relaxation for high bias values. Through use of independent component analysis of the data, we show that the relaxation consists of an exponential component, along with a separate 'disorder' component. The strength of the disorder component appears to be inversely proportional to the piezoelectric activity at that (x,y) location, providing a clue as to the underlying physics of the system, which, in this case is linked to relative ease of a field-induced phase transition.

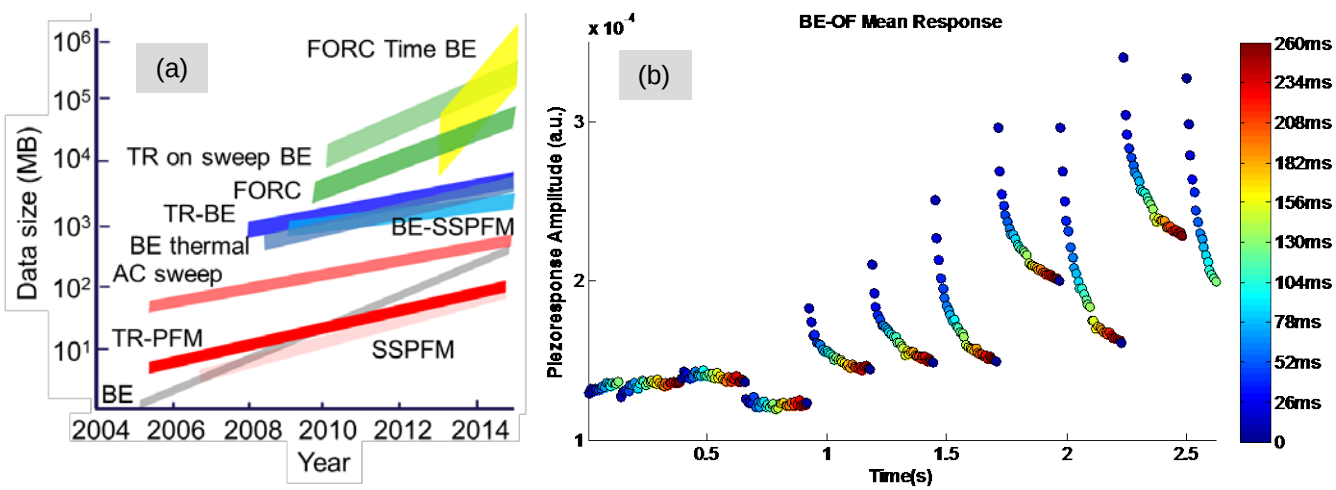


Fig 1: (a) Growth in size of data files in band-excitation spectroscopy in the last decade. **(b)** Average PFM response from a relaxation BE experiment on a ferroelectric relaxor single crystal sample.

Full information acquisition and analysis of surface diffraction data during oxide growth

Rama K. Vasudevan,[1,2] Alexander Tselev,[1,2] Anthony G. Gianfrancesco,[1,2,3]
Arthur P. Baddorf[1,2] and Sergei V. Kalinin[1,2,3]

[1]Center for Nanophase Materials Sciences and [2]Institute for Functional Imaging of Materials,
Oak Ridge National Laboratory
[3] UT/ORNL Bredesen Center, University of Tennessee

Reflection high energy electron diffraction (RHEED) has become an indispensable tool for *in-situ* monitoring of the growth of thin films and nanostructures by molecular beam epitaxy or pulsed laser deposition. The RHEED image sequence data can contain information pertaining to dynamic growth parameters (such as growth rate) as well as static and quasi-static information relating to surface roughness, characteristic size of growth islands, island or step density, surface reconstructions and others. However, much of this information is discarded or rarely analyzed, and typically only specular reflection variation over time is considered, largely as a result of sub-optimal geometry of RHEED, and the dynamic nature of the scattering that makes development of necessary microscopic models difficult. Here, we attempt to overcome this obstacle through both full information capture of the RHEED image sequence, and subsequent analysis by a variety of statistical algorithms, including principal component analysis, k-means clustering, as well as Fourier methods. The principal component analysis is useful in isolating 2D growth modes from 3D growth modes, and also can be used to negate the effects of a thermal drift in the system. K-means clustering indicates the onset of a clear transition from a disordered surface to a more ordered surface, about half way through the deposition of the epitaxial film. Interestingly, Fourier methods show that the third harmonic is largest in intensity at reflections typically associated with (undesirable) 3D growth. These studies suggest the potential for use of these methods *in-operando*, to predict final film quality, and may allow for improved control over film growth processes. Moreover, such studies highlight that "big-data diffraction" can be a key growth area in unearthing material physics.

A HIERARCHICAL MODEL REDUCTION ALGORITHM FOR NEUTRON SCATTERING OPTIMIZATION PROBLEM

FENG BAO , RICHARD ARCHIBALD , DIPANSHU BANSAL , AND OLIVIER DELAIRE

Abstract. The goal of a neutron experiment that uses neutron scattering is to solve for the scattering function, where final measured intensity of neutron scattering is represented as a high-dimensional convolution of the resolution and scattering functions. Determining the scattering function, given the measured intensity of neutron scattering, is a challenging problem because of the dimension of the convolution and the number of unknowns in neutron scattering experiments. In this presentation, we introduce a physical structure based model reduction method which searches for the global optimal of the scattering function hierarchically.

Poster Withdrawn

Atom by atom 3D chemical imaging using atom probe tomography

Baishakhi Mazumder* and Jonathan Poplawsky
Center for Nanophase Material Sciences, Oak Ridge National Laboratory, Oak Ridge, TN
**mazumderb@ornl.gov*

Atom probe tomography (APT) is a nano-analytical tool used to analyze materials at the atomic scale[1]. An APT setup integrates a field-ion microscope with a 2D spatially sensitive time-of-flight mass spectrometer. Based on the field evaporation principle, atoms are removed one-be-one, which provides the ability for 3D elemental characterization of the solid. The mass-to-charge states of individual field-evaporated ions with their *x*-, *y*-, and *z*-coordinates are determined with up to subnanometer resolution. This technique is capable of collecting large data sets from hundreds of millions of atoms in a short period of time. These data sets are reconstructed and analyzed with sophisticated software programs. In addition to visualization capabilities, many quantitative capabilities are available such as interface abruptness, roughness, particle size distribution, proximity histogram analysis etc. Data from APT can be integrated with other techniques such as scanning transmission electron microscopy (STEM), electron beam induced current (EBIC) and electron energy loss spectroscopy (EELS).

This presentation will deal with big data sets collected from two different semiconducting materials using the above mentioned characterization techniques. The first part will discuss about the data generated and assimilated from a InGaN/GaN quantum well (QW) LED structure. The analysis provides valuable information about In fluctuation within the QWs that impact the transport behavior in LEDs which governs the turn-on voltage of these devices. The second part will discuss an important and fundamental solar cell material problem analyzed and solved with the help of STEM-EBIC and APT. CdTe is an important solar material, which requires a post-deposition Cl diffusion step for activation. This crucial activation step improves the solar cell efficiency by more than an order of magnitude. The aforementioned experimental techniques have been integrated to show the nanoscale changes that improve the device efficiency after this treatment. The results using these techniques include Cl doping profiles at grain boundaries and interfaces that are responsible for inverting these regions n-type, and creating internal electric fields that aide in increasing photo-induced currents.

Ref:
1] T.F. Kelly and M.K. Miller, Rev. Sc. Instrum. 78 (2007) 031101
2]P. Bas, A. Bostel, B. Deconihout and D. Blavette, Appl. Surf. Sc. 87 (1995) 298

Acknowledgement: This research was supported by ORNL's Center for Nanophase Materials Sciences (CNMS), which is a DOE Office of Science User Facility.

Statistical Inference Techniques for Integrating Neutron Scattering Experiments with Molecular Dynamics Simulations to Characterize Intrinsically Disordered Protein Ensembles

Arvind Ramanathan,[1*] Christopher B. Stanley,[2*] and Chakra S. Chennubhotla, [3*]

[1]*Computational Science and Engineering Division, Health Data Sciences Institute, Oak Ridge National Laboratory, Oak Ridge, TN 37831*

[2]*Biology and Soft Matter Division Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831*

3Department of Computational and Systems Biology, University of Pittsburgh, Pittsburgh, PA 15260

[*]*{ramanathana@ornl.gov, stanleycb@ornl.gov, chakracs@pitt.edu}*

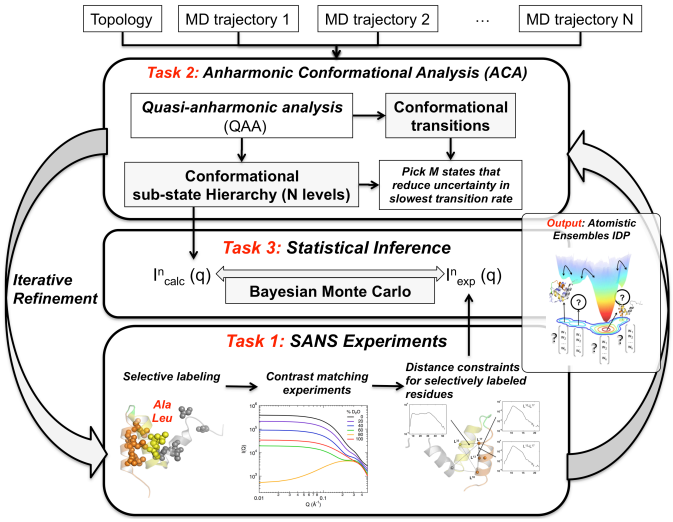


Figure 1: Statistical inference techniques for integrating neutron scattering experiments with molecular dynamics simulations.

Elucidating structural details of protein conformations and its relation to protein function is a forefront area in structural biology. In this project, we have investigated an intrinsically disordered protein (IDP), implicated in acute myeloid/lymphatic leukemia (AML/ALL), nuclear co- activator binding domain (NCBD), which has the propensity to adopt extended conformations in unbound form and undergoes synergistic folding with substrate specific conformations when bound. Since IDPs like NCBD are highly flexible, their full conformational range is not observable by any one structure determination technique. Therefore, we have developed a novel, integrated experimental and computational technique to elicit high-resolution structural details of such IDP ensembles. Specifically, we have (1) designed methods to prepare amino-acid-type selectively deuterated NCBD and utilize SANS contrast variation techniques to refine high-resolution conformational ensembles; (2) built parallel ensemble simulation strategies on heterogeneous computer architectures to generate millisecond timescale atomistic simulations; and (3) developed Bayesian inference techniques for statistical characterization of IDP conformational ensembles. The integrated approach enables us to accurately understand, simulate and predict how NCBD binds to its targets under physiological conditions. The integrated framework proposed here has broader applications for SANS analysis of flexible and order-disorder phenomena in polymer and materials science research. ***Funding***: This project is funded by ORNL SEED 7278.

Generations of spatial constraints for electron tomographic reconstruction

Suhas Sreehari[1,*], S. V. Venkatakrishnan[2], Lawrence F. Drummy[3], Jeffrey P. Simmons[3], and Charles A. Bouman[1].

