Quantum Materials Fabrication and Analysis at BNL User Facilities

Mingzhao Liu

Co-design Center for Quantum Advantage (C^2QA)

Center for Functional Nanomaterials

Workshop on DOE User Facilities for Quantum Information Science

January 12-13, 2022

@BrookhavenLab

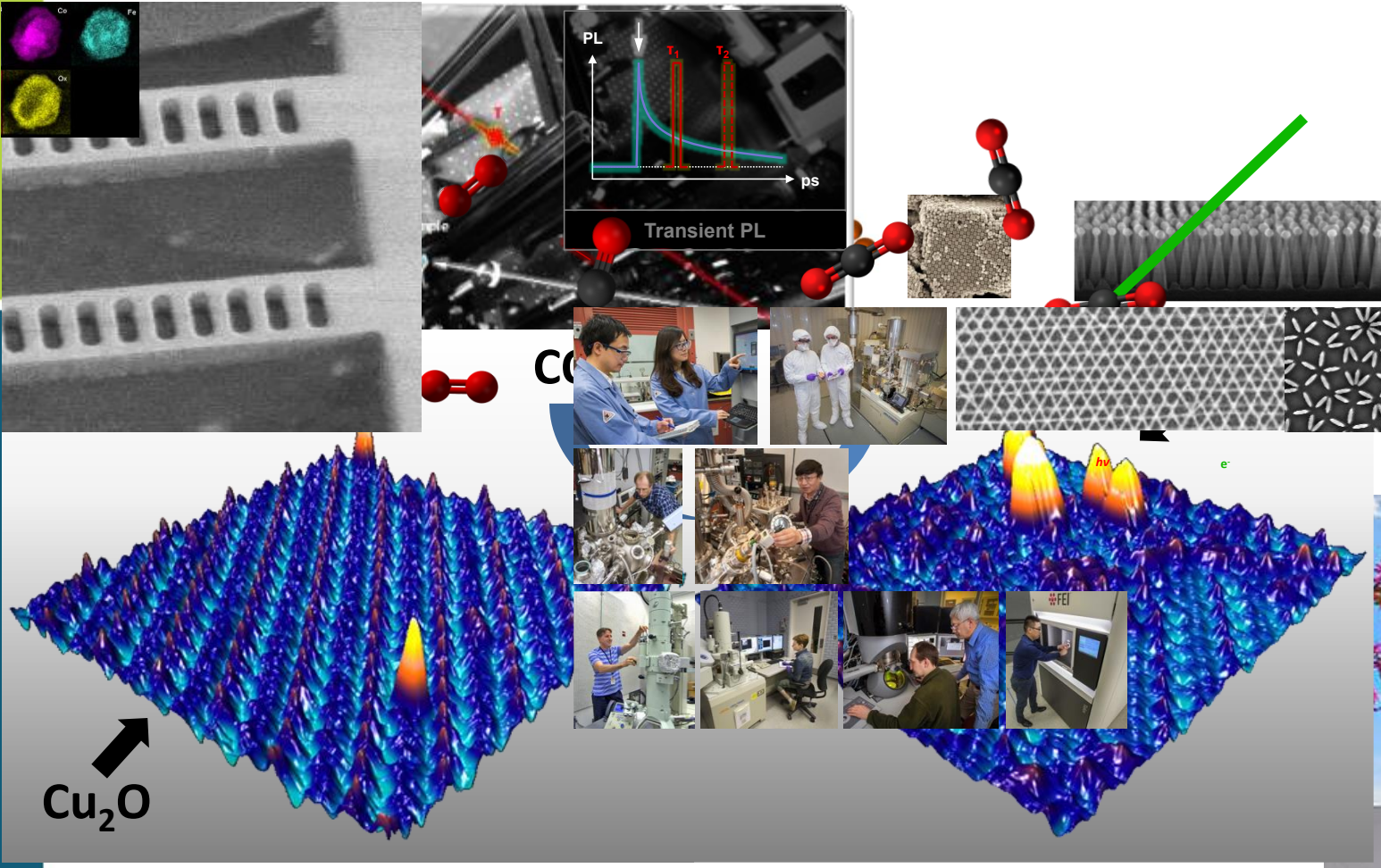
BNL team for QIS user support

NSLS II

A. Barbour, G. L. Carr, S. Hulbert, A. Hunt, I. Jarrige, M. K. Liu, Q. Shen, E. Stavitski, A. Walter, I. Waluyo, C. Weiland

CFN

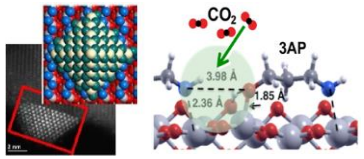
C. T. Black, G. Doerk, A. Head, S. Hwang, K. Kisslinger, M. Z. Liu, S. Park, J. Sadowski, D. Stacchiola, A. Stein, K. Yager



- One of 5 DOE Nanoscience Research Centers (NSRCs)

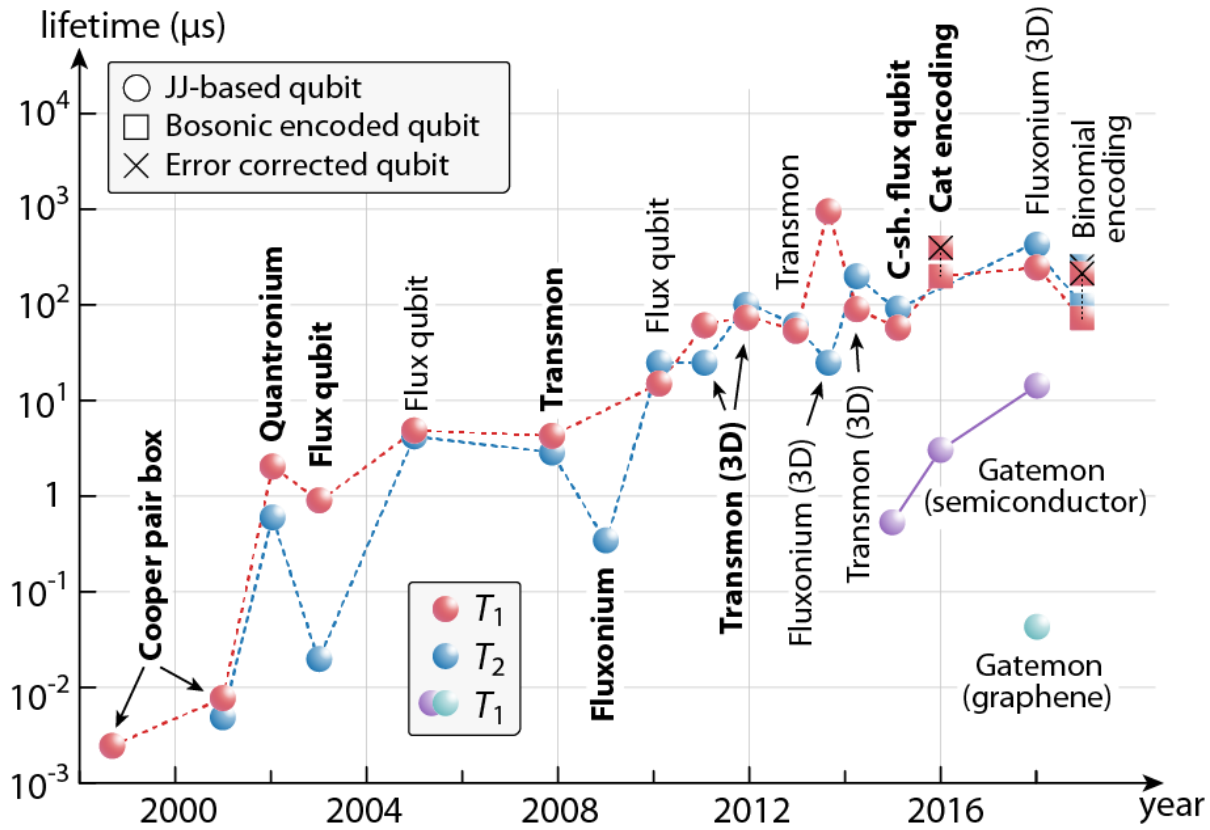
Advanced X-ray and UV Probes

Theory and Computation



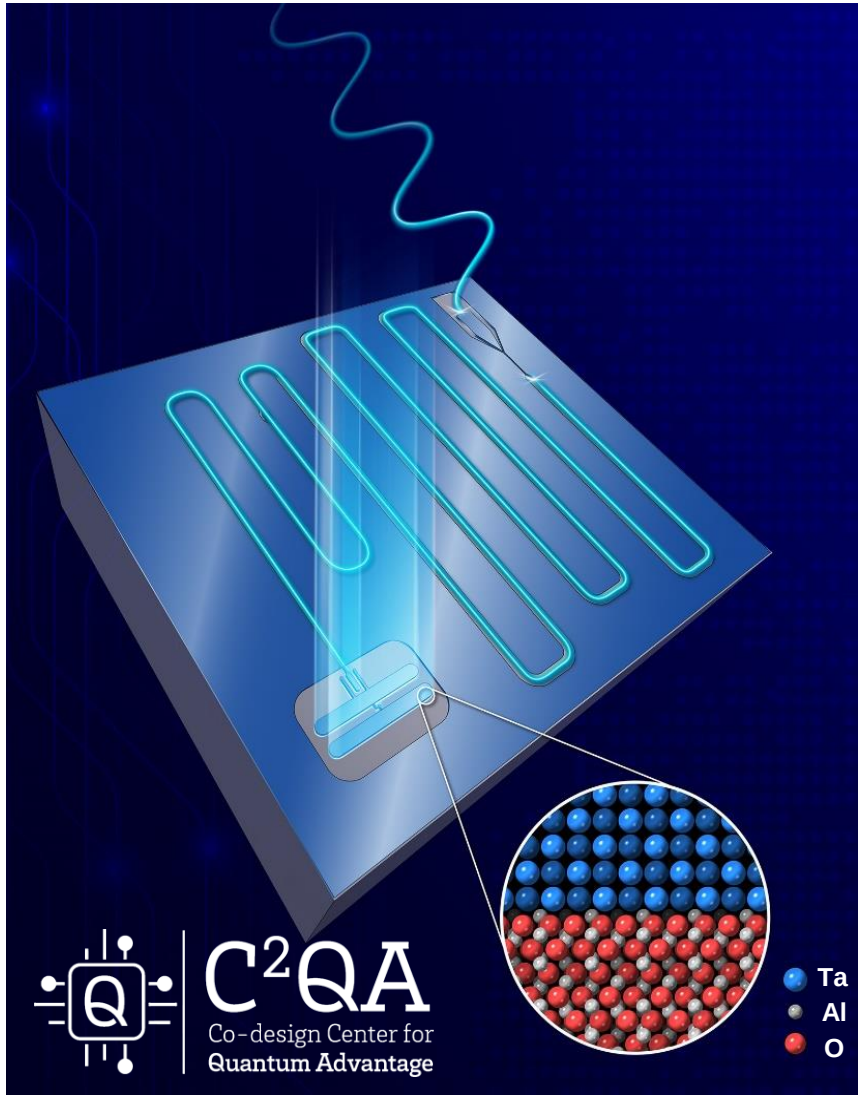
Brookhaven National Laboratory

Understand and mitigate superconducting qubits loss



Kjaergaard *et al*, arXiv:1905.13641v1

- T_1 for 2D transmon qubits had been limited to ~0.1 ms due to TLS loss at capacitor/resonator surface



