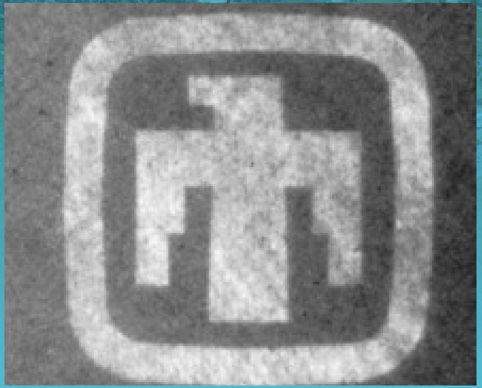
SAND2022-10363C

Sandia National Laboratories

Expanding the Energy Range from eV to MeV ~~and Fabrication of Sources~~ Enabling Novel Focused Ion Beam Nanofabrication and Modification



Michael Titze, Aaron Katzenmeyer, Vigneshwaran Chandrasekaran, Anthony Flores, Barney Doyle, Yongqiang Wang, Han Htoon, Edward Bielejec

08/02/2022

U.S. DEPARTMENT OF ENERGY NNSA

SAND2022-2513 C

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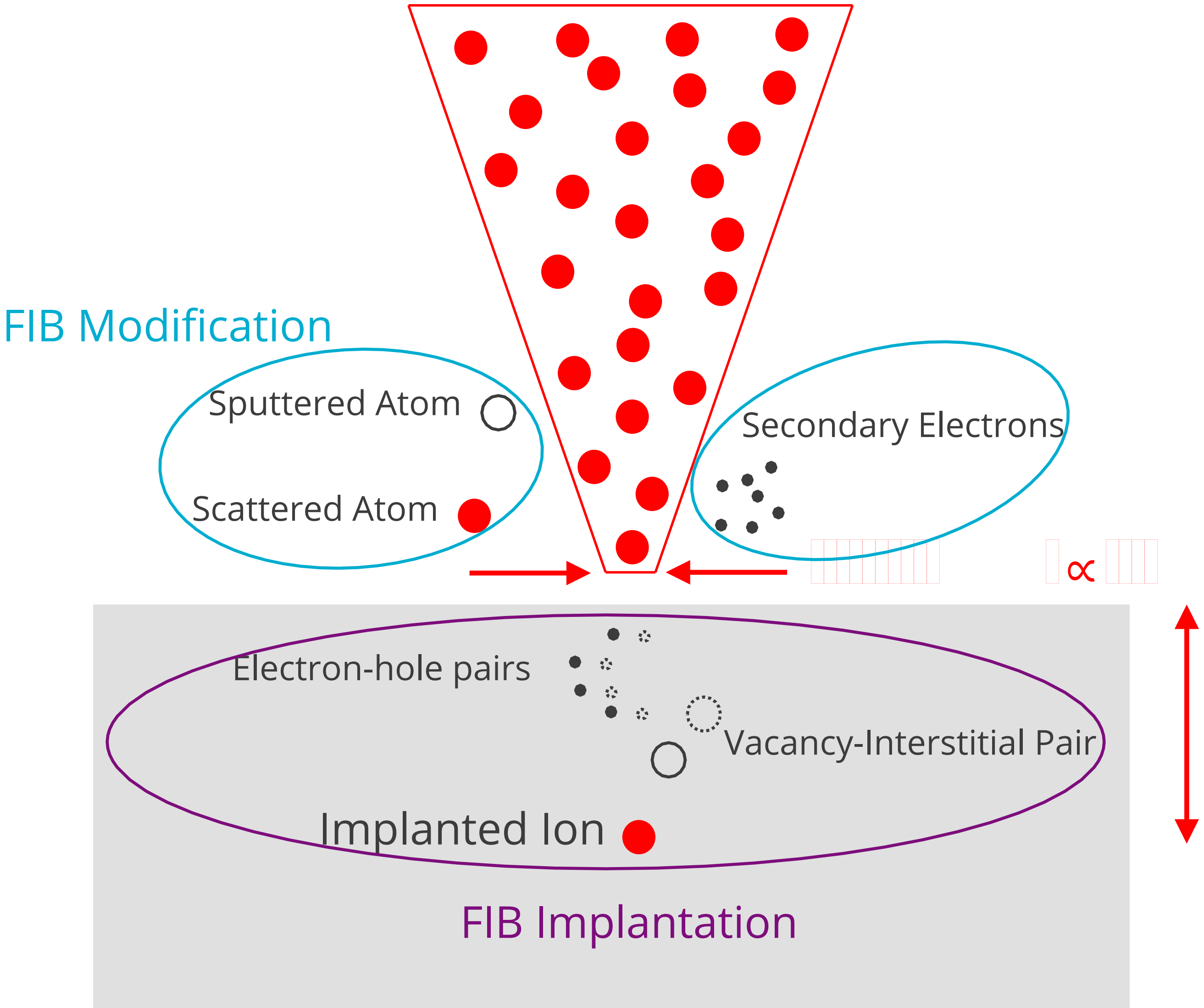
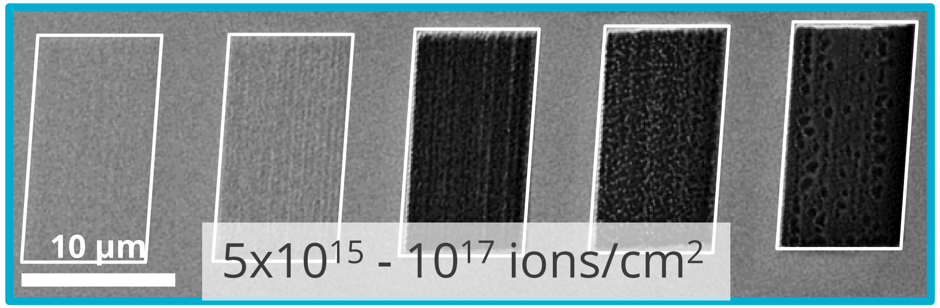
Outline

- Overview of the Sandia Ion Beam Laboratory
  - Focused Ion Beam Capabilities
- Expanding the Energy Range for Ion Implantation
  - High Energy Range (≥1 MeV)
  - Medium Energy Range (10 – 200 keV)
  - Low Energy Range (< 10 keV)
- Conclusion

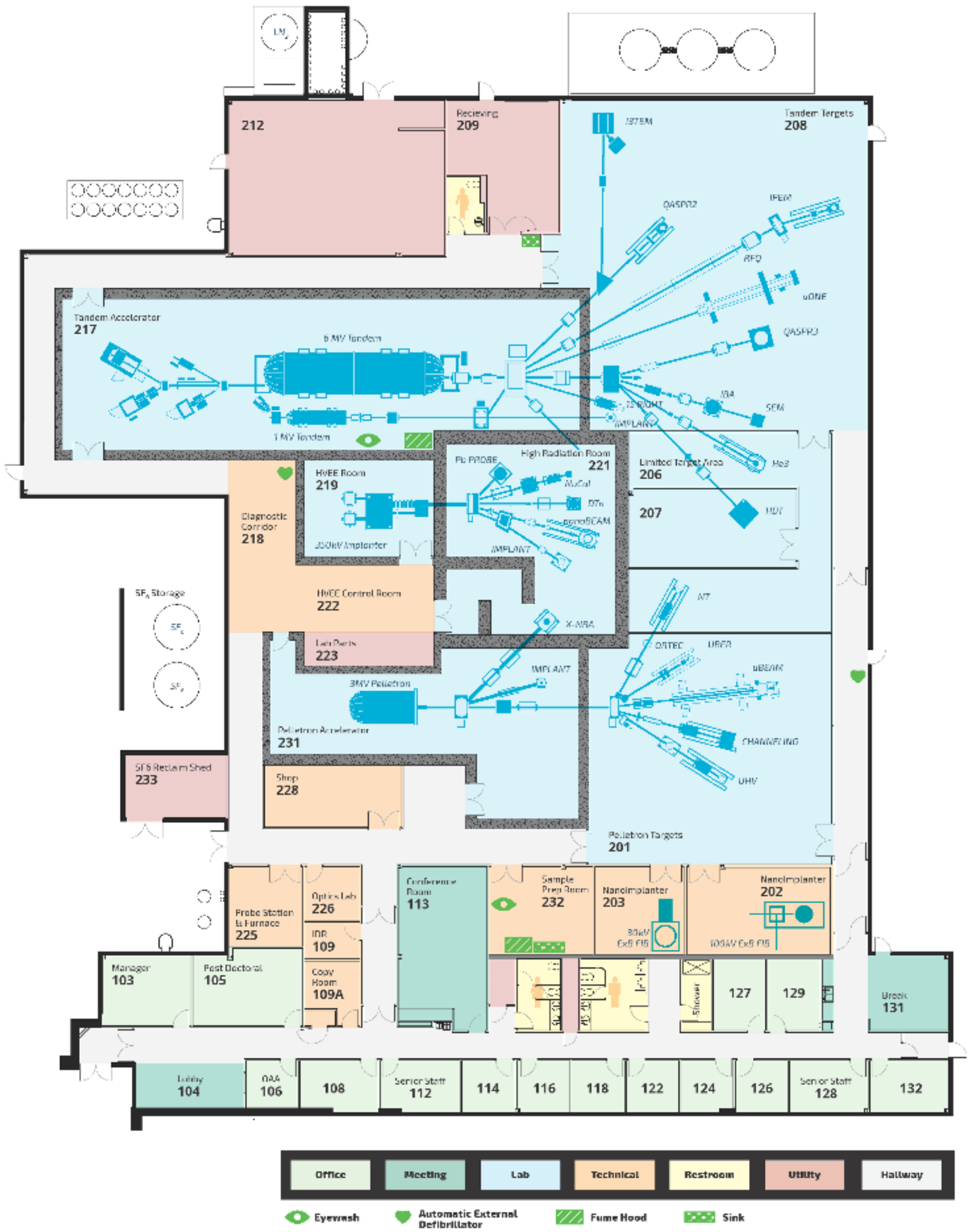
Ion Beam Modification versus Ion Implantation





Vastly different dose regimes
- Single atom devices
- (almost) No beam curernt

building 720
ION BEAM LABORATORY



Sandia's Ion Beam Laboratory (IBL)

7 Operational Accelerators and >25 end-stations

(including *in-situ* DLTS, PL, TEM, SEM, 1200°C heating, etc...)

Operational

(1) 6 MV Tandem Accelerator

(2) 3 MV Pelletron Accelerator

(3) 1 MV Tandem Accelerator

(4) 350 kV HVEE Implanter

High energy focused micobeams 1 μm

(5) 100 kV ExB FIB nanoImplanter

(6) 35 kV ExB FIB Raith Velion

(7) 35 kV Zeiss HeIM

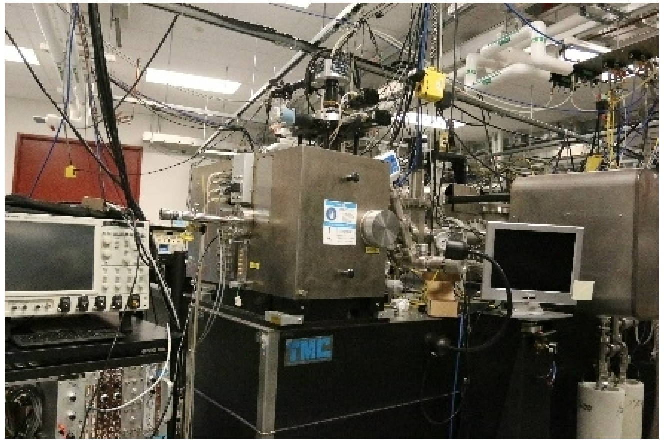
Low energy focused nanobeams <1 to 20 nm

Installing

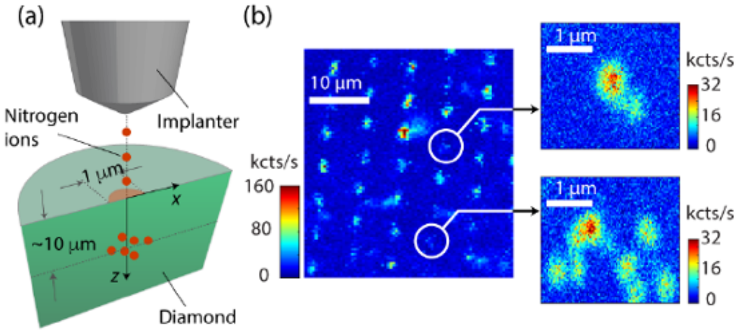
(8) 35 kV Plasma FIB

High Energy Focused Microbeams ~ 1 µm Spot Size

- 6 MV Tandem microbeam (microONE)

