

Success Stories from PARADIM

Darrell G. Schlom

*Department of Materials Science and Engineering
Cornell University*



PARADIM

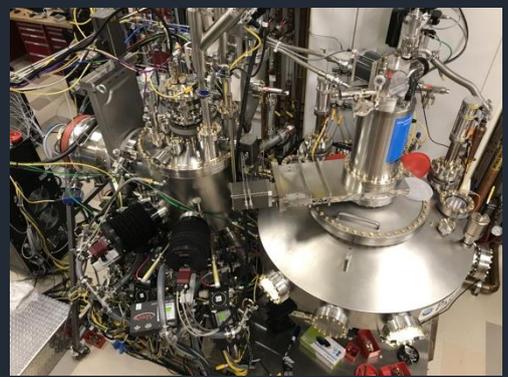
PLATFORM FOR THE ACCELERATED REALIZATION, ANALYSIS & DISCOVERY OF INTERFACE MATERIALS

Democratize materials discovery in the U.S.A.

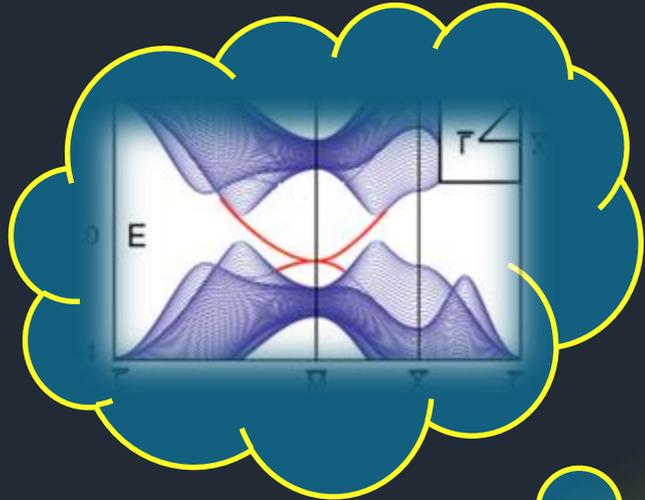
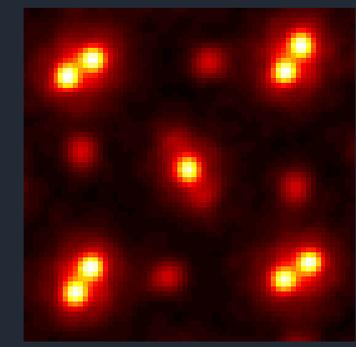
by enabling a more effective way of pursuing materials research, one that accelerates materials discovery by establishing a materials discovery ecosystem—
a national community of practitioners—
and equipping them with theoretical and experimental methods that enable them to reduce to practice the inorganic materials of which they dream

Platform

Thin Film Growth



Electron Microscopy



External Users
 Local Users
 Scientific Staff
 Platform Collaborators
 In-House Research Team

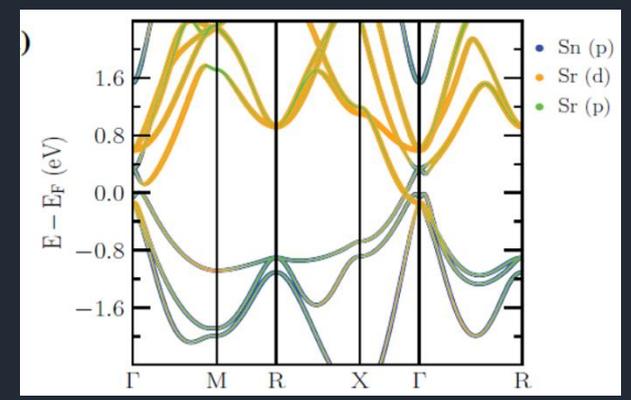
Bulk Crystal Growth



Community of Practitioners



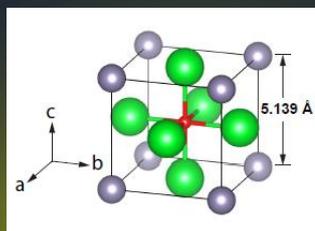
Theory & Simulation



Platform Example

three Scientific Staff
two In-House Faculty
two Platform Collaborators
one In-House Ph.D. student

User's Vision



Community of Practitioners

Prof. Eom
+ Postdoc
+ Ph.D.
student



External Users

Local Users

Scientific Staff

Platform Collaborators

In-House Research Team

Y. Ma, A. Edgeton, H. Paik, B.D. Faeth, C.T. Parzyck, B. Pamuk, S-L. Shang, Z.K. Liu, K.M. Shen, D.G. Schlom, and C.B. Eom, "Realization of Epitaxial Thin Films of the Topological Crystalline Insulator Sr_3SnO ," *Advanced Materials* 32 (2020) 2000809.

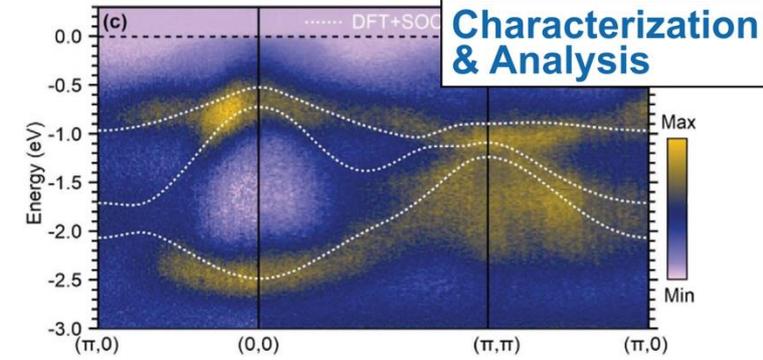
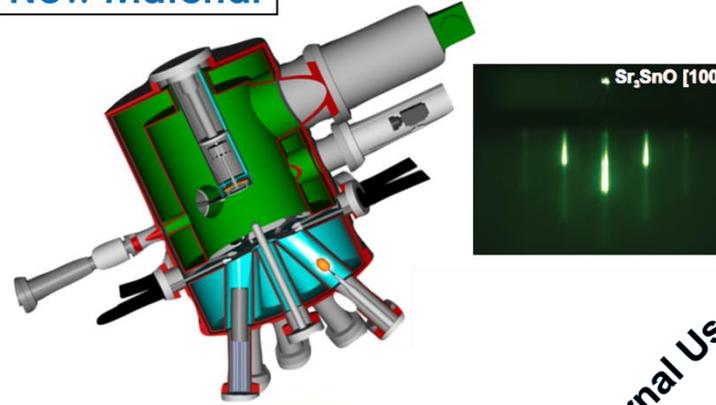
An enlightened and seasoned scientist had a bold vision—to make a new type of topological insulator in a form that could be relevant to technology. The dream compound involved a material predicted to have the desired band structure, but it had never been realized. The group tried for two years to make a simpler version of the material in their own lab, but could not find the right conditions. The group came to PARADIM and in two visits totaling six weeks of round-the-clock molecular-beamtime, they succeeded!

Guided by thermodynamic calculations from Platform collaborators at Penn State, a growth protocol was designed and implemented on PARADIM's signature tool that enabled exquisite control of the sample growth. All relevant measurements were made without exposing the sample to air to assess and optimize its crystalline perfection and electronic structure. The bandstructure of the new material was measured and compared to calculated expectations provided by PARADIM's Theory & Simulation Facility. This new type of topological insulator should be more readily tuned by the underlying substrate and layering to provide desired and technologically useful properties than traditional topological insulators.

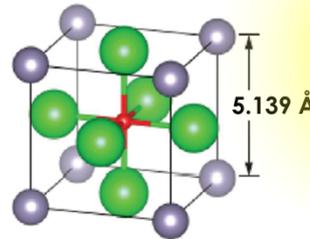
Y. Ma, et al., *Adv. Mater.* **32**, 2000809 (2020).

C.B. Eom, University of Wisconsin-Madison; Z.-K. Liu, Penn State University; K.M. Shen, D.G. Schlom, Cornell University

Realization of New Material



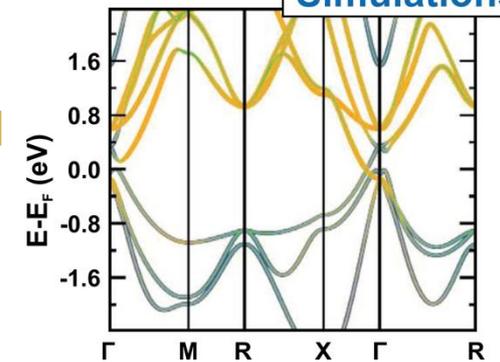
User's Vision



Community of Practitioners

External Users
Local Users
Scientific Staff
Platform Collaborators
In-House Research Team

Theory & Simulations



Boron carbide (B₄C) is an extremely hard material, with a melting temperature above 2000° C; its electronic properties vary strongly with the precise composition.

