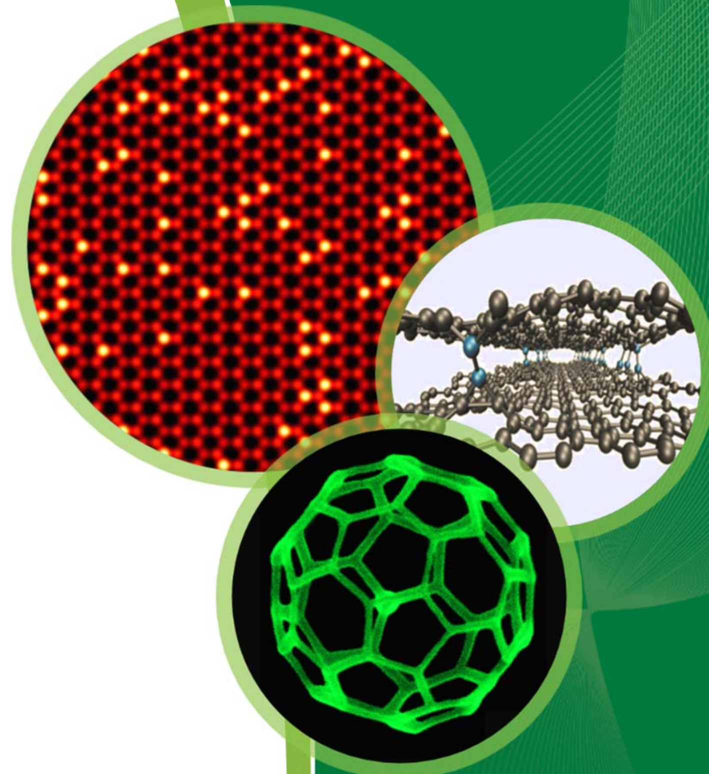
The Center for Nanophase Materials Sciences:

Technical Capabilities and Research Areas



Hans Christen

CNMS Director

Biological and Environmental Research Advisory Committee (BERAC) Meeting

April 20, 2017

ORNL is managed by UT-Battelle
for the US Department of Energy

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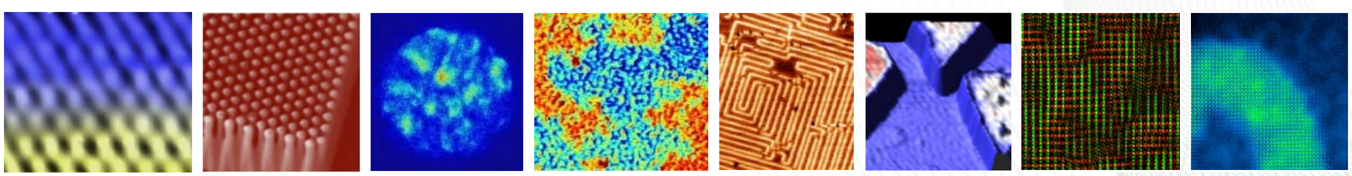
Some of the most fundamental materials science challenges are intrinsically nanoscience challenges

BESAC report "Challenges at the Frontiers of Matter and Energy: Transformative Opportunities for Discovery Science" (November 2015)

1. Mastering Hierarchical Architectures and Beyond-Equilibrium Matter
2. Beyond Ideal Materials and Systems: Understanding the Critical Roles of Heterogeneity, Interfaces, and Disorder
3. Harnessing the Coherence in Light and Matter
4. Revolutionary Advances in Models, Mathematics, Algorithms, Data, and Computing
5. Exploiting Transformative Advances in Imaging Capabilities across Multiple Scales



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Key challenges in nanoscience: Understanding formation and function

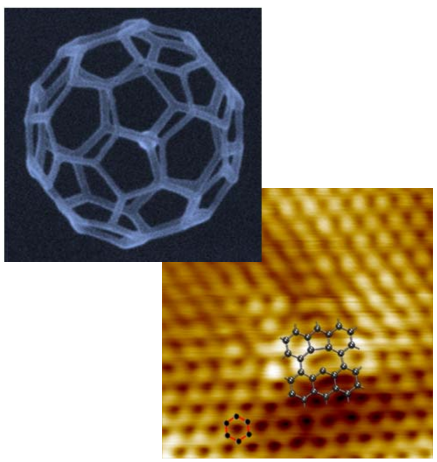
Understanding and Controlling Formation

l atoms
where we want them to be

inate
individual defects

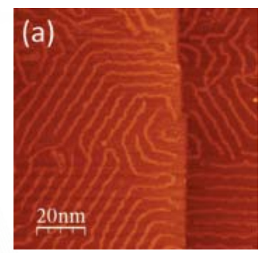
How do we control and direct
self-assembly

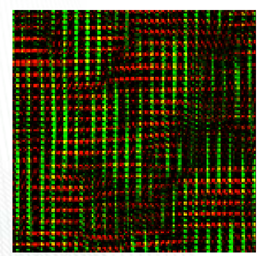
How do we reproducibly and
scalably produce complex and
hierarchical matter



How do defects and nanostructure
influence energy transport and
energy conversion (electrons,
photons, excitons, phonons)?

low can we understand and direct
mass transport (ionic motion,
deformations, droplets)





Understanding and Controlling Function

A user facility is a powerful resource

- Nanoscience is an integral part of very broad areas of science:
  - A User Facility can add the missing pieces to a research team
  - Users are often “experts elsewhere”
- Adequate staffing serves a dual purpose:
  - Maintenance of instrumentation, quality control, training of users
  - Scientific vision, expertise to adapt capabilities to specific applications

DOE Basic Energy Sciences User Facilities: Approximately 15,000 Users (FY2016)

Light Sources, Neutron Sources, and Nanoscale Science Research Centers (NSRCs); located at National Laboratories

- Resources available at no cost to researchers who intend to publish results
- External peer review
- Coordinated access to co-located facilities
- Strong collaborative environment with facility scientists

Five Nanoscale Science Research Centers (NSRCs):
Approx. 3,000 Users (FY2016)
(Three Electron Beam Microcharacterization Centers (EBMCs) were merged into the NSRCs in 2015)

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Five NSRCs provide specific focus areas and ties to co-located facilities

BERKELEY LAB MOLECULAR FOUNDRY

Center for Functional Nanomaterials
Brookhaven National Laboratory

OAK RIDGE National Laboratory
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CINT
Los Alamos · Sandia National Laboratories