[1.] *Electrical and Computer Engineering, Purdue University, West Lafayette, IN.*
[2.] *Lawrence Berkeley National Laboratory, Berkeley, CA.*
[3.] *Air Force Research Laboratory, Dayton, OH.*
* *E-mail: ssreehar@purdue.edu*

Electron tomography has been a widely used technique to image the 3D structure of nano-scale materials and biological structures alike. Transmission electron microscope (TEM) images tend to suffer from low signal-to-noise ratio (SNR) especially in the case of low-dose imaging, rendering electron tomography typically harder than other tomographic problems. Fortunately, in this area of nano-scale materials imaging, we are presented with many similar or identical structures, e.g. boundaries, crystallographic habits, intensity gradients. We have developed the "plug-and-play" technique that allows us to use denoising algorithms like non-local means (NLM) to capture such self-similarities in TEM images of materials to offset the low-SNR problem, and have produced much clearer and sharper tomographic inversions compared to emerging methods like q-generalized Gaussian Markov random field (qGGMRF) smoothing based iterative reconstruction and baseline methods like filtered backprojection (FBP).

Let us take a step back and briefly look at the evolution of tomographic reconstruction techniques – starting with filtered backprojection and quickly moving on to generations of spatial constraints that have greatly improved the quality of reconstruction.

Filtered backprojection is a process of inverting the tomographic projections acquired as the beams (electron beams in this case) pass through the object being imaged. This process assumes nothing about the underlying characteristics of the image, thereby rendering the reconstruction completely dependent on what little is acquired through the tomographic projections. This makes it extremely challenging to obtain good reconstructions when there is a limited set of tilts with low SNR. This explains the poor quality reconstruction seen in Figs. 1(b) and 2(b).

A major leap in tomographic reconstruction quality is made possible by recognizing that most images can be characterized by some sort of spatial dependence. When such spatial dependence can be formulated accurately, the resulting reconstruction can be of much higher quality than afforded by filtered backprojection. Markov random field (MRF) is one such way to formulate spatial constraints that forces each pixel/voxel to have an intensity that is close to intensities of pixels/voxels in a finite-sized neighboring window. The q-generalized Gaussian Markov random field is based on the simpler Markov random field, but is known to model the edges better. The application of qGGMRF as a spatial constraint is by means of "model-based iterative reconstruction" (MBIR). MBIR solves for the maximum *a* posteriori (MAP) estimate of the image given the projections; the MAP estimate is the optimization of a single cost function that is composed of a log likelihood term (based on the data) and a log prior probability term (based on the spatial constraints).

Below are two examples illustrating the vast improvement in reconstruction quality brought by qGGMRF as the prior model in the MBIR framework. Both are low-dose real datasets: the first one is a biological specimen dataset of ferritin molecules, while the second one is a materials dataset containing aluminum spheres.

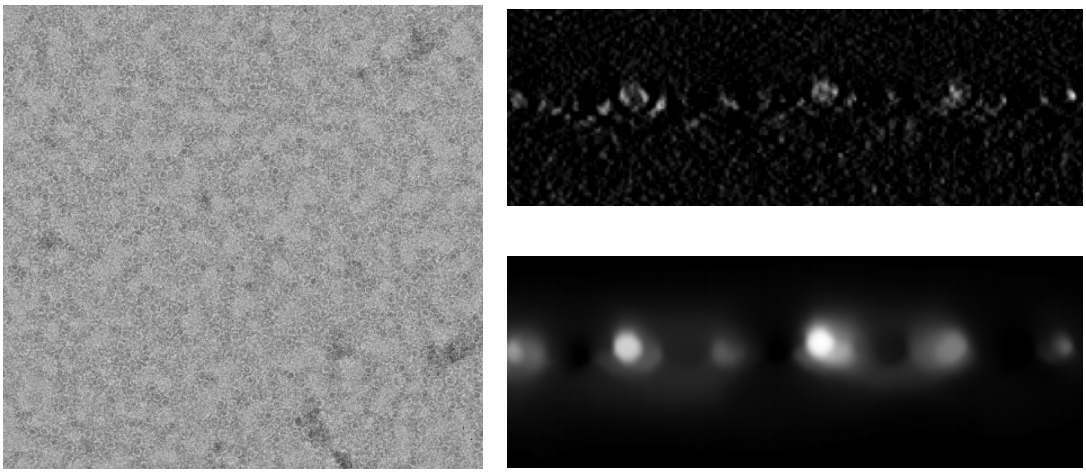


Figure 1. (Clockwise from left) (a) Contrast-stretched version of the 0° tilt of the ferritin structures bright field TEM dataset; (b) FBP reconstruction; (c) qGGMRF reconstruction – all of slice #1149 of the dataset.

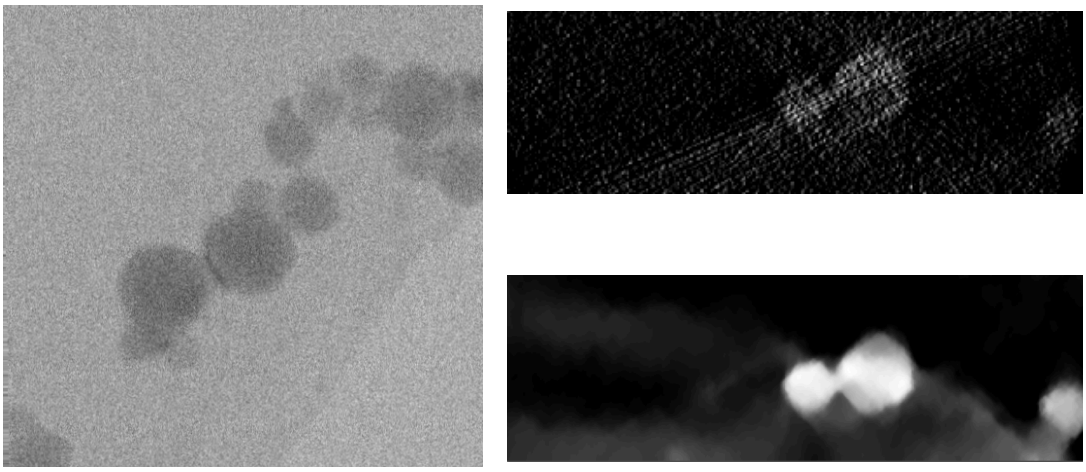


Figure 2. (Clockwise from left) (a) 0° tilt of the aluminum spheres bright field dataset; (b) FBP reconstruction; (c) qGGMRF reconstruction – all of slice #307 of the dataset.

The image similarities present in the TEM images of material structures and biological molecules are typically non-local in nature. qGGMRF being a local smoothing model is inadequate in capturing such non-local redundancies. A certain class of denoising algorithms, like BM3D and non-local means (NLM), naturally exploits long-range image similarities. However, typical implementations of MBIR formulate the log likelihood data term and the spatial constraint term in one tightly coupled cost function. Since these denoising algorithms are not explicitly formulated as solutions to optimization problems, it is unclear as to how to use them in the MBIR framework.

To address this concern, we developed what we call the "plug-and-play" framework [3] that allows us to plug any denoising operator into MBIR – hassle-free. The plug-and-play technique effectively decouples the log likelihood term from the log prior probability term. We then define two operators – the inversion operator (based just on the data) and the denoising operator (based just on the prior model) – that are applied alternatingly until convergence. We specifically use 3D non-local means (NLM) as the prior model in the plug-and-play framework [4], and we showcase high quality tomographic reconstructions of two real datasets of ferritin structures and aluminum spheres. In all the results, we observe that smear artifacts are visibly suppressed, and that edges are preserved. NLM-based reconstructions are also sharper than reconstructions that use the standard qGGMRF [1, 2] as prior model. Both NLM and qGGMRF-based reconstructions are significantly better than the filtered backprojection reconstruction.

The figure below is representative of the preliminary plug-and-play reconstruction results corresponding to the experiments of Figs. 1 and 2.

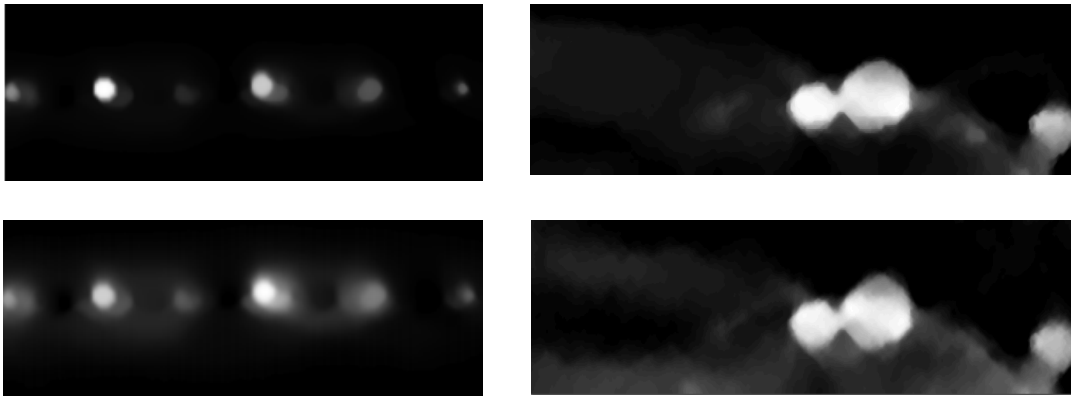


Figure 3. (Clockwise from top left) (a) 3D NLM reconstruction of slice #1149 of the ferritin dataset, (b) 3D NLM reconstruction of slice #307 of the aluminum spheres dataset, (c) qGGMRF reconstruction of slice #307 of the aluminum spheres dataset, and (d) qGGMRF reconstruction of slice #1149 of the ferritin dataset; (c) and (d) are repeated to provide immediate comparison.

NLM-based plug-and-play reconstructions, like in Fig. 3(b), show less smear artifacts than the qGGMRF reconstruction in Fig. 3(c). Fig. 3(a) highlights clearer reconstruction of the ferritin core than in Fig. 3(d). NLM denoising offers a practical way of capturing the redundancies shown in Figs. 1(a) and 2(a).

This marks the beginning of a string of research efforts that (a) fully exploit and refine the capabilities of the plug-and-play framework (including convergence properties), and (b) explore and extend modern denoising operators as "prior models" within the promising plug-and-play framework.

References:

[1] C. A. Bouman, "Model based image processing."

[2] S.V. Venkatakrishnan *et al*, "Model-Based Iterative Reconstruction for Bright-Field Electron Tomography," *accepted to the IEEE Transactions on Computational Imaging.*

[3] S. V. Venkatakrishnan *et al*, "Plug-and-play priors for model based reconstruction," *IEEE Global Conference on Signal and Information Processing,* 2013.

[4] S. Sreehari, S. V. Venkatakrishnan, L. F. Drummy, J. P. Simmons, and C. A. Bouman, "Advanced prior modeling for 3D bright field tomography," *To appear in the Proceedings of Computational Imaging XIII, SPIE,* February 2015.