PARADIM's tilting laser diode floating zone furnace with machine learning (ML) guidance has enabled users to grow the world's largest and most structurally perfect B₄C single crystals. The *in-situ* monitoring of gas-phase species during the growth provides insights on the underlying mechanism by which the composition changes, and in combination with ML based real-time guidance on zone stability, enables the successful growth of single crystals with controlled stoichiometry.

Various growth rates were explored to mitigate microstructure defects (including stacking faults, SF) and zone refinement allowed for a significant reduction of trace impurities. Laser light was found to induce graphitization in the presence of air, enabling a new process to fabricate electronic structures in boron carbide with micron or nanoscale precision.

This user project contributed to the development of, and benefitted from, PARADIM's first deployment of deep learning-assisted crystal growth. The algorithm tracks important variables along the lag of synthesis parameters and automatically identifies zone shape and stability—easing synthesis for all users of PARADIM.

M. Straker, *et al.* [J. Cryst. Growth](#) **543**, 125700 (2020).

M.V.S. Chandrashekhara, *et al.* [U.S. Patent No. US10981836-B2](#) (2021).

M.V.S. Chandrashekhara and Michael Spencer, Morgan State University, D. Elbert *et al.*, Johns Hopkins University & N. Schuster, Army Research Laboratory

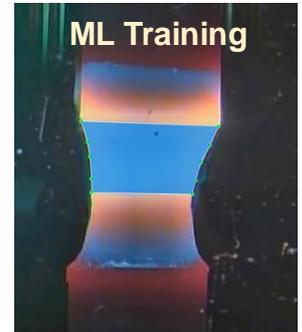
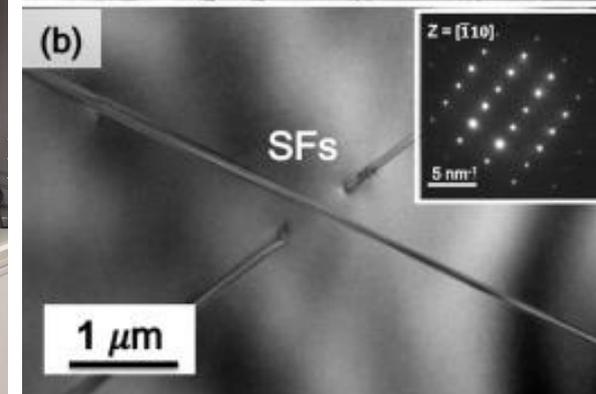
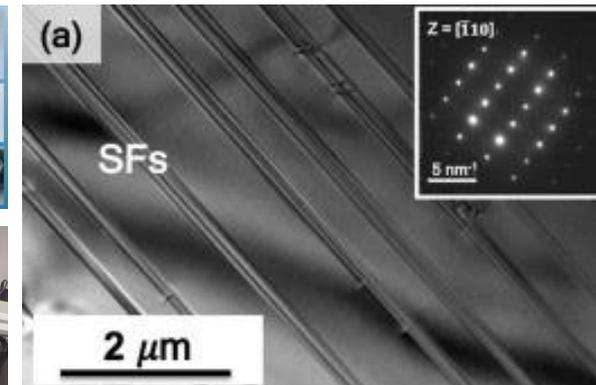


Image segmentation rapidly identifies the melt zone (labeled in red) and classifies synthesis parameters, assisting B₄C growth stabilization.

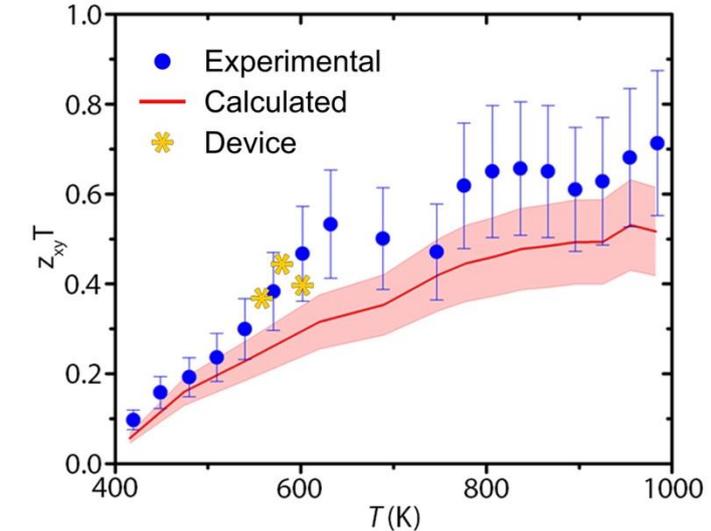
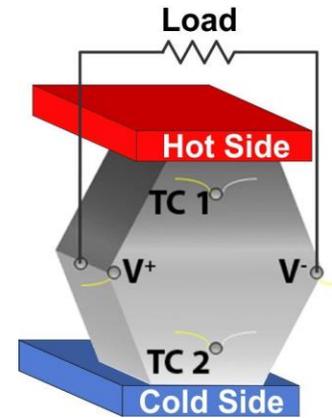
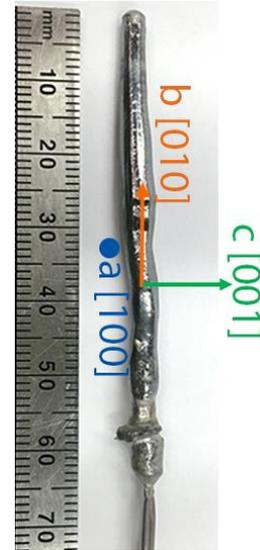
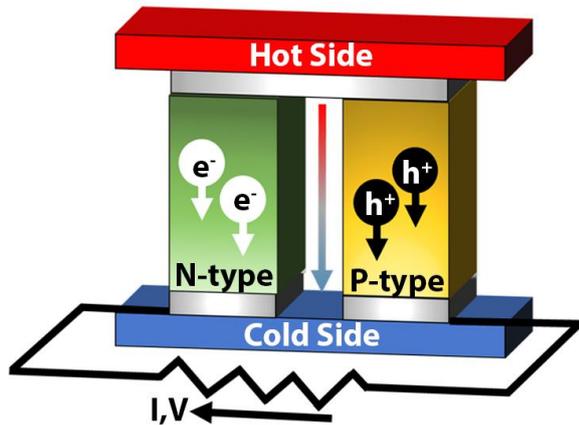


Transverse Thermoelectrics—A Route to more Efficient Energy Conversion

The direct conversion between heat and electricity that allows for active cooling or the use of waste heat for power generation is enabled by thermoelectric devices.

Joshua Goldberger, Ohio State University, with W. Windl and J.P. Heremans (both OSU) and D. Cahill (UIUC)

Traditional thermoelectric modules utilize hundreds of pairs of p-type and n-type semiconductors, where heat flows from top to bottom and the electric current meanders through the whole structure. The sheer number of metal-semiconductor contacts causes large efficiency losses compared to the thermoelectric efficiency of the bare semiconductor. Also, the need for stable contacts at the hot side limits operating temperatures and device lifetimes.



In “transverse” thermoelectric devices—developed at Ohio State with the discovery of goniopolarity—a voltage develops perpendicular to the temperature gradient, requiring only one active material and no contacts near the hot side, avoiding a major source of energy loss and device degradation.

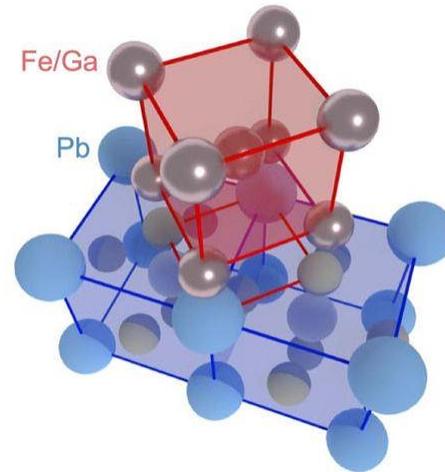
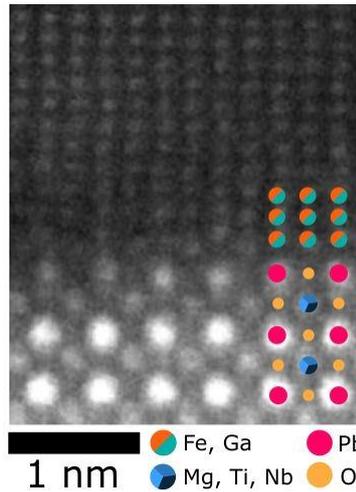
The group came to PARADIM’s bulk crystal facility to grow sufficiently large single crystals of rhenium silicide (Re_4Si_7), a material with record transverse thermoelectric efficiency. Constructing a transverse thermoelectric generator from a single crystal, the group confirmed that the device efficiency (yellow stars) shows no losses compared to the bare material (blue dots) and the calculated efficiency (red curve). The present work establishes transverse thermoelectrics as a viable technology, and rhenium silicide as the “gold-standard” transverse thermoelectric.