Argonne NATIONAL LABORATORY
Center for Nanoscale Materials

Co-Located Facilities

ALS

NERSC

HFIR

SNS SPALLATION NEUTRON SOURCE

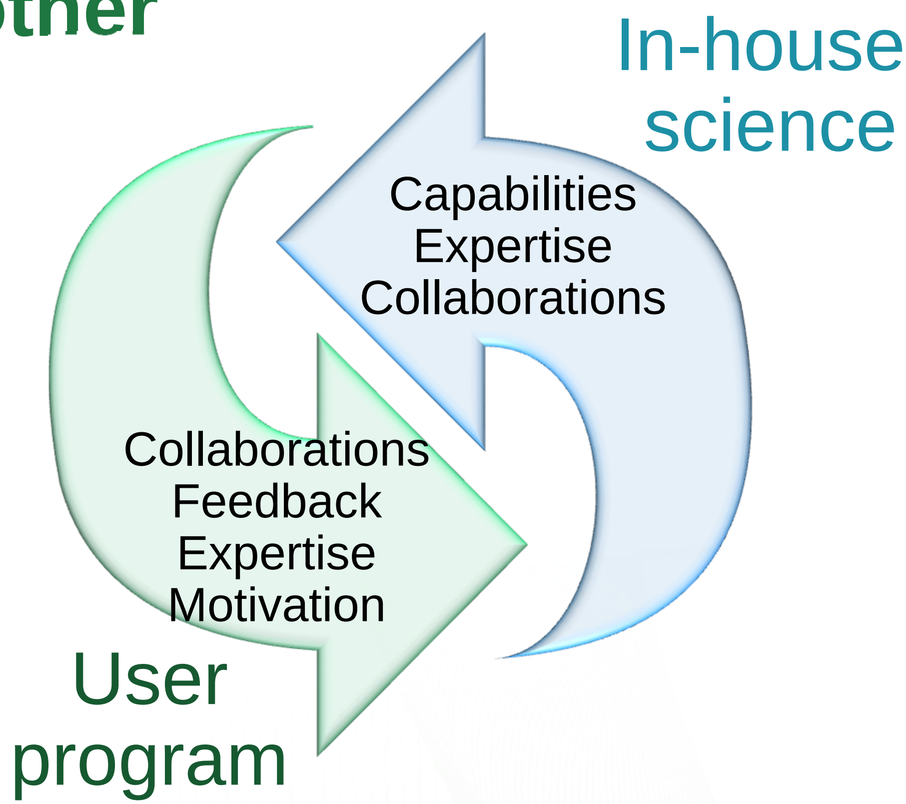
NATIONAL HIGH MAGNETIC FIELD LABORATORY

Expertise and Capabilities

- Each NSRC emphasizes specific areas of synthesis, fabrication, imaging/characterization, and theory/modeling/simulation ("*make, characterize, and understand*")
- Users may request multiple capabilities at an NSRC, or perform work at more than one NSRC
- See NSRC Portal: nsrcportal.sandia.gov

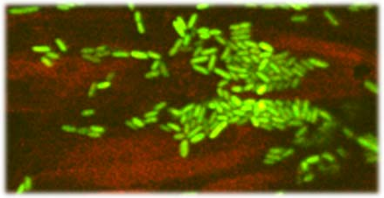
NSRC in-house science and user program benefit from each other

- All staff members dedicate 50% of their time to work with users and 50% to in-house research
- In-house research is key to developing capabilities and expertise
- 80% of instrument time is dedicated to the user program



CNMS: laboratories, a gateway to neutrons and computing, direct interactions with staff

- CNMS building:
  - Total 80,000 sq. ft., includes 32 laboratory modules and a 10,000 sq. ft. cleanroom (Class 1000; Class 100 in e-beam lithography suite)
- Ultra-quiet space for electron and scanning probe microscopy
- Close ties to ORNL's two neutron facilities (Spallation Neutron Source [SNS] and High Flux Isotope Reactor [HFIR]) and to the Oak Ridge Leadership Computing Facility
- Bio-affiliate laboratories for users with biological sample requirements



CNMS delivers impactful science

FY2016 numbers:

- 601 unique users (575 on-site)
  - Average stay at CNMS: ~13 days
  - 50% from US academic institutions
  - 38% faculty; 24% postdocs; 38% students
- 435 refereed regular papers published that acknowledge CNMS
  - 51% in journals with IF>5
  - 36% in journals with IF>7
  - 70% co-authored by users
- 18% of CNMS (FY13-FY15) users are also SNS/HFIR users
- ~$24M from DOE-BES-SUFD

93 US institutions from 36 states and territories
44 foreign institutions from 19 foreign countries

CNMS journal covers (past 18 months)

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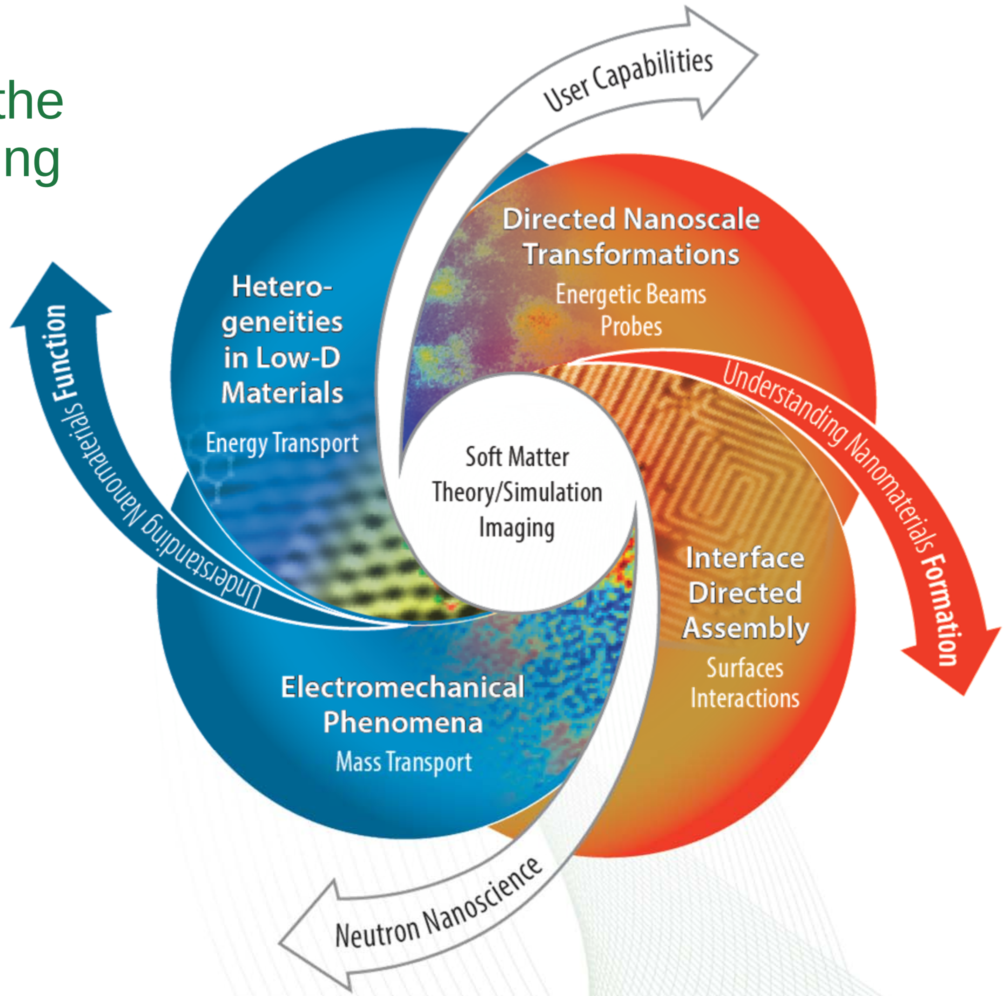
CNMS executes a focused in-house research effort to advance our understanding of nanomaterials function and formation