Brookhaven National Laboratory

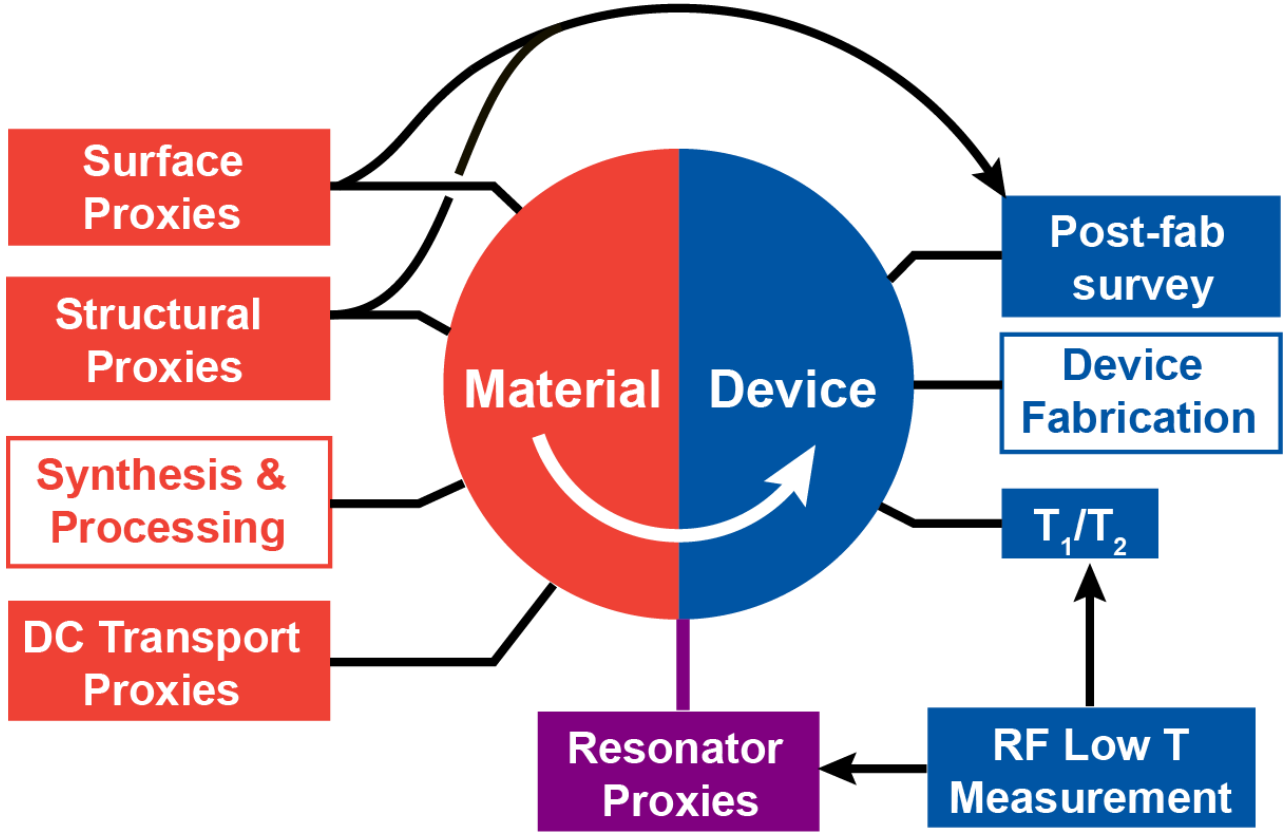
Correlate material properties with qubit performance

Synchrotron
X-ray spectr.

TEM;
X-ray diffraction

Magnetron
sputtering

PPMS

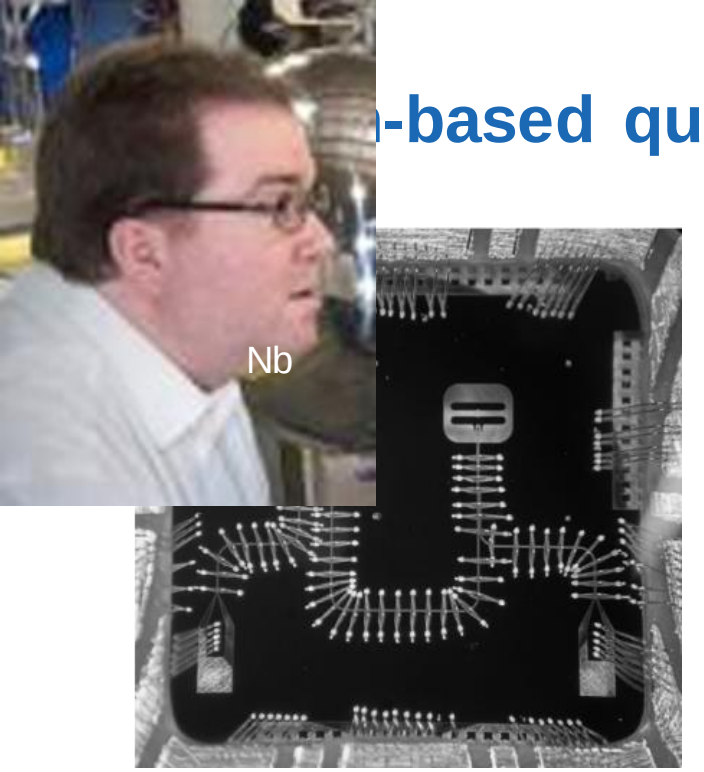


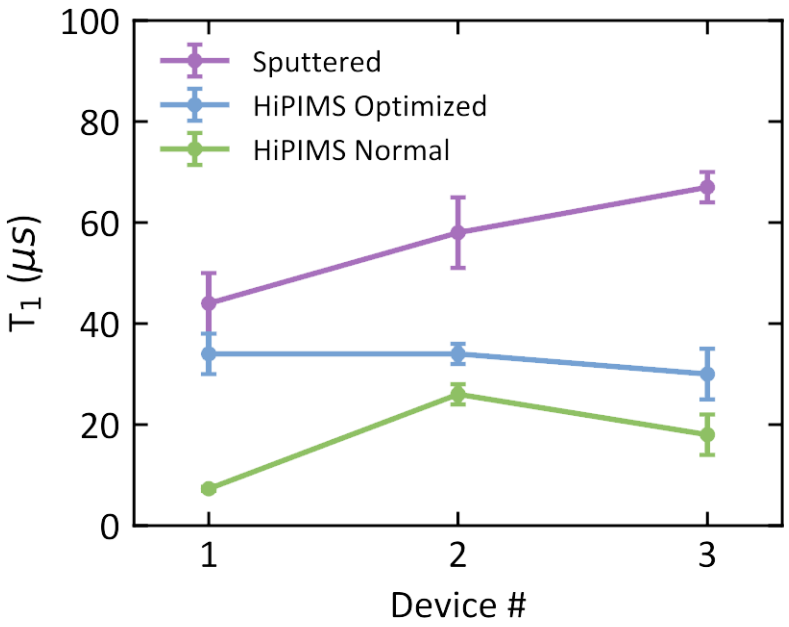
Cleanroom
facilities

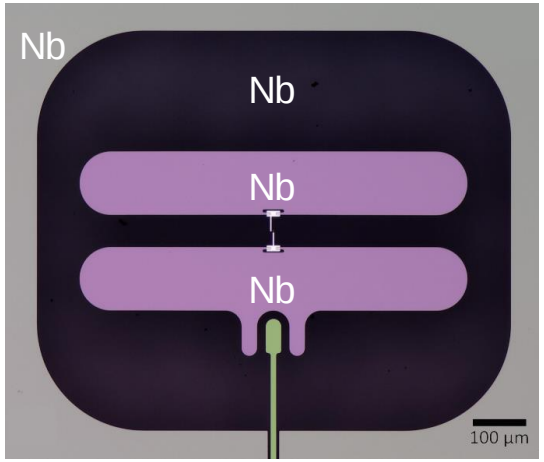
Dilution refrigerator
(Princeton, Yale)

- Identify key material proxies to infer qubit coherence at low T
- Build stronger synergy between BNL and C^2QA device efforts

-based qubits

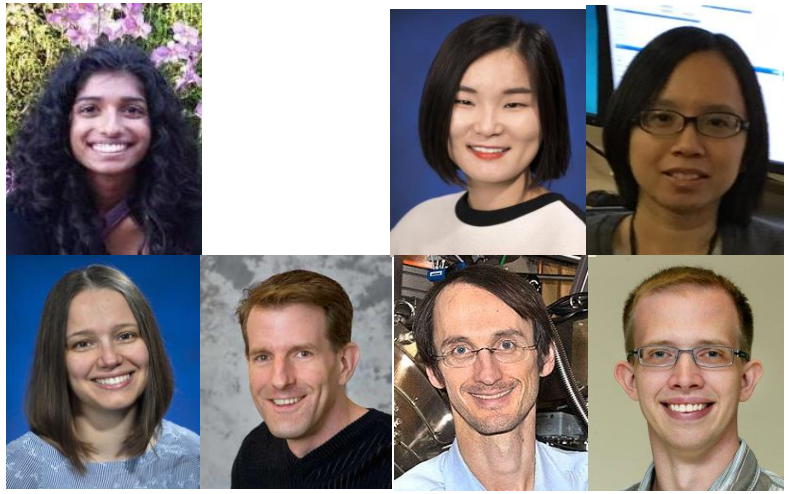






Deposition	T_1 (µs)
Sputtering	55
HiPIMS opt	35
HiPIMS norm	17

Princeton-BNL team

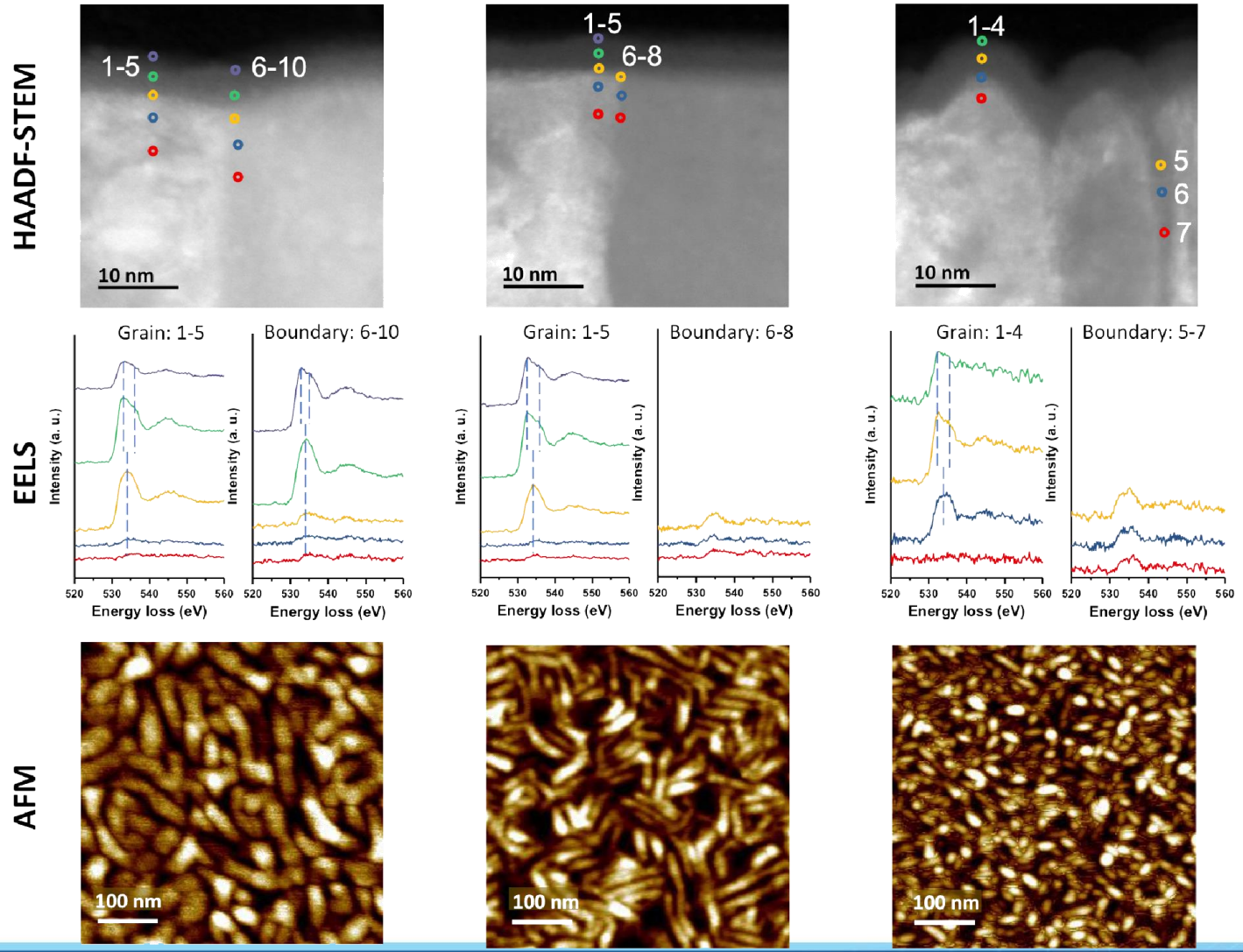


A. Premkumar et al, *Commun. Mater.* **2**, 72 (2021)

- Qubit device with Nb capacitor/resonator and AlO_x/Al/ AlO_x JJs
- T_1 dependence on Nb deposition technique
  - HiPIMS = High-power impulse magnetron sputtering