M.R. Scudder, *et al.*, [Energy Environ. Sci. 14 \(2021\) 4009-4017](#); covered in [NSF Science Nation video](#).

Beyond Terfenol-D

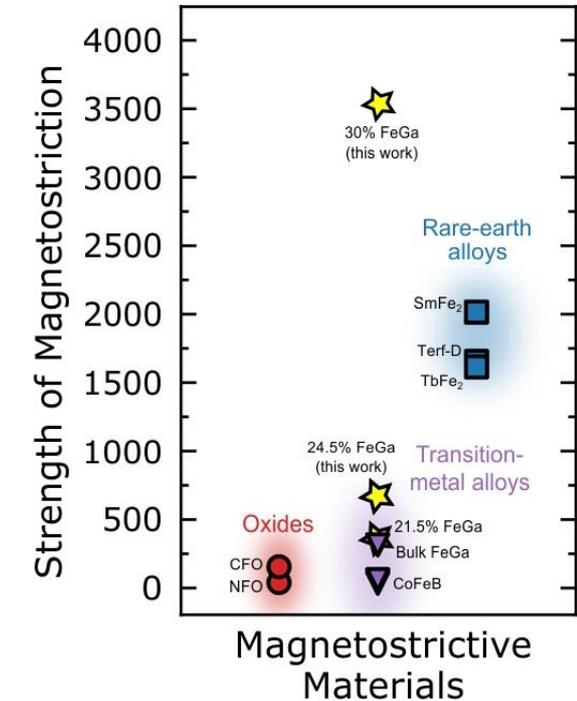
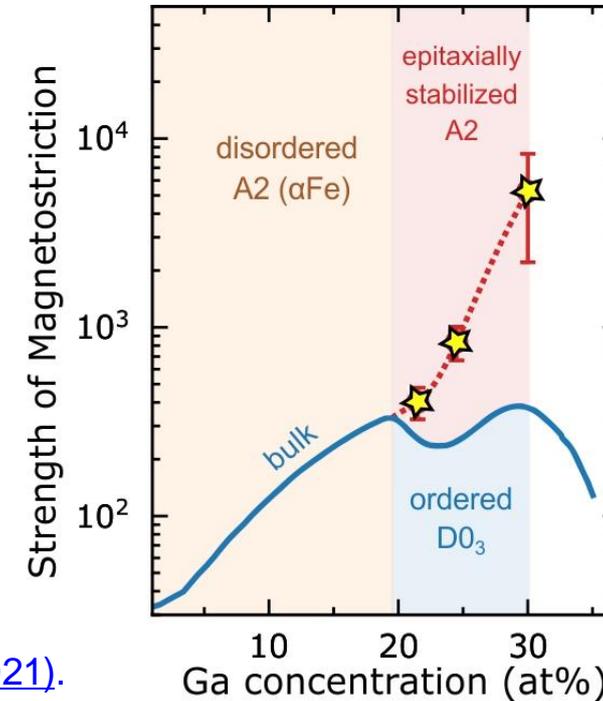
For nearly 50 years, Terfenol-D ($Tb_xDy_{1-x}Fe_2$) has reigned as the material for which an applied magnetic field results in the greatest change in shape, a property known as magnetostriction. A distant second to Terfenol-D is Galfenol ($Fe_{1-x}Ga_x$), the best magnetostrictor free of rare-earth elements.

When a magnetostrictive material is combined with a piezoelectric material, the resulting composite enables electrical control of magnetism at room temperature. Such composites were first made by gluing a high-performance piezoelectric (e.g., PMN-PT) to a magnetostrictor.



P.B. Meisenheimer, et al. [Nat. Commun. 12, 2757 \(2021\)](https://doi.org/10.1038/s41467-021-25757-2).

R. Hovden, E. Kioupakis, J.T. Heron, University of Michigan + 14 other institutions (including Intel)



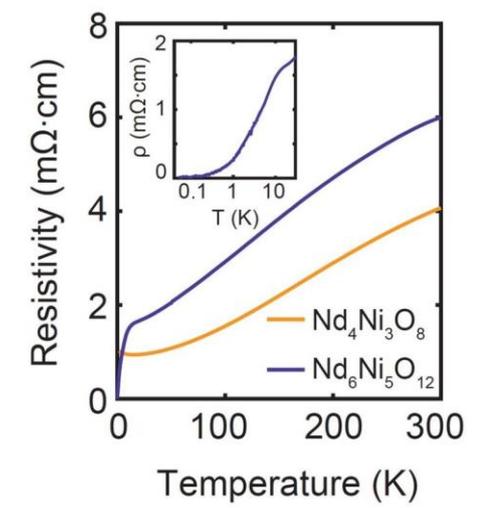
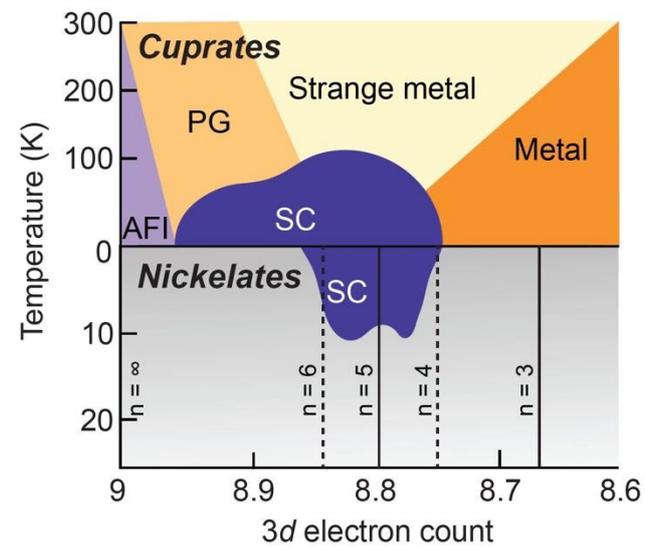
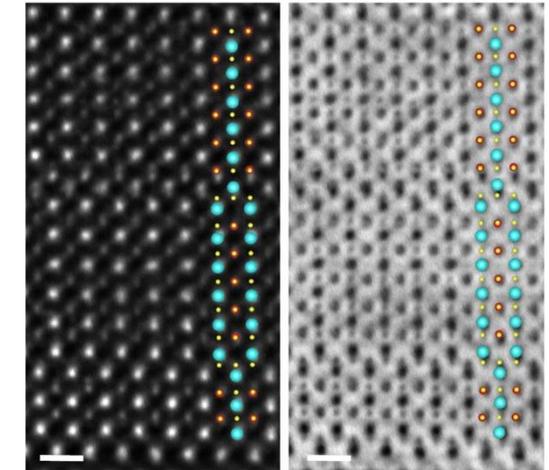
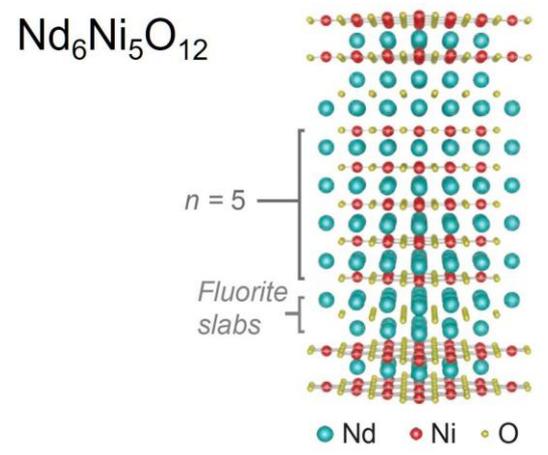
PARADIM user John Heron wanted to eliminate the glue in such a composite to achieve better coupling and higher performance. So, he came to PARADIM to make an atomically abrupt epitaxial composite using PMN-PT as a single-crystal substrate onto which he deposited Galfenol. Not only did he achieve epitaxy, but the epitaxy enabled him to stabilize the high-performance A2 magnetostrictive phase of Galfenol to higher gallium concentrations than ever before achieved. Importantly, his measurements showed that the magnetostriction in this metastable A2 phase continued to increase—becoming **10x higher than bulk Galfenol and nearly twice as high as Terfenol-D, i.e., a new record magnetostrictor has been born!** The composites utilizing this record magnetostrictor also have superb performance and calculations indicate that when optimally scaled they will provide non-volatile functionality with switching energies of ~80 aJ/bit. These epitaxial composites are thus relevant to beyond CMOS devices.

Since the discovery of high-temperature superconductivity in copper-based oxides (cuprates), there has been a sustained effort to understand its origin and to discover new superconductors based on similar building principles. Indeed, superconductivity has recently been discovered in the doped rare-earth nickelate $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$. Undoped NdNiO_2 is the infinite-layer end member of a larger family of layered nickelates, which can be explored by molecular beam epitaxy (MBE).