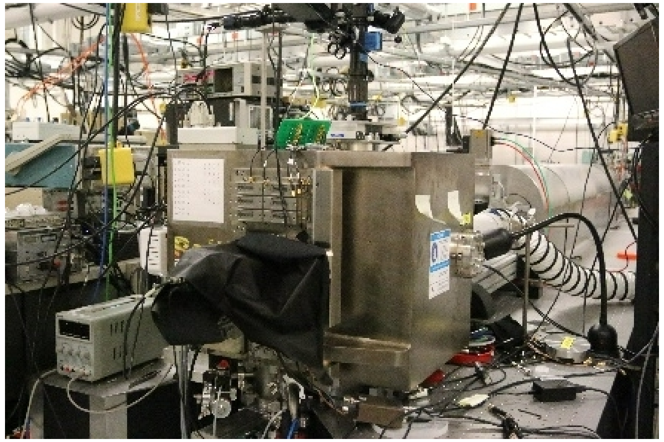


- High resolution laser stage
- Spot size <1 µm
- Energy 0.8 – 70 MeV
- H to Au

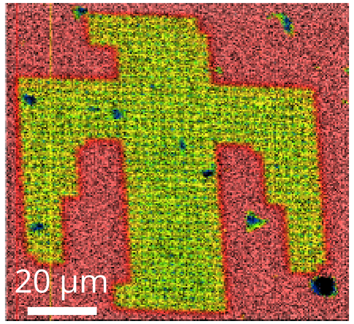


A. Lozovoi et al., Nat. Electron., 4, 717 (2021)

- 3 MV Pelletron microbeam (Light Ion Microbeam)

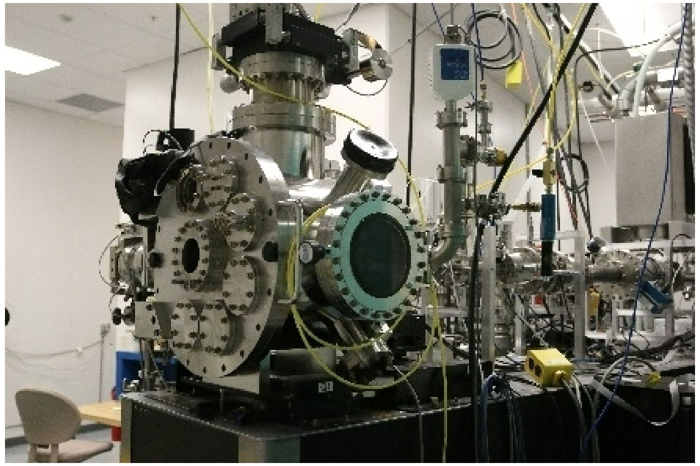


- High resolution laser stage
- Spot size <600 nm (H)
- Energy 0.25 – 3 MeV
- H, He, N, Ar, Xe, …

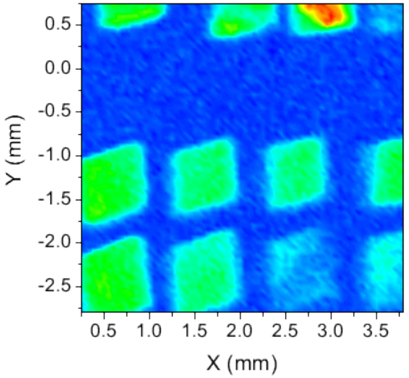


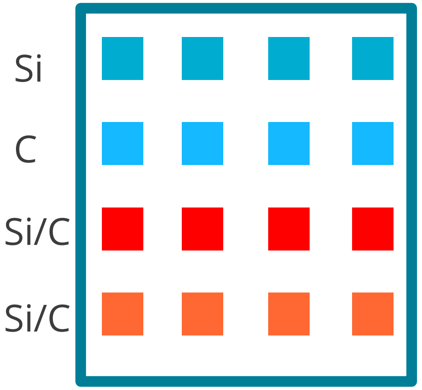
IBIC on PIN diode

- 350 kV HVEE microbeam (NanoBeamLine)



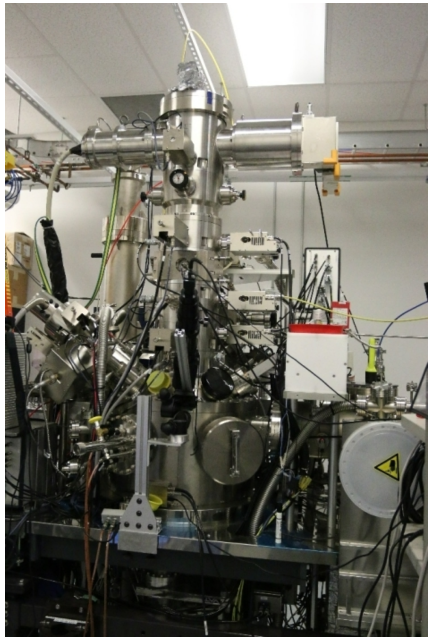
- Piezo stage
- Spot size <1 µm
- Energy 20 – 350 keV
- H to Au





Low Energy Focused Nanobeams < 1 nm – 20 nm Spot Size

- 100 kV A&D FIB100NI (nanoImplanter)

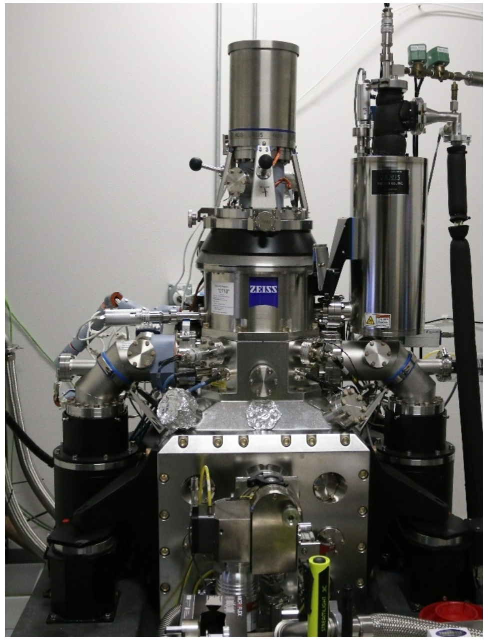


- High resolution laser stage
- Spot size <10 nm (Ga)
- Energy 10 – 200 keV
- 1/3 periodic table

- 35 kV Raith Velion (Velion)

- High resolution laser stage
- Spot size <6 nm (Ga)
- Energy 1 – 70 keV
- 1/3 periodic table

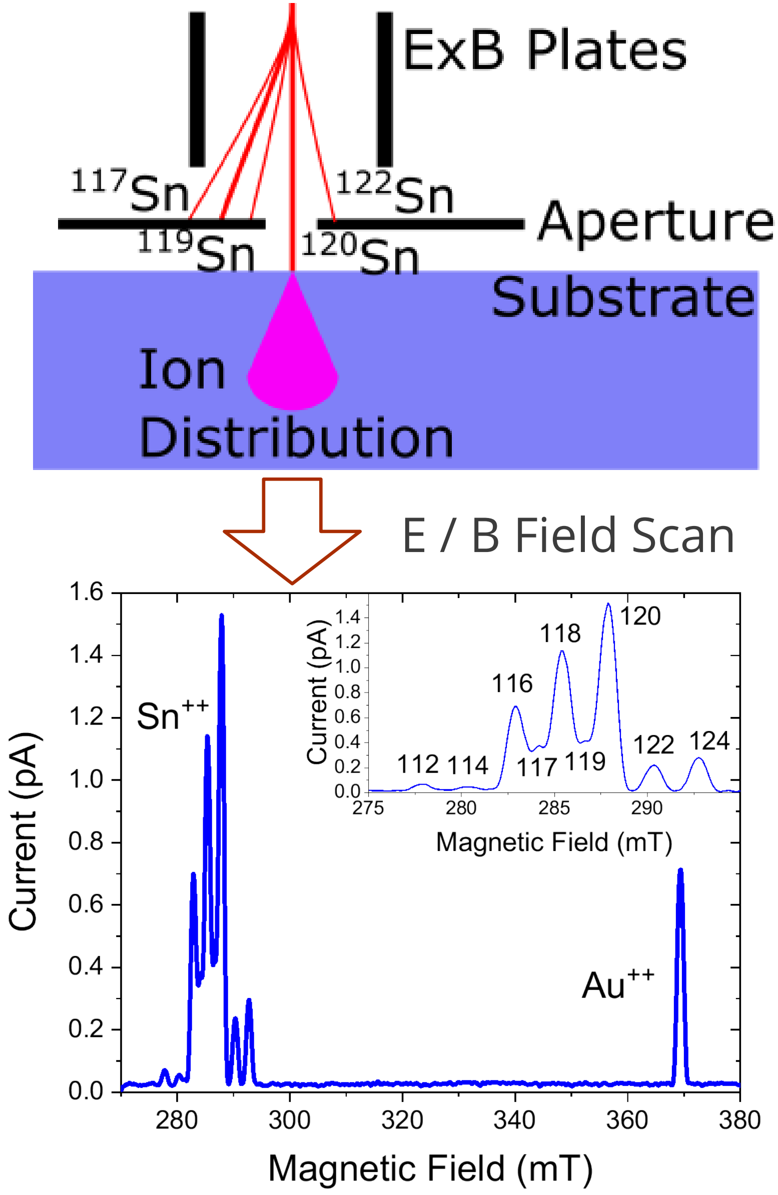
- 35 kV Zeiss Orion Plus (HeIM)



- Piezo stage
- Spot size <1 nm
- Energy 10 – 35 keV
- He

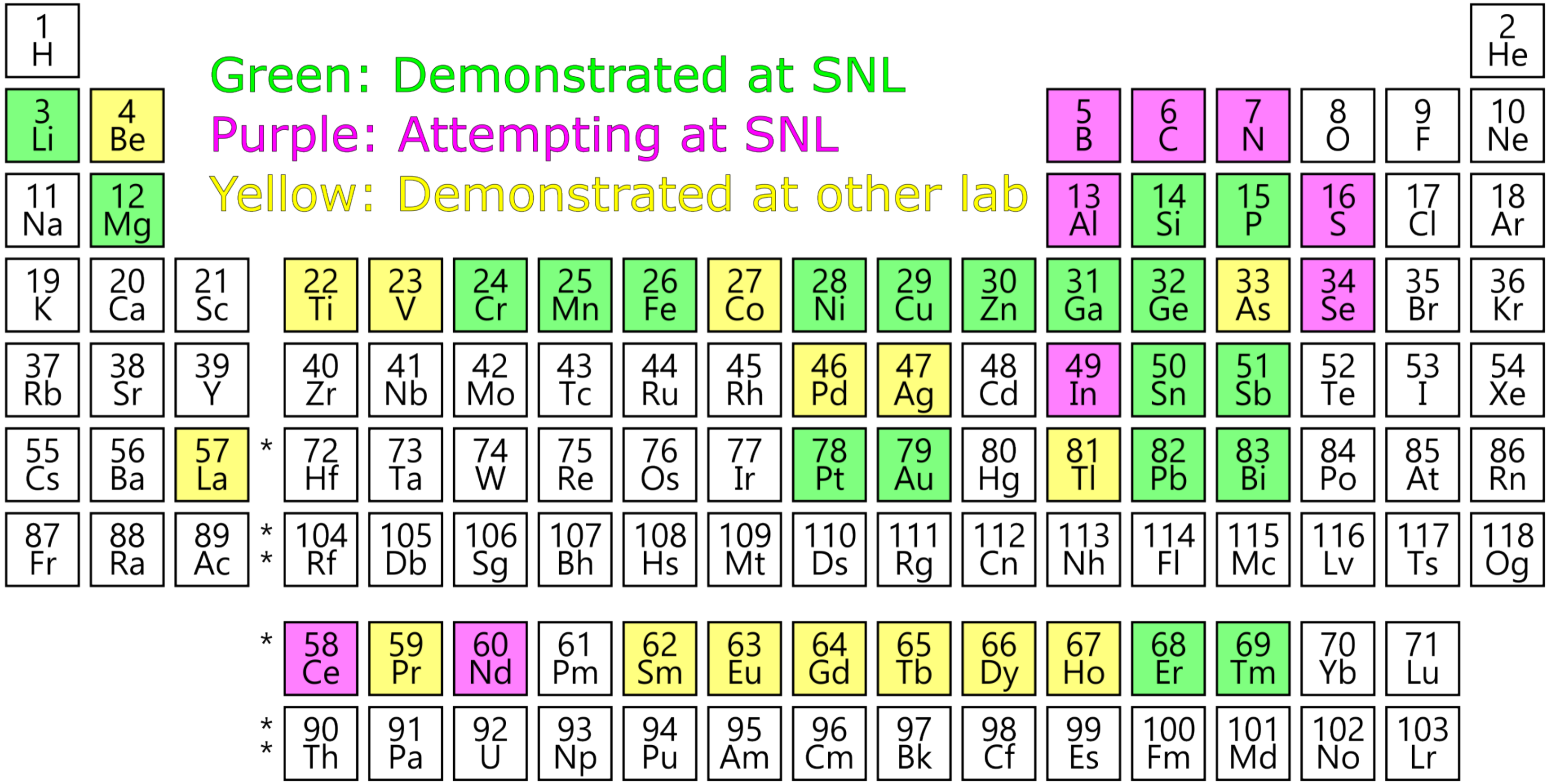
All equipped with Lithography Software for Patterning