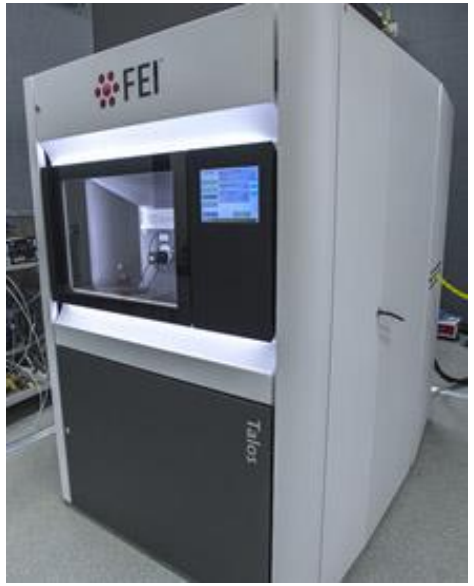
Oxidation at surface and grain boundaries

Best — Sputtered | HiPIMS Optimized | **Worst** — HiPIMS Normal

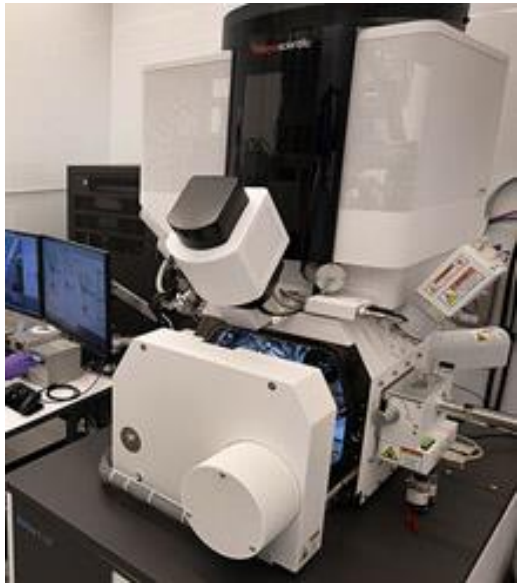


- HiPIMS Normal (worst) film has smallest grains
- Native oxide and oxygen diffusion along grain boundaries

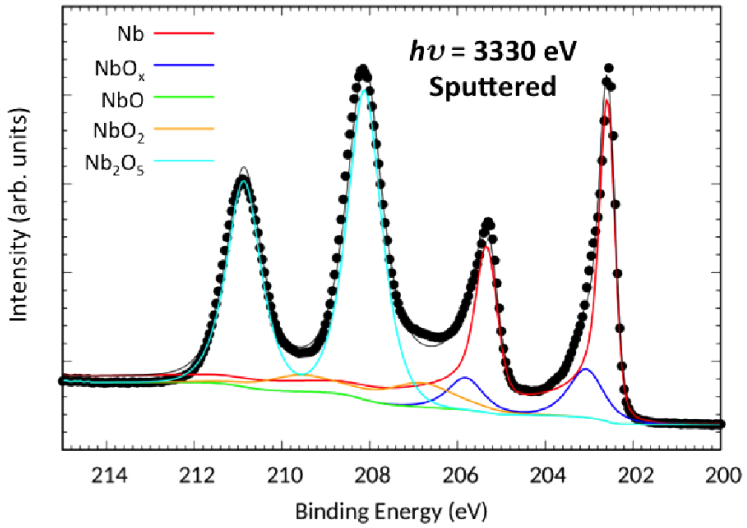
FEI Talos F200X (CFN)

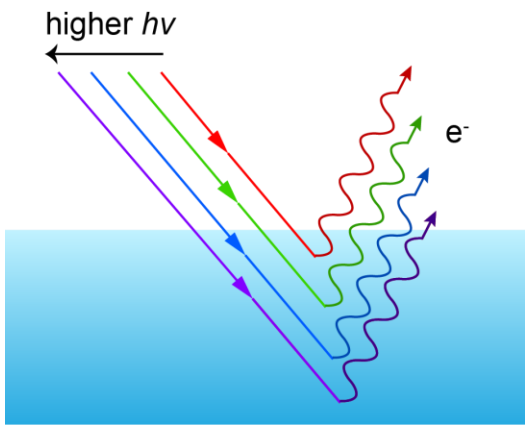


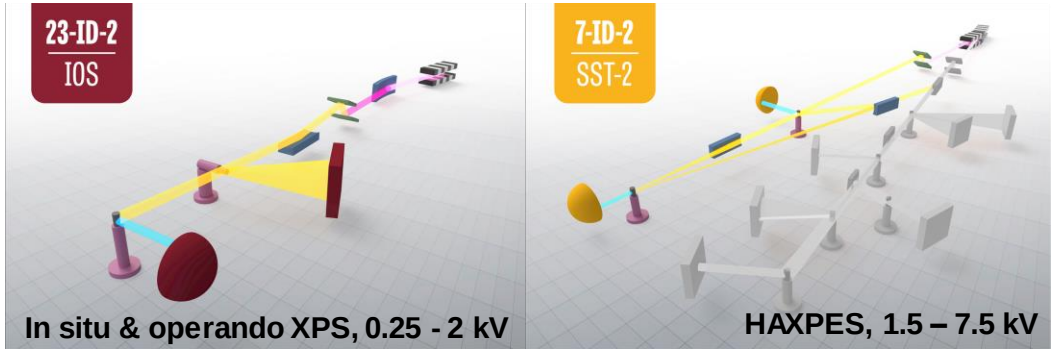
FEI HELIOS FIB (CFN)

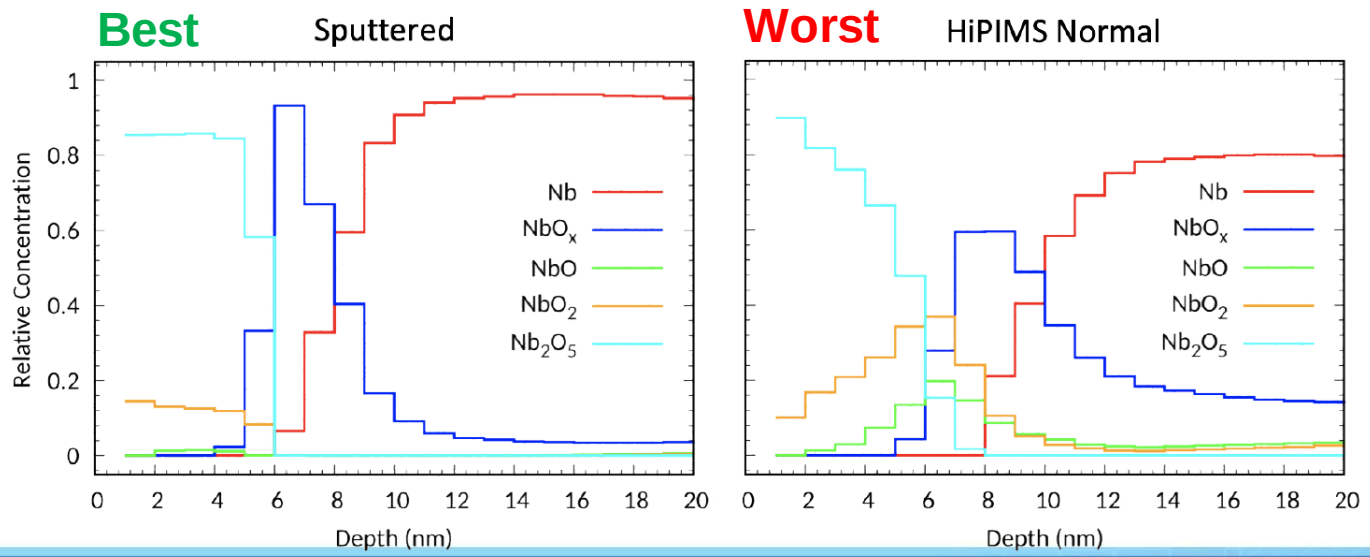


Nb surface chemistry studied by XPS depth profiling



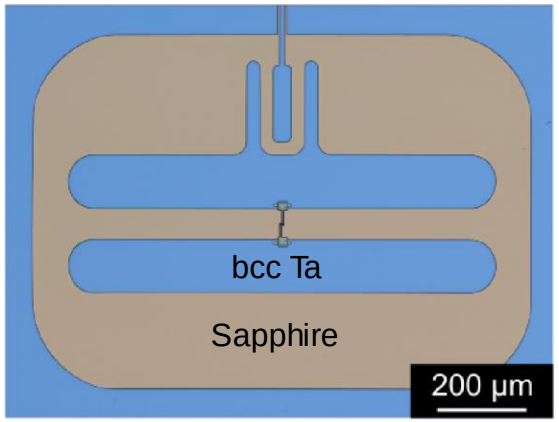


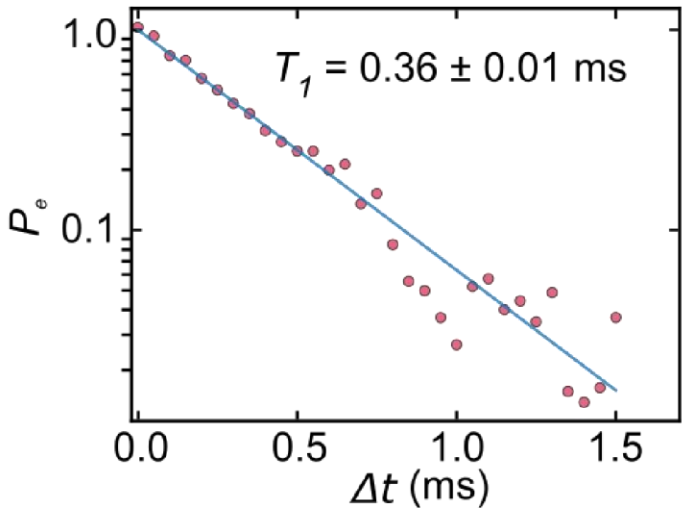




- XPS profiling with 7 different incident energies
- Native oxide on Nb is about 10 nm thick
- Nb forms suboxides beyond Nb_2O_5
- Worst film forms most diffusive oxide layer with high fraction of suboxides