Acknowledgment of support:

This work was supported by AFOSR/MURI grant #FA9550-12-1-0458, by UES Inc.

Title: 2D/3D Visualization and Analysis of Neutron Imaging Data

Jean Bilheux[1], Steven Hahn[1], Steve Miller[1], Hassina Bilheux[2]

[1] Neutron Data Analysis and Visualization Division
[2] Chemical and Engineering Materials Division
Oak Ridge National Laboratory, Oak Ridge, Tennessee

The High Flux Isotope Reactor (HFIR) CG1-D Neutron Imaging beam line studies a large variety of samples such as additive manufacturing (AM) pieces, vehicle components, plants, biological tissues and soils. A rotation stage allows the collection of radiographs, which are later used to produce computed tomographic (CT) data sets. Detailed analysis of these CT data requires visualization capability in 2 and 3 dimensions in order to identify features of interest.
The commercial visualization software VG Studio Max is currently being used. This software performs direct 3D comparisons of neutron CT with engineering drawings. Algorithms such as segmentation, data co-registration, porosity and length evaluation are also available analysis in this software package. However these CT data sets can be several tens of GBs and will be even larger with the next generation of instrumentation that will be built at the Spallation Neutron Source. The Versatile Neutron Imaging Instrument at the Spallation Neutron Source (VENUS) will generate large data sets that will require a parallel processing backend for responsive volumetric visualization. Thus, we are testing the ability of ParaView to provide such capability. We present an overview of the visualization and analysis of AM turbine blades obtained with VG Studio and ParaView.

4-D Data Acquisition in STEM: A Universal Detector for Sensitive Imaging

Miaofang Chi,[1,3] Stephen Jesse,[1,3] Albina Y. Borisevich,[2,3] Sergei V. Kalinin,[1,3] and Andrew R. Lupini[2,3]

[1] Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN 37831
[2] Materials Science & Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831
[3] Institute for Functional Imaging of Materials, Oak Ridge National Laboratory, Oak Ridge, TN 37831

Scanning Transmission Electron Microscopy (STEM), especially with aberration-correction, provides a wealth of information encoded in the phase, amplitude, angles, and radii of the scattered beams that can be used for exploring the nature of the materials. Detectors with varying geometries are being developed to generate images that offer differing sensitivities to specific materials characteristics. For example, with distinct inner and outer angles, annular detectors are routinely being used in STEM experiments: bright field (BF) for phase contrast imaging, annual bright field (ABF) for imaging light elements, low angle annular dark field (LAADF) for strain-field mapping, and high angle annular dark field (HAADF) for Z-contrast imaging. These detectors, which are based on a preconfigured hardware design, have intrinsic limitations in terms of simultaneous image acquisition, selectivity of scattering electrons, and signal rejection in imaging. Here we demonstrate the recent development of a 4D data acquisition and analysis system, which integrates a high-speed direct electron camera, a home-built external beam control and acquisition system, and a sub-Å electron probe. Such a configuration is validated to serve as a universal detector for STEM imaging. In fact, not only can conventional STEM images be acquired, but more importantly, new imaging approaches, which are relatively challenging or not achievable with current hardware configurations, can be investigated for improved efficiency. Furthermore, this method, combined with multivariate statistics, will provide enhanced opportunities to reveal "hidden" materials structural and physical characteristics, offering new perspectives in materials design and improvements.

Research supported by Oak Ridge National Laboratory's Center for Nanophase Materials Sciences (CNMS), which is an Office of Science User Facility (MC, SJ, SVK), and by the Materials Sciences and Engineering Division (ARL, AYB), Office of Basic Energy Sciences, U.S. DOE.

Current Efforts and Future Needs for Recording, Visualizing and Analyzing Multidimensional Datasets in Transmission Electron Microscopy

Colin Ophus,* Jim Ciston and Peter Ercius
The National Center for Electron Microscopy, Molecular Foundry, Lawrence Berkeley Natl. Lab., Berkeley, CA, USA
*clophus@lbl.gov

The original transmission electron microscopes (TEM) recorded imaging and diffraction data using photographic plates or film, and analysis was carried out by hand. When digital technology became widespread, the TEM community responded by scanning in film experiments and using computer analysis tools. Eventually CCD detector technology (with a scintillator to convert electrons to photons) supplanted film, allowing for all-digital recording and processing of microscopy data. CCD detectors also allowed video-rate time series data to be collected for *in-situ* experiments. Initially computer processing power could not keep up with the new higher data rates, but due to Moore's law this issue faded away. Recording three-dimensional datasets is now commonplace, having expanded from time series to include tilt series (tomography), through-focus series (to reconstruct the phase of electron plane waves), energy-filtered image stacks (spectroscopic techniques) and others.

However, another TEM detector technology revolution is currently underway: the widespread adoption of direct electron detectors (DEDs). Using CMOS technology to directly record incoming electrons, DEDs allow for much larger fields of view, and much higher recording rates. TEM researchers are now recording higher dimensional datasets, such as full 2D images of a diffracted electron probe over a 2D grid of probe positions (4D-STEM patterns). Figure 1 shows a 4D-STEM experiment, which could be used for example for ptychography. Other 4D datasets include tilt-defocus series for tomography, coherent diffraction imaging, virtual apertures, direct strain mapping and more. The element of time can also be added to the previous experiments, creating 5D datasets.

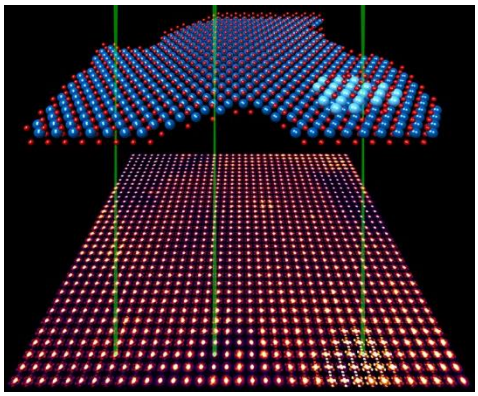


Figure 1 – 4D-STEM experiment on a MoS_2 2D crystal, 420 GB uncompressed.

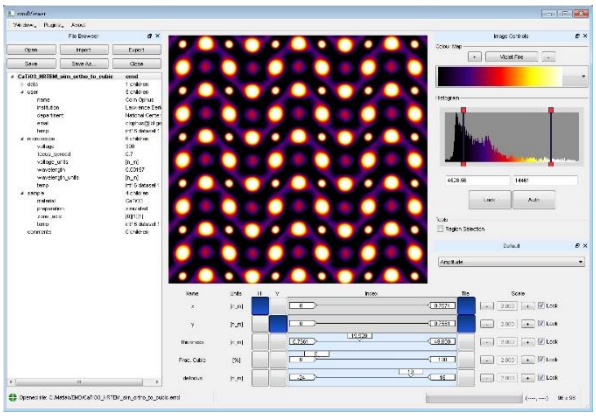


Figure 2 – Screenshot of the free high-dimensional data viewing software developed at NCEM called emdViewer, (http://emdatasets.lbl.gov). This software also reads proprietary TEM formats and we are developing a plug-in framework to allow fast processing of larger datasets.

This talk will discuss how a DoE user facility, the National Center for Electron Microscopy, is dealing with the new influx of data generated by DEDs. It will briefly cover automated methods for analyzing and compressing data, methods for visualizing this data, and new experimental possibilities. We will also talk about TEM image simulations, which have inherently very high dimensionality due to the ability to vary experimental parameters such as structure, composition, thickness, microscope optical configuration, electron scattering collection angle, sample tilt and other parameters. We have developed a data format called "Electron Microscopy Datasets" (.emd) for arbitrarily complex TEM data, based on the open source HDF format. We offer free software, shown in Figure 2, for slice visualization of high dimensionality data that can also read and convert proprietary vendor data formats into our open source format that maintains all relevant metadata.

Automatic image segmentation for 3D FIB/SEM reconstructions through the use of the trainable WEKA segmentation plug-in in Imagej

O. Dyck[1*], A. Papandrew[1]

[1] Chemical and Biomolecular Engineering, University of Tennessee, Knoxville

[*]odyck@utk.edu

3D object reconstruction in a FIB/SEM is performed by serially slicing away material with the focused ion beam (FIB) and imaging the new surfaces with the scanning electron microscope (SEM). Most modern FIB/SEMs are equipped to perform this task in a semi-automated fashion. Once the image stack is obtained, the challenge of performing the reconstruction remains. There are two critical steps: registration (aligning each image properly) and segmentation (determining the location of the various features). Modest sample drift can be corrected for during registration as well as the artifact caused by the angle between the sample face and SEM beam. Segmentation is the most difficult part of the reconstruction. While manual segmentation is possible, this is extremely time consuming. Various thresholding techniques exist where a specific gray value is chosen as the discriminator between two materials[1]. This can quickly and easily be applied to an image stack, however the fidelity of the reconstruction depends heavily upon this value and small variations can cause significant changes in the reconstructed structure[1]. In lieu of thresholding we investigated the ability of the trainable Weka segmentation plug-in[2] in Imagej to automatically segment image stacks for 3D reconstructions of the porous cathodes of solid acid fuel cells (Figure 1). We demonstrate that segmentation problems due to gray level gradients and overlapping gray level peaks in the image histogram can be alleviated. Moreover, we show that sections composed of both high and low intensity (i.e. unfilled pore space) can be fairly reliably detected and segmented which is not possible using the thresholding technique.

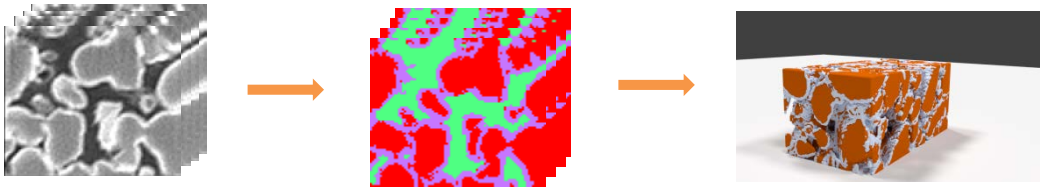


Fig 1. Schematic showing the segmentation steps. Left: a representative aligned image stack. Middle: Weka segmented image stack. Right: a rendering of the 3D reconstruction.