Here, experimentalists and theoreticians **connecting at PARADIM** engineered superconductivity through dimensional confinement in these layered systems without the need for chemical doping. Using **PARADIM's signature MBE**, the team grew various stacked films each incorporating a precisely tuned number of layers as confirmed experimentally in **PARADIM's Electron Microscopy Facility**. They demonstrated superconductivity for the 5-layer system with a critical temperature $T_c = 13$ K. The theoretical analysis reveals that the electronic structure of the 5-layer system can be understood as qualitatively intermediate between that of the cuprates and the infinite-layer nickelate. Engineering a distinct superconducting nickelate using a closed Materials-by-Design loop paves the way to explore nickelates as a new family of superconductors, which can be tuned by dimensionality and doping.

G.A. Pan, *et al.*, [available online Nat. Mater. \(2021\)](#).

Antia S. Botana (Arizona State U.) and Julia A. Mundy (Harvard)



The Highest Resolution Microscope, enabled by a new detector technology, reaches an ultimate resolution limit – the vibrations of atoms themselves

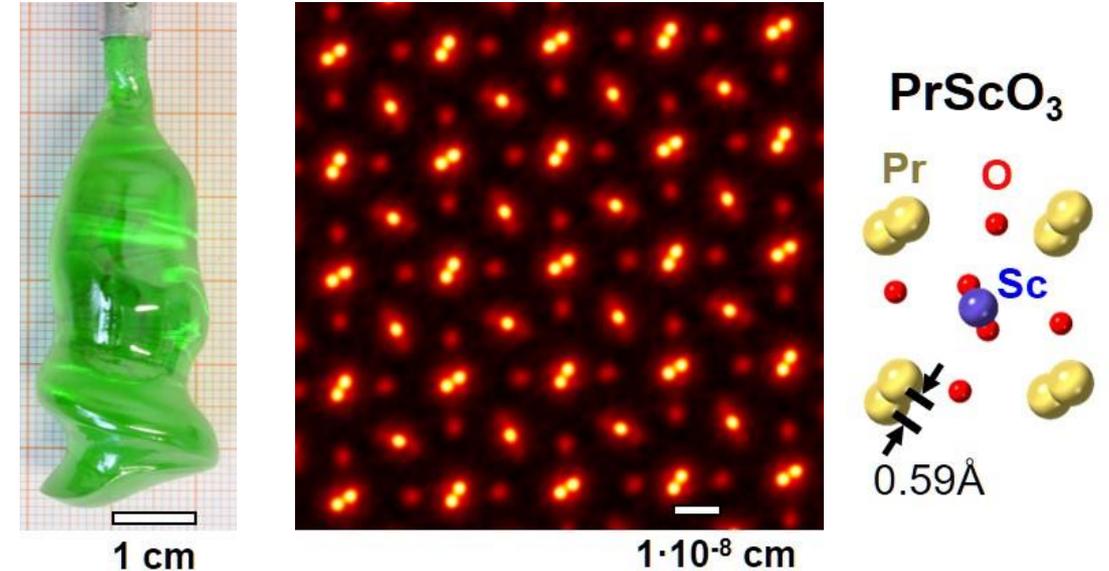
Electron microscopy is a widespread and often essential tool for structural and chemical analysis at the atomic level. Image resolution is dominated by the energy (or wavelength) of the electron beam and the quality of the lens. By combining our new design of electron microscope pixel array detector (EMPAD), which has the dynamic range to record the complete distribution of transmitted electrons at every beam position and a phase retrieval algorithm to process the data, PARADIM's in-house research team has increased the spatial resolution well beyond the traditional lens limitations, setting a world record in 2018 for the highest resolution microscope (0.39 Å Abbe resolution [1]) at the same dose and imaging conditions where conventional imaging modes reach only 0.98 Å. The EMPAD is the culmination of over a decade of detector development at Cornell, supported by NSF (through CHESS, CCMR), DOE, the WM Keck Foundation, and the Kavli Institute, and has been commercially licensed by ThermoFisher Scientific and is now manufactured and sold at scale.

Our next-generation EMPAD prototype, with an order of magnitude increase in speed and data rate, has made it possible to image thicker samples at double the spatial resolution (<0.20 Å Abbe resolution), limited mainly by the random thermal motions of the atoms themselves [2]. **This new super-resolution imaging is available to PARADIM users** utilizing the new EMPAD detector in combination with multislice ptychography in (and only in) PARADIM's electron microscopy user facility.

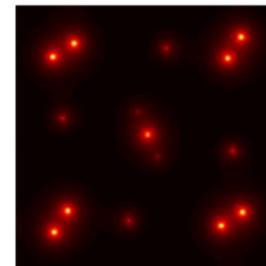
[1] Y. Jiang *et al.*, [Nature 559 \(2018\) 343–349](#);
[2] Z. Chen *et al.*, [Science 372 \(2021\) 826-831](#).

David A. Muller, Cornell University

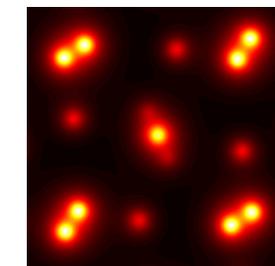
Highest resolution microscope image. Left a PrScO₃ crystal and microscope image zoomed in 100 million times.



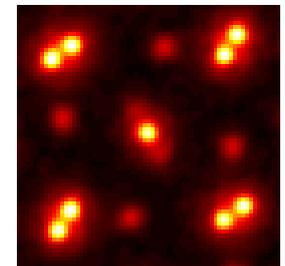
Simulation of static atoms



Simulation with thermal vibrations



Experiment

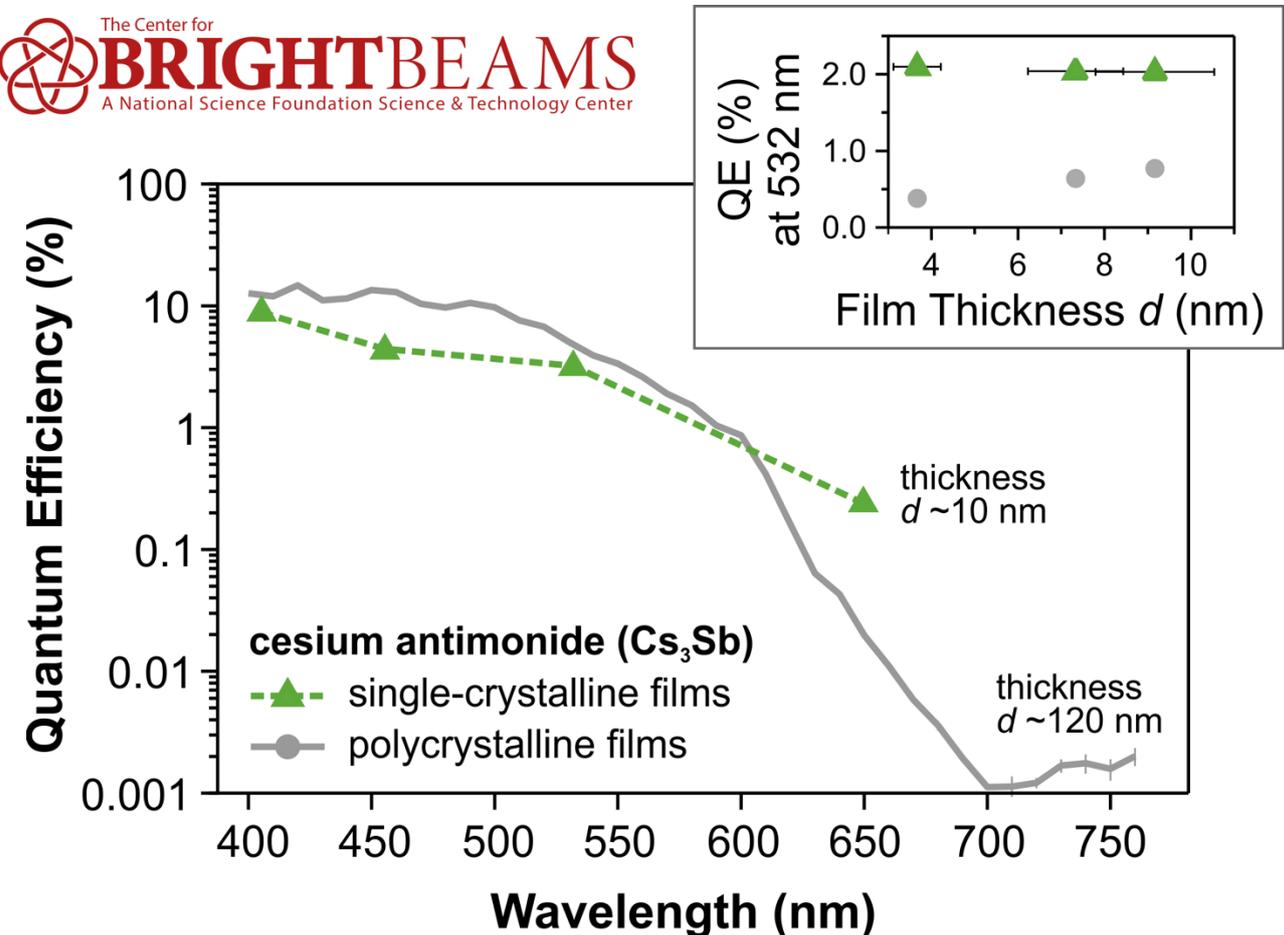


The ultimate performance of some of the most powerful characterization tools including x-ray free electron lasers, ultrafast electron microscopes, and particle accelerators are determined by the ability of their electron sources to emit electrons. This small, yet vital element of these multimillion to multibillion dollar systems, has the potential to be improved greatly; the performance of commonly used electron sources pales in comparison to the theoretical limit due to roughness, disorder, and polycrystallinity. The path to maximally efficient electron sources is thus believed to lie with single-crystal films, where the smoothness, homogeneity, and termination can be controlled at the atomic level. Unfortunately, the most desired materials for electron sources contain highly reactive species like cesium, which has stymied the preparation of single-crystal films of these desired electron sources—until now.