Tantalum-based qubits



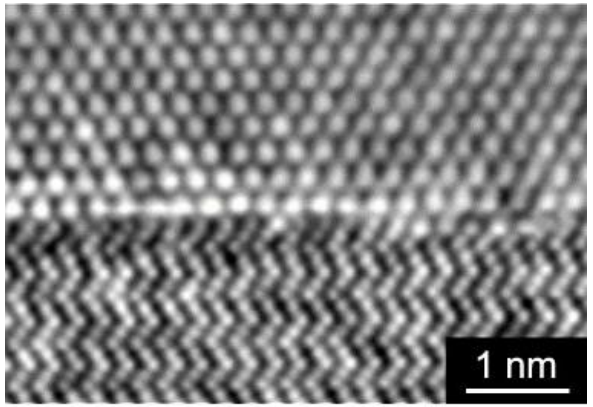


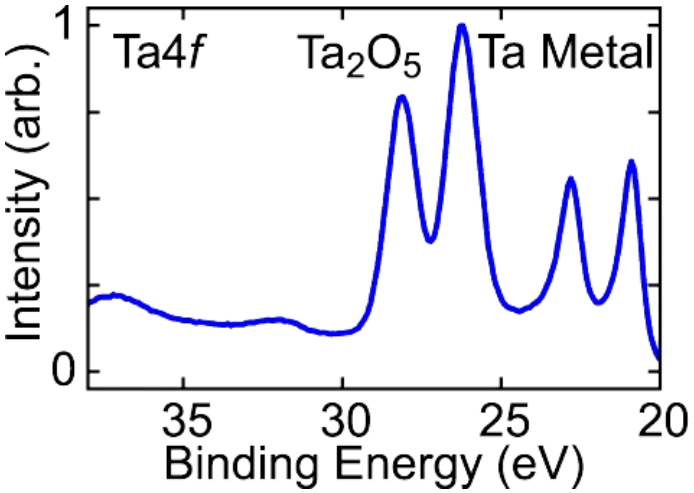
R. Cava N. de Leon A. Houck

A. Place et al, *Nature Commun.* **12**, 1779 (2021)

Ta

Al_2O_3





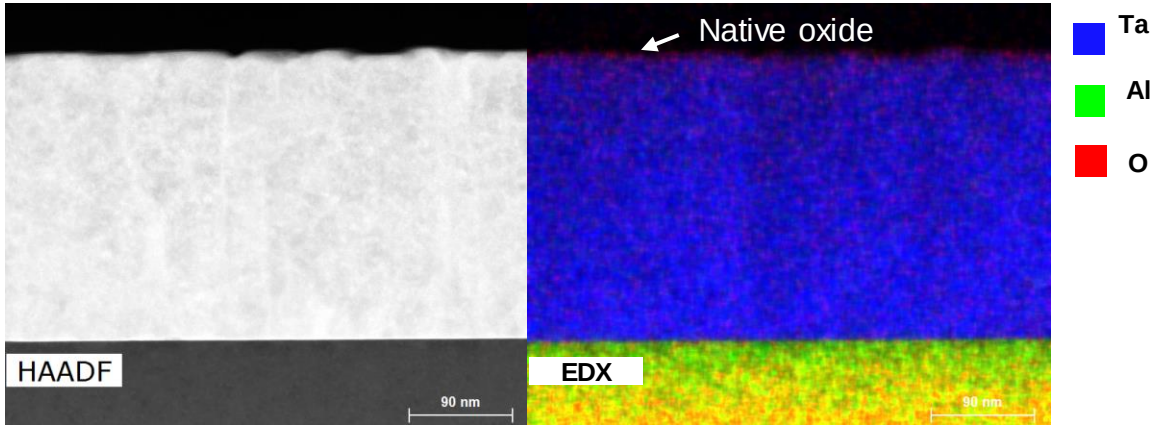
- Replacing Nb with *bcc*-Ta (T_c = 4.3 K) brings 3x longer T_1
- Ta can grow epitaxially over sapphire
- Native oxide on Ta is thin and simple (Ta^{5+} dominated), but still an oxide (likely amorphous)

Results from Princeton

Ta still oxidizes

Resonator *Q* vs chemical processing

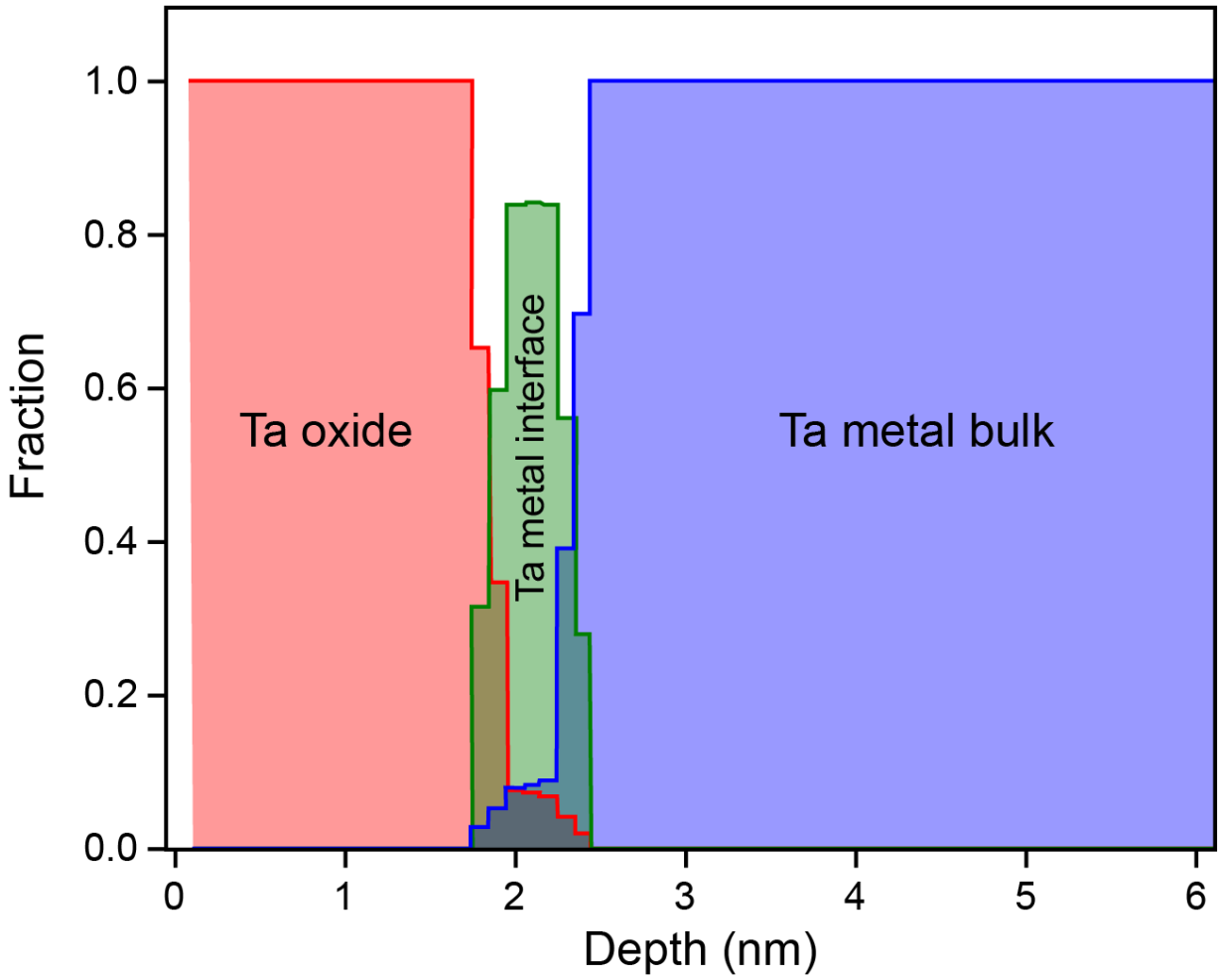
de Leon, Houck (Princeton)



FEI Talos F200X (CFN)

- Forming native oxide of 2-3 nm thick, universal across all sources
- Amorphous, host of TLS
- Surface treatment may lower loss
- Resonator *Q* as proxy for qubit lifetime

In-situ/ex-situ XPS depth profiling



Princeton-BNL team

A. Barbour, N. de Leon, A. Dutta, S. Hulbert, A. Hunt, M. Liu, A. Walter, I. Waluyo, C. Weiland, Y. Jia

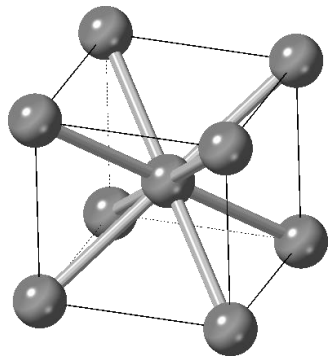
7-ID-1 / SST-1 — **HAXPES 0.15– 2.2 keV**

7-ID-2 / SST-2 — **HAXPES 1.5– 7.5 keV**

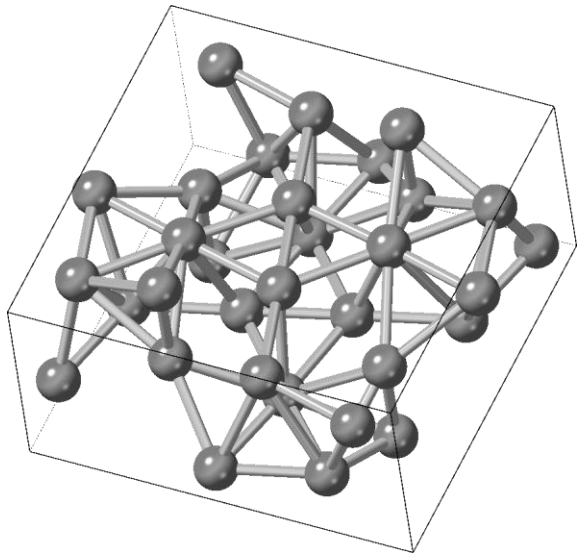
23-ID-2 / IOS — **In situ and operando XPS 0.25 - 2 keV**

- XPS profiling w/ 17 incident energies
- Study of oxide variation by wet/dry chemical treatments
- In situ study of oxide removal/regrowth during annealing