Liquid Metal Alloy Ion Sources – Available Ions



Added 8 new elements over past 3 years

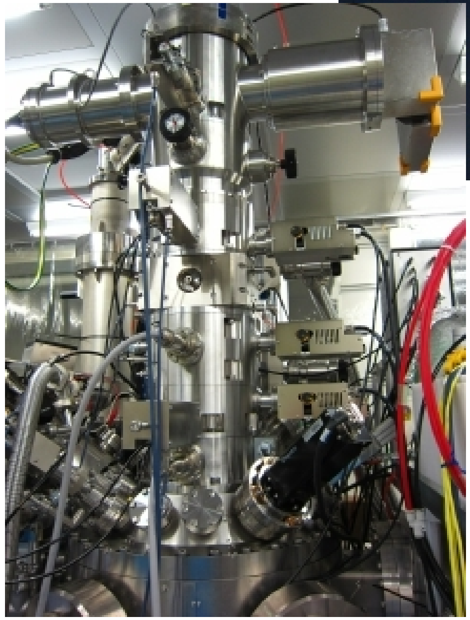
Green: Demonstrated at SNL
Purple: Attempting at SNL
Yellow: Demonstrated at other lab

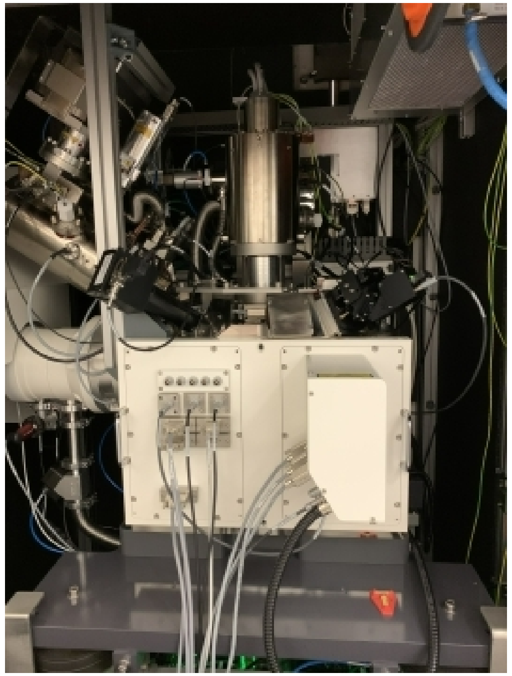


Adapted from L. Bischoff et al., Appl. Phys. Rev., 3 (2016)

Energy Ranges for Focused Ion Beam Implantation

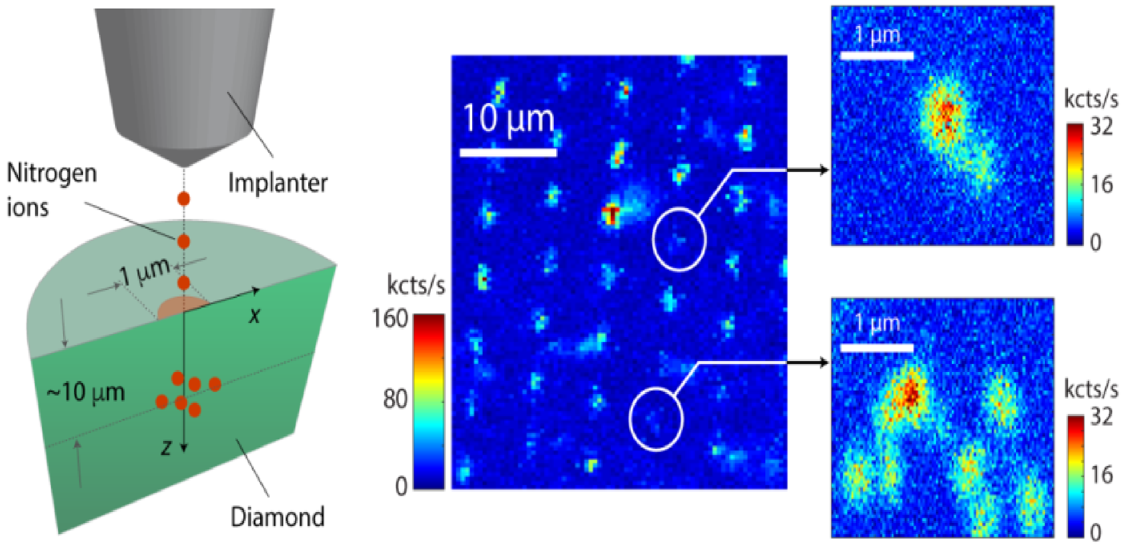
- High Energy (> 1 MeV)
  - Specialized high energy ion accelerators
  - Best achievable spot sizes ~ 1μm
- Medium Energy (10 – 200 keV)
  - Where FIB systems like to run
  - SED imaging / Single ion counting possible
  - Best achievable spot sizes ~ 10 nm
- Low Energy (< 10 keV)
  - Using FIB in non-standard energy regime
  - Targeting single (few) atomic layers
  - Targeting resolution ?
  - How to get there

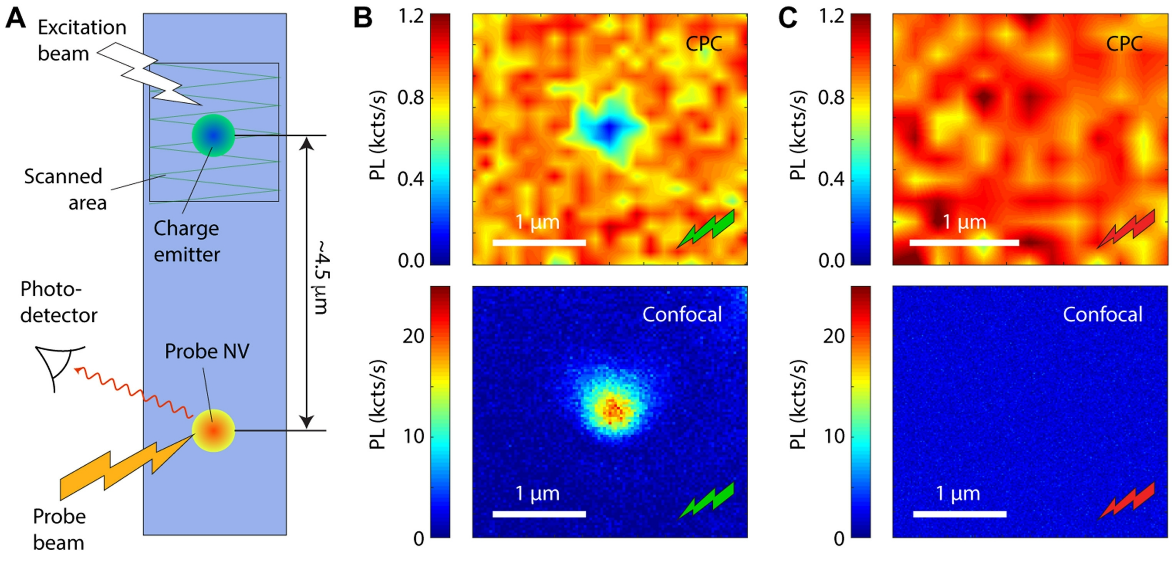




Example of High Energy Ion Implantation – NV Quantum Sensing

- 20 MeV N ions
- Microbeam on 6 MV Tandem

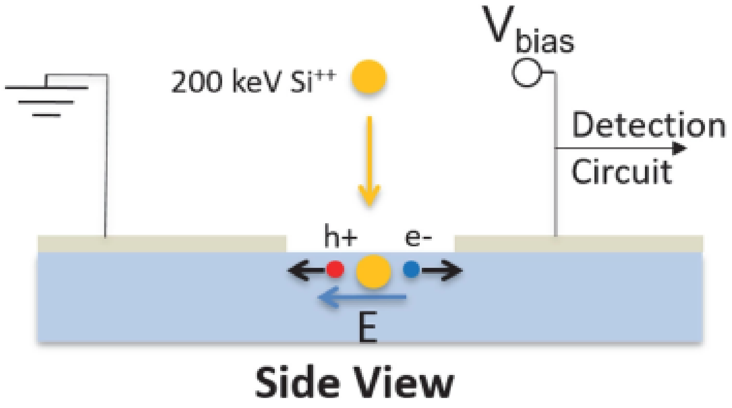




Deep Implantation to shield the implanted ion from surface noise
→ NV sensing its crystal environment

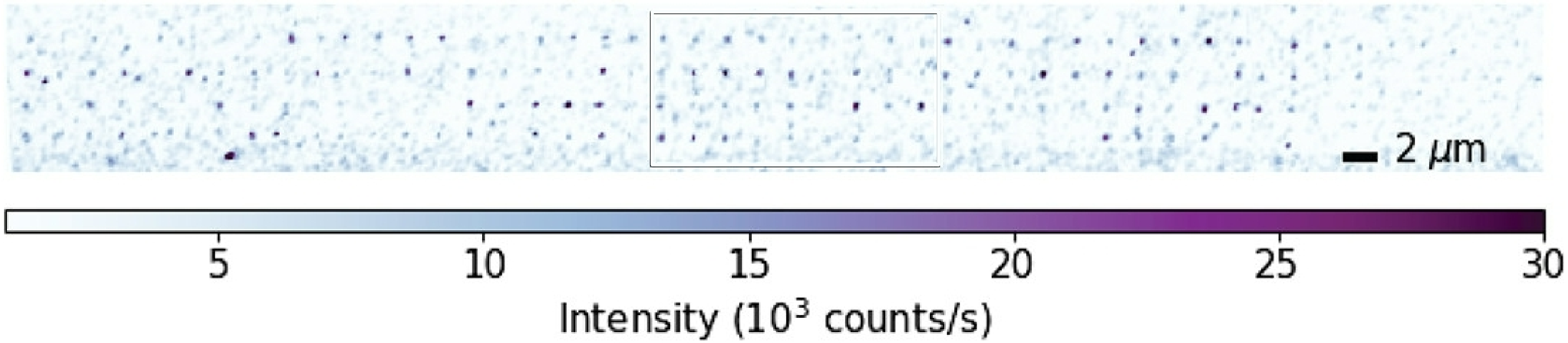
Example of Medium Energy Implantation – Single Ion Counting

- 200 keV Si^{++}
- 100 kV A&D nanoImplanter



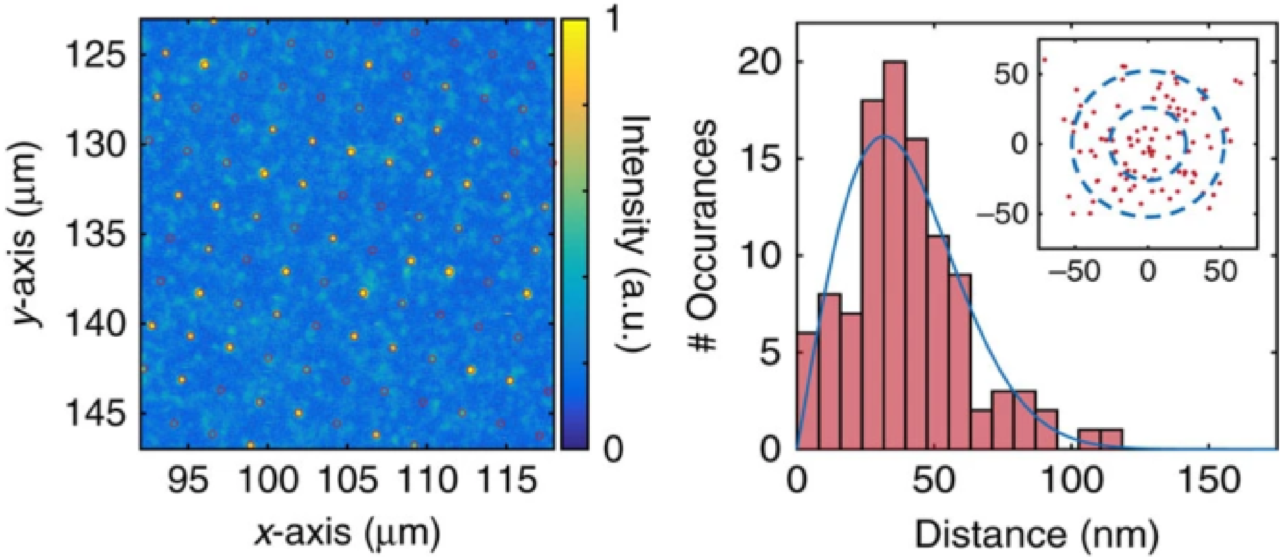
Deterministic placement of single ions
- +4/-1 % error bar on ion #
- < 40 nm targeting resolution