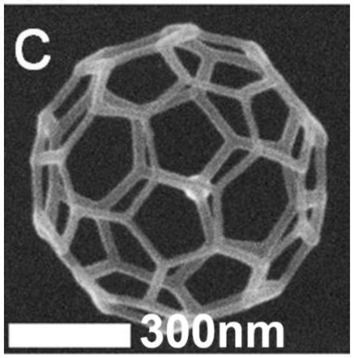
These research activities drive the development of the corresponding necessary capabilities that then become available to users.

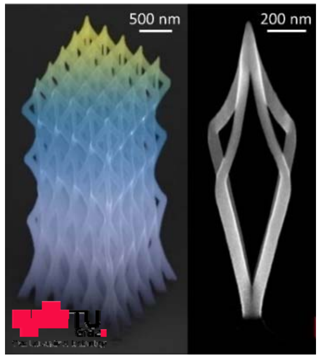


Example of capability development:

3D direct-write nanofabrication using focused electrons, ions, or photons

E-beam induced deposition (EBID)

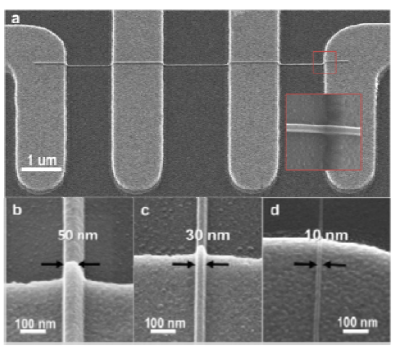




J.D. Fowlkes *et al*., *ACS Nano* (2016)

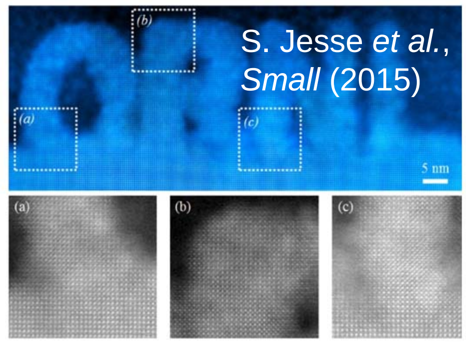
R. Winkler *et al*., *ACS Appl. Mat. Interf.* (2016)

Ion-beam induced deposition (IBID)



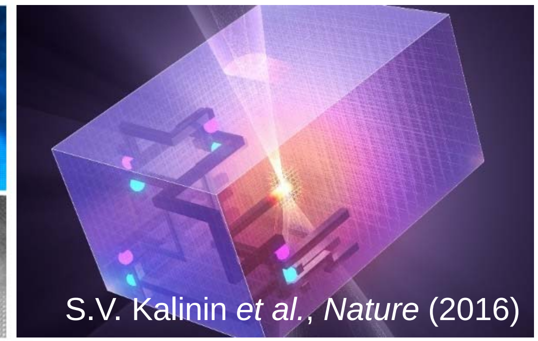
IBID benefits from smaller minimum probe diameter and beam penetration

Materials modification and deposition using the Scanning Transmission Electron Microscope



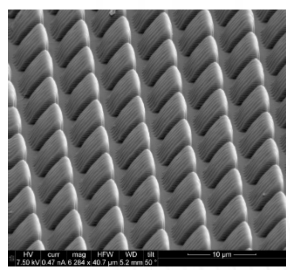
S. Jesse *et al.*, *Small* (2015)

Selective crystallization of $SrTiO_3$

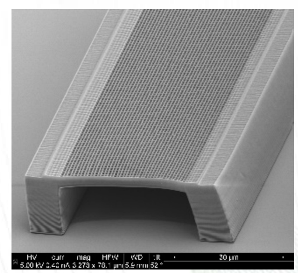


S.V. Kalinin *et al.*, *Nature* (2016)

Direct Laser Write based on 2-photon polymerization (liquid, solid precursors)



Asymmetric wettability



Fluidic structures, size-based separations

Nanoscribe Photonics Professional GT

These approaches rely on a close integration of theory/modeling/simulation and the development of precursors.

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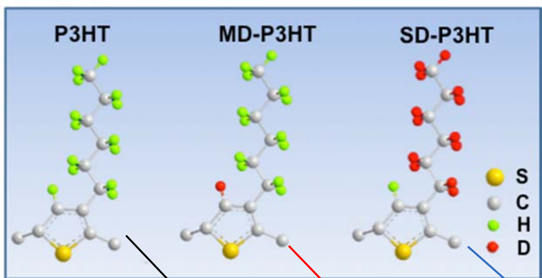
Example of capability development:

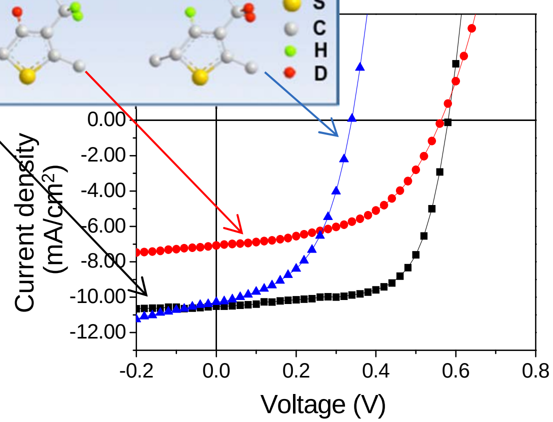
Linking precise synthesis, computing, and neutrons for soft matter research

Selective deuteration introduces a surprising way to modify optoelectronic and structural properties