Polymorphism of tantalum

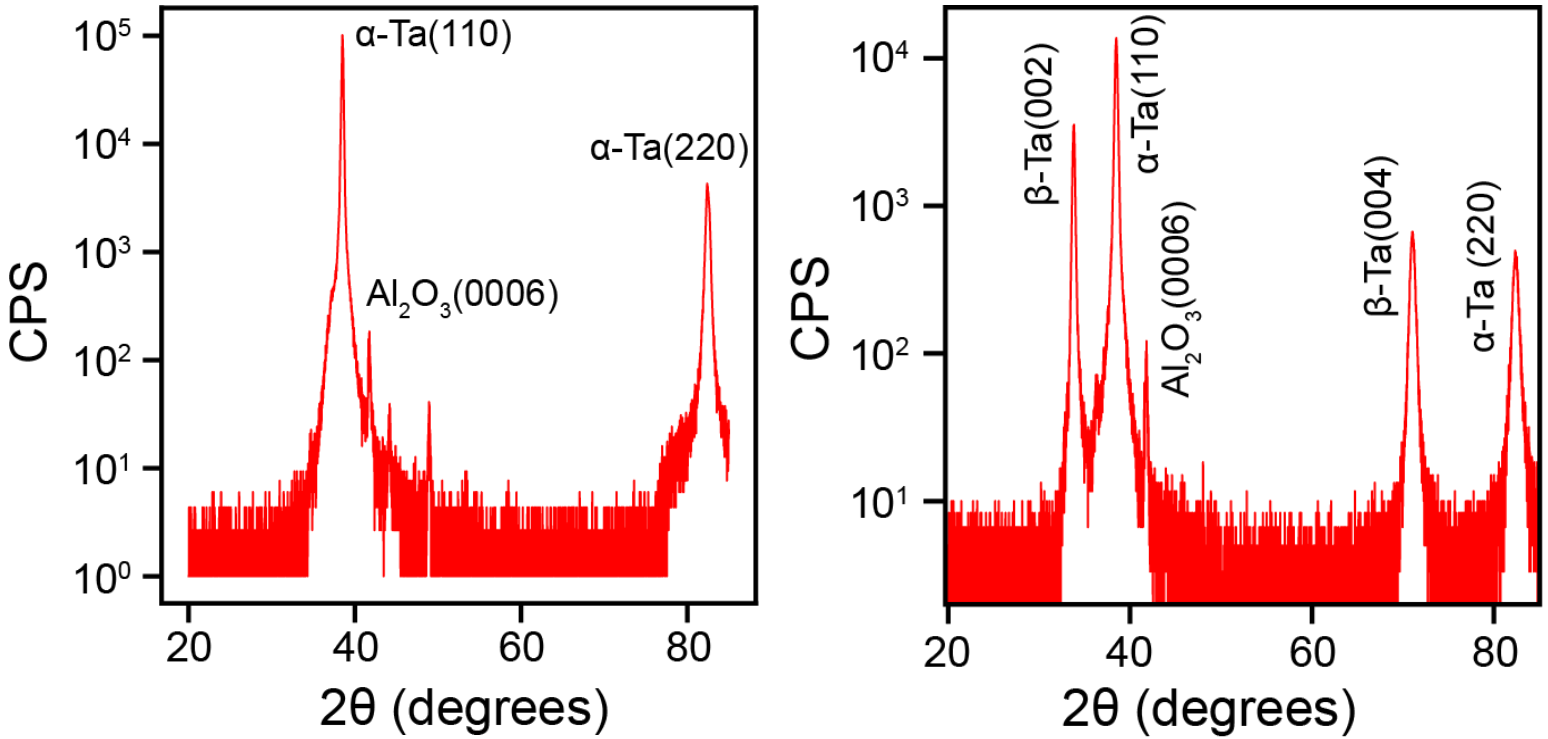


α-Ta (bcc)



β-Ta (tetragonal)

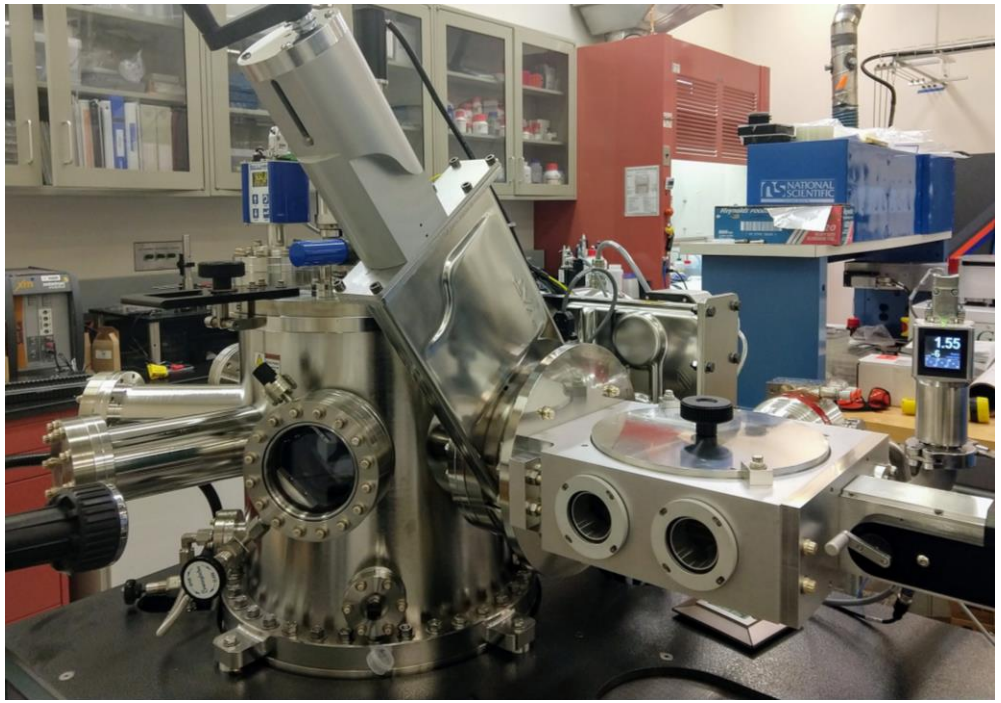
Two Ta films deposited at similar conditions



- Sputter deposition of Ta easily leads to a metal stable β–phase
- A superconductor with Tc ~0.8 K
- Undesired due to more quasiparticle contribution

Robust fabrication of crystalline, bcc-Ta film

AJA Orion Sputter (CFN)



Rigaku SmartLab XRD(CFN)

- Yale-Princeton-BNL team
- Film growth + XRD survey
- High sensitivity to substrate temperature

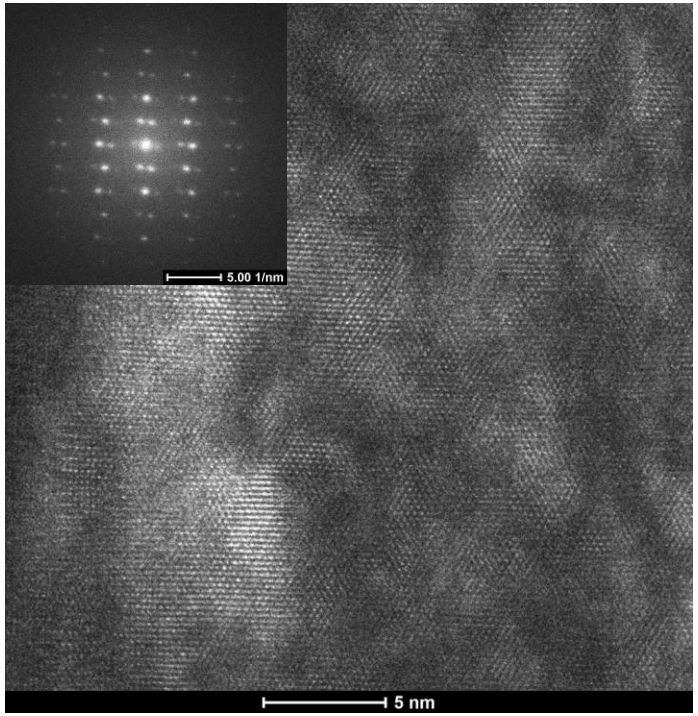
Brookhaven National Laboratory

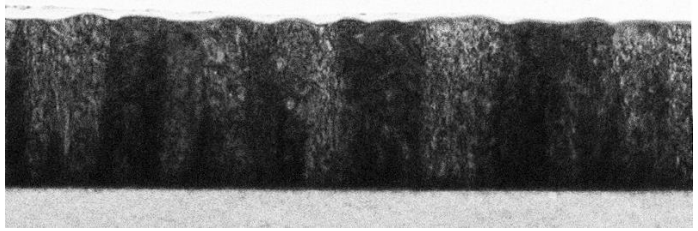
Electron microscopy

Electron Microscopy (CFN)

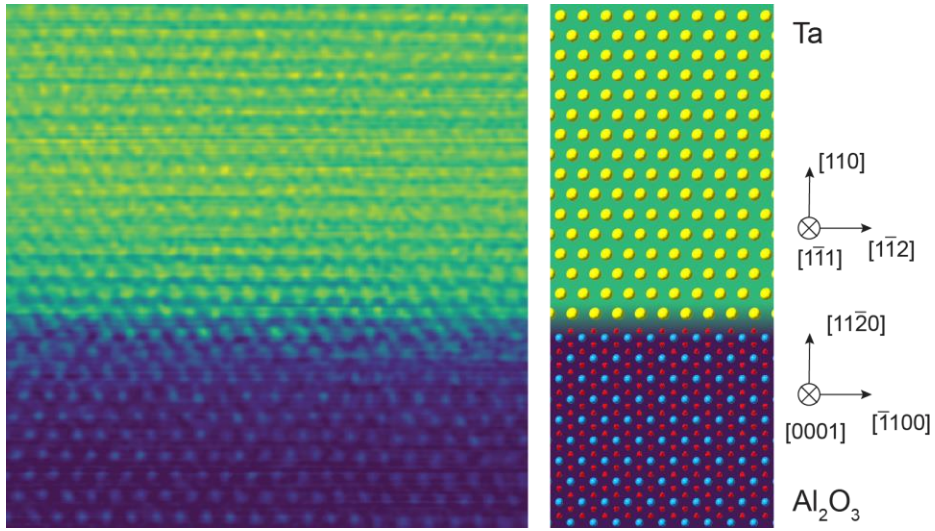
S. Hwang

K. Kisslinger



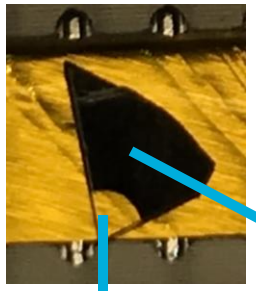


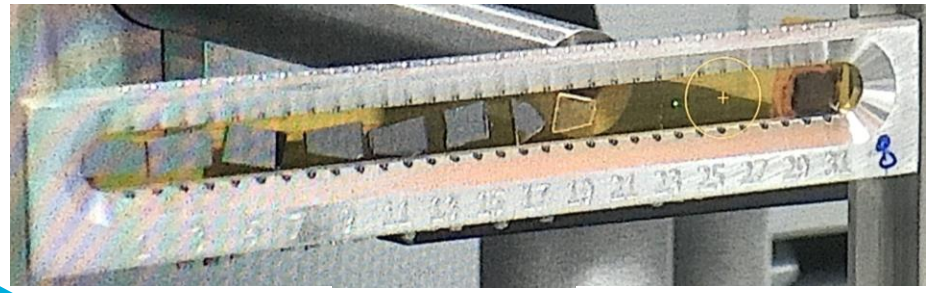
BNL Ta film, (110) orientation



- Fully epitaxial growth of Ta on *a*-plane sapphire (CFN) -- most previous studies was on *c*-plane sapphire