[1] J. Joos, T. Carraro, A. Weber, and E. Ivers-Tiffée. Journal of Power Sources, Volume 196, Issue 17, (2011), Pages 7302-7307

[2] M. Hall, E. Frank, G. Holmes, B. Pfahringer, P. Reutemann, I. H. Witten, SIGKDD Explorations, Volume 11, Issue 1 (2009)

Atomic Resolution Image as a Markov Random Field: Seeing through Noise

Artem Maksov[1], Oleg Ovchinnikov[2], Sergei Kalinin[3], Bobby G. Sumpter[4]

[1] *UT/ORNL Bredesen Center, University of Tennessee, Knoxville, TN 37996*
[2] *Vanderbilt University Department of Physics And Astronomy, Nashville, TN 37240*
[3] *The Institute for Functional Imaging of Materials, Oak Ridge National Laboratory, Oak Ridge, TN 37831*
[4] *Center for Nanophase Materials Science, and Computer Science and Mathematics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831*

**amaksov@vols.utk.edu*

Knowing the structure of a material is crucial to understanding its chemical and physical properties. Atomic resolution imaging allows great insight into structural and functional information about materials derived from identification of atomic positions and lattice symmetry. However, current methods of identifying atomic positions often rely on global properties of the image, such as crystal lattice parameters derived from Fourier space analysis, as well as image filtering methods, which alter the original data. While this works for special classes of images, often such methods lead to loss of local information, which is crucial to better understanding the properties of researched material. We propose a method of identifying atoms from locally available information by modeling the image as a Markov Random Field (MRF), which is often used for image segmentation in other fields. Our assumption is that atoms can be completely disordered; yet still identifiable on the image based just on local pixel-to-pixel contrast ratio transitions, thus obeying Markov property of being independent of other elements. Thus we can segment image into regions that contain atoms and regions that do not. In order to obtain optimal parameters for the pairwise potentials of the MRF model we performed testing on simulated images with varying noise levels. Additionally, the method was tested on a library of simulated and experimental images with known atomic positions. Our proposed approach allows robust atomic identification in high-noise images without introducing additional assumptions based on long-range structural parameters or global filtering methods.

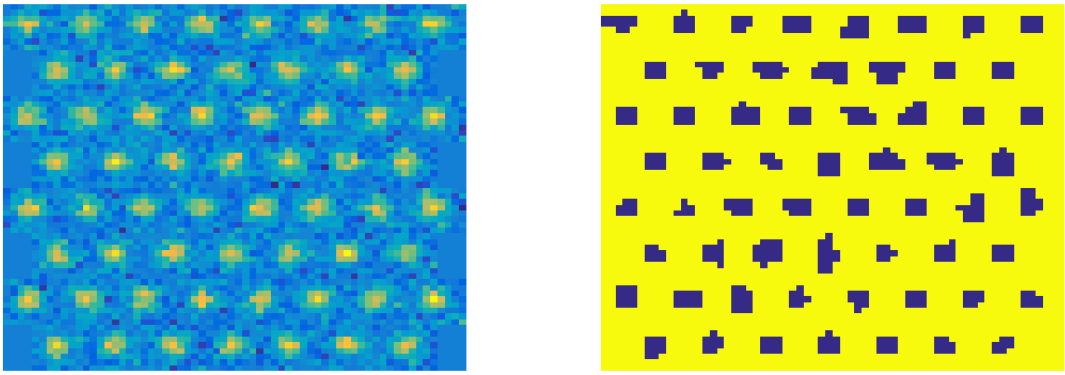


Figure 1: Simulated noisy atomic resolution image (left frame) and its MRF decoding (right frame)

Possibility of modulating a frequency comb with $4x10^9$ harmonics For extremely high data rates or secure communication

Thomas E. Henage[1,*], Joel D. Rasmussen[1], Mark J. Hagmann[1], and Dmitry A. Yarotski[2]

[1]*NewPath Research L.L.C., Salt Lake City, Utah*
[2]*Los Alamos National Laboratory, Los Alamos, New Mexico*
[*]*thenage@gmail.com*

Introduction

When terahertz radiation is generated with a mode-locked ultrafast laser the spectrum contains extremely large numbers of harmonics at integer multiples of the pulse repetition frequency of the laser. We have demonstrated this effect in two systems:
(1) Oscillating electron-hole dipoles formed at the surface of a semiconductor [1], and
(2) Optical rectification in a ZnTe(110) crystal where there are approximately $4x10^9$ harmonics from 500 Hz to 2 THz [2].

The large number of harmonics and their unusually small linewidth (< 1 Hz) may be understood because of the quasi-periodic excitation by the laser [3].

Numerical simulations of modulation and detection with the measured frequency comb

The following procedure was used in numerical simulations of modulation using data for the waveform that was measured using the second method.
(1) One or more harmonics were deleted after transforming to the frequency domain.
(2) Gaussian noise was added after transforming back to the time domain.
(3) The modified waveform was transformed to the frequency domain,
(4) Decoding was accomplished by taking the ratio of the modified and unmodified spectra.

Analytical simulation of modulation and detection with a finite sequence of pulses

The four-step procedure just described was also used in analytical simulations where the unmodulated waveform was defined as a finite sequence of pulses. All Fourier transforms were evaluated analytically, and again, decoding was accomplished in the presence of Gaussian noise.

Conclusions

(1) Coding by removing 1 or more harmonics, followed by decoding in the presence of Gaussian noise, was demonstrated both in numerical simulations based on the measured frequency comb and analytically with a finite sequence of pulses.
(2) Communication is possible with a series of finite sequences of N pulses where each sequence conveys different information. Increasing N reduces the linewidth to enable increasing the number of harmonics but reduces the symbol rate, thus providing the same data rate.
(3) In possible applications to secure communications the information could be coded on 4×10^9 harmonics, which is comparable with the number of leaves of grass on a golf course.

References:

1. M.J. Hagmann, S. Pandey, A. Nahata, A.J. Taylor and D.A. Yarotski, Appl. Phys. Lett. 101 (2012) 231102.
2. M.J. Hagmann, T.E. Henage, A.K. Azad, A.J. Taylor and D.A. Yarotski, Electron Lett. 23 (2013) 1459-1460.
3. M.J. Hagmann, F.S. Stenger and D.A. Yarotski, J. Appl. Phys. 114 (2013) 223107.

Approximate Bayesian Computation for Censored Data and its Application to Assess the Reliability of Nanocomponents

Nader Ebrahimi[1] and Kristin McCullough[2]

[1] Northern Illinois University, DeKalb, IL 60115
[2] Grand View University, Des Moines, IA 50316
* kmccullo4@gmail.com

Bayesian analysis focuses on obtaining the posterior distribution of an unknown parameter by updating the prior distribution of that parameter using the likelihood function, which expresses the probability of observed data under a particular statistical model. For complex models the likelihood function may be difficult to obtain or computationally costly to evaluate. Approximate Bayesian computation (ABC) refers to a family of algorithms that perform Bayesian analysis while bypassing the evaluation of the likelihood function. We propose using ABC to assess the reliability of nanocomponents in nanosystems. This research is motivated by the need to study the reliability of nanowires acting as the sensing elements in a specific hydrogen gas nanosensor when nanosensor lifetimes are censored. Censoring is a key analytical problem that occurs when some information about system failure, but not the exact failure time, is known. Since existing ABC algorithms are not applicable to censored data, we first present modifications to the existing ABC algorithms that will enable us to use ABC with censored data. Then we demonstrate how the modified algorithms can be used to infer nanocomponent reliability.

Multiscale Material Modeling with Mesoscopic Models

Pablo Seleson[1,*]

[1]*Oak Ridge National Laboratory*
One Bethel Valley Road, P.O. Box 2008, MS 6164, Oak Ridge, TN 37831
[*]*selesonpd@ornl.gov*

Multiscale material modeling has been recognized in recent years as an important research field towards accurate yet efficient simulation of complex systems. Large disparities in time and length scales, between different components of many systems of interest, make fully microscopic simulations of such systems unfeasible. Multiscale modeling offers, however, the possibility to bridge between scales by employing reduced-order methods and coupling algorithms. In this presentation, we will discuss the role of mesoscopic models in multiscale modeling and their connection to the imaging sciences.

Challenges in Physically Based Modeling and the Need for Data Driven Approach for Structure-Processing-Property Linkages in Materials

B. Radhakrishnan

Computer Science and Mathematics Division, ORNL

The talk will address a broad area in materials for which physically based modeling of processing-structure-property linkages becomes very challenging and will present the need for a concerted effort on data-driven modeling approaches where the model data is only but an important part of a multi-dimensional database including processing and characterization data. The example deals with modeling the phenomenon of nucleation in solid-state transformations. Nucleation is a complex event that depends on the presence of structural defects, thermodynamic driving force, interfacial energies and strain energy, in addition to other unknown factors. Currently, it is not possible to predict a priori what is the phase or the variant of the phase or a cluster that can nucleate in a material in its local microstructural environment under a set of given processing conditions. A possible modeling approach and the associated challenges and how imaging data can be used in conjunction with model to develop a predictive knowledge base for this important problem will be presented.

Denoising Training for Parameter Estimation via Bilevel Optimization

Richard C Barnard,[1,*] Karl Kunisch[2]

[1]*Oak Ridge National Laboratory*
[2]*Karl-Franzens-Universität Graz*
[*]*barnardrc@ornl.gov*

In the general frameworks of image denoising and control optimization, one often wants to minimize a function containing two kinds of terms: terms penalizing the output's performance or fidelity to measured data, and terms penalizing the structure of the input. These terms have to be assigned relative weights. We look at the following problem: given a set of noisy measurements, a physical law (in the form of a partial differential equation) giving the input-to-output process, and a set of input penalty functions, what is the best set of weights that lead to a denoising problem for reconstruction of the input? We present the relevant algorithms for several systems as well as some numerical results.

Scalable Machine Learning Approaches for Material Imaging

Ranga Raju Vatsavai,[1,2]

[1]*North Carolina State University*
[2]*Oak Ridge National Laboratory*
[*]*rvatsavai@acm.org*

Probabilistic graphical models provide a unifying framework for capturing complex dependencies among random variables. Markov Random Fields (MRFs), which are undirected graphical models, are widely used in computer vision and image analysis. These models can also be applied for nanoscale material imaging. Recent studies have applied MRFs for electron microscopy image segmentation to inverse problems in X-ray crystallography. Though versatile, MRFs are computationally expensive. In this poster, we present scalable implementations of MRFs using various computational models.

Anomaly Detection and Probabilistic Modeling for Image Data

Erik M. Ferragut,[1] Jason Laska,[1] Robert A. Bridges,[1]

[1]*Oak Ridge National Laboratory,*
Computational Sciences and Engineering Division

[*]*ferragutem@ornl.gov*

A number of data analytics fields have explored the need to discover unexpected anomalies within large data sets. Recent research in these areas has led to a number of remarkable improvements in the theory of anomaly detection. The new methods adopt a probabilistic modeling approach, which has several advantages. These include (1) detection of interesting features using purely unsupervised methods, (2) proven optimality in the Neyman-Pearson sense, (3) regulation of false positive rate, (4) meaningful comparisons of anomaly scores between disparate and heterogeneous distributions, (5) automatically providing context by using a constellation of detectors. This work has been applied primarily on streaming cyber and cyber-physical data and on medical records data. However, there is a significant potential for application of the same methods (with different probabilistic models) to materials image data. Possible paths forward for application of these methods to analysis of materials data will be discussed.