Selective deuterations on backbone or side-chain of P3HT in P3HT/PCMB photovoltaics

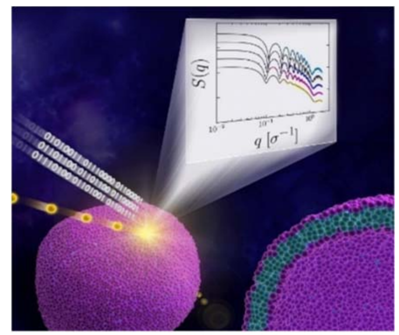


M. Shao *et al.*, *Nat. Commun.* (2014)



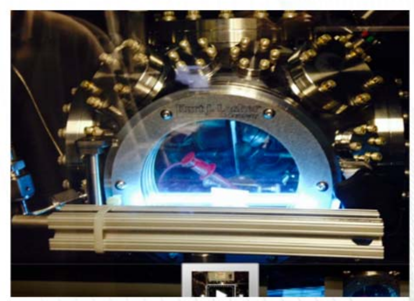
Computational capability for users: quantum calculations to treat electron-phonon interactions

Coarse-grained molecular dynamics calculations enable detailed neutron studies of lipid bilayer membranes



J.-M. Carrillo *et al.*, *J. Chem. Theory & Comp.* (2017)

CNMS develops sample environments to study the formation of polymer systems (combining neutrons with optical probes)

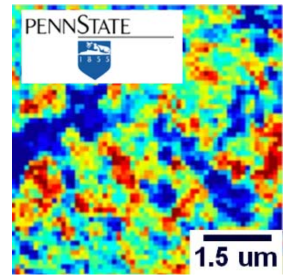


J. Zhu *et al.*, *Nanoscale* (2015); N. Herath et al., *Scientific Reports* (2015)

Example of capability development:

Pushing the limits of force-based scanning probe microscopy

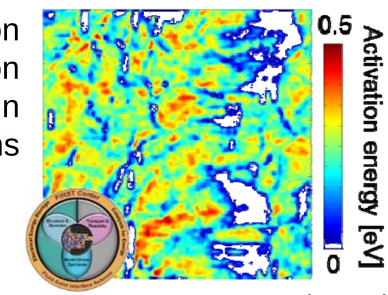
- Excitation with a band of frequencies renders the technique quantitative (spectroscopic)
- EFRC collaboration lead to Electrochemical Strain Microscopy: chemical modifications are tracked as topography changes
- Development of data analytic methods yields meaningful information



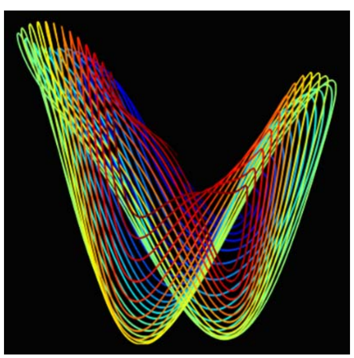
Quantitative map of material nonlinearity at the nanoscale.

Bintachitt *et al*, PNAS (2010)

Mapping of activation energy for Li-ion transport in $LiCoO_2$ thin films

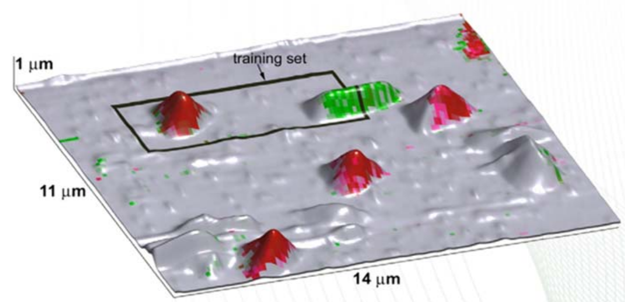


N. Balke *et al.*, *NanoLett.* (2012)



Piezoelectric hysteresis loops captured 3000x faster in “General-mode AFM” (full information capture without imposed operator bias).

S. Somnath *et al.*, *Nature Commun.* (2016)



ural network training
ntifies bacteria based on
ctromechanical response

M.P. Nikiforov *et al.*, *Nanotechnol.* (2009)

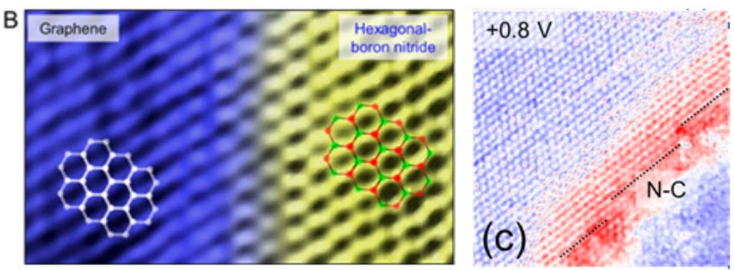
Example of capability development:

Understanding electronic, magnetic, and transport properties at the nanoscale

Development of STM and 4-probe STM based imaging and spectroscopy modes

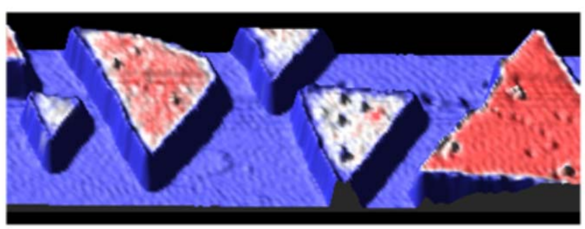
- tunneling thermopower microscopy
- scanning tunneling potentiometry
- spin-polarized STM

STM image and spectroscopy revealing confined electronic states at Gr/h-BN heterojunctions



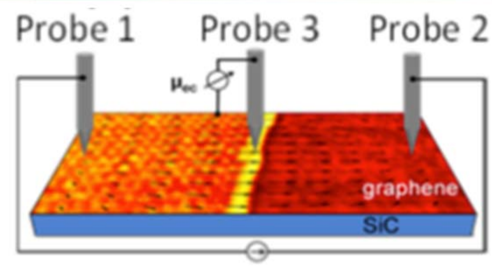
J. Park et al, *Science* (2014); *Nature Comm.*(2014)

SP-STM reveals spin polarization of Co/Cu



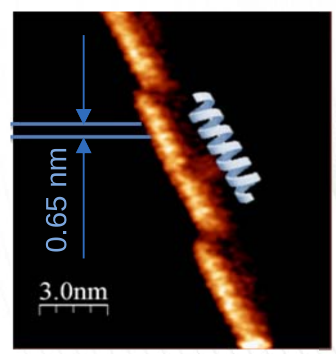
J. Park et al, *Nano Lett*. (2017)