Pathway to creating scalable single photon emitters



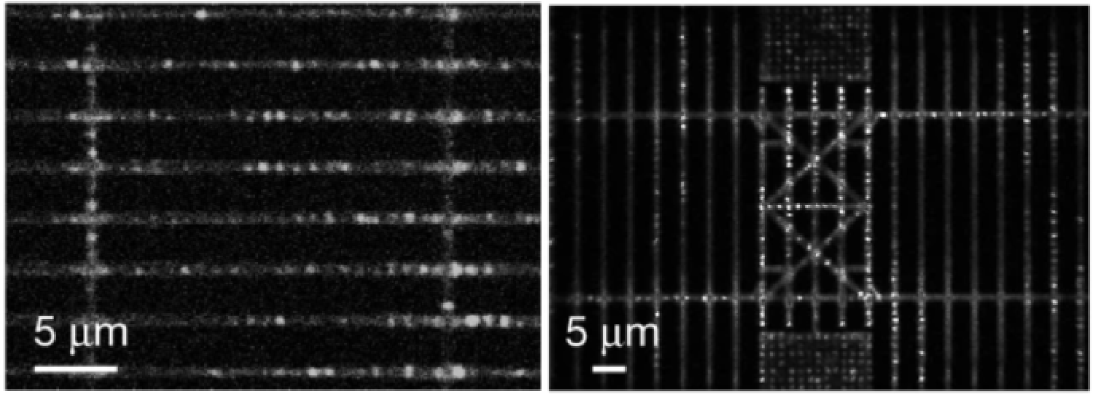
FIB Enabled Quantum Optics

< 50 nm Targeting Resolution



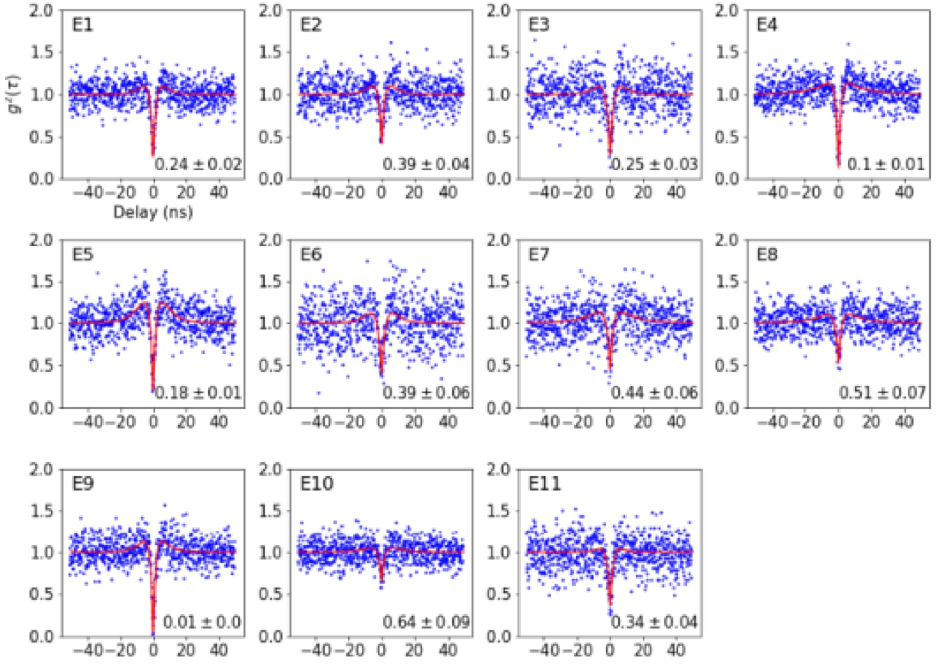
T. Schroder et al., Nat. Communs., 8, 15376 (2017)

Scalable Implantation



N. Wan et al., Nature, 583, 226 (2020)

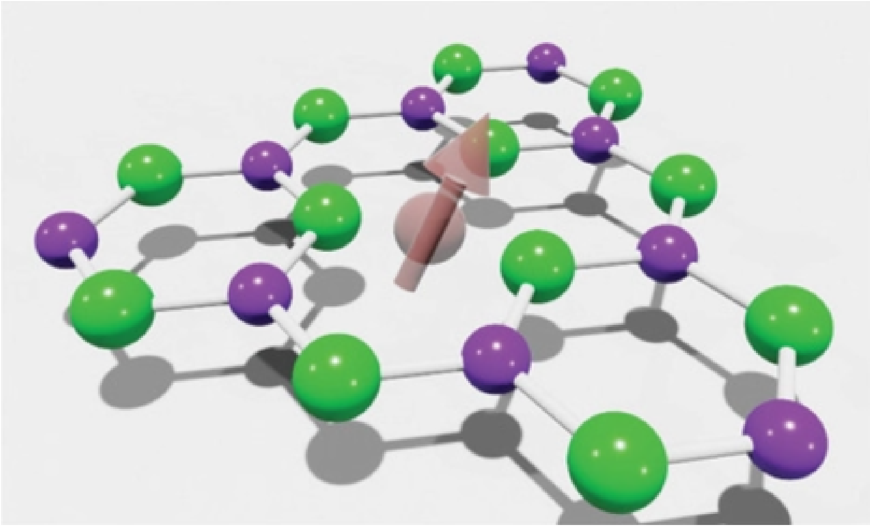
Forming Single Photon Emitters



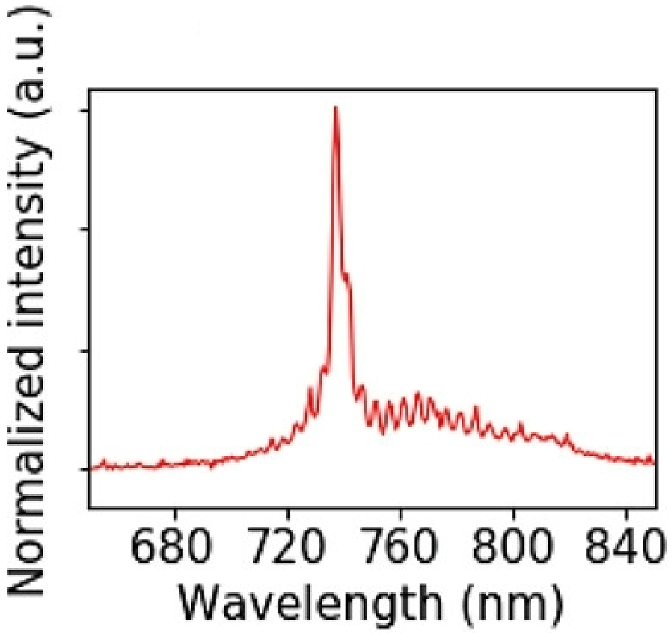
M. Titze et al., Nano Lett. (2022)

Low Energy Implantation - Motivation

- 2D materials are attractive material class for CMOS-integrable quantum optics
- Deterministic placement of **impurity**-type emitters is challenging
  - Stopping in single atomic layer
  - Minimize damage to surrounding lattice
  - Introduce non-native atom



N. Mendelson et al., Adv. Mater., 34, 2106046 (2022)

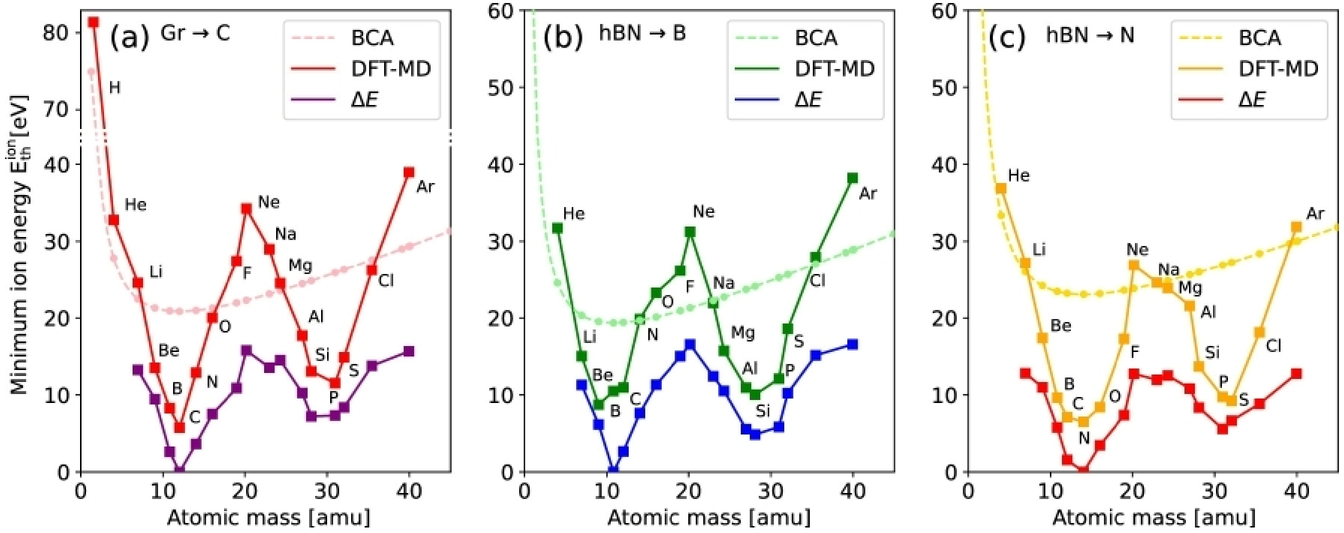


M. Titze et al., Nano Lett. (2022)

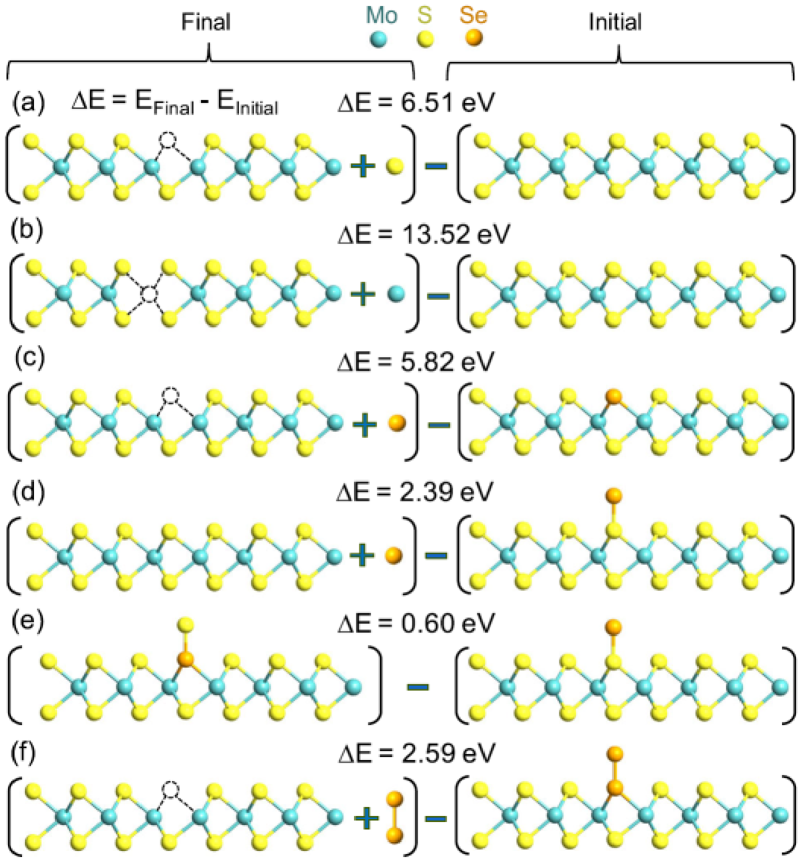
Ion Implantation into 2D Materials

- Formation of vacancies depends on ion energy
- Incorporation of impurities requires ultra-low energy
- Deterministic Placement by Focused Ion Beam Implantation