Fast and Scalable Image-Analysis 'Apps' on Big Data Architectures

Sreenivas R. Sukumar, [1,*] Seung-Hwan Lim[1], Chris Symons[1] and Bronson Messer[2]

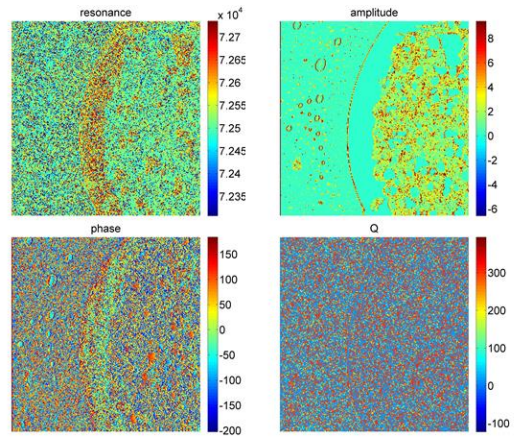
[1]*Computationsl Sciences and Engineering Division*
[2]*NationalCenter for Computational Sciences*
[*]*sukumarsr@ornl.gov*

This presentation will addresses the challenges in speeding-up and scaling-up algorithms such as Fourier, Wavelet, and Hilbert-Huang transforms and machine learning algorithms such as support vector machines, unsupervised clustering and association-rule mining in data-intensive compute environments. These algorithms are critical to the analysis of large datasets (particularly images) generated by scientific instruments and simulations [1,2]. The transform methods enable discovery of scientific laws and processes by revealing periodic, self-similar and multi-scale behaviors while machine learning algorithms learn from examples and features (that may be extracted using the transform methods) to predict patterns of future outcomes/events. Existing parallel implementation of the transform methods assume that the problem of interest is modeled as a system of theoretical-equations (algebraic, differential, integral, etc.) and scales well on HPC architectures. Traditional HPC architectures that handle such compute-intensive equations are an order of magnitude slower (if not worse) when a similar system has to be analyzed from sensory observations (e.g. microscopy, neutron scattering, etc.). This is because architectures that support computational solvers are not good data-intensive solvers. Data-intensive solvers require complex iterative back-and-forth intra-node disk-memory-compute interaction, and inter-node communication between distributed compute and storage resources. The patterns of interactions depend on the design of the algorithm and characteristics of the data.

Based on our experience, building high-performance analytical workflows of image-analysis algorithms has been daunting because (i) the Big Data hardware and software architecture landscape is constantly evolving, (ii) newer architectures impose new programming models, and (iii) data-parallel kernels of analysis algorithms and their performance facets on different architectures are poorly understood. To address this problem, we will present the idea of 'Mini-Apps' as an enabler for scientific discoveries. We will also present .preliminary results of scalable data-parallel kernels of popular image analysis algorithms using different programming models (e.g. MapReduce, Bulk-synchronous) along with performance benchmark results in diverse architectures (Rhea, Urika-XA, Amazon EC2, etc.) on a DOE-centric image-analysis use-case. One such experimental result of speed-up is illustrated in the Figure below.

We will conclude with ideas on how similar scale-up and speed-up can be achieved for custom domain-specific image analysis workflows – using the "Mini-Apps" being developed for Big Data architectures hosted within the Compute and Data Environment (CADES) infrastructure.

Instrument **Data**



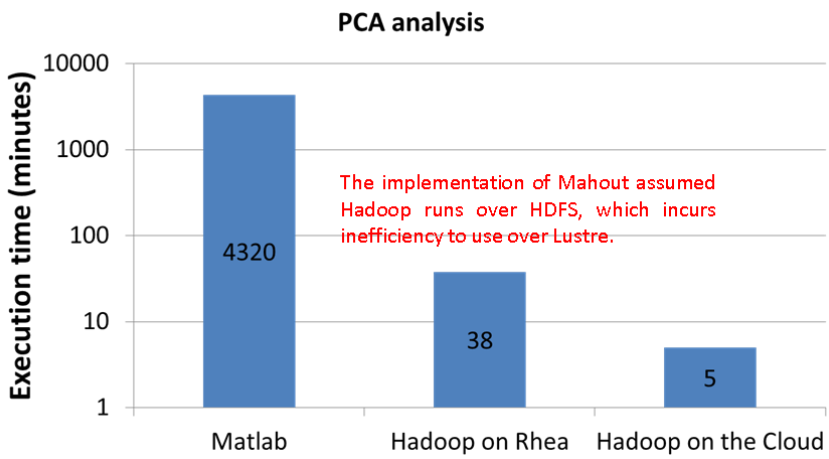


Figure 1: Results of running principal component analysis algorithm on a stream of 16,000 images obtained from a microscope at CNMS. We were able to improve the performance of the algorithm by several orders of magnitude by choosing the optimal architecture for the image analysis.

Acknowledgements: The authors thank Alex Belianinov, Stephen Jesse and Mahmut Okatan for sharing datasets and educating us on the methods used in image-analysis workflows at CNMS.

Use of BigData Analytics to Identify High-Temperature Coatings for Nuclear Cladding Materials

Jonathan Kenneth Bunn[1,2], Randy L. Fang[1,2], Mark R. Albing[1,2], Apurva Mehta[3], Matthew. J. Kramer[4], Matthew. F. Besser[4], Jason R. Hattrick-Simpers[1,2*]

[1] Department of Chemical Engineering, University of South Carolina Columbia, SC 29208

[2] SmartState Center for the Strategic Approaches to the Generation of Electricity, University of South Carolina Columbia, SC 29208

[3] Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Laboratory, Menlo Park, CA 94025

[4]Ames Laboratory, Iowa State University, Ames, IA 50011

[*]*simpers@cec.sc.edu*

High-temperature alloy coatings that can resist oxidation are urgently needed as nuclear cladding materials to mitigate the danger of hydrogen explosions during meltdown. Here we apply a combination of computationally guided materials synthesis, high-throughput structural characterization and big data analysis tools to investigate the feasibility of coatings from the Fe-Cr-Al alloy system. Composition-spread samples were synthesized to cover the region of the phase diagram previous bulk studies have identified as forming protective oxides. The metallurgical and oxide phase evolution were studied via *in situ* synchrotron x-ray diffraction at temperatures up to 690 K. A composition region with an Al concentration greater than 3.08 at%, and between 20.0 at% to 32.9 at% Cr showed the least overall oxide growth. Subsequently, a series of samples were deposited on stubs and their oxidation behavior at 1373 K was observed, several promising compositions were identified that withstood oxidation without failure for up to 6 hours.

Large-scale Fiber Tracking from Microscopic Composite Images

Youjie Zhou,[1] Hongkai Yu,[1] Jeff Simmons,[2] Yuewei Lin,[1] Yang Mi,[1] Song Wang[1,*]

[1]*Department of Computer Science & Engineering, University of South Carolina, SC*
[2]*Materials and Manufacturing Directorate, Air Force Research Labs, OH*
[*]*songwang@cec.sc.edu*

SiC/SiC continuously reinforced Ceramic Matrix Composites (CMC's) are being aggressively developed for advanced hypersonic applications. It is hypothesized that the lifetimes of mechanical systems such as SiC/SiC composites are controlled by flaws that occur in the microstructure. In order to test this hypothesis, it is necessary to characterize the microstructures developed, the flaws being characteristic of processing parameters. In practice, this often means reconstructing the fibers from serial section datasets and modeling the structures formed by the fibers. With conventional cross-correlation methods, we have been able to achieve accuracies in the low 90% range. By their nature, flaws are rare, which requires that the fibers be reconstructed with a much higher degree of accuracy. This work has a goal of developing tracking algorithms capable of achieving 99+% accuracy for samples containing on the order of 10,000 fibers.

To address this, we propose an algorithm that first detects the fiber cross sections on 2D serial section slices and builds a fiber correspondence between adjacent slices by constrained non-rigid registration. We then concatenate and smooth the inter-slice fiber correspondence using an online Bayesian filter, which generates a large set of the desired 3D fiber tracks. To verify the effectiveness of proposed algorithm, we compare its performance against several state-of-the-art multi-target tracking algorithms, including integer linear programming based tracking (DPNMS), motion dynamics based tracking (SMOT), continuous energy based tracking (CEM), and individual detection linking using the Viterbi algorithm (KTH).

For the quantitative performance evaluation, we constructed a large-scale microscopy image dataset (3,600 images with dimension 1292x968), on which more than 10,000 fibers were manually annotated as the ground truth. A comprehensive set of widely-used metrics are utilized for assessing the tracking performance, including the Multiple Object Tracking Precision (MOTP), the Multiple Object Tracking Accuracy (MOTA), and the average running time. We also study each tracking method’s performance when increasing the inter-slice distance – larger inter-slice distance means faster image acquisition and the capability to analyze larger material samples.

Selected results of tracking algorithms and quantitative comparisons of their accuracy for SiC/SiC CMC's will be presented. Proposed algorithm outperforms several comparison methods in terms of both tracking accuracy and time efficiency. We also found that tracking large-scale fibers from composite images is still a very challenging problem, especially with large inter-slice distance.

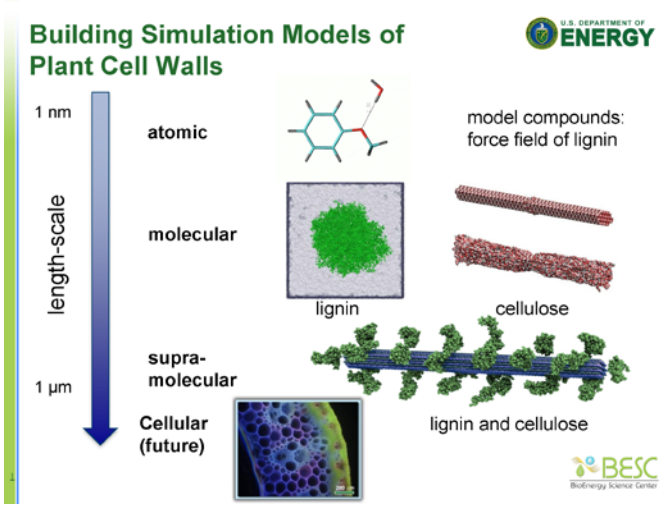
Integrated Methods for Imaging of Cellulose Composites

Barbara R. Evans[1,*], Riddhi Shah[2], Hugh O'Neil[2], Sai Venkatesh Pingali[2], Daisuke Sawada[2], Volker Urban[2], Loukas Petridis[3], Jeremy C. Smith[3], Arthur J. Ragauskas[3], and Brian Davison[3]

[1]Chemical Sciences Division; [2]Biology and Soft Matter Division; [3]Biosciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

[*]evansb@ornl.gov

Cellulose is an abundant and widely used biological polymer, most notably as a major component of lignocellulosic biomass produced by plants. The hierarchical assembly of individual glucan chains into a unique crystalline fibrous structure and its combination with other polymers to form both natural and synthetic composites is challenging to elucidate and visualize. Examination of cellulose composite structure at atomic, molecular, and cellular levels is complicated by chemical similarity of the component polymers. To distinguish component polymer interactions, we are using assembly of model composites that strive to emulate chemical and physical properties of lignocellulosic materials. We have developed methods for deuteration of bacterial cellulose and plant lignocelluloses to assist in elucidation of the molecular structure of cellulose and its composites. By isotopic labeling of both natural and synthetic composites with deuterium, individual components can be examined in the composite with FTIR, $^{1}H^{2}H$-NMR, and small angle neutron scattering. High-resolution microscopy can provide the surface and spatial dimensions to relate the data from these bulk methods. Combination of the resolution of atomic force microscopy with chemical force and other techniques can potentially provide nanoscale localization of chemical composition required to understand the properties of these complex materials. Utilizing these data to produce integrated images of composite hierarchical materials requires concomitant development of computer algorithms to analyze data sets. Combination of information from these complementary techniques will enable development of structural models by computational simulation.