Scientists from NSF's Center for Bright Beams, an NSF-STC, made **use of PARADIM's molecular-beam epitaxy system and expertise in safely handling alkali metals in vacuum** to explore the growth of cesium antimonide (Cs_3Sb) thin films in single-crystal form. The precisely controlled fluxes in combination with important feedback from advanced in-situ characterization tools contributed to the success of this work.

The team, with support from PARADIM researchers, achieved films that display unusually high efficiency at thicknesses as low as 4 nm. These single-crystal films open the door to dramatic brightness enhancements for electron sources via increased efficiency, reduced surface disorder, and the possibility of engineering new photoemission functionality at the level of single atomic layers.

Jared M. Maxson (Cornell University)



C.T. Parzyck, *et al.*, *Phys. Rev. Lett.* **128**, 114801 (2022),
Access to data: doi.org/10.34863/6d5f-aj24.

Modulating Catalytic Activity using a Ferroelectric

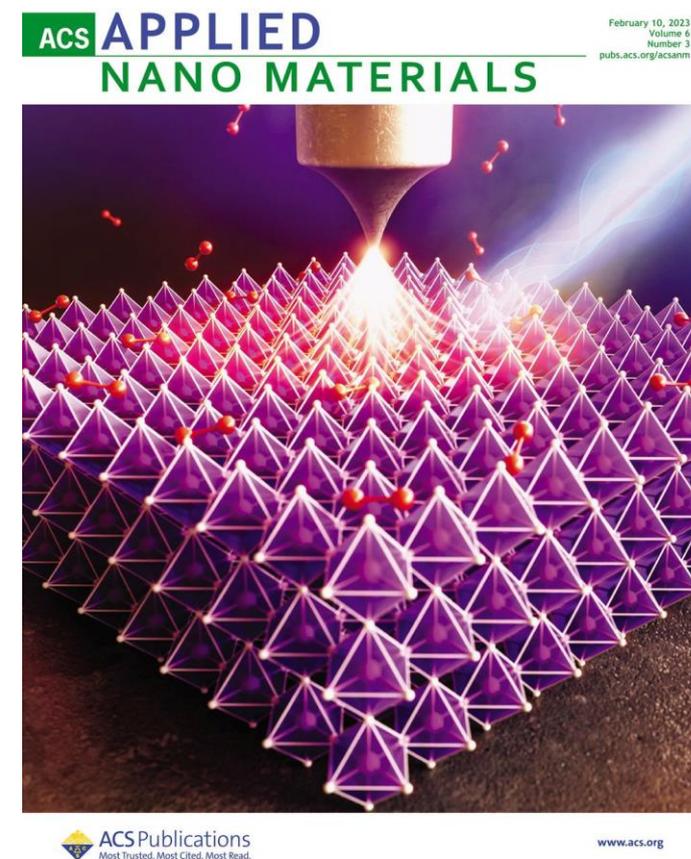
T.A. Pascal and D.P. Fenning (UC San Diego),
D.G. Schlom (Cornell), and V. Rose (Argonne National Laboratory)

Ferroelectric materials are widely utilized in nonvolatile memory, sensors, and actuators. But as a PARADIM user has recently demonstrated, the switchable structure at the surface of a ferroelectric can alter its electronic and interface properties—providing an excellent opportunity to modulate catalytic activity.

With the help of PARADIM, Fenning's group at UCSD has explored the use of MBE-grown epitaxial thin films of BaTiO_3 —a ferroelectric model compound—to study the role of polarization on the hydrogen evolution reaction (HER) by surface spectroscopy and *ab initio* DFT+U calculations [1]. The work indicates that an upward-polarized (001) surface reduces the work function relative to the downward polarization leading to a smaller HER barrier, in agreement with higher catalytic activity observed experimentally.

To further elucidate the effect of polarization switching on surface structure and chemistry the researchers teamed up **with scientists from Argonne National Lab** to study the BaTiO_3 thin films by synchrotron X-ray scanning tunneling microscopy (SX-STM), a unique method that integrates nanoscale surface imaging and chemically sensitive spectroscopy [2].

In combination with *ab initio* calculations a stronger binding strength of a model reactant (here O_2) to the upward-polarized surface is observed. The work advances the understanding of the surface chemistry and electronic structure of ferroelectrics.



[1] P. Abbasi, *et al.* [Nano Letters](#) **22**, 4276-4284 (2022).

[2] P. Abbasi, *et al.* [ACS Applied Nano Materials](#) **6**, 2162-2170 (2023).

Access to data: [10.34863/80nw-gm95](https://doi.org/10.34863/80nw-gm95).

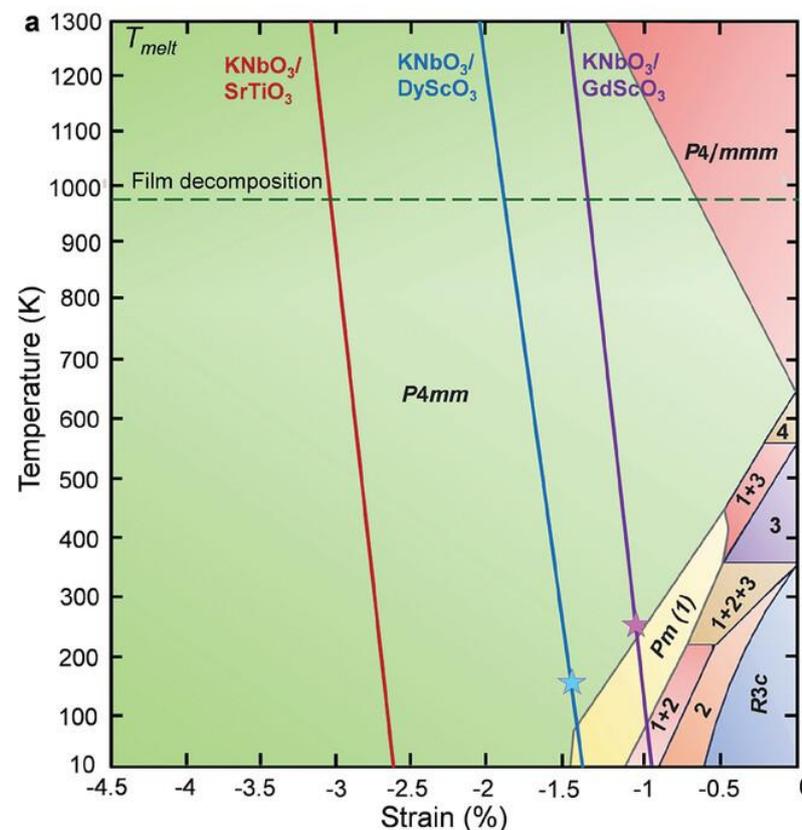
Venkatraman Gopalan, Long-Qing Chen, Susan Trolier-McKinstry, Roman Engel-Herbert (all Penn State University), David A. Muller, and Darrell G. Schlom (Cornell U)

Like people, the behavior of materials can change dramatically when put in a tight squeeze. Sometimes materials even get better when squeezed as is the case for the ferroelectric KNbO₃, a material with naturally built-in electric polarization and the opportunity to serve as an environmentally friendly and safe replacement to current state-of-the-art lead-based ferroelectrics. As the figure shows, KNbO₃ is predicted to be far more sensitive to being squeezed than common ferroelectrics. Aided by PARADIM, researchers from Penn State have been able to put these predictions of “strain tuning” to the test. The group used PARADIM’s signature molecular-beam epitaxy system (MBE) to grow KNbO₃ thin films by spraying potassium, niobium, and oxygen atoms on an underlying crystal with the same structural motif, but slightly smaller spacing between its atoms. The achieved strains exceeding 1% are found to stabilize the polarization over a much wider temperature range in agreement with theory.