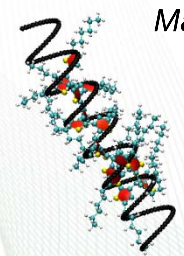
Scanning tunneling potentiometry to map conductivity across grain boundaries



K.W. Clark et al, *Nano Lett*. (2013); *PRX* (2014)

STM helps determine structure of helical polymer





H. H. Zhang et al., *Macromolecules* (2016)

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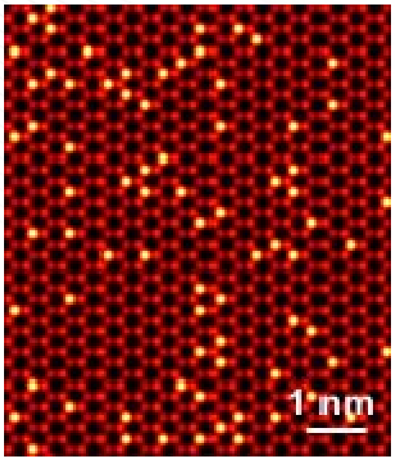
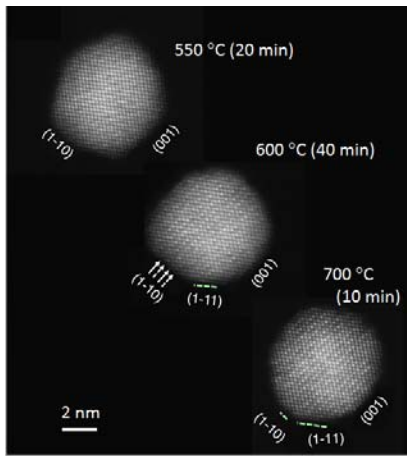
CNMS emphasizes aberration-corrected Scanning Transmission Electron Microscopy (AC-STEM) and Electron Energy Loss Spectroscopy (EELS)



Identifying individual dopant atoms $Mo_{1-x}W_xSe_2$

X. Li *et al.*,
Adv. Mater. (2016)

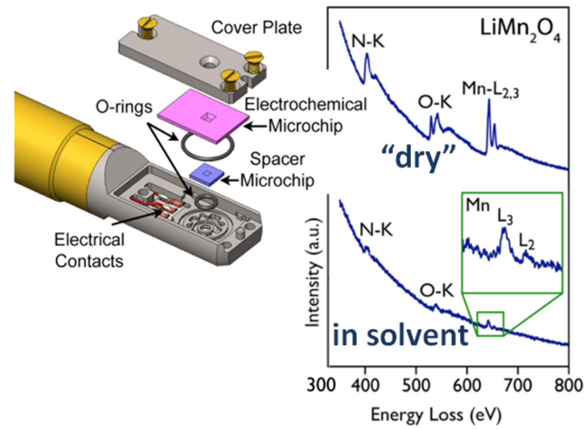


Tracking a single Pt_3Co at high temperature

M. Chi *et al.*,
Nature Commun. (2015)

Local determination of magnetic properties using controlled aberrations

Rusz *et al.*,
PRL (2014)



Quantitative EELS measurements in liquids

R.R. Unocic *et. al.*,
ChemComm (2015)

– Nion UltraSTEM Cs-corrected STEM (60-100kV)
– FEI Titan S Cs-corrected STEM/TEM (60-300kV)
– Access to Nion HERMES Monochromated AC-STEM
– Access to Hitachi HF3300 TEM/STEM

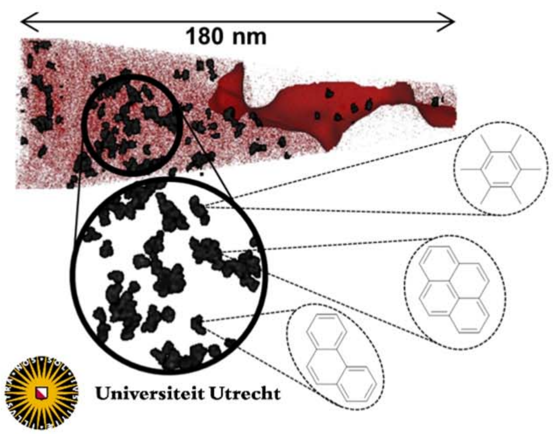
With ~1Å probe and <10meV energy resolution, the MAC-STEM becomes the nanoscale/real-space counterpart to neutron scattering (e.g., phonons)

Atom Probe Tomography and Chemical Imaging

Atom Probe Tomography

Laser-LEAP (local electrode atom probe):
complete 3D reconstruction of atomic positions (within $1 nm^3$);

Applied to non-metallic samples.
Sensitive to any element.

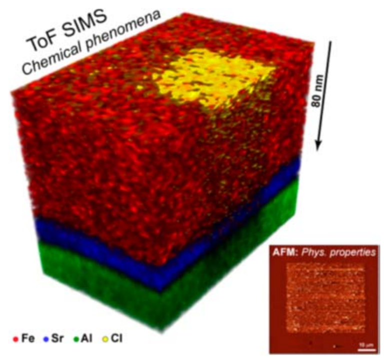


Example: Coke formation in zeolite catalyst

J.E. Schmidt, *et. al.*, *Angew. Chem. Int. Ed.* (2016)

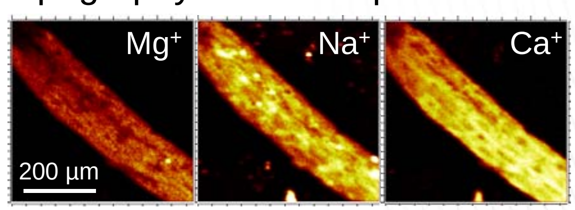
Access to AFM/ToF-SIMS

Developing methodologies to combine chemical imaging and functional mapping



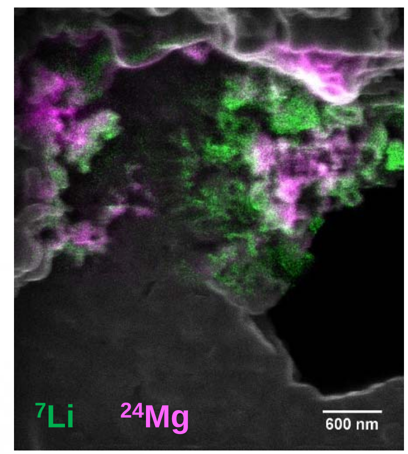
Chemical effects of ferroelectric switching
A.V. Ievlev *et. al.*, *ACS Appl. Mater. Interf.* (2017)

Chemical composition and topography of Arabidopsis root



Secondary Ion Mass Spectrometry (SIMS) – Helium Ion Microscopy (HIM)