→ Theory predicts a range of energies for incorporation



S. Kretschmer et al., J. Phys. Chem. Lett., 13, 2, 514 (2022)



M. N. Bui et al., NPJ 2D Mater. Appl., 6, 42 (2022)

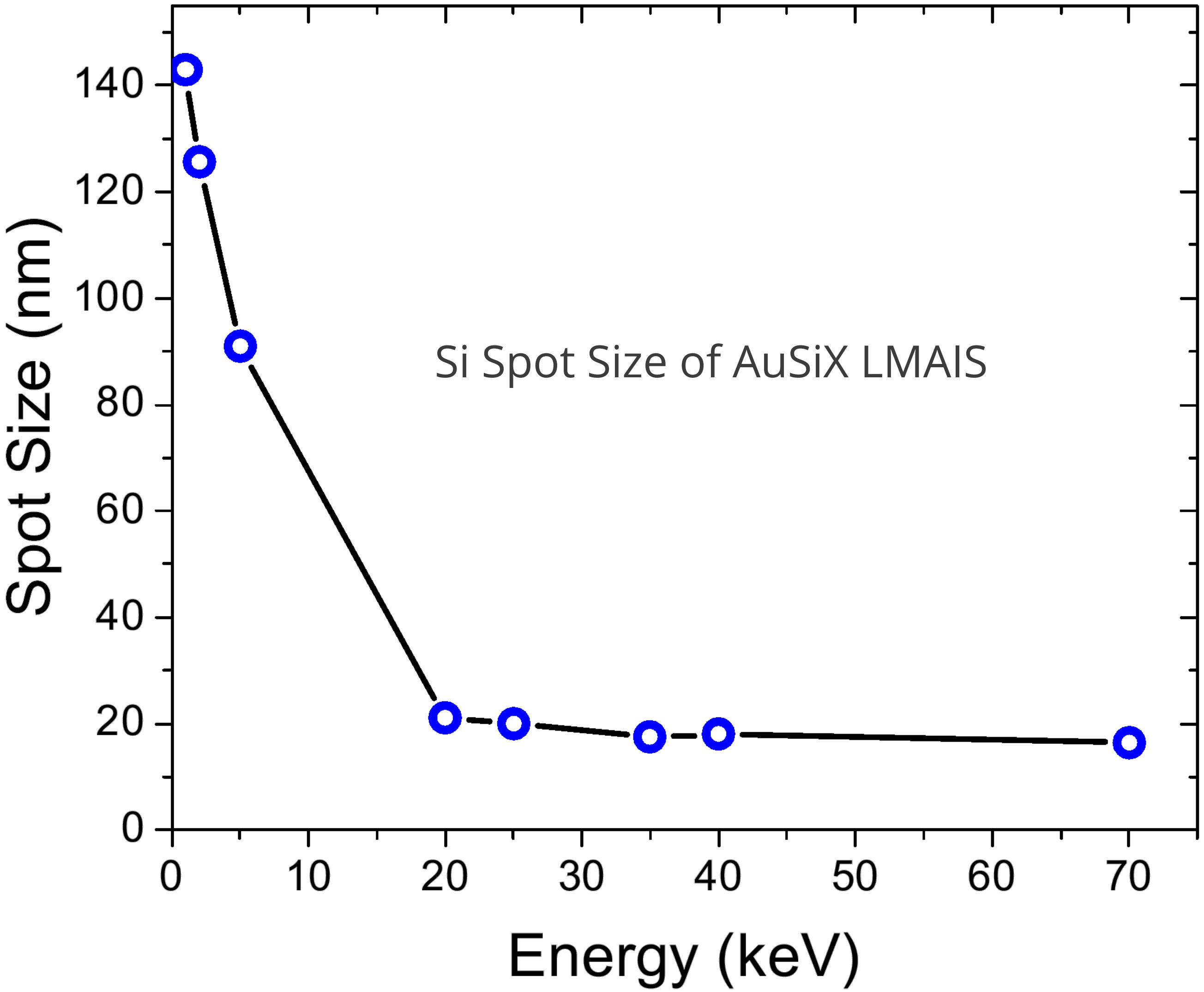
Challenges with FIB-Based Ultra-Low Energy Ion Implantation

- Spot Size increase due to chromatic abberations
- Range Prediction vs Experiment
- How to reduce Energy to stop in atomic monolayer

Caveats of Low Energy Focused Ion Beam Implantation

- Spot Size
- Range Prediction vs Experiment
- How to reduce Energy to stop in atomic monolayer

  - Chromatic aberrations become more prominent for lower energy ions
    → Increased spot size
    - LMAIS have ~15 eV energy spread
      → Landing energy >> 15 eV
  - Imaging becomes more difficult with low energy ions, since fewer e-h pairs



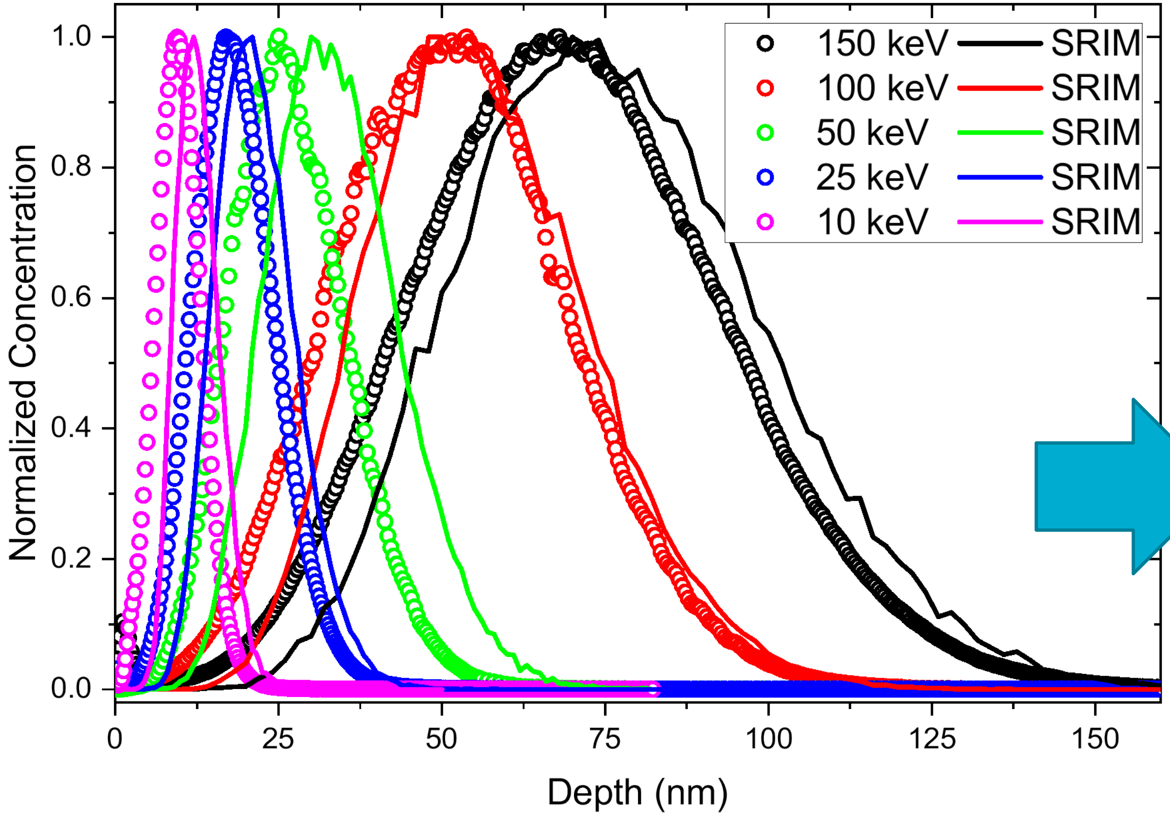
Binary Collision Approximation and Experiment - Range Comparison

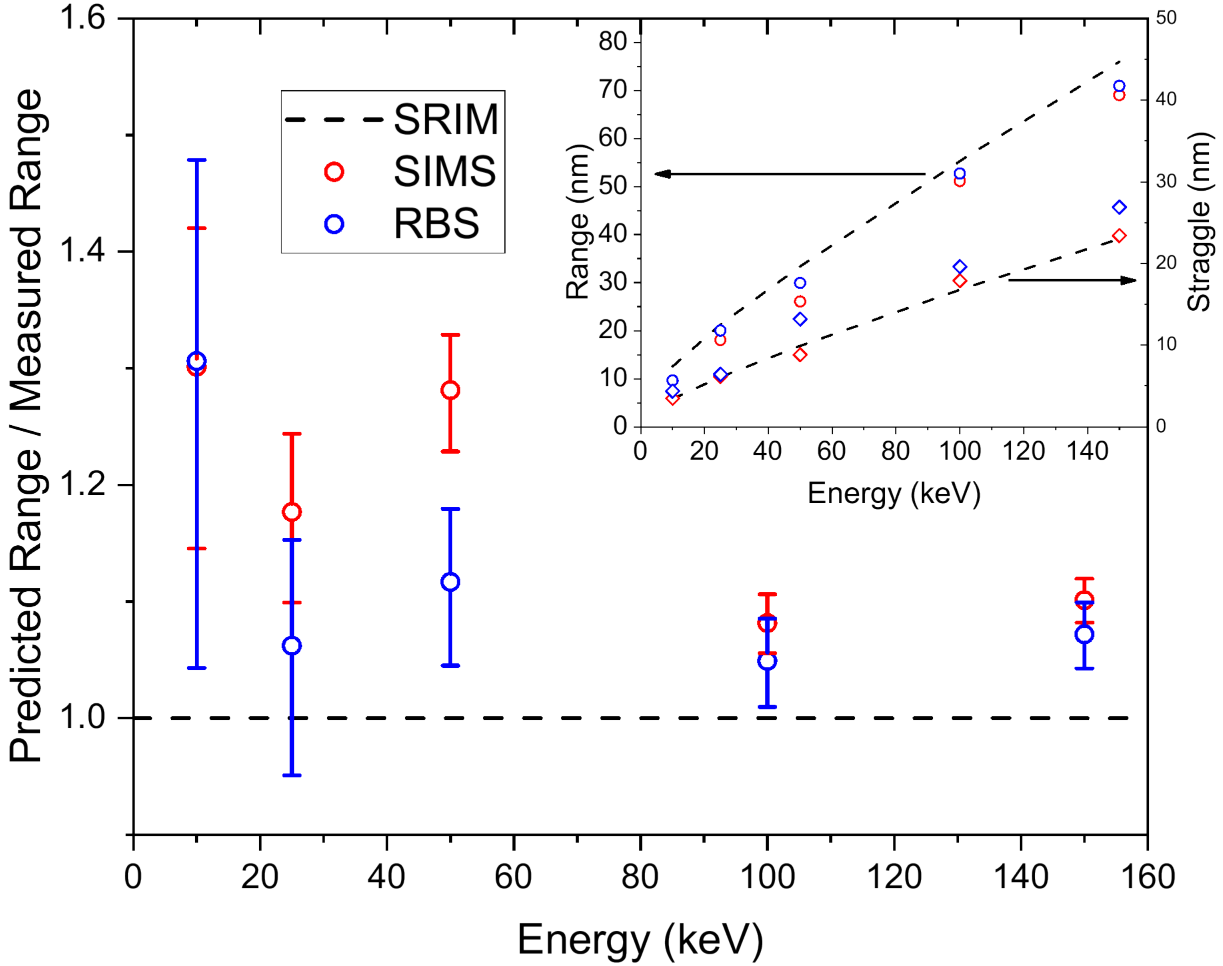
- Spot Size
- Range Prediction vs Experiment
- How to reduce Energy to stop in atomic monolayer

- Three characterization techniques of ion range
  - Secondary Ion Mass Spectrometry (SIMS)
  - Rutherford Backscattering Spectrometry (RBS)
  - Atom Probe Tomography (APT)

Technique	Sample Requirements	Depth Resolution	Sensitivity	Probe Technique
SIMS	Best for Si	2 nm	1 ppm	Sequential (sputtering)
RBS	Any planar sample	2 nm	1 monolayer	Full depth (~50 um)
APT	Sharp tip geometry	2 A	1 monolayer	Sequential (field extraction)