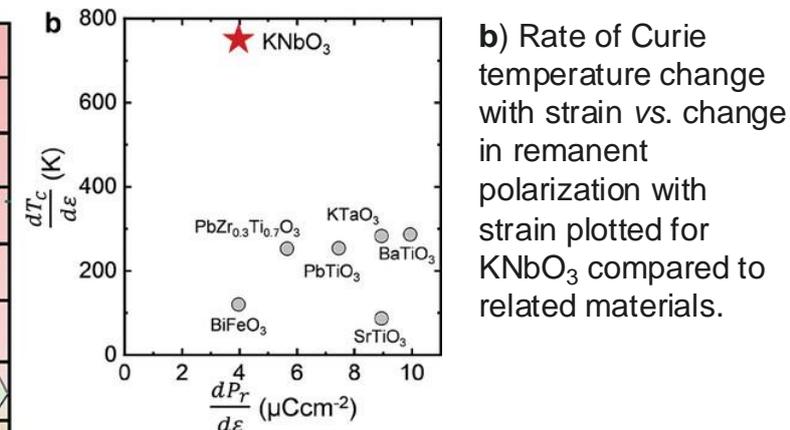
All data associated with the use of PARADIM facilities to grow and characterize the new material—some 133 GB are publicly available to fuel future materials discovery.

S. Hazra, *et al.* [available online Adv. Mater. \(2024\)](https://doi.org/10.1002/adma.202307000).

Access to data: <https://doi.org/10.34863/fs5e-s772>.

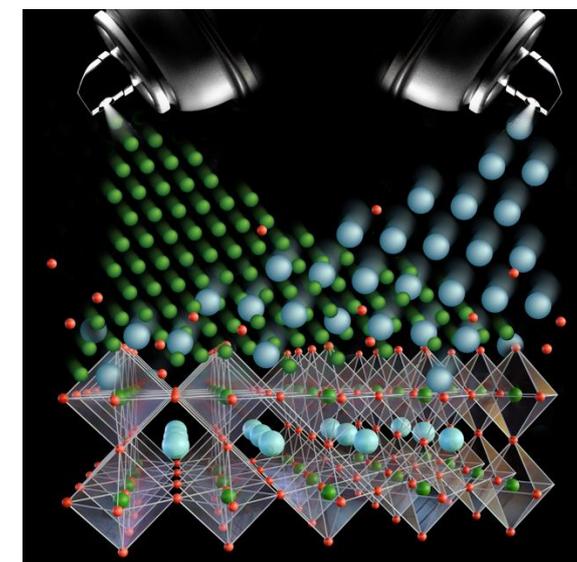


a) Thermodynamic phase-field simulations of biaxially compressed KNbO₃ on SrTiO₃, DyScO₃, and GdScO₃ marked by red, blue, and violet lines. Blue and violet stars indicate the onset of the tetragonal-to-monoclinic transition temperature observed experimentally.

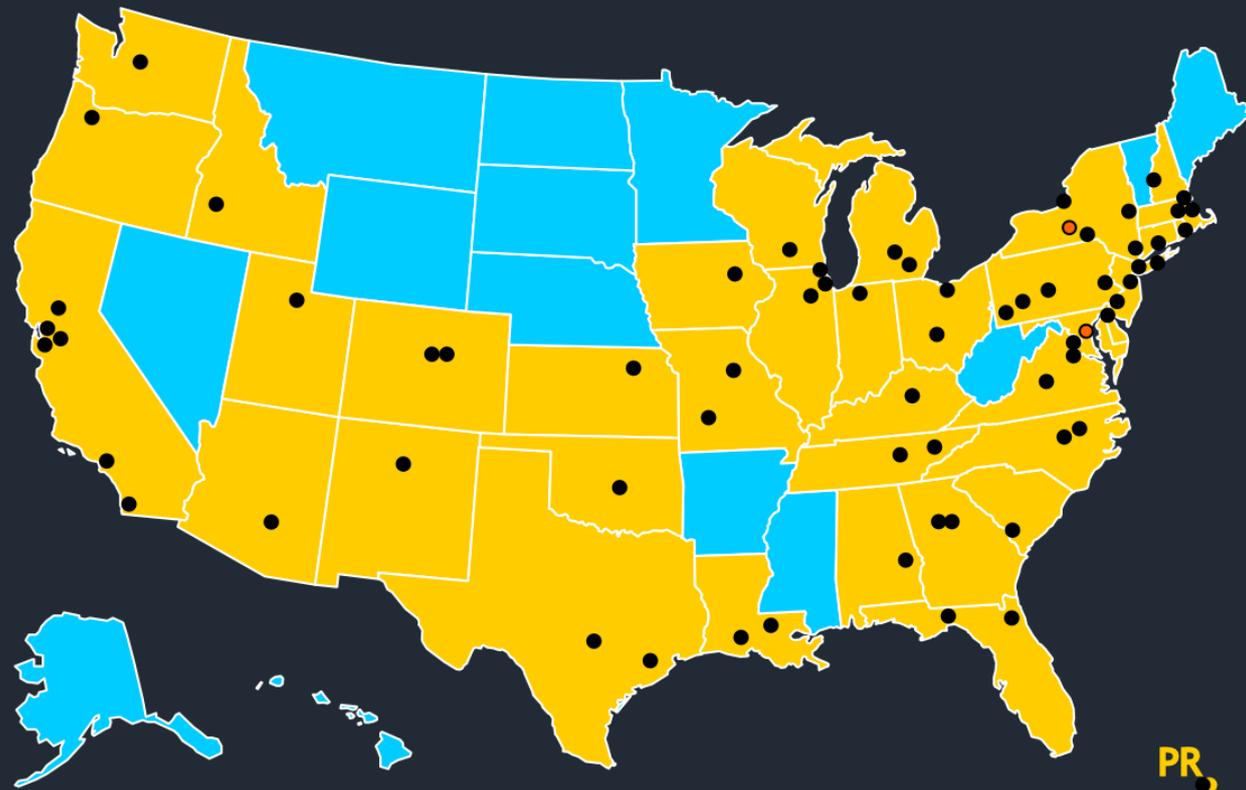


b) Rate of Curie temperature change with strain vs. change in remanent polarization with strain plotted for KNbO₃ compared to related materials.

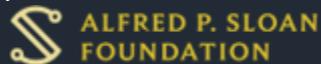
Cartoon of MBE ≈ “atomic spray painting” in action



From 36 states, DC, and Puerto Rico



Daniel Bediako (UC Berkeley)
NSF-CAREER 2022
Sloan Fellow 2024



Antia Botana (Arizona State University)
NSF-CAREER 2020 Sloan Fellow 2022



A. Shoji Hall
(Johns Hopkins University)
NSF Career 2020



Ryan Comes (Auburn University)
AFOSR YIP 2019 NSF-CAREER 2020



Robert Hovden
(University of Michigan)
DOE BES 2023



Serena Eley (Colorado School
of Mines, now University
of Washington)
NSF-CAREER 2020



Huiwen Ji (University of Utah)
NSF Career 2021



Lauren Garten (Georgia Tech)
AFOSR YIP 2021 ONR YIP 2022



Jason Kawasaki
(U. Wisconsin-Madison)
AFOSR YIP 2020



Martin Mourigal (Georgia Tech)
NSF-CAREER 2017
DOE-BES



Jian Shi
(Rensselaer Polytechn. Inst.)
NSF-CMMI
NSF-EPMD
ARO-STIR



Samaresh Guchhait
(Howard University)
NSF-MRI



Henry La Pierre (Georgia Tech)
Sloan Fellow 2022



Julia Mundy (Harvard)
DOE-BES 2021
Sloan Fellow 2022
NSF CAREER 2023



Vladan Stevanovic
(Colorado School of Mines)
NSF-CAREER 2019

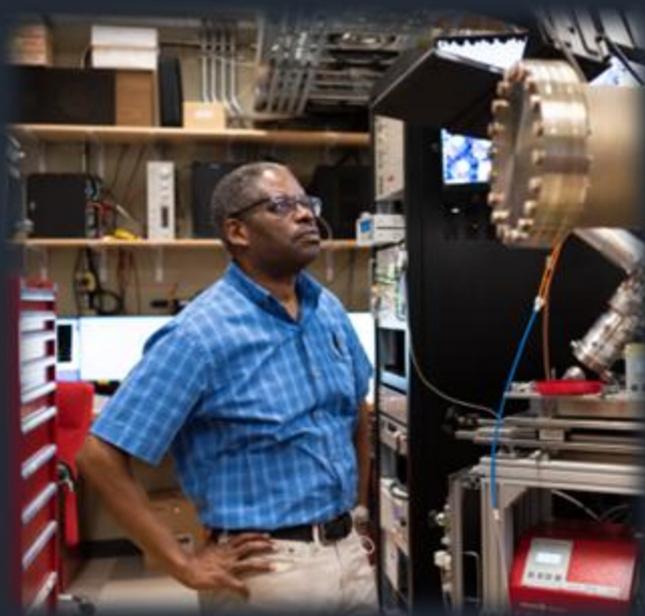




115+ REUs
(74% female or URM or non-R1)