Combining SIMS with nm-scale ion beam imaging



Zeiss Orion NanoFab:
- Outperforms SEM for imaging (especially for insulating samples)
- Highest-resolution ion milling tool

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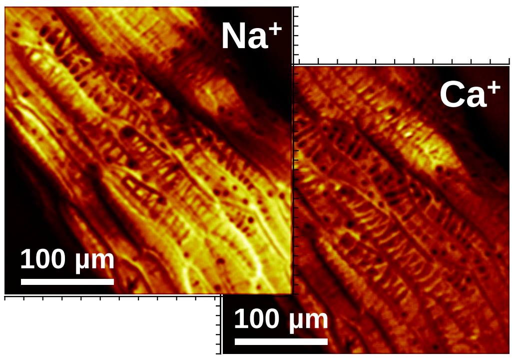
A wide user community benefits from CNMS imaging capabilities

Chemical maps of Sphagnum leaves

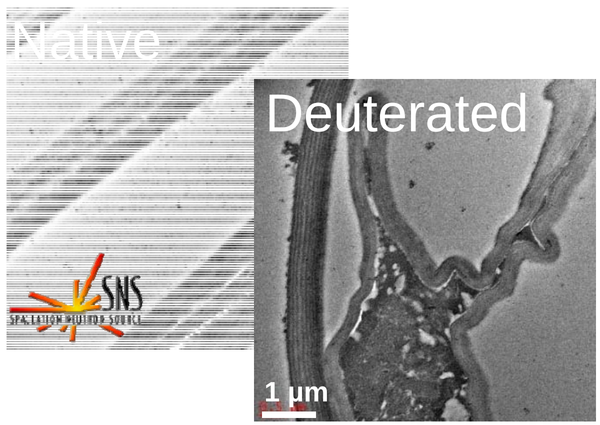


ToF-SIMS imaging

Multi-scale mass spectrometry based capabilities at CNMS:

- MALDI-ToF Imaging
- ToF-SIMS
- AFM thermal desorption MS
- HIM-SIMS

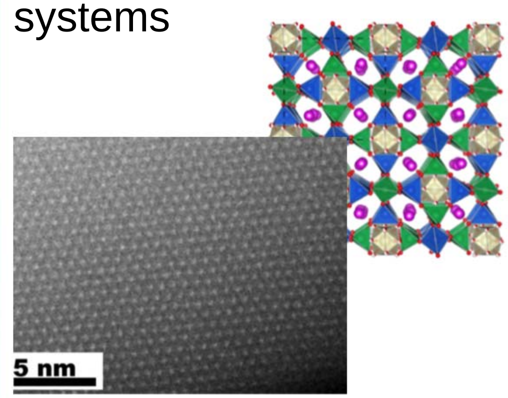
Growth-stress induced abnormal lignin distribution in cell walls of deuterated switchgrass



Low-dose TEM imaging provides critical complementary information to neutron scattering

S. Bhagia *et al.* (2017)

Study of perrhenate sodalite for ^{99}Tc immobilization from contaminated subsurface systems



STEM imaging to confirm equal distribution / lack of clustering.

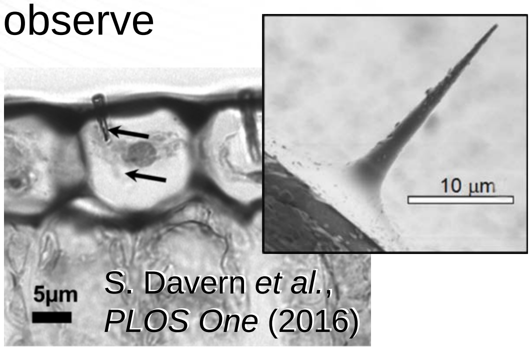
E.M. Pierce *et al.*, *ES&T* (2016)

SNS Spallation Neutron Source | SLAC | Argonne National Laboratory

Oak Ridge National Laboratory | Center for Nanophase Materials Sciences

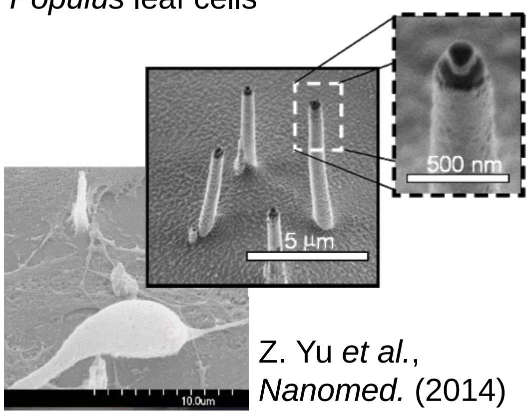
Users benefit from unique nanofabrication capabilities and CNMS-developed platforms

Carbon fiber arrays to manipulate and observe



S. Davern *et al.*, *PLOS One* (2016)

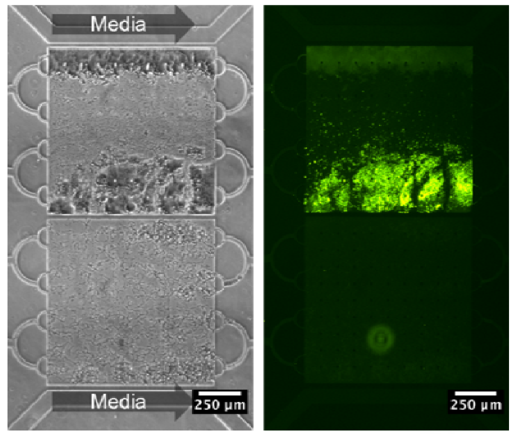
Delivering femtomole to picomole quantities of fluorescent or radiolabeled molecules into *Populus* leaf cells



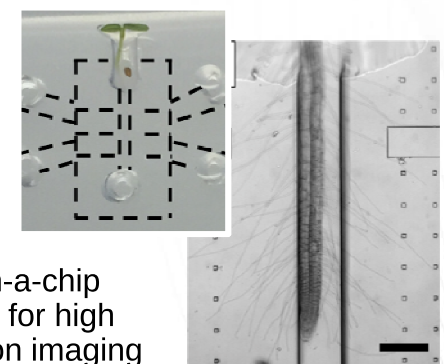
Z. Yu *et al.*, *Nanomed.* (2014)