Synchrotron X-ray diffraction

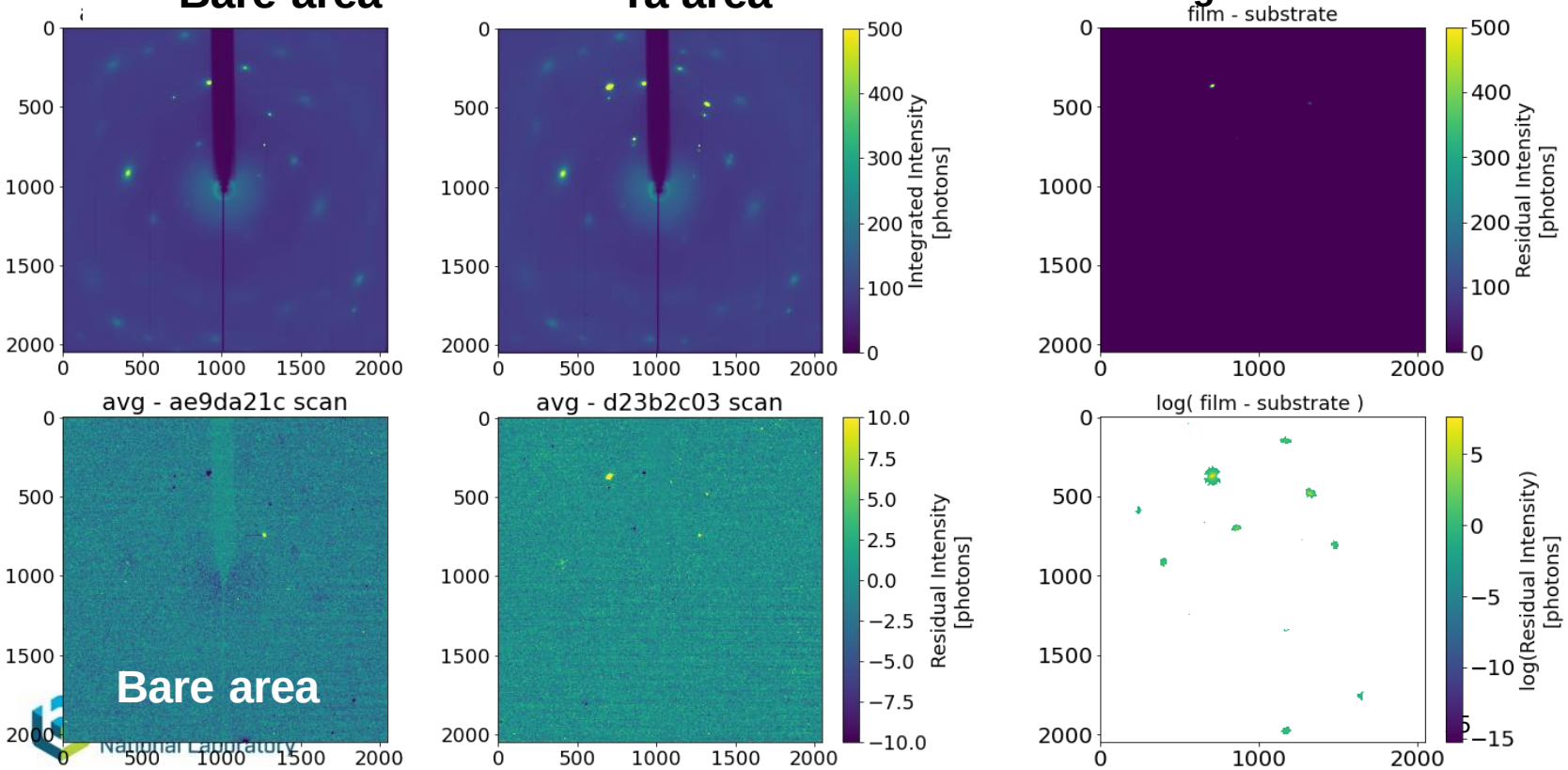




Bare area

Ta area

Average Difference

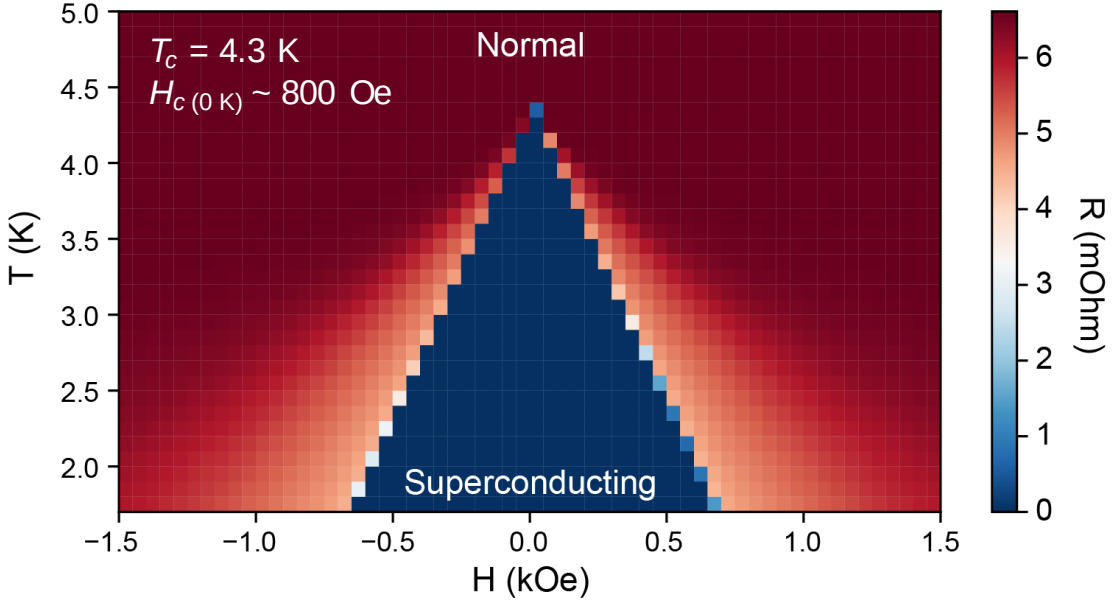


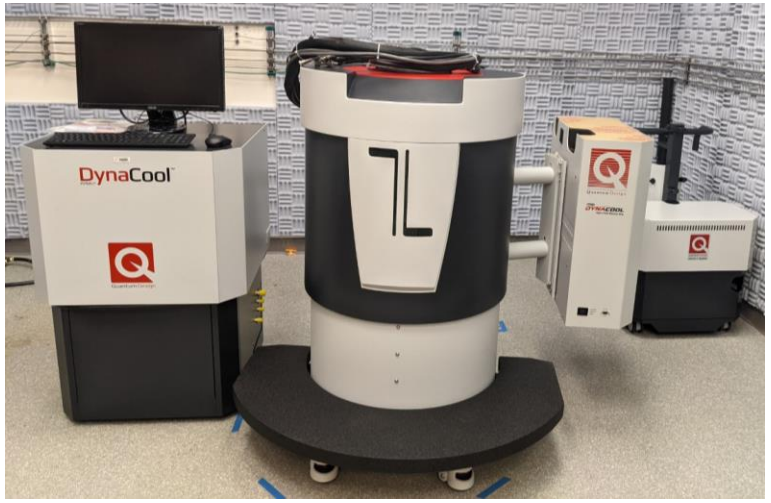
28-ID-1 PDF

A. Barbour D.Olds

- X-ray powder diffraction (XPD) & pair-distribution function (PDF)
- Film ordering & homogeneity w/ 500 µm spot
- Both in-plane and out-of-plane ordering
- High-throughput screening

Low temperature transport properties



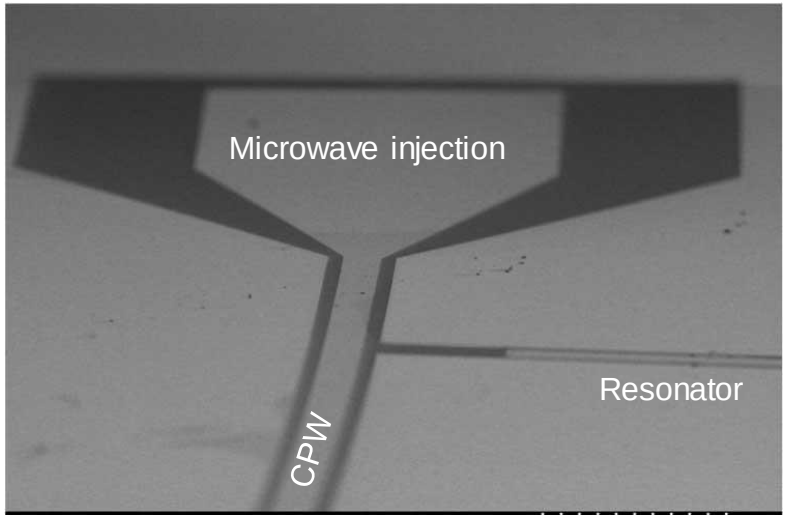


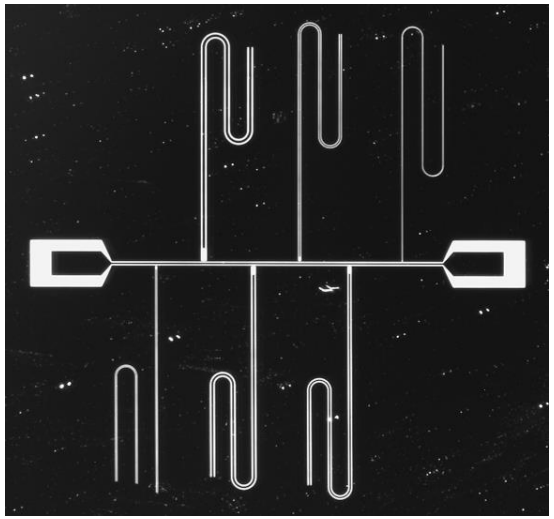
Dynacool 12 T PPMS (CFN)

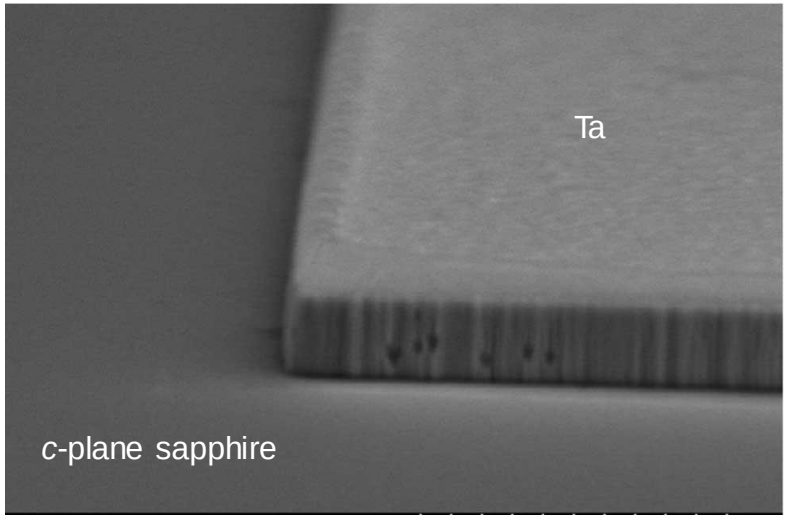
- New facility at CFN: Quantum Design DynaCool PPMS with 12 T magnet
- ^{3}He sub-Kelvin insert (~350 mK base)
- Multi-function probe for custom experiments

Brookhaven National Laboratory

Resonator device fabrication







JEOL JBX-6300FS 100 keV EBL

NanoFab (CFN)

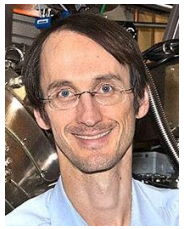
Y. Jia (CFN) | A. Stein (CFN) | A. Dutta (Princeton) | Y. Gang (Princeton)

- Hanger type Ta CPW (coplanar waveguide) resonator fabricated in CFN Cleanroom
- Using Princeton patterns
- Reactive ion etching gives sharp, straight edge profile

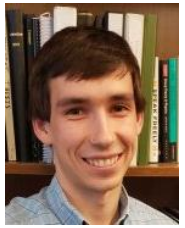
LEEM/XPEEM on qubit device

21-ID-2
ESM-XPEEM

J. Sadowski
(CFN)



I. Jarrige
(NSLS II)



A. Place
(Princeton)

XPEEM/LEEM:

- Surface potential mapping
- Local work function
- Surface chemical composition mapping
- Local chemistry (µXPS)

TM silicides for superconducting qubits

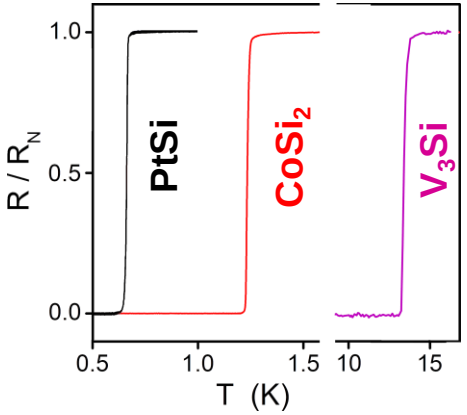
Y. Jia R. Li A. Stein K. Kisslinger A. Bollinger (CMPMS) C. Black