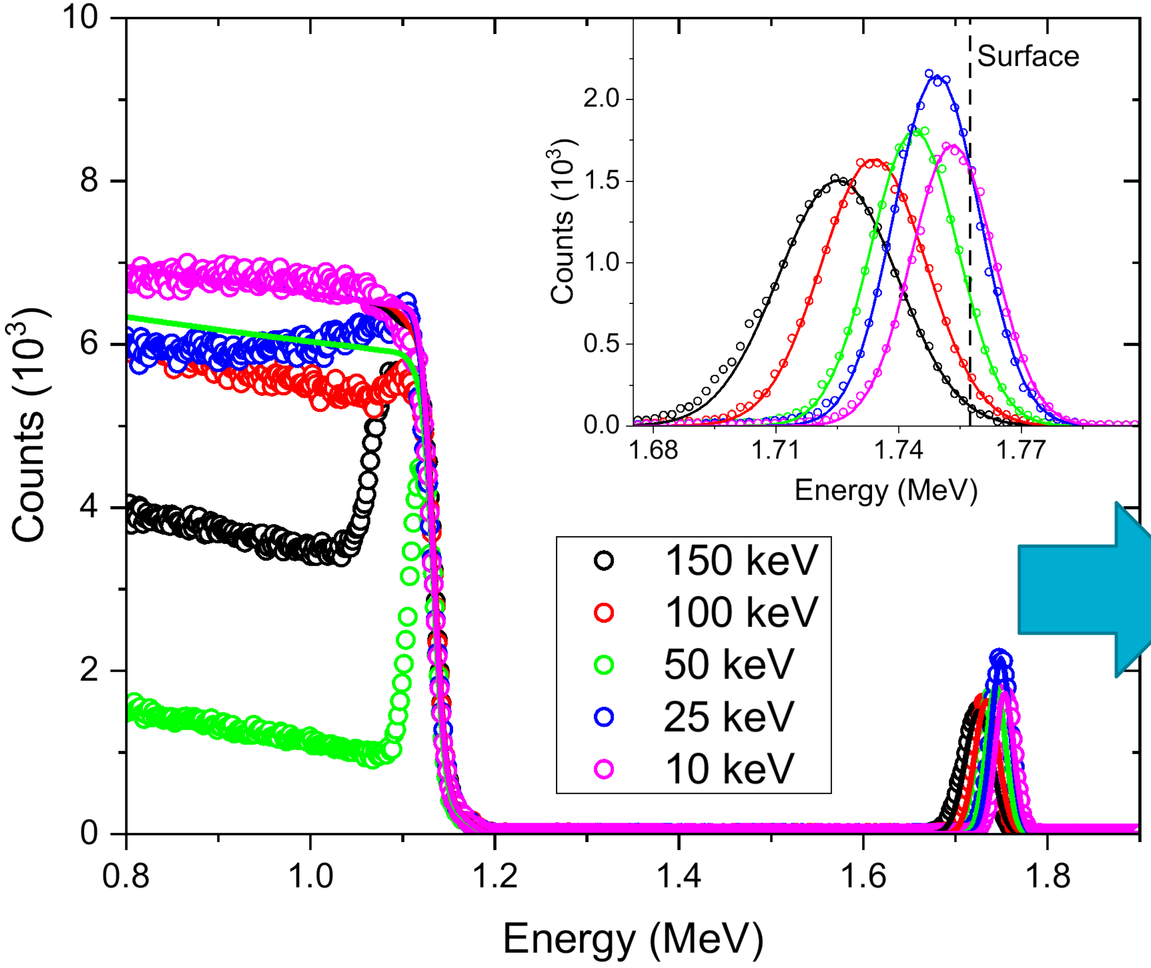
Implant Depth Determination - SIMS

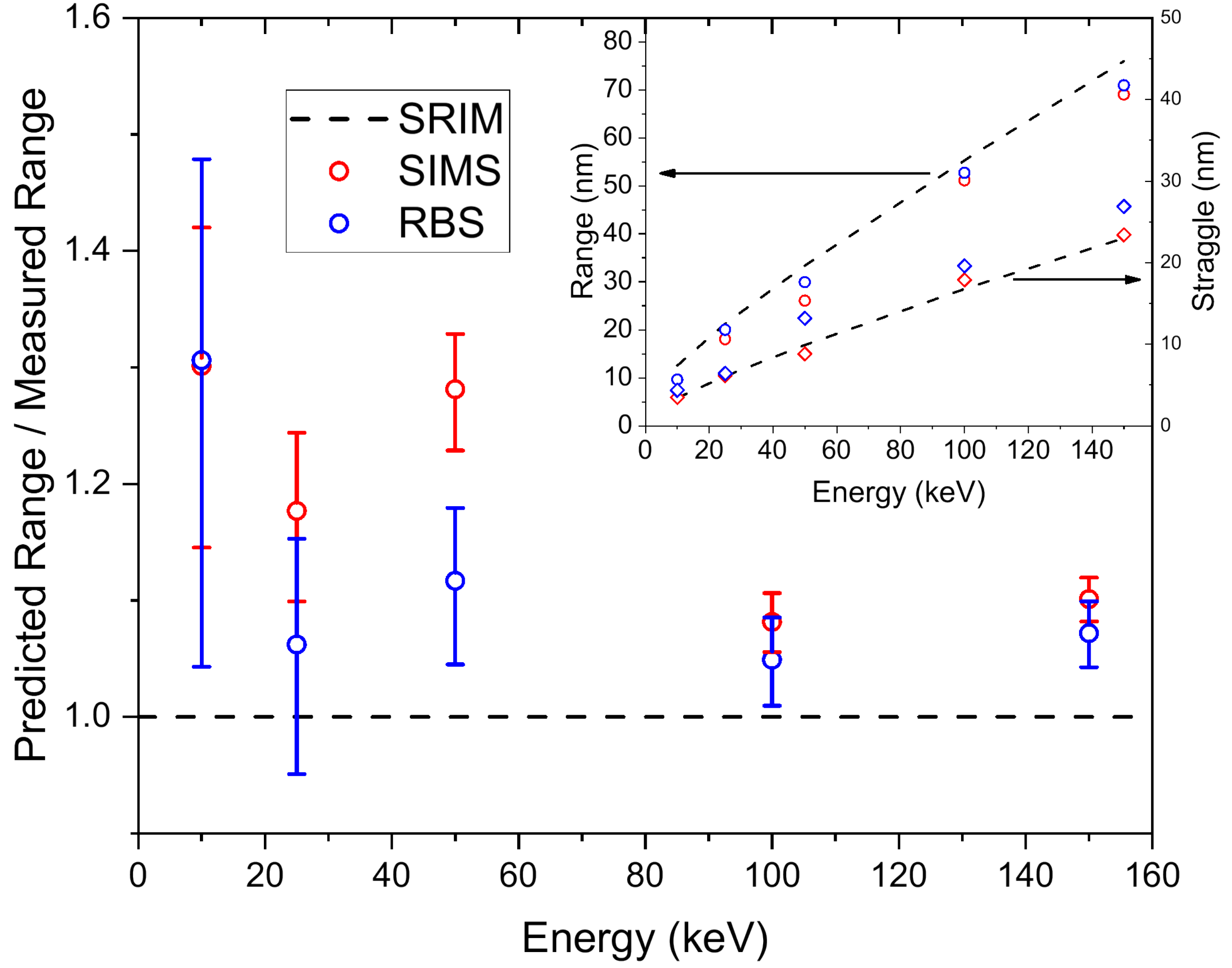




Implant Depth Determination - RBS

2 MeV 1024 ch. He RBS → Need high resolution data to achieve 2 nm depth resolution

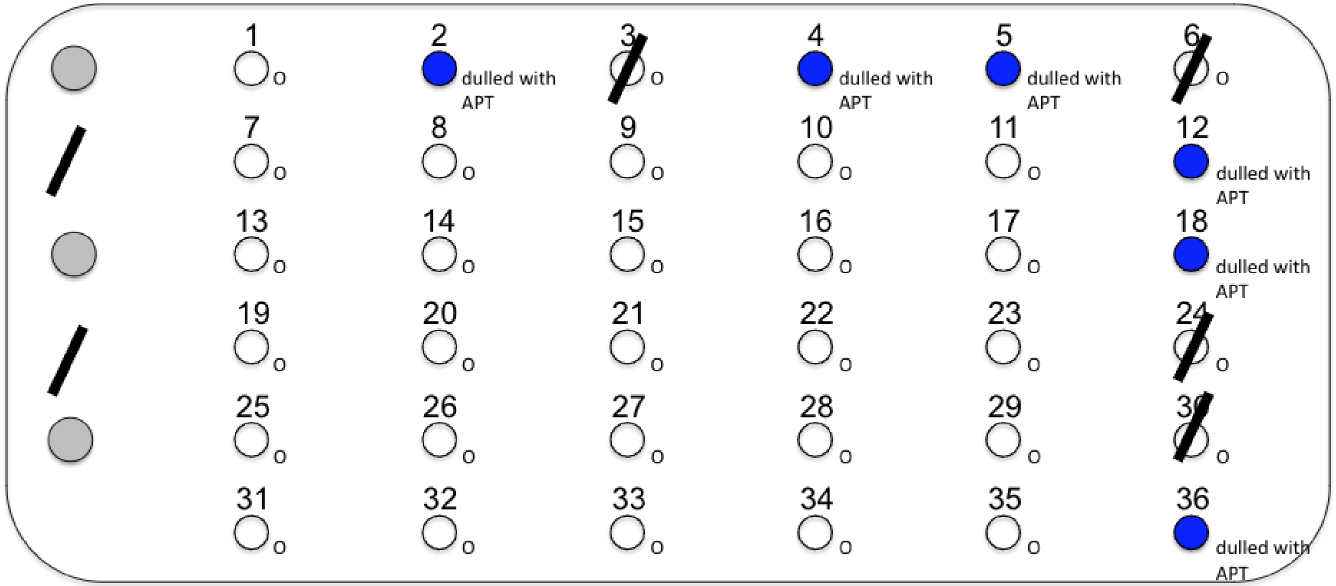


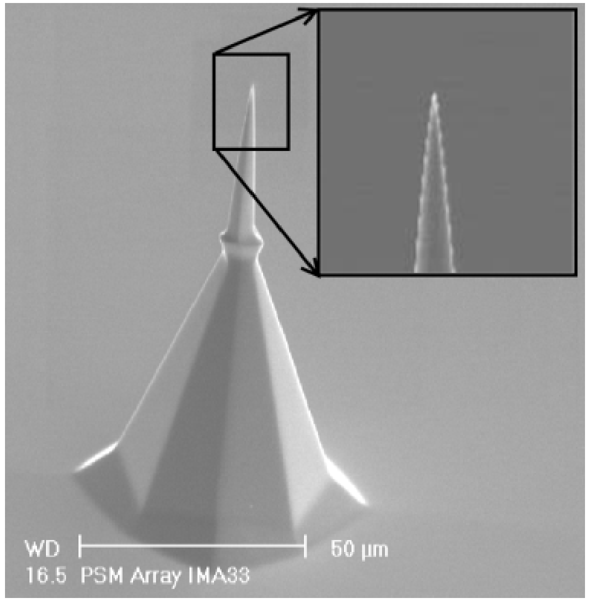


Sample Preparation for Ultra-Low Energy Ion Implantation

- Start out with APT tip
- Pre-dull tip with APT
- Implant with low-energy Au

Implant Plan



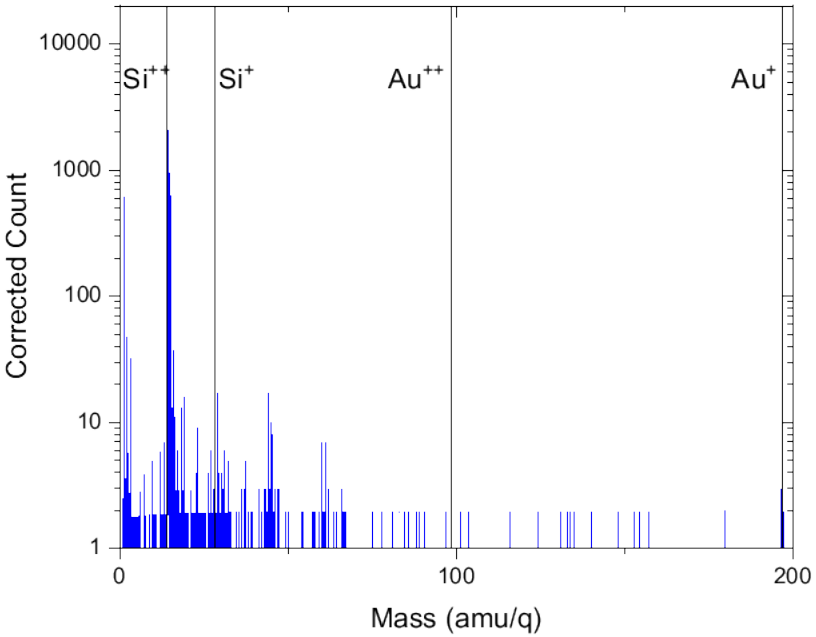


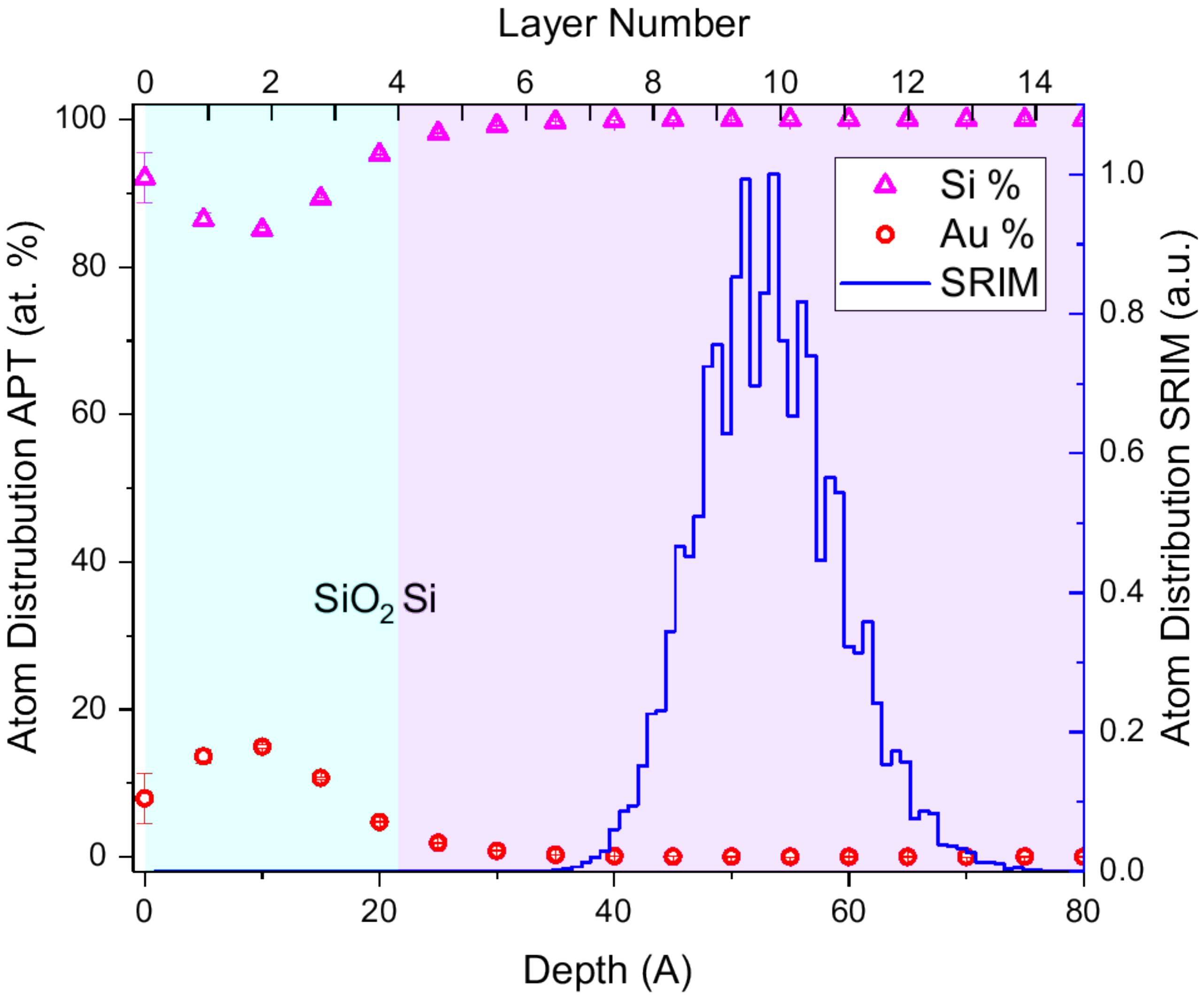
Tip #	Energy
2	25 keV
4	10 keV
5	5 keV
12	3 keV
18	2 keV
36	1 keV