Vertically aligned carbon nanofiber electrodes as a nano-neuron interface

Complex fluidic platforms to study microbial interactions



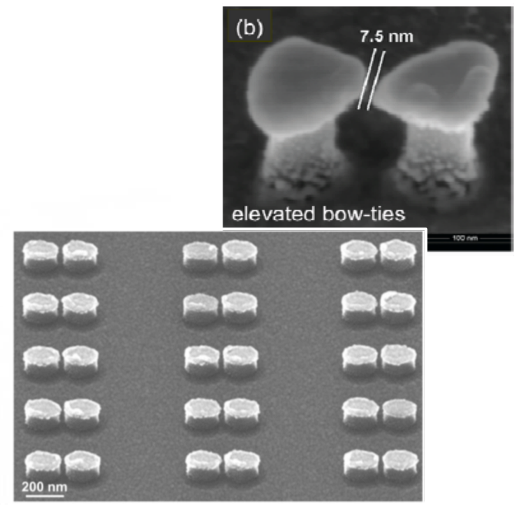
Co-culture and communication between two microbial communities



Plant-on-a-chip platform for high resolution imaging of plant-microbe interactions in real time

Nanostructures for Raman Spectroscopy

CNMS-developed fabrication methods have been used by a number of users



Example: Trace-level perchlorate analysis of impacted groundwater by elevated gold ellipse dimer nanoantenna surface-enhanced Raman scattering

A.M. Jubb *et al.*, *J. Raman Spectr.* (2016)

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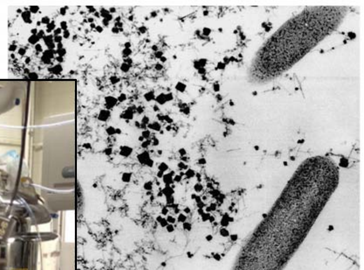
User research has a broad impact

Characterization of nanofermented quantum dot materials

Graduate students start company to commercialize ORNL nanoferrmetation technology

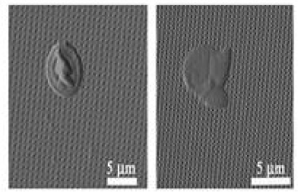


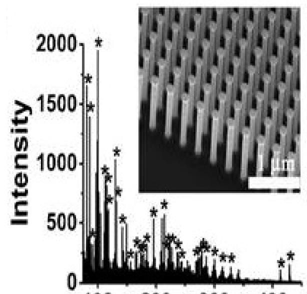
nano elements source, LLC

CNMS user projects to characterize and process the nanomaterials.

Single cell mass spec

LDI-MS analysis of single cells (~30 fL volume) on nanofabricated post arrays.





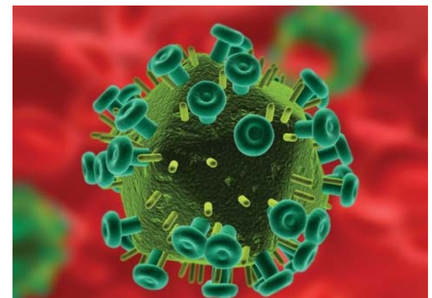
Akos Vertes, George Washington University

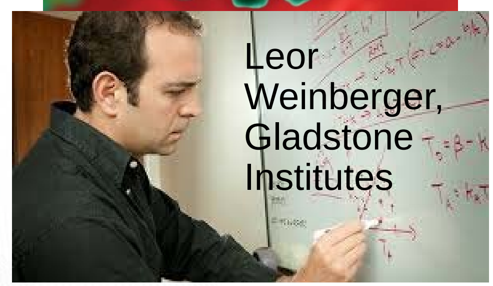
The researchers performed nanofabrication and device characterization at CNMS. The work led to the commercial availability of the REDIchip.

B. Walker et al. *Angew. Chem. Int. Ed.* (2013)

Understanding proviral latency in HIV

Understanding how stochastic fluctuations in small molecular populations lead to proviral latency in HIV, the primary clinical problem in AIDS treatment





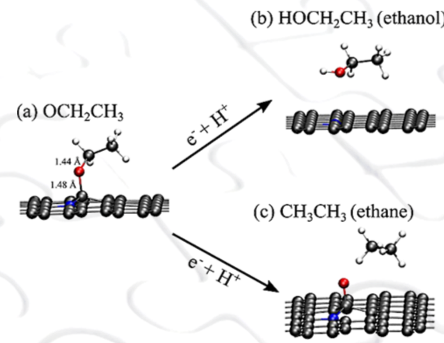
Leor Weinberger, Gladstone Institutes

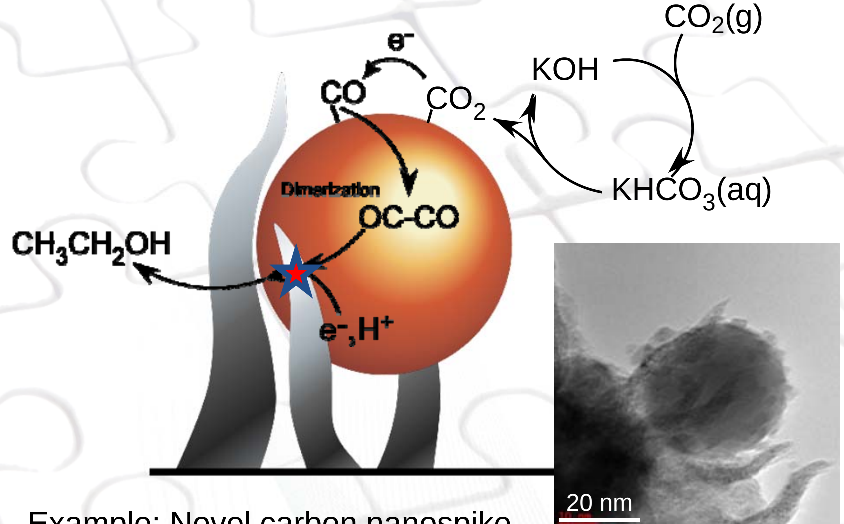
Work at CNMS focused on understanding the fluctuations using time-lapse noise spectroscopy techniques developed at the CNMS.

R. Dar *et al.*, *PNAS* (2012)

The multiple pieces fit together to form a center

- “Make, characterize, understand”
- Broad range of synthesis and fabrication
- Suite of functional measurements
- Strongly integrated theory effort
- Data analytics to underpin imaging and spectroscopy of complex systems
- Strong ties to neutron scattering and high performance computing
- Interactions with the other NSRCs





Example: Novel carbon nanospike catalysts for electrochemical conversion of CO_2 to ethanol. Theory shows how N-dopants introduce the necessary curvature and suitable binding sites.