References:

He et al. (2014) *Cellulose* 21(2), 927.

Langan et al., (2012) *Industrial Biotechnology* 8, 209–216.

Linder et al., (2013) *Biomacromolecules* 14, 3390–3398.

Acknowledgments: This research is supported by the U. S. Department of Energy, Office of Biological and Environmental Research, under contract SFA Biofuels FWP ERKP752.

Image Correlation of Scanning Probe, Electron and Fluorescence Microscopy Images for Advanced Materials in Energy

Dane W. de Quilettes, Jeff Harrison, Durmus Karatay, Sarah M. Vorpahl and David S. Ginger
University of Washington, Department of Chemistry and Department of Physics

Our research requires cross-correlating combinations of scanning probe microscopy with optical and electrical excitation to elucidate the nanostructural properties of materials for energy applications, including hybrid perovskites, kesterites, and polymer thin films semiconductors. We study the local functional properties of these materials using scanning-probe-based measurements of local carrier lifetime, photocurrent, surface potential, and ferroelectricity. We correlate these data with local optical spectroscopy, including photoluminescence lifetimes and spectra, in order to understand the microstructural factors affecting carrier recombination and photocurrent collection. Additionally, we use scanning electron microscopy and mass spectrometry techniques to characterize local elemental composition. We use unsupervised and supervised machine learning algorithms to understand correlations and causations between different material properties, as well as classification of microstructures found in these materials. By applying these methods, we seek to understand and elucidate underlying mechanisms for various electronic and photonic properties that are not readily understood from individual methods alone.

Crystal Structure Manipulation of ZnO particles using organic ligands: electrochemical and photoelectrochemical characterization and EXAFS studies for solar energy harvesting applications

Mitk'El B. Santiago-Berríos *,María de los A. Cepeda-Pérez, Astrid M. Rodríguez-Negrón, Natalia I. Román-Montalvo, Cristina M. Reyes-Marte, Svetlana Gelpí-Domínguez, Valerie Ann Carrasquillo, William A. Muñiz, Edgar J. Trujillo, Rahul Singhal† and Harry Rivera**

**Universidad Metropolitana; School of Science and Technology*
PO Box 21150, San Juan, PR 00928-1150
msantiago326@suagm.edu
† Central Connecticut State University
Department of Physics and Engineering Physics, New Britain, CT-06050
singhal@ccsu.edu
*** Inter American University of Puerto Rico*
Bayamón Campus, Carr 500 Dr John W. Harris, Bayamón, P. R. 00957

Zinc oxide (ZnO) is a white inorganic semiconductor that has been proposed for different applications, which include, but is not limit to, medicine, sensors, and solar energy harvesting applications. ZnO a wide gap semiconductor have been proposed for the construction of solar cells due to their high electron mobility and ease of the construction for a transparent electrode. Our group has synthesized different ZnO crystal structures by simple wet chemistry synthesis. We tested the effect of two different ligands (3-mercaptopropionic acid, and benzoic acid) during the ZnO synthesis. Both ligands are mild acids, which compete with the Lewis acid of the reaction, which is Zn^{2+}. The ZnO materials are characterized by different spectroscopic and analytical techniques such as X-Ray Powder Diffraction (XRPD), Scanning Electron Microscope (SEM), Energy Disperse Spectroscopy (EDS), UV-vis, and IR Spectroscopy. The functionalized materials were also characterized using EXAFS. Preliminary results shows evidence that addition of benzoic acid to zinc oxide particles induces the formation of zinc oxide rods. However, the materials functionalized with 3-mercaptopropionic acid have a film-like structure. These materials have demonstrated to form from rods to thin films, which allow to us to devise strategies for the construction of solar cells. This presentation will focus on the recent advances made in the Nanoscience Laboratory at Universidad Metropolitana.

Correlation of Structure and Chemical Environment of the Iron in Magnetosomes Residing in Magnetotactic Bacteria

Emre Firlar and Tanya Prozorov

Emergent Atomic and Magnetic Structures, Division of Materials Sciences and Engineering, Ames Laboratory, Ames, IA 50011, USA

*efirlar@ameslab.gov

Magnetotactic bacteria produce magnetite magnetosomes to orient themselves along the magnetic field on earth. Formation of these magnetosomes requires uptake of soluble iron and biomineralization in the intracellular membrane vesicles. While the crystal structure of magnetosome magnetite is well-established, far less is known about the chemical environment of the biomineral at the early stages of magnetosome formation. To this end, we employed the combination of electron diffraction and EELS analyses to probe the crystallinity and chemical bonding in the nascent magnetosome particles. Use of SAED analysis revealed that newly formed magnetosome particles with the mean size of 10 nm are mostly amorphous, whereas the fully grown magnetosomes of ~50 nm are unambiguously indexed to magnetite. We have also identified the narrow "magnetosome transition range" corresponding to the onset of crystal lattice formation. We utilized the peak fitting analysis of EEL spectra to monitor the evolution of chemical bonding in magnetosomes and correlated these findings with the emergence of crystalline lattice(**Figure 1**). Specifically, we interpreted the FeL_3 edge peaks in terms of the interplay between octahedral and tetrahedral iron coordination in growing magnetosome particles.

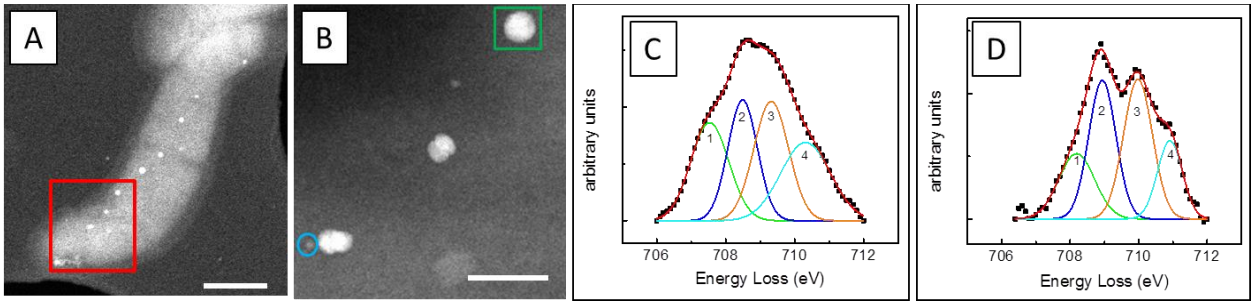


Figure 1- A) HAADF-STEM image of a bacterial cell obtained from the sample 30 minutes after induction. B) Magnified image of the area shown with red square in figure A. Fe L_3 core EEL spectra of the particles shown C) by the green square in figure B with size: 46.4 nm x 49.4 nm and D) by the blue circle in figure B inset with size: 9.5 nm x 10.9 nm. The ratio of the areas of peak 1 to peaks (2+3), corresponding to Fe^{2+} (octahedral) to Fe^{3+} (tetrahedral + octahedral) ratio, is around 1:2.1 for large particle and 1:3.3 for small particle. Scale bars: A: 500 nm, B: 100 nm.

Magnetosome Magnetite Biomineralization: Making the Sense of it All

Tanya Prozorov[1*], Taylor J. Woehl[1,2], Emre Firlar[1]

[1]*Emergent Atomic and Magnetic Structures, DMSE Ames Lab, Ames IA 50011*
[2]*Currently at NIST*
[*]*tprozoro@ameslab.gov*

Magnetotactic bacteria produce chains of magnetosomes, the intracellular nanometer-sized magnetic crystals surrounded by a thin phospholipid bilayer membrane. Magnetosomes exhibit nearly perfect crystal structures with narrow size distribution and consistent species-specific morphologies, leading to desirable magnetic properties. As a result, the magnetite biomineralization in these microrganisms is of fundamental interest to diverse disciplines, from biotechnology to astrobiology [1]. Current studies point to a number of steps involved in magnetosome growth, including cellular uptake of soluble iron, its complexation with membrane proteins, and nucleation and growth of a mature magnetosome. However, due to a dearth of *in vivo* observations of magnetosome growth, the molecular mechanism of biomineralization in magnetotactic bacteria and the magnetosome nucleation and growth process remains unclear. Recently, we demonstrated a correlative electron and fluorescence microscopy technique for imaging *live* magnetotactic bacteria in liquid with nanometer resolution using an environmental fluid cell, and identified imaging conditions under which the microorganisms did not sustain observable electron beam damage [2]. Current effort is focused on probing the magnetosome magnetite biomineralization both *in vivo* and in *vitro* by using the combination of correlative STEM-FM imaging, electron diffraction, and analytical spectroscopy.

[1] Prozorov, T.; Bazylinski, D. A.; Mallapragada, S.K.; Prozorov, R. "Novel Magnetic Nanomaterials Inspired by Magnetotactic Bacteria: Topical Review" *Mater. Sci. Eng. R.*, **2013**, *173*, 133-172.
[2] Woehl, T. J., Kashyap, S.; Perez-Gonzalez, T.; Faivre, D.; Trubytsyn, D. ; Bazylinski, A. D.; Prozorov, T. "Correlative Electron and Fluorescence Microscopy of Magnetotactic Bacteria in Liquid: Toward *In Vivo* Imaging" **2014**, *Sci. Rep.,* 4, 6854.

Driven Reaction Front Dynamics at Mineral-Aqueous Interfaces

Numan Laanait
Center for Nanophase Materials Sciences, Oak Ridge National Lab

Abstract:

Reaction-Diffusion processes are prevalent in nature. They permeate the non-equilibrium configurations of systems that are of interest to diverse disciplines such as chemical physics, biology, and geochemistry. Reaction-Diffusion processes span multiple spatiotemporal scales, requiring local and dynamical information on the behavior of the system, and precise experimental control over its thermodynamic state. Elucidating the dynamical and spatially resolved mechanisms that underlie the behavior of systems in non-equilibrium conditions is one of the biggest challenges facing the imaging community, for it necessitates a seamless integration between experimental probes, image analysis, and physical theory/modeling. Concurrently, it is the next frontier in imaging science.

In this talk, I will show that modern structural beam-probes such as in situ electron and X-ray microscopes, with their exquisite spatial and spectral properties, offer unprecedented opportunities to simultaneously *observe and control* reaction-diffusion phenomena. In particular, I describe a new approach to controllably drive the dissolution of the calcium carbonate-aqueous interface by the intense radiation fields of synchrotron X-rays, and simultaneously probe the dynamics of propagating surface reaction fronts using surface X-ray microscopy. Evolving structures of the reactive surface are related to the time-dependent solution composition by a reaction kinetics model. Reaction front instabilities are observed at extreme disequilibria with front velocities of tens of nanometers per second, and are identified as a dynamical signature of transport-limited reaction dynamics. The presented approach enables new opportunities in investigating the reactive behavior of materials under extreme environmental conditions; conditions that are simulated and finely tuned by high-energy microscopic beams.