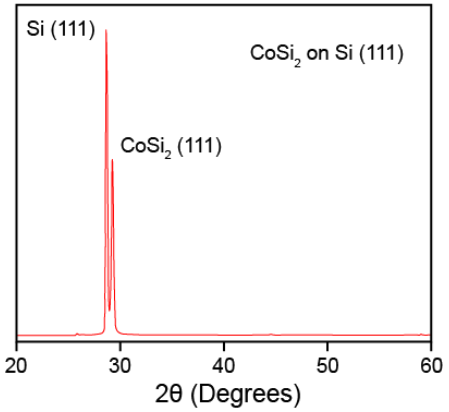
Materials Synthesis (CFN)

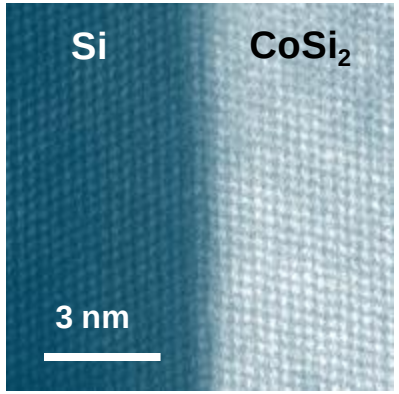
NanoFab (CFN)

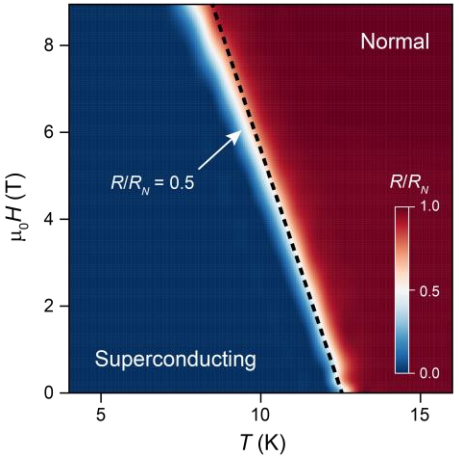
- Superconducting qubits on silicon substrate
- Silicides form on silicon-metal interface
- Superconducting TM silicides -- compatible with CMOS
- Use superconducting silicides as for silicon-compatible Josephson junctions and resonators

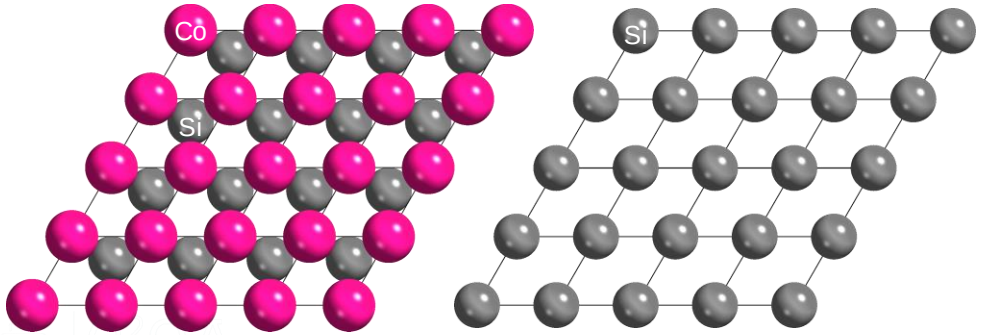
Silicide	T_c (K)	Silicide	T_c (K)
Nb_3Si*	19	$Sc_5Ir_4Si_{10}$	8.4
V_3Si	17	$LaRu_3Si_2$	7.3
Ba_8Si_{46}	8.07	AlCaSi	6.2
α-$ThSi_2$	3.16	$Lu_2Fe_3Si_5$	6.1
W_3Si_2	2.84	Al_2CaSi_2	5.8
$LaSi_2$	2.3	Li_2IrSi_3	3.7
$CaSi_2$	1.6	LaPtSi	3.5
$CoSi_2$	1.4	$LaRhSi_2$	3.4
Mo_3Si	1.4	$LaRhSi_2$	2.3
PdSi	0.93	ZrIrSi	1.7
PtSi	0.88	$LaIr_2Si_2$	1.6





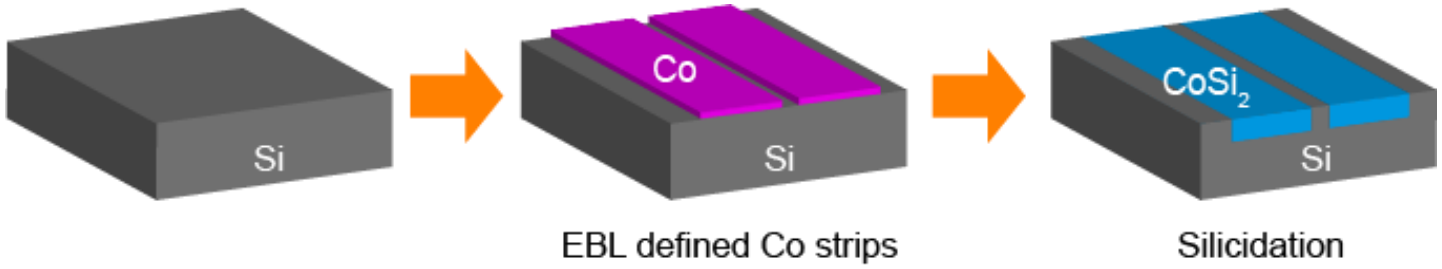


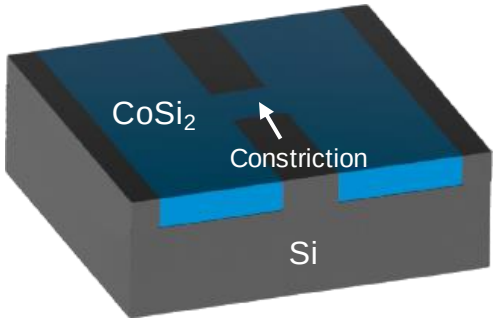


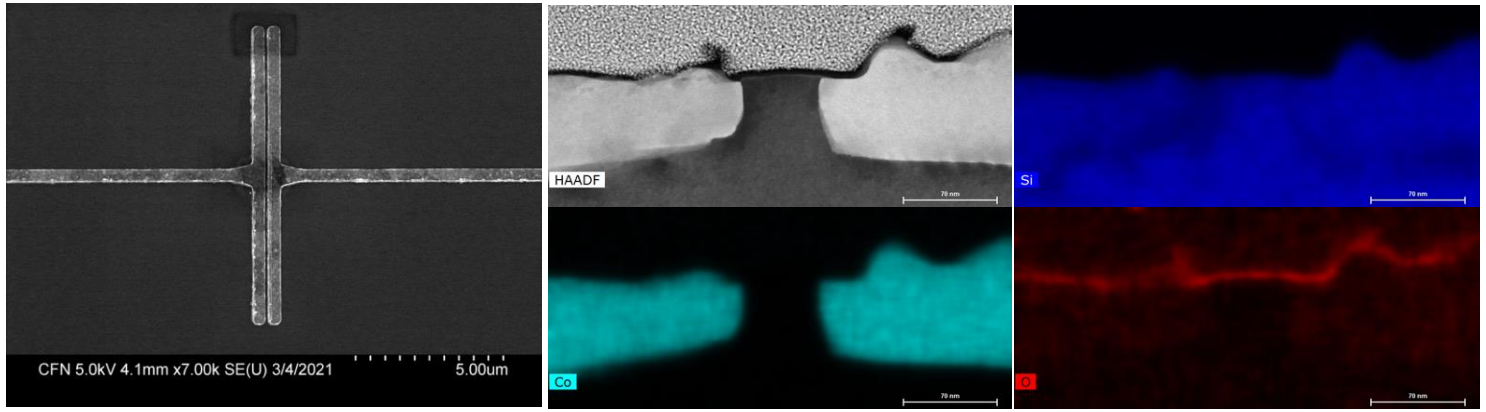


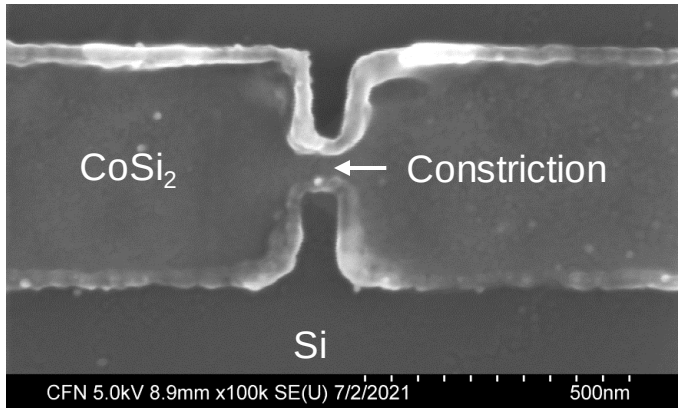
Brookhaven National Laboratory

$CoSi_2$-based Josephson junction







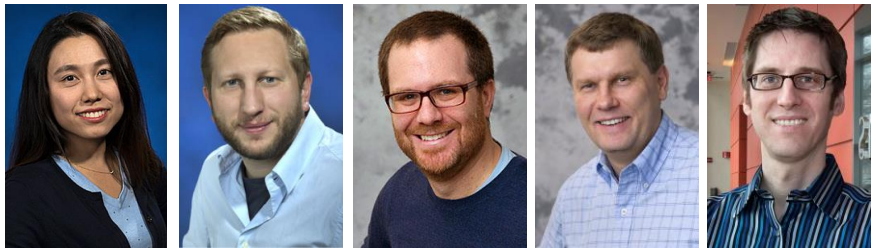


- $CoSi_2$-Si-$CoSi_2$ coplanar Josephson junctions
- On p^{++} Si wafer (no carrier freeze out) for S-N-S junction
- Junction width *W* below 100 nm

- $CoSi_2$ constriction for S-c-S type junctions
- On intrinsic Si wafer

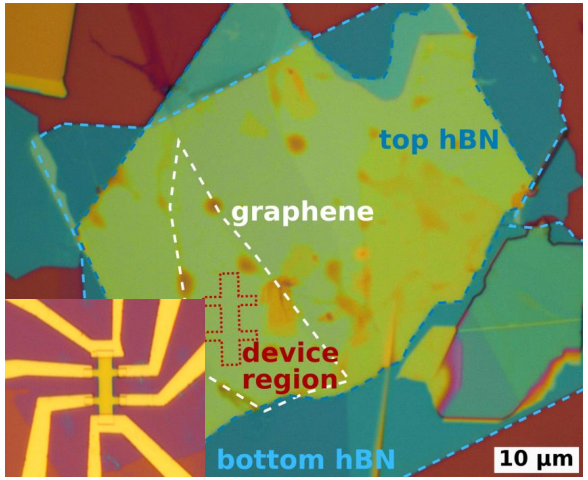
Brookhaven National Laboratory

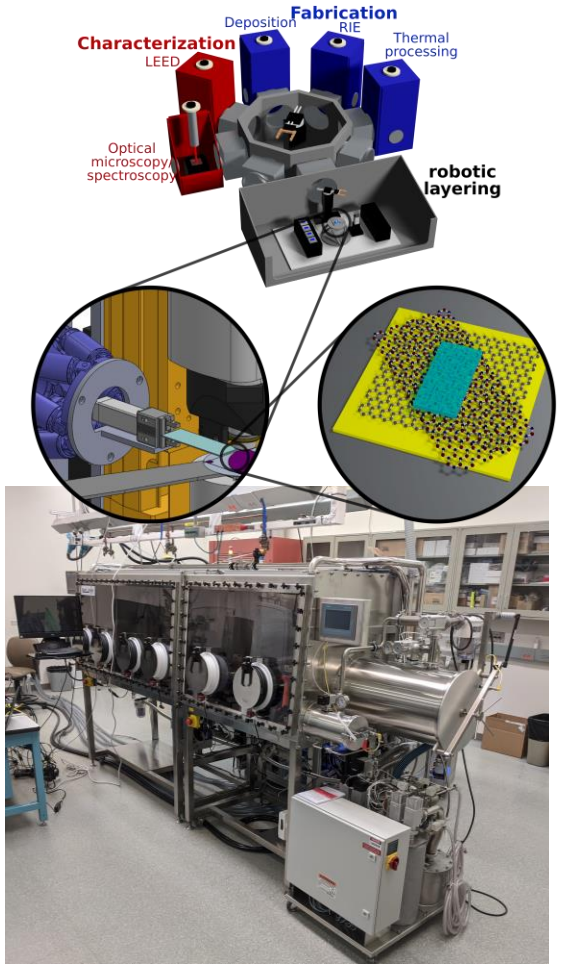
CFN quantum material press (QPress)