APT Measurements done by Jonathan Poplawsky at ORNL

Atom-Probe Tomography on Ultra-Low Energy Implanted Sample

- APT needs to be run in Voltage-mode
- SRIM overestimates range by **6X**
- Indicates that 100s of eV will be sufficient to target single atomic layers
  - Can do (at cost of spot size) with ~15 eV energy spread





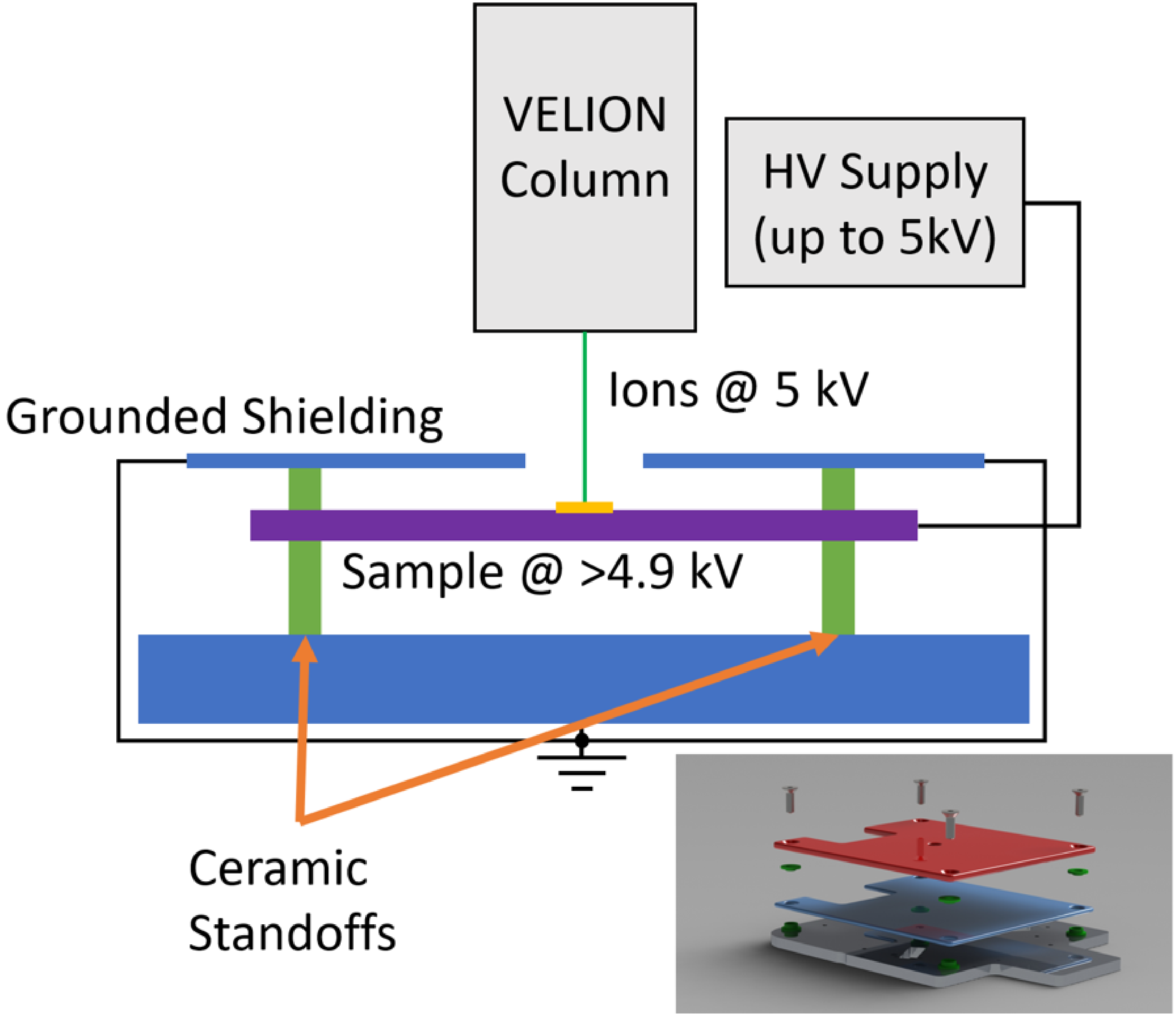
Expanding the Energy Range Further

- Spot Size
- Range Prediction vs Experiment
- How to reduce Energy to stop in atomic monolayer

  - 1 keV Au stops within 4 atomic layers of SiO_2
  - To target atomic layers of 2D materials
    - Stopping in a single layer
    - Lower mass ions interesting for their optical properties

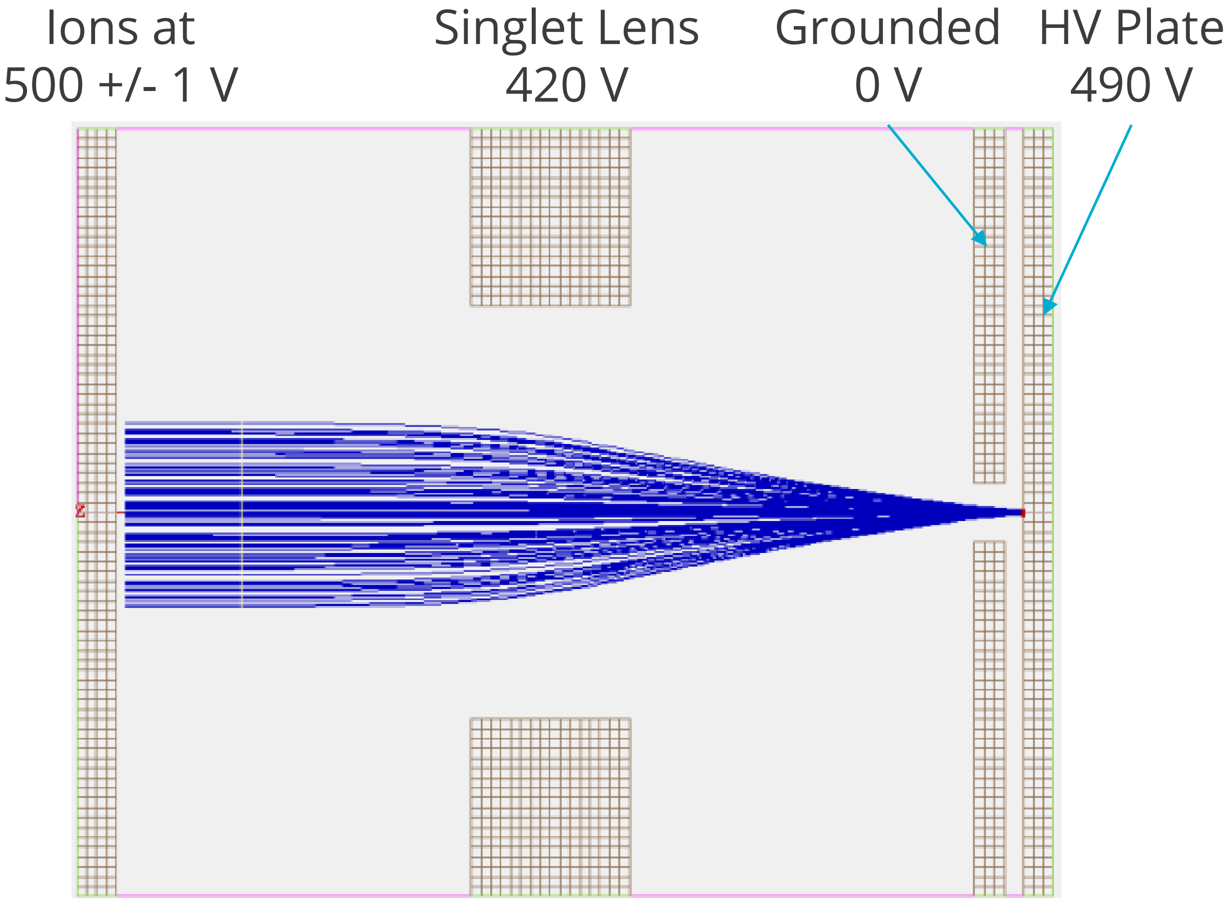
  → Need even lower energy

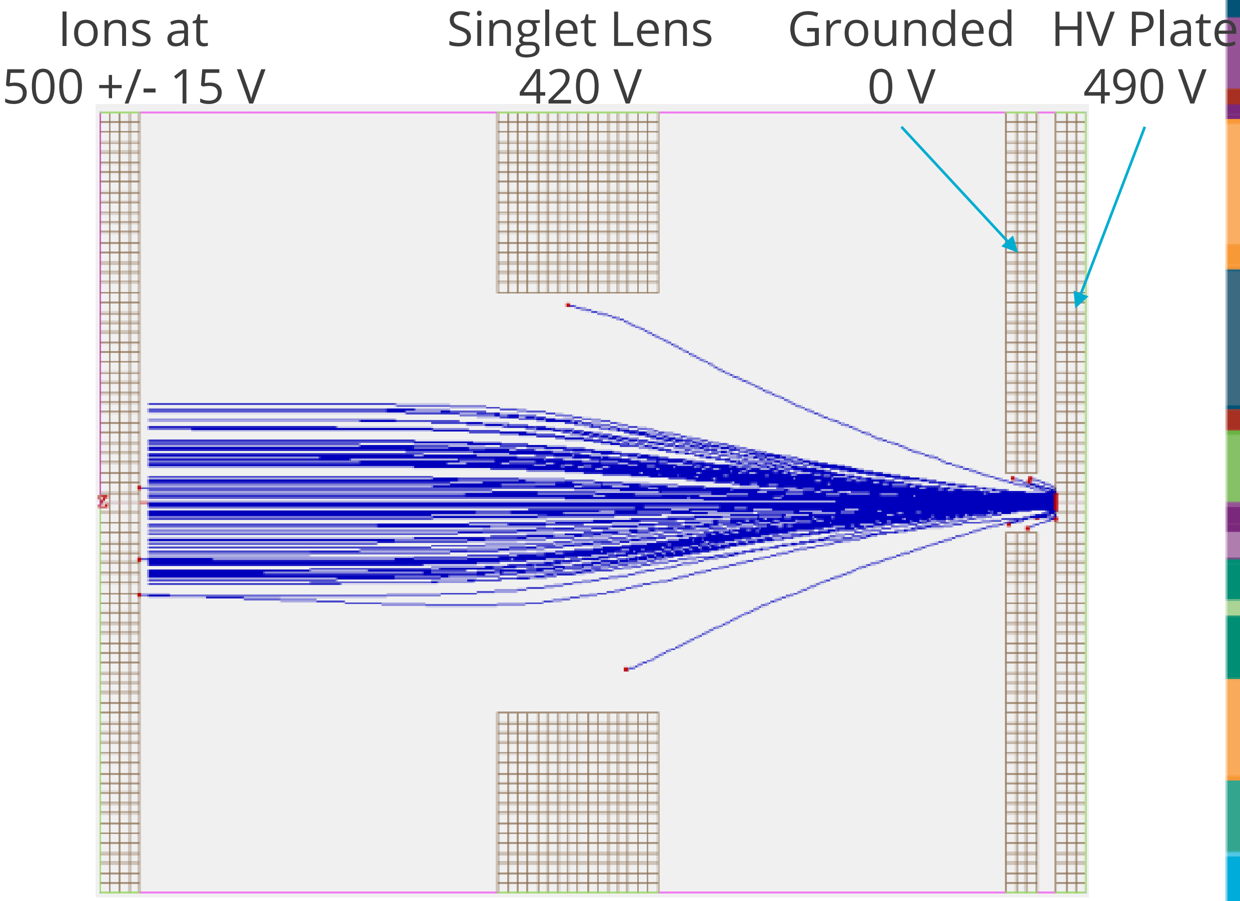
  - Biased sample holder to decelerate ions