Humidity and Temperature effects on piezoresponse of $Pb(Fe_{0.5}Nb_{0.5})O_3$

Dhiren K. Pradhan[1], Rama K. Vasudevan[2], Evgheni Strelcov[2] ,Venkata S. Puli[3], Sergei V. Kalinin[2], Ram S. Katiyar[1]

[1]Department of Physics and Institute of Functional Nanomaterials, University of Puerto Rico, San Juan-00936, PR, USA

[2]Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

[3]Department of Physics and Engineering Physics, Tulane University, New Orleans, Louisiana, LA 70118

Piezoresponse Force Microscopy (PFM) has emerged as a powerful tool for experimental investigations of ferroelectric materials. In the imaging mode, PFM allows visualization of static domain structures with nanometer spatial resolution. Application of a sufficiently large voltage through a conductive scanning probe microscopy (SPM) tip can induce local polarization switching and can be extended for creation of tailored domain structures and ferroelectric data storage. Finally, acquisition of the piezoresponse signals during polarization reversal allows measurement of local hysteresis loops, which can be used for characterization of the switching process in the nanoscale area in the vicinity of the tip. The broad application of PFM for probing domain structures and polarization reversal in ferroelectrics demands deep understanding of the basic mechanisms involved. PFN ($Pb(Fe_{0.5}Nb_{0.5})O_3$) is a well known multiferroic material with high dielectric constant, very good ferroelectric properties, low dielectric loss and low magnetization value at room temperature.PFN thin films were grown by optimized pulsed laser deposition (PLD) .The thicknesses of PFN thin films are found to be ~ 70 nm. The highly c-axis oriented growth containing only (00l) diffraction peaks of PFN films along with in plane epitaxial relationship were confirmed by high resolution X-ray diffraction measurements. From the atomic force micrographs it was observed that all the films were densely packed, smooth, free from microcrack and particulates with uniform grain-size distributions. The existence of ferroelctricity confirmed by band excitation PFM. The local polarization reversal by an electric field produced by a conductive SPM tip as a function of the relative humidity and temperature in an SPM chamber has been studied. The decrease of piezoresponse is observed with increase of relative humidity and temperature. The observed phenomena are attributed to the existence of a water meniscus in the vicinity of the tip–surface contact. The ferroelectric phase transition is also probed by the temperature dependence of piezoresponse studies. In addition to the temperature

dependence of piezoresponse studies, the phase transition is also confirmed by temperature dependent dielectric spectra. Detailed studies on effect of humidity and temperature on piezoresponse of PFN will be discussed in the meeting.

Participants

Rick Archibald
ORNL
archibaldrk@ornl.gov

Arthur Baddorf
ORNL
baddorfap@ornl.gov

Feng Bao
ORNL
baof@ornl.gov

Eduard Barnard
Lawrence Berkeley National Lab
esbarnard@lbl.gov

Richard Barnard
ORNL
barnardrc@ornl.gov

Alex Belianinov
ORNL
ba8@ornl.gov

Tom Berlijn
ORNL
tberlijn@gmail.com

Jean Bilheux
ORNL
bilheuxjm@ornl.gov

Dan Bocek
Asylum Research, Oxford Instruments
dan.bocek@oxinst.com

Albina Borisevich
ORNL
albinab@ornl.gov

Nigel Browning
Pacific Northwest National Laboratory
nigel.browning@pnnl.gov

Jihua Chen
ORNL
chenj1@ornl.gov

Hans Christen
ORNL
christenhm@ornl.gov

Jim Ciston
Lawrence Berkeley National Lab
jciston@lbl.gov

Valentino Cooper
ORNL
coopervr@ornl.gov

Mary Comer
Purdue University
comerm@purdue.edu

Cory Czarnik
Gatan
cczarnik@gatan.con

Eduardo D'Azevedo
ORNL
dazevedoef@ornl.gov

Farcsco De Carlo
Argonne National Laboratory
decarlo@anl.gov

Michael Demkowicz
MIT
demkowicz@mit.edu

Dane deQuilettes
University of Washington
dwd2@uw.edu

Peter Doak
ORNL

doakpw@ornl.gov

Benjamin Doughty
ORNL
doughtybl@ornl.gov

Larry Drummy
Air Force Research Laboratory
lawrence.drummy.1@us.af.mil

Margaret Drouhard
University of Tennessee
Meg.drouhard@utk.edu

Joseph Dycus
North Carolina State University
jhdycus@ncsu.edu

Jacob Eapen
North Carolina State University
jeapen@ncsu.edu

Barbara Evans
ORNL
evansb@ornl.gov

Erik Ferragut
ORNL
efn@ornl.gov

Emre Firlar
ScienAcmees Laboratory
efirlar@ameslab.gov

Michael Fitzsimmons
ORNL
fitzsimmonsm@ornl.gov

Miguel Fuentes-Cabrera
ORNL
fuentescabma@ornl.gov

Zheng Gai
ORNL
zgai@yahoo.com

Panchapakesan Ganesh
ORNL
ganeshp@ornl.gov

Garrett Granroth
ORNL
granrothge@ornl.gov

Anthony Gianfrancesco
University of Tennessee
plamsnt@gmail.com

Adrienne Greene
Sandia National Laboratory
acgreen@sandia.gov

Michael Groeber
Air Force Research Laboratory
michael.groeber@us.af.mil

Doga Gursoy
Argonne National Laboratory
dgursoy@aps.anl.gov

Mark Hagmann
NewPath Research L.L.C.
mhagmann@newpathresearch.com

Steven Hahn
ORNL
Hahnse@ornl.gov

Jeffrey Harrison
University of Washington
jarrisonwins@gmail.com

Todd Hastings
University of Kentucky
todd.hastings@uky.edu

Qian He
ORNL
heq1@ornl.gov

Thomas Henage
NewPath Research L.L.C.
thenage@gmail.com

Dale Hensley
ORNL
hansleydk@ornl.gov

Holland Hysmith
ORNL
holland.hysmith@gmail.com

Vighter Iberi
ORNL
iberivo@ornl.gov

Anton Ievlev
ORNL
ievlevav@ornl.gov

Russell Jacobs
Self
rjacobs@yahoo.com

Yuanwen Jiang
University of Chicago
yuanwen@uchicago.edu

Waine Jouber
ORNL
joubert@ornl.gov

Sergei Kalinin
ORNL
sergei2@ornl.gov

Durmus Karatay
University of Washington
ukaratay@uw.edu

Kerstin Kleese van Dam
Pacific Northwest National Laboratory
kerstin.kleesevandam@pnll.gov

Paula Kotula
Sandia National Laboratory
pgkotul@sandia.gov

Ivan Kravchenko
ORNL
kravchenkoii@ornl.gov

Mikhail Kravchenko
University of Tennessee
mkravche@bols.utk.edu

A. Gilad Kusne
NIST
aaron.Kusne@nist.gov

Nouamane Laanait
ORNL
laanaitn@ornl.gov

James LeBeau
North Carolina State University
jmlebeau@ncsu.edu

Steven Lee
U.S. Department of Energy
steven.lee@science.doe.gov

Lianshan Lin
ORNL
linl@ornl.gov

Eric Lingerfelt
ORNL
lingerfeltej@ornl.gov

Arthur Maccabe
ORNL
maccabeab@ornl.gov

Jason Maldonis
University of Madison, Wisconsin
maldonis@wisc.edu

Artem Maksov
ORNL
maksovab@ornl.gov

George Maracas
U.S. DOE - Office of Science
george.maracas@science.doe.gov

Baishakhi Mazumder
ORNL
mazumderb@ornl.gov

Kristin McCullough
Grand View University
kmccullo4@gmail.com

Ian McNulty
Argonne National Laboratory
mcnulty@anl.gov

Stephen Mick
Gatan
smick@gatan.com

Dean Miller
Argonne National Laboratory
miller@anl.gov

Rohan Mishra
Vanderbilt University
rohan.mishra@vanderbilt.edu

Eva Mutunga
University of Tennessee
emutunga@vols.utk.edu

D Frank Ogletree
Lawrence Berkeley National Lab
dfogletree@lbl.gov

M. Baris Okatan
ORNL
m.b.okatan@gmail.com

Hung O'Neill
ORNL
oneillhm@ornl.gov

Colin Ophus
Lawrence Berkeley National Lab
clophus@lbl.gov

Jewook Park
ORNL
parkj1@ornl.gov

Srikanth Patala
North Carolina State University
spatala@ncsu.edu

Robert Patton
ORNL
pattonrm@ornl.gov

Thomas Potok
ORNL
potokte@ornl.gov

Dhiren Pradhan
University of Puerto Rico
dhirenkumarp@gmail.com

Thomas Proffen
ORNL
tproffen@ornl.gov

Tanya Prozorov
Ames Lab
tprozoro@ameslab.gov

Weiju Ren
ORNL
renw@ornl.gov

Robert Sacci
ORNL
saccirl@ornl.gov

Ritesh Sachan
ORNL
sachanr@ornl.gov

Jerzy Sadowski
Brookhaven National Laboratory
sadowski@bnl.gov

Subramanain Sankaranarayanan
Argonne National Laboratory
skrssank@anl.gov

P James Schuck
Lawrence Berkeley National Lab
pjschuck@lbl.gov

Pablo Seleson
ORNL
selesonpd@ornl.gov

Fatih Sen
Argonne National Laboratory
fsen@anl.gov

Jeff Simmons
Air Force Research Laboratory
jeff.simmons.3@us.af.mil

Mary Jane Simpson
ORNL
simpsonm@ornl.gov

David Skinner
Lawrence Berkeley National Lab
deskinner@lbl.gov

Patrick Smith
University of Texas/Austin
patrick.smith@beg.utexas.edu

Suhas Sreehari
Purdue University
ssreehar@purdue.edu

Eric Stach
Brookhaven National Laboratory
estach@bnl.gov

Chad Steed
ORNL
steedca@ornl.gov

Andrew Stevens
Duke University
ajs104@duke.edu

Joshua Stuckner
Virginia Tech
stuckner@vt.edu

Bobby Sumpter
ORNL
sumpterbg@ornl.gov

Christopher Symons
ORNL
symonsct@ornl.gov

Ho Ming Tong
University of Florida
gabriel.tong@ufl.edu

Bryan Thompson
SYSTAP, LLC
bryan@systap.com

Raymond Unocic
ORNL
unocicrr@ornl.gov

Daniela Ushizima
Lawrence Berkeley National Lab
dushizima@lbl.gov

Rama Vasudevan
ORNL
rvv@ornl.gov

Ranga Vatsavai
North Carolina State University
rrvatsav@ncsu.edu

Mario Viani
Asylum Research, Oxford Instruments
mario.viani@oxinst.com

David Vine
Argonne National Laboratory
dvine@anl.gov

Sarah Vorpahl
University of Washington Seattle
svorpahl@uw.edu

Paul Voyles
University of Wisconsin-Madison
paul.voyles@wisc.edu

Song Wang
University of South Carolina
songwang@cec.sc.edu

Jack Wells
ORNL
wellsjc@ornl.gov

Jim Werner
Los alamos National Laboratory
jwerner@lanl.gov

Tracy Whitaker
ORNL
whitakerth@ornl.gov

Kai Xiao
ORNL
xiaok@ornl.gov

Kyle Yakal-Kremski
National Academy of Sciences
kyle.yakal-kremski.ctr@nrl.navy.mil