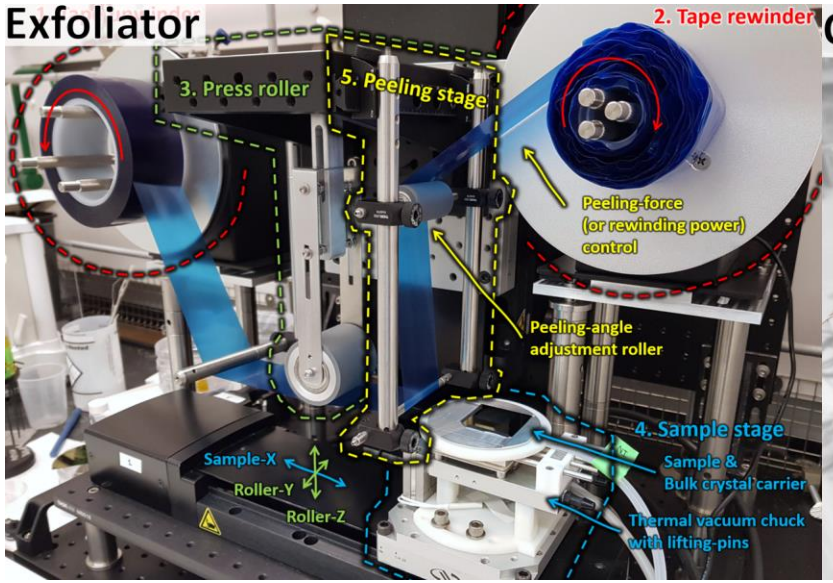
S. Park G. Doerk A. Stein J. Sadowski K. Yager

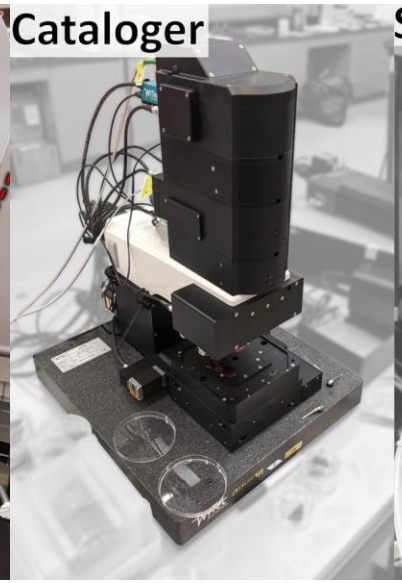
- **Automate study of 2D heterostructures**
  - 3-year DOE QIS project (since FY 2019)
  - Reliably fabricate complex stacks of atomically-thin (2D) materials
  - Understand physics of exfoliation
  - Study QIS materials
  - Non-expert access to frontier synthesis
- **Partnerships**
  - P. Kim, A. Yacoby (Harvard)
  - D. Shahrjerdi (NYU)
  - Broader user community

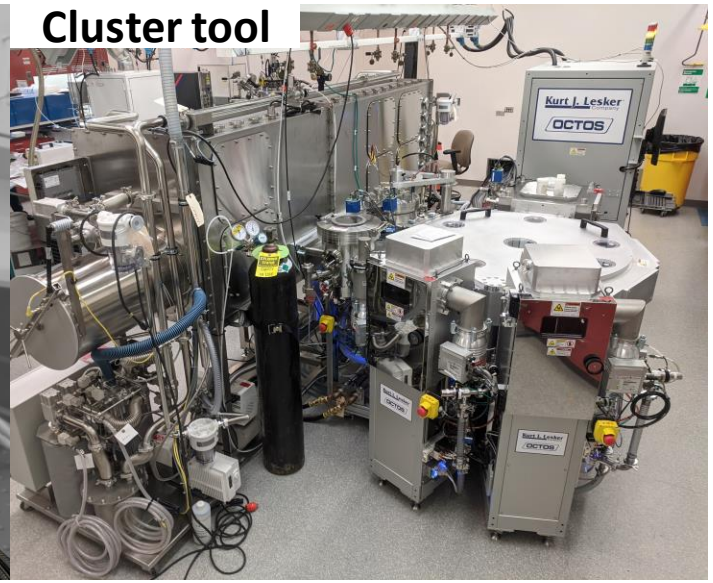




QPress modules







Mechanical exfoliator

- CFN custom roll-to-roll design
- Reproducible control of exfoliation process
- Enables rapid generation of exfoliated substrates

Cataloger

- Optical microscope with integrated Raman imaging/AFM
- Machine-vision identifies/classifies flakes
- Generate “flake libraries”

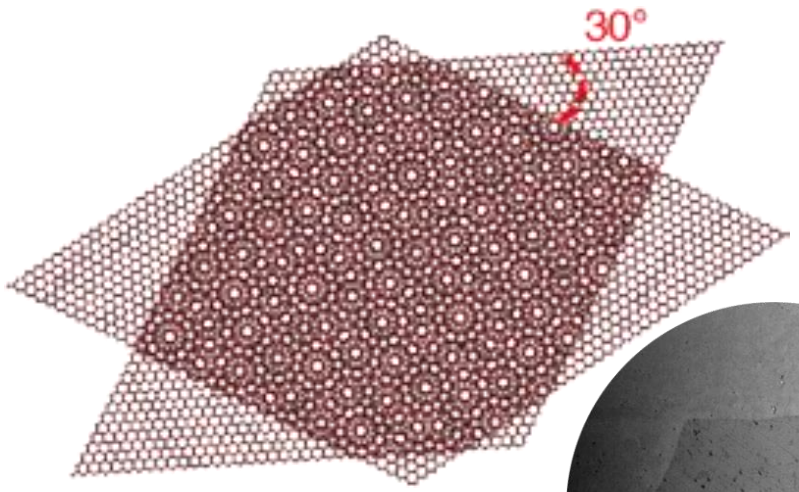
Stacker

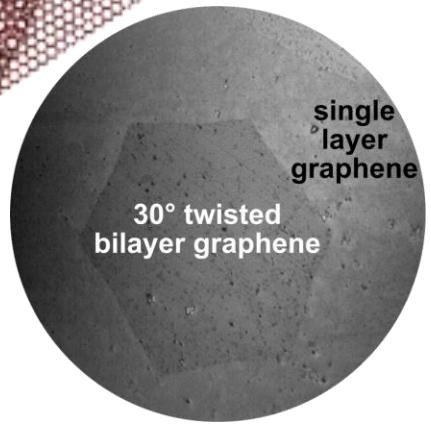
- Robotic control of stamp and sample
- Build heterostructure stacks, such as graphene/hBN

Cluster tool

- Custom design from Lesker
- Installed and operating in CFN
- Robotic transfer between stations
  - Through vacuum/inert atmosphere (glovebox)
  - Thermal/plasma processing
  - E-beam deposition (multiple targets)

QPress science progress





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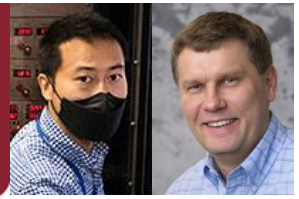
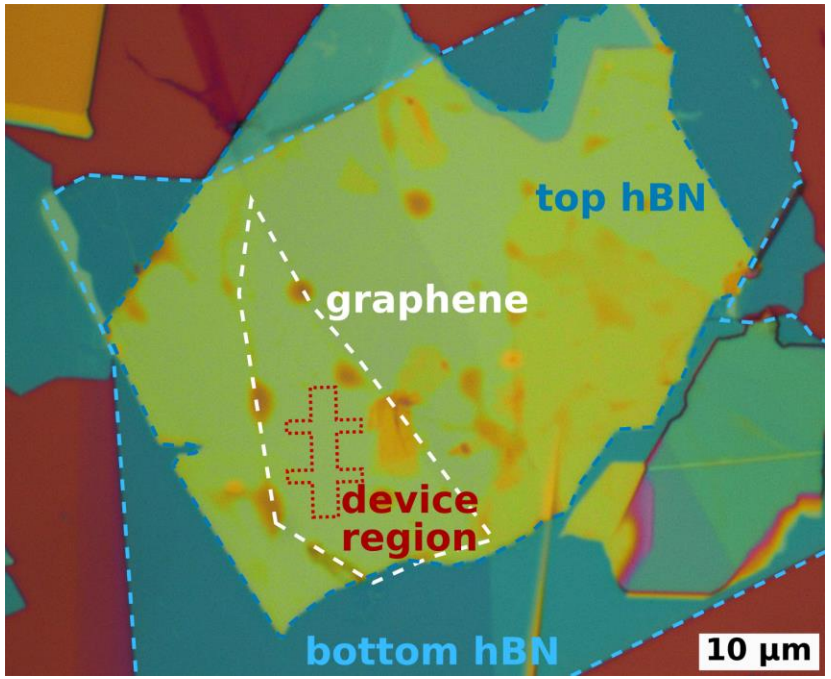


Z. Dai J. Sadowski



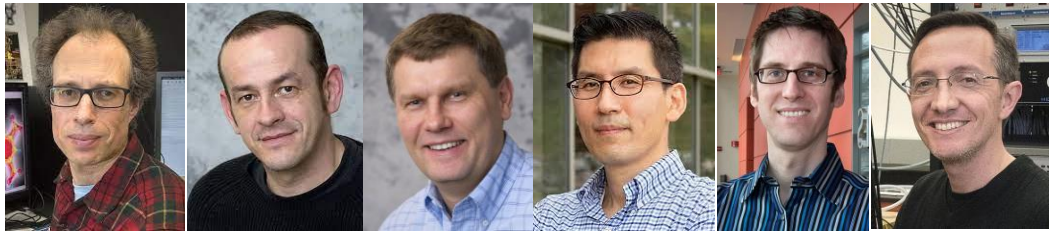
D. Shahrjerdi (NYU)

- **Bound States in Graphene Heterostructure**
  - Identify new quantum confinement resonance in twisted graphene bilayers
  - Developed special TiO_x-coated SiO_x substrates

Dai et al. *Phys. Rev. Lett.* **2021**, 127, 086805.

- **Build heterostructures**
  - Generate graphene library using QPress exfoliator
  - Built hBN/graphene/hBN heterostructures using QPress stacker (~6× faster fabrication)
  - Demonstrated high-performance FETs from heterostructures

Future QPress



P. Zahl M. Cotlet J. Sadowski C. Nam K. Yager D. Stacchiola

- **Complete commissioning/integration**
  - Continue to refine modules
  - Integrate into single software framework
  - Deploy data tools for automated experiments
- **Facility expansion**
  - Integrated Multimodal Characterization and Processing (QM-IMCP;$6.9M, 3 years since FY '22)
  - Processing heterostructure materials with atomic level precision at a new atomic layer etching tool
  - Characterization of electronic states, magnetic imaging, and chemical analysis at the upgraded synchrotron-based microscope
  - Analysis of carrier dynamics at an upgraded optical ultrafast-microscope
  - Mapping the electrostatic potential of atomic defects and single spin detection at the new scanning probe microscope (SPM) with quantum sensors, making direct connection with C^2QA efforts

QM-IMCP