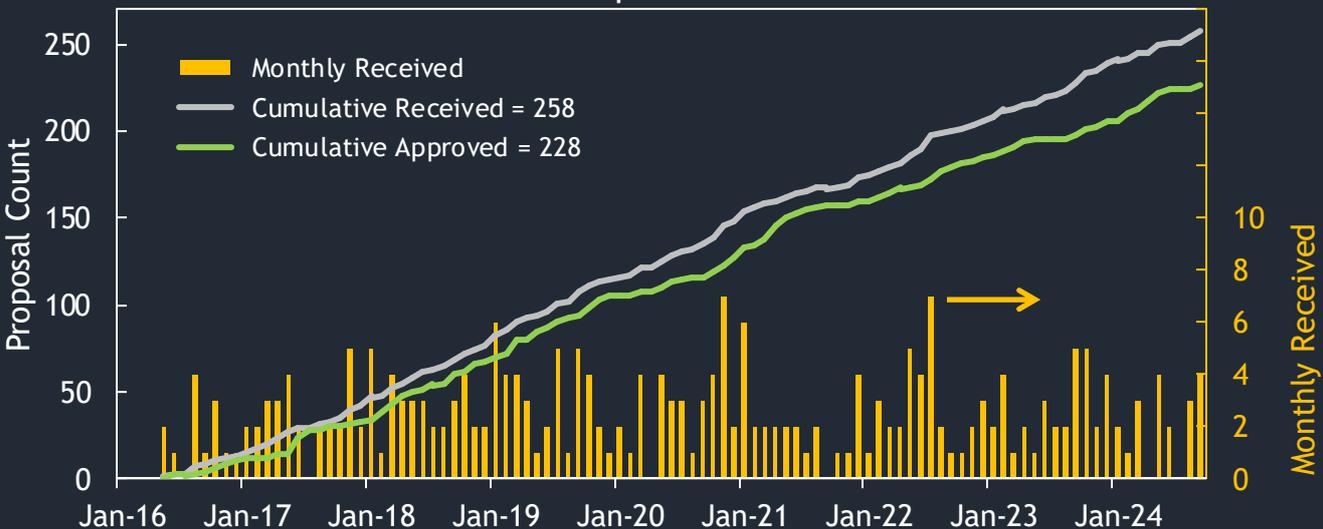
475+
on-campus
Summer School
participants to date



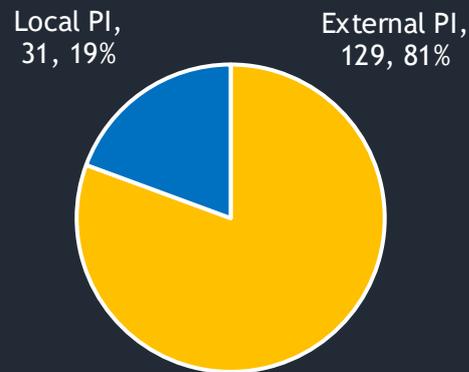
additional REUs from PREM
Kavli-PARADIM Summer/Sabbatical Fellowship
→ PREM publications (MBE + SIMS + Mössbauer)

PARADIM by the Numbers

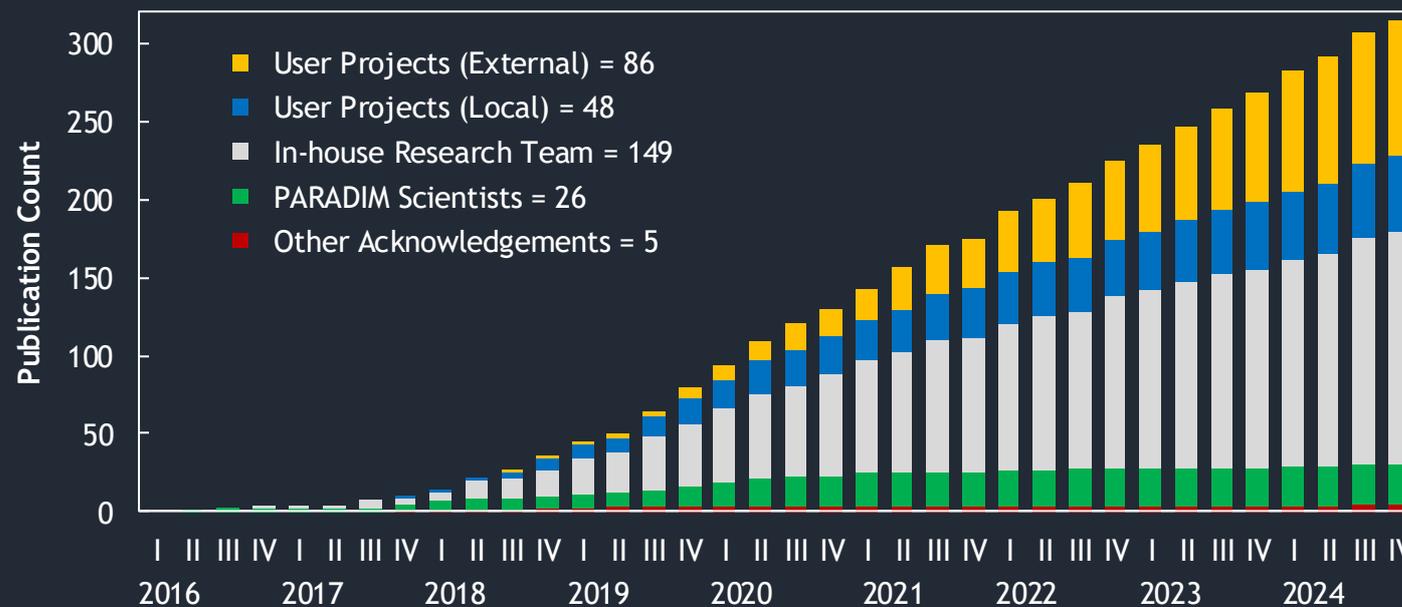
PARADIM Proposals over Time



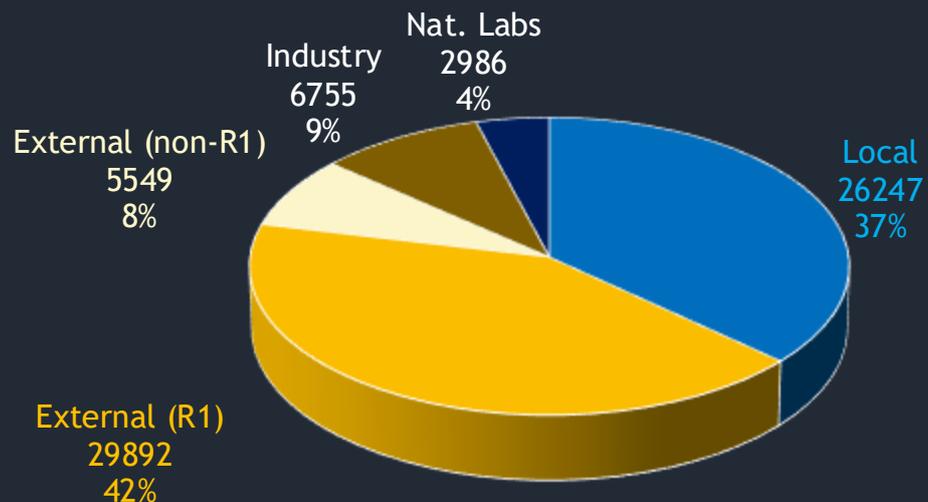
Origin of Proposal PI



PARADIM Publications over Time (Total = 314)



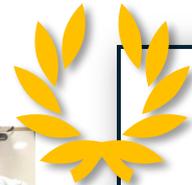
Usage of PARADIM, Total = 71429 h

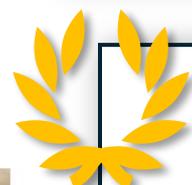


REU Participants 2016-2024

Jim Overhiser, PARADIM Director of Education and Outreach



 74% Diversity

 49% continue in STEM fields

 17 co-authors on publications (20 publications)

Participants in the PARADIM REU summer program are immersed in a vast PARADIM community that is part of larger research efforts in materials innovation. PARADIM REUs work alongside graduate students, postdocs, and Platform faculty sharing a common goal to accelerate the discovery or perfecting of electronic materials. As a result of embedding them in established networks of practitioners this diverse group of students become part of a professional, community-building program which creates a welcome pathway for these under-graduates to not only interact with the broader network but to become active participants.

PARADIM REU Alumni Success

PARADIM continues to track the success of our REU alumni. Many of our [former participants](#) are the recipients of prestigious scholarships that help them to continue their work in STEM fields and in their pursuit of higher degrees.