Sangmo Yang
ORNL
Yangs@ornl.gov

Wei-Chang Yang
NIST
david.yang@nist.gov

Hongkai Yu
University of South Carolina
yu55@email.sc.edu

Dimitri Zakharov
Brookhaven National Laboratory
dzakharov@bnl.gov

Amanda Zetans
ORNL
zetansac@ornl.gov

Jiaming Zhang
Hewlett Packard labs
jiamingzhang98@gmail.com

Minghao Zhang
UCSD
miz016@eng.ucsd.edu

Wu Zhou
ORNL
zhouw1@ornl.gov

Youjie Zhou
University of South Carolina
zhou42@email.sc.edu

ORNL Visitor Wireless Network

As a visitor to Oak Ridge National Laboratory (ORNL), access to the ORNL wireless network is available to you in many of our buildings. To gain access, you must have a PC or other portable device with wireless capability. If your PC or portable device does not have wireless capability, a limited number of wired-to-wireless Ethernet bridges may be available from your host organization.
In ORNL building 5100, National Institute for Computational Sciences (NICS), the wired-Ethernet may also be used by visitors in the same manner as described in this document.

Logging into the ORNL Wireless Network
You must log into the ORNL Wireless Network by opening a window in a Web browser. Attempting to access the network through an email client or other network application will not redirect you to the Log-in Page.
1. Complete the following steps to log in to the ORNL Wireless Network.
2. Connect to the ORNL Wireless Network via wireless connection or via ORNL Visitor wired connection (where available.) (Network name = ornl-visitor)
3. Open a Web browser and attempt to access any website. The browser automatically redirects you to the login page.
4. Enter the appropriate information in the fields provided (i.e., Name, Contact Number, and Email Address.)
5. Review and accept the "Accepted Use Policy." (Required)
6. Click **Connect**.

Note: You will be brought to a window with a countdown. This is normal and will take approximately one minute to connect you to the network. Due to content caching, some smart phones may display the registration page even after being registered. Restarting the phone's browser or turning the phone off and back on should resolve this issue.*Revision 3.0, April 26, 2012*

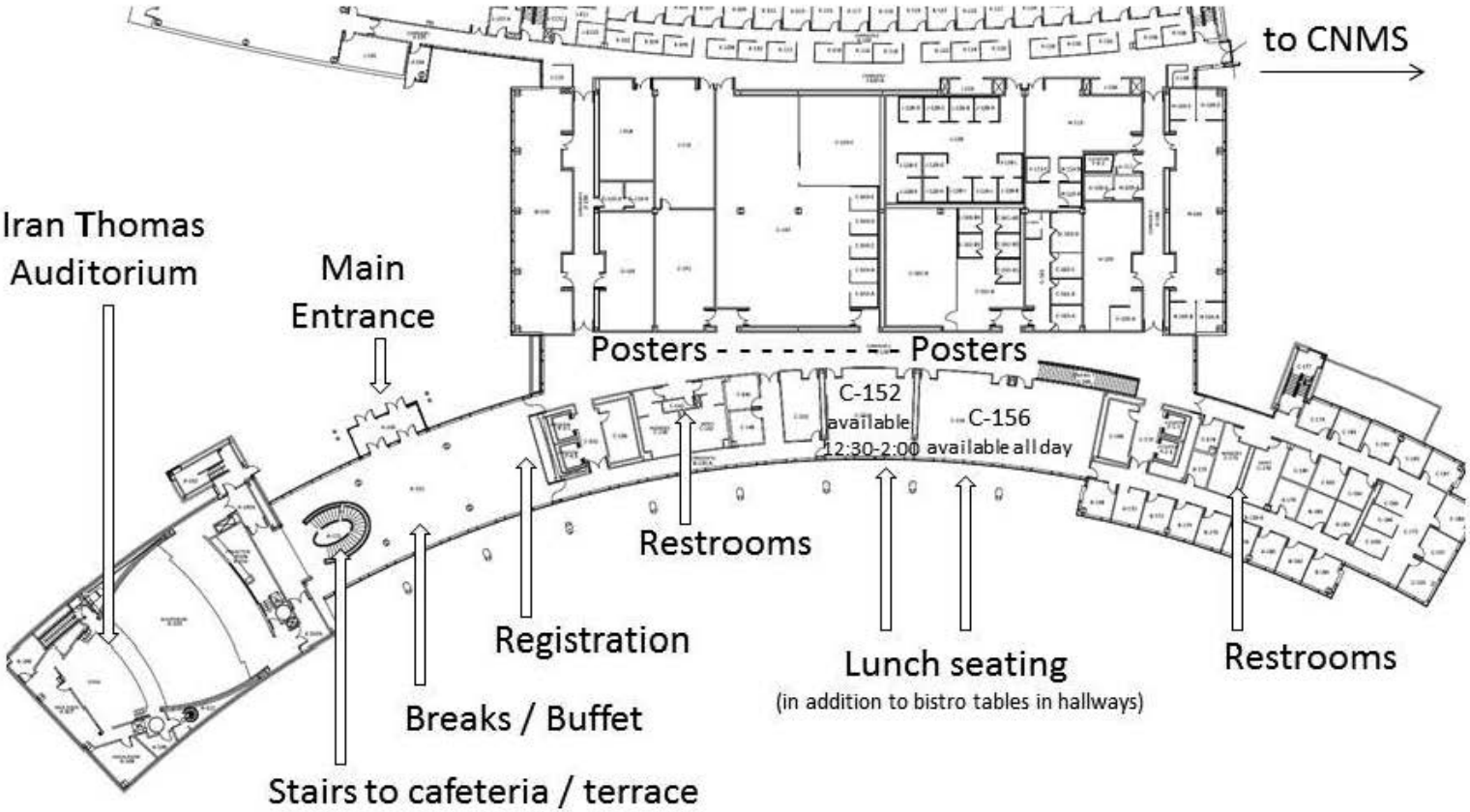
ORNL Wireless Network Access Acceptable Use Policy
This policy is a guide to the acceptable use of the ORNL Wireless Network. Any individual connected to the ORNL Wireless Network in order to use it directly, or to connect to any other network(s), must comply with this policy. By using the ORNL Wireless Network you agree that you understand there is no expectation of privacy for your activities on the ORNL Wireless Network. ORNL retains the right to monitor all network traffic, to access the contents of any network traffic, and to disclose all or part of information gained to authorized individuals or agencies, all without prior notice to, or consent from, any user, sender, or addressee. Please note that web content filtering is utilized on the ORNL Wireless Network to flag or outright block certain categories of content that ORNL has deemed malicious, objectionable or counterproductive to conducting business at ORNL (e.g. pornography, hacking tools, gambling sites, etc.)
The following guidelines will be applied to determine whether or not a particular use of the ORNL Wireless Network is appropriate:

• Users must observe the privacy rights of others. Users shall not intentionally seek information on, or represent themselves as, another user unless explicitly authorized to do so by that user. Executing any form of network monitoring which will intercept data not intended for the User is not acceptable.
• Users must comply with the legal protection applied to programs, data, photographs, music, written documents and other material as provided by copyright, trademark, patent, licensure and other proprietary rights mechanisms.
• Users shall not intentionally develop or use programs that harass other users or infiltrate any other computer, computing system or network and/or damage or alter the software components or file systems of a computer, computing system or network.
• Use of the ORNL Wireless Network for malicious, fraudulent, or misrepresentative purposes is not acceptable. Transmission of any material that violates applicable laws or regulations, including, but not limited to, any copyrighted material, material protected by a trade secret or material or messages that are unlawful, harassing, libelous, abusive, threatening, harmful, vulgar, obscene or otherwise objectionable in any manner or nature or that encourages conduct that could constitute a criminal offense, give rise to civil liability or otherwise violate any applicable local, state, national or international laws or regulations is not acceptable.
• The ORNL Wireless Network may not be used in a manner that impairs network access by others. Nor may the ORNL Wireless Network be used in a manner that impairs access to other networks connected to the ORNL network. Port or security scanning is not acceptable. Introduction of malicious programs into the network or server (e.g., viruses, worms, Trojan horses, e-mail bombs, etc.) is prohibited. Using any program/script/command to intentionally circumvent or disable ORNL's security infrastructure is not acceptable.
Revision 3.0, April 26, 2012

• Unsolicited advertising is not acceptable. Making fraudulent offers of products, items, or services originating from any ORNL network resource is not acceptable. Sending unsolicited email messages, including the sending of "junk mail" or other advertising material to individuals who did not specifically request such material (email spam) is not acceptable.
• Repeated, unsolicited and/or unwanted communication of an intrusive nature is not acceptable. Continuing to send e-mail messages or other communications to an individual or organization after being asked to stop is not acceptable. Sending any form of harassment via email, telephone or paging, whether through language, frequency, or size of messages is not acceptable.

By using the ORNL Wireless Network you are becoming a party to and agree to be bound by this Acceptable Use Policy. By logging on, you agree to release, hold harmless, and indemnify ORNL and its managing and operating contractor for any damages that may result from your access to the Internet or inappropriate usage. The intent of this policy is to identify certain types of uses that are not appropriate, but this policy does not necessarily enumerate all possible inappropriate uses. You furthermore acknowledge that you have read and understand the Acceptable Use Policy and agree to be bound by the Acceptable Use Policy in effect and as updated by us from time to time. If you do not agree to be bound by the Acceptable Use Policy, you may not use the ORNL Wireless Network and you should cancel your request. If you do not agree to be bound by the Acceptable Use Policy, ORNL will be under no obligation to provide you with access to the ORNL Wireless Network.
The Services may be terminated due to a breach of the Acceptable Use Policy at any time.

DOE Standard Warning Banner

This is a Federal network and is the property of the United States Government. It is for authorized use only. Users (authorized or unauthorized) have *no explicit or implicit expectation of privacy*. Any or all uses of this network and all files on this network may be intercepted, monitored, recorded, copied, audited, inspected, and disclosed to authorized site, Department of Energy, and law enforcement personnel, as well as authorized officials of other agencies, both domestic and foreign. *By using this network, the user consents* to such interception, monitoring, recording, copying, auditing, inspection, and disclosure at the discretion of authorized site or Department of Energy personnel. Unauthorized or improper use of this network may result in administrative disciplinary action and civil and criminal penalties. *By continuing to use this network you indicate your awareness of and consent to these terms and conditions of use*. DISCONNECT IMMEDIATELY if you do not agree to the conditions stated in this warning.

Building 8600 – First Floor