Ion Source Energy Spread Disallows Focusing

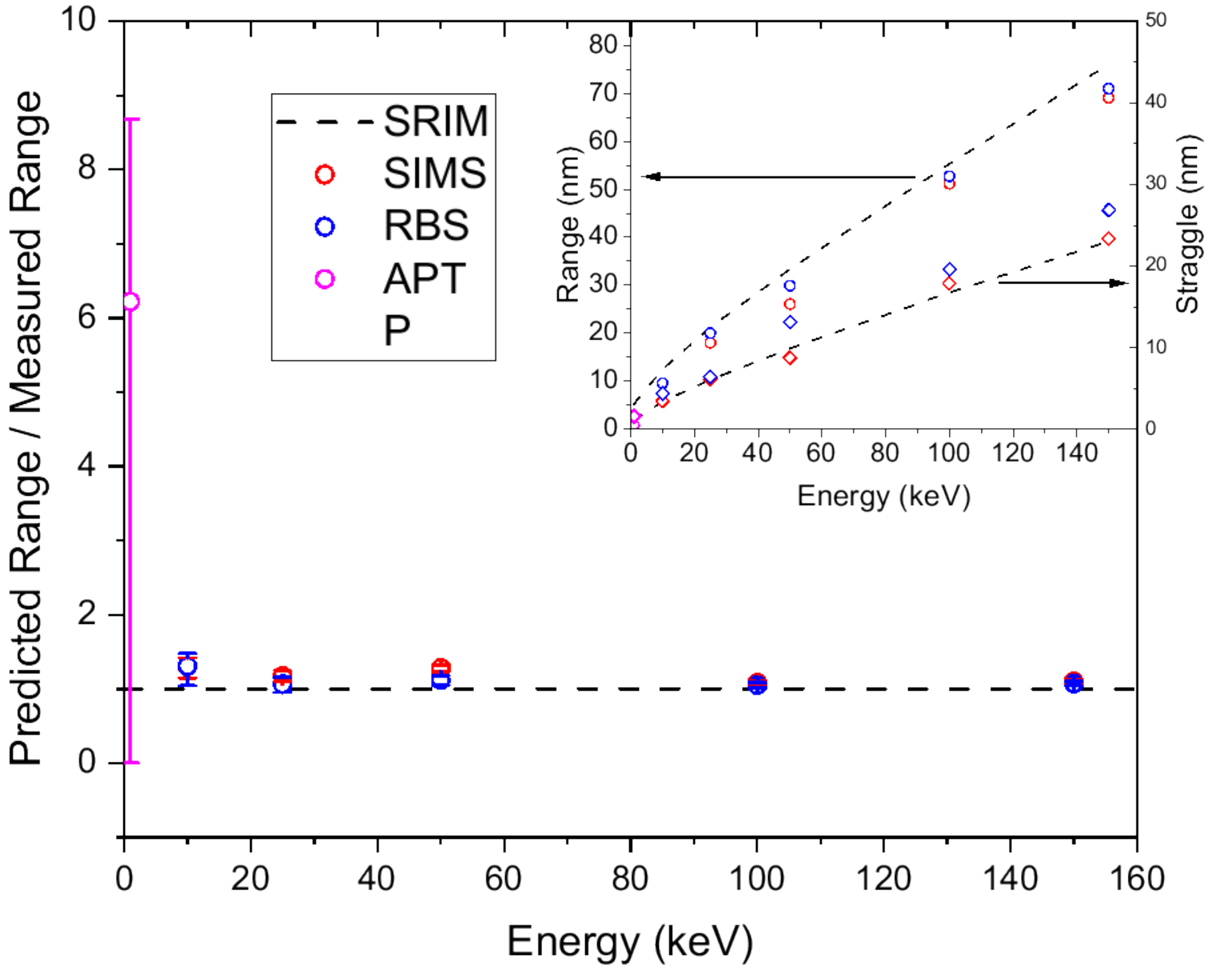
- SIMION Simulations show the ion deceleration leads to increased spot size
- When landing energy ~ energy spread ions get reflected + spot size increases





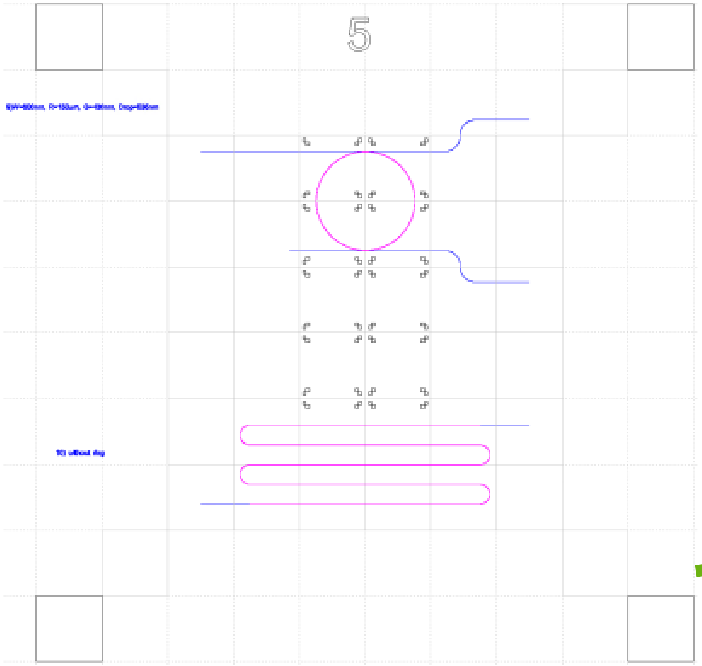
Conclusion

- FIB implantation can likely be performed into single atomic layers
- Source energy spread main contributor to spot size
- SRIM is off 6X at 1 keV (vs. 30 % error at 10 keV)

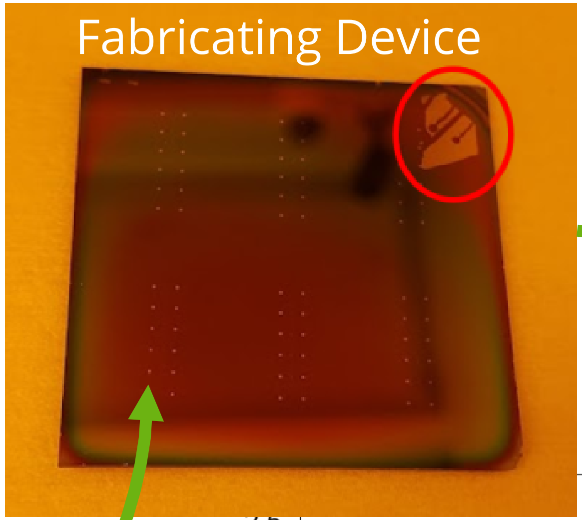


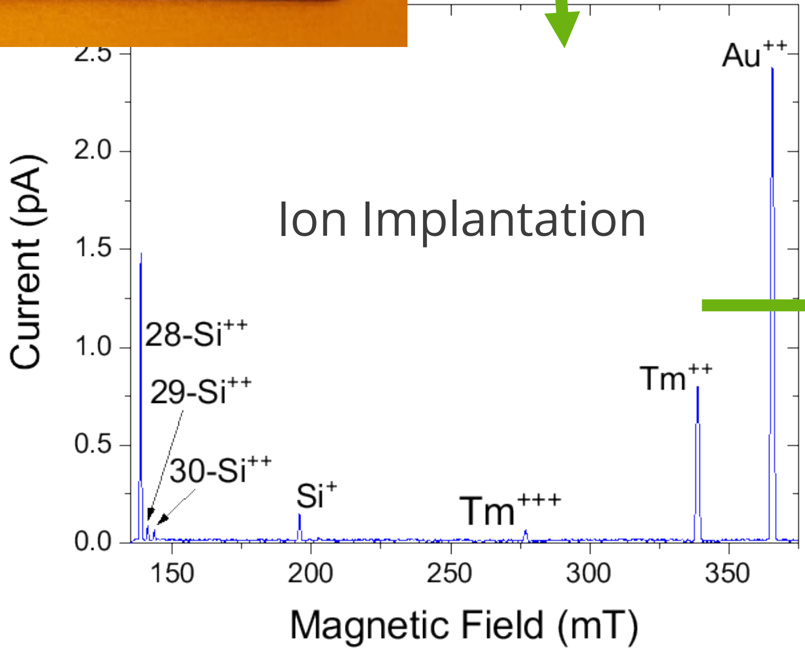
Interaction with CINT Users – Design to Manuscript

M. Hosseini @ Purdue



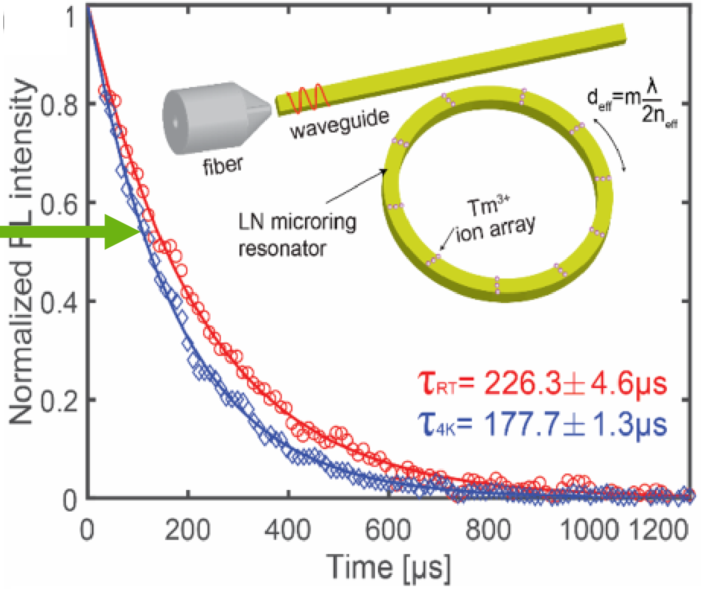
Discussion of design feasibility





Tm for $LiNbO_3$ Photonics

Post-Processing + Measurement



Dongmin Pak, ..., M. Titze et al. (in review)

How can YOU access these capabilities? CINT User Proposal

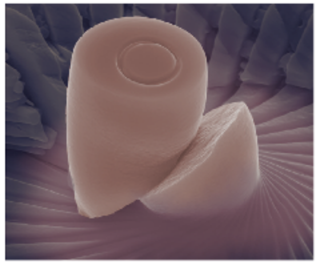
THE CENTER FOR INTEGRATED NANOTECHNOLOGIES

One Scientific Community Focused on Nanoscience Integration

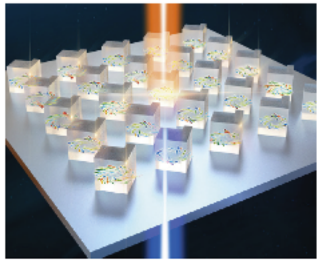
The Center for Integrated Nanotechnologies (CINT) is a Department of Energy Office of Science Nanoscale Science Research Center. CINT offers world-leading scientific expertise and specialized capabilities to create, characterize, and integrate nanostructured materials at a range of length scales, from the nano- to meso-scale. It is jointly operated by Los Alamos and Sandia national laboratories and leverages the unmatched scientific and engineering expertise of the host labs.

Science Thrusts

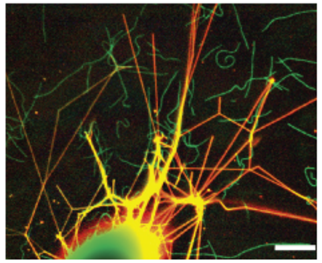
Integration is the key to exploiting the novel properties of nanoscale materials and creating new technologies. CINT's scientific staff and capabilities are organized around four interdisciplinary science thrusts which address different challenges in nanoscience integration.



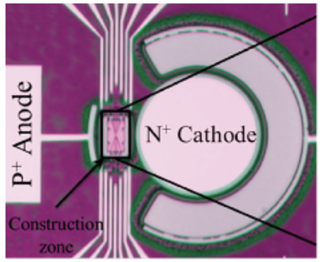
In-Situ Characterization and Nanomechanics
Developing and implementing world-leading capabilities to study the dynamic response of materials and nanosystems to mechanical, electrical, or other stimuli.



Nanophotonics & Optical Nanomaterials
Synthesis, excitation, and energy transformations of optically active nanomaterials and collective or emergent electromagnetic phenomena (plasmonics, metamaterials, photonic lattices).



Soft, Biological & Composite Nanomaterials
Synthesis, assembly, and characterization of soft, biomolecular, and composite nanomaterials that display emergent functionality.



Quantum Materials Systems
Understanding and controlling quantum effects of nanoscale materials and their integration into systems spanning multiple length scales.

User Program

CINT is an Office of Science national user facility. CINT helps the international research community perform cutting-edge research in the areas of nanoscience and nanotechnology, and is available free of charge for open science. As a user facility, CINT has the structure and mission to collaborate widely across academia, industry, and within DOE labs. Access is via peer-reviewed technical proposals. Proprietary research may be conducted in accordance with Federal regulations for full-cost recovery. CINT cannot provide funding to users.

The Center for Integrated Nanotechnologies (CINT)

https://cint.lanl.gov/