Y. Song *et al.*, *ChemistrySelect* (2016)

See www.ornl.gov/facility/cnms and nsrcportal.sandia.gov

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Center for Nanophase Materials Sciences

A DOE User Facility for Creating, Characterizing, and Understanding Nanomaterials

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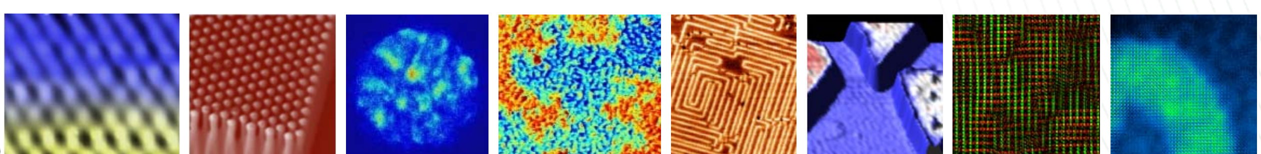
Providing access to staff expertise and equipment at no cost to users who intend to publish the results.

Access to CNMS:

- Two proposal calls per year; proposals for short-term projects are accepted continuously
- Simple 2-page proposal
- Joint proposals with neutron sources (SNS, HFIR)
- Located at Oak Ridge National Laboratory, near Knoxville, TN

Research areas:

- **Synthesis** – Soft matter (precision synthesis, selective deuteration), 2D materials, hybrid structures, epitaxial oxides
- **Nanofabrication** – Direct-write (3D) fabrication, e-beam lithography, multiscale fluidics, 10,000 sq. ft. cleanroom
- **Advanced Microscopy** – AFM, STM, aberration-corrected and *in situ* TEM/STEM, He-ion microscopy, atom-probe tomography
- **Chemical Imaging** – Multiple approaches based on mass spectrometry or optical spectroscopies
- **Functional Characterization** – Laser spectroscopy, transport, magnetism, electromechanical phenomena
- **Theory/Modeling, Data Analytics** – Including gateway to leadership-class, high-performance computing



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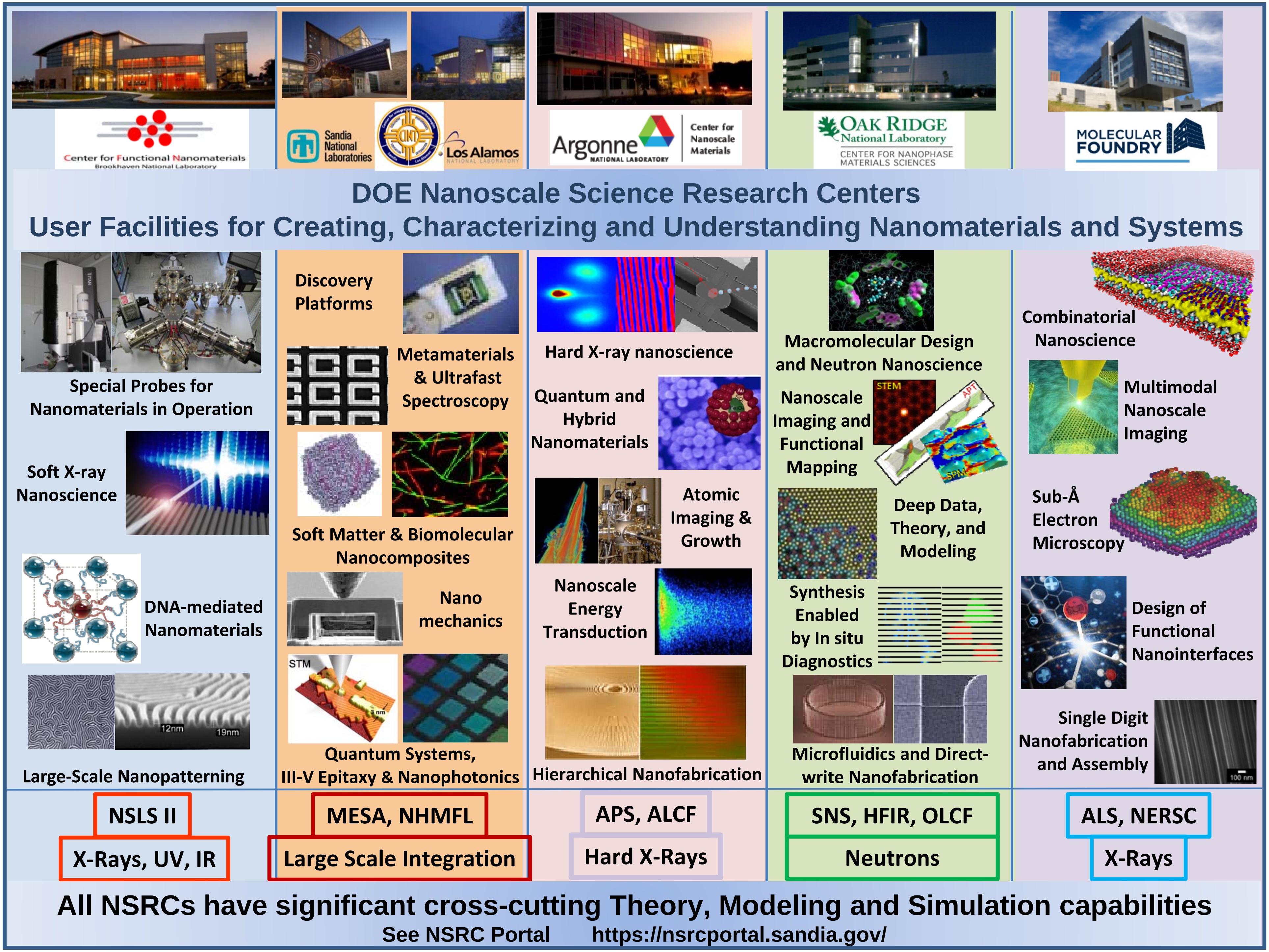
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19nm
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Metamaterials & Ultrafast Spectroscopy
Soft Matter & Biomolecular Nanocomposites
Nano mechanics
STM
Quantum Systems, III-V Epitaxy & Nanophotonics
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Quantum and Hybrid Nanomaterials
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Macromolecular Design and Neutron Nanoscience
Nanoscale Imaging and Functional Mapping
STEM
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Synthesis Enabled by In situ Diagnostics
Microfluidics and Direct-write Nanofabrication
Combinatorial Nanoscience
Multimodal Nanoscale Imaging
Sub-Å Electron Microscopy
Design of Functional Nanointerfaces
Single Digit Nanofabrication and Assembly
100 nm
NSLS II
X-Rays, UV, IR
MESA, NHMFL
Large Scale Integration
APS, ALCF
Hard X-Rays
SNS, HFIR, OLCF
Neutrons
ALS, NERSC
X-Rays
All NSRCs have significant cross-cutting Theory, Modeling and Simulation capabilities
See NSRC Portal https://nsrcportal.sandia.gov/