[NSF Graduate Research Fellowship Program \(GRFP\)](#)

Kira Martin ('23), Luc Capaldi ('21), Veronica Show ('21), Xin 'Jason' Zhang ('20), Patrick Singleton ('20), Zubia Hasan ('20), Jessica Dong ('20), Priscilla Santiesteban Navarro ('19), Cesar Lema ('18), Stephanie Eberly ('18), Heather Calcaterra ('17), and (Honorable mention) Arthur McCray ('16)

[Barry Goldwater Scholarship](#)

Ethan Ray ('22), Luke Omodt ('23)

[Rackham Merit Fellow Scholarship](#)

Priscilla Santiesteban Navarro ('19)

[GEM Fellowship](#)

Anthony Coleman ('18)

[SOROS Fellowship for New Americans](#)

Zubia Hasan ('20)

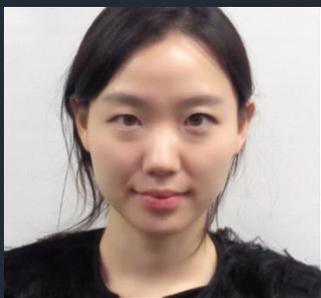
These recipients (year indicates experience at PARADIM) represent 15% of all PARADIM REUs through 2023, since the inception of the Platform in 2016. Among the 15 awardees, 6 are women (40%), 4 are URM (27%), and 6 pursued their undergraduate degrees at non-R1 institutions (40%).

Jim Overhiser, PARADIM Director of Education and Outreach



Alumni: Training the next generation

S. Chae



Oregon State

C. Chang



Seoul Nat. U.

M. Holtz



Colorado School
of Mines

K. Kang



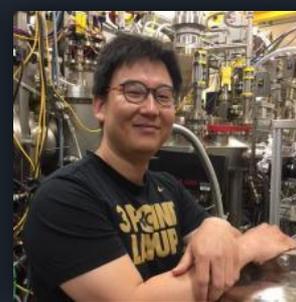
KAIST

A. Mannix



Stanford

H. Paik



U. Oklahoma

B. Pamuk



Williams
College

Assistant Professors

D. Samarakoon



Northwestern
State

S. Xie



Princeton

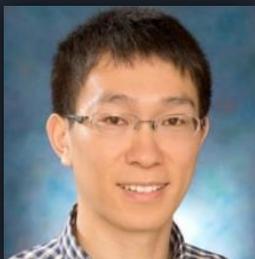
S. Yang



U. Chicago

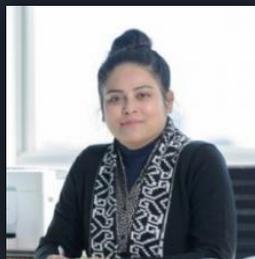
Associate Professors

Z. Chen



Tsinghua U.

H. Das



Tokyo
Inst. Tech.

G. Olsen



Norwegian U.
Sci. Tech.

Staff Scientists at National Labs

B. Godge



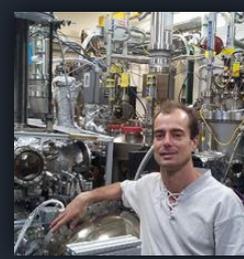
Schmidt Fellow,
MPI Phys.
Chem. Solids

A. Mesaros



CNRS

L. Moreschini



LBL

W. A. Phelan



LANL

K. Spoth



Hauptman-
Woodard

Alumni: Training the next generation

Post Docs

I. Baggari



Harvard

T. Berry



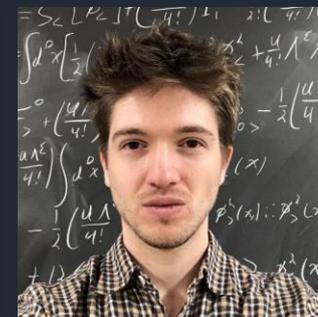
Princeton

A. Ferrenti



U. British
Columbia

E. Gerber



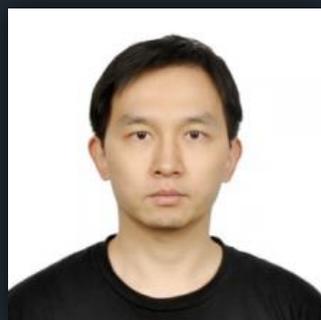
Universite Paris-
Saclay

F. Hensling



MPI Solid-State
Res.

C. Hu



LBNL

E. Lochocki



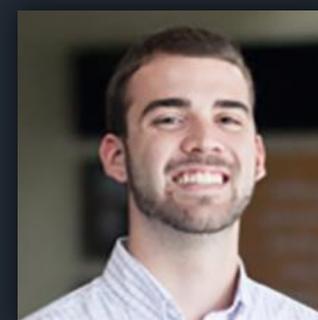
U. Illinois

M. Matty



Lincoln Labs

L. Pressley



ORNL

M.
Smeaton



NREL

Alumni: Training the next generation

Industry

E. Barr



Micron
Technology

N. Carey



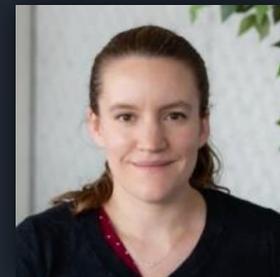
Data
Consultant

M. Kahn



Intel

J. Nelson



First Solar

N. Ng



Intel

N. Schreiber



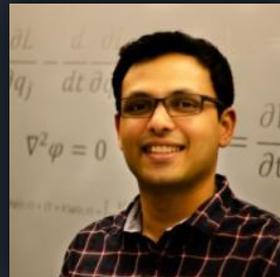
Diodes Inc.

M. Sinha



Intel

N. Sivadas



Samsung US

E. Turgut



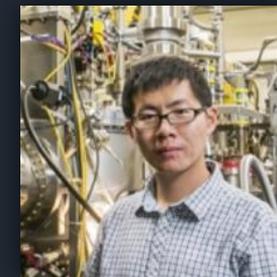
Infinera

J. Venderley



Eli Lilly

Z. Wang



ChiQuant



PARADIM

AN NSF MATERIALS INNOVATION PLATFORM

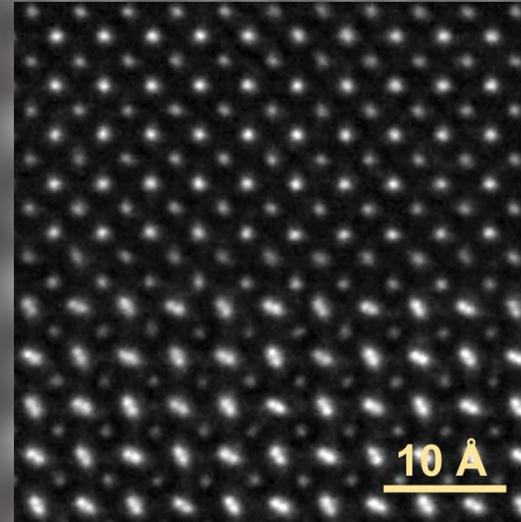
The Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials is a mid-scale research infrastructure sponsored by the National Science Foundation

Where **any scientist** can learn to create **any electronic material**



Thin Film Growth

62-element MBE (and **S-MBE**)
 T_{sub} up to 2000 °C
in vacuo spin- and angle-resolved photoemission spectroscopy (MBE+ARPES)



Electron Microscopy

World-record resolution STEM with in-house developed detector (EMPAD) and cryogenic capabilities, site-specific analysis down to a single atom

National User Facility

Free access and staff support for domestic academics on approved proposals for the discovery of Novel Electronic Materials (travel support available for non-PWI)

Summer Schools

Free annual programs offering training in the Materials-by-Design process and the unique tools available at PARADIM

Research Experience for Undergraduates (REU)

Each summer, a 10-week opportunity to immerse in hands-on research in our world-class research facility

Kavli-PARADIM Summer Fellowship

A program to bring faculty and their students from HBCU or other URM-serving institutions to PARADIM to build lasting connections

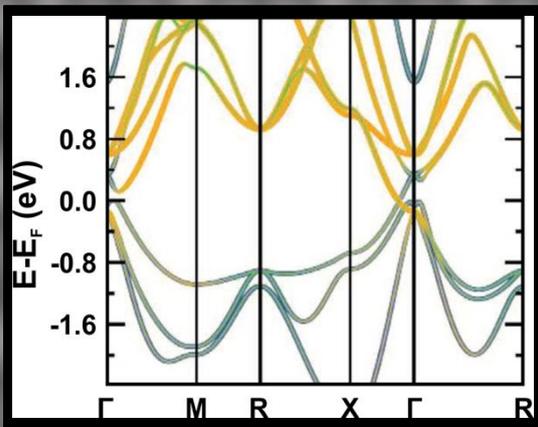
Free Access National User Facility

Theory & Simulation

Molecular dynamics and ab initio approaches, Materials-by-design toolbox, cloud-based analytics, big data infrastructure, and codes & tutorials

Bulk Crystal Growth

World-leading suite with all floating zone techniques including high-pressure (300 bar), *in situ* monitoring and machine learning (ML)-assisted growth



www.paradim.org