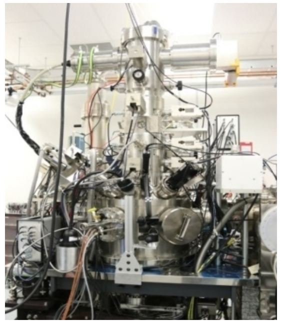
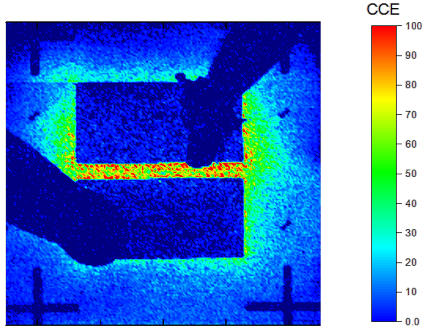
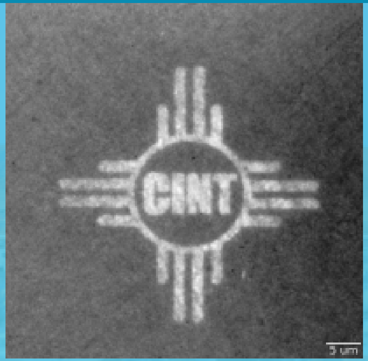
SAND2023-05154C

Sandia National Laboratories

Towards Deterministic Defect Center Fabrication Using Sandia's Focused Ion Implantation Capabilities







E. Bielejec

Sandia National Laboratories, Albuquerque, NM 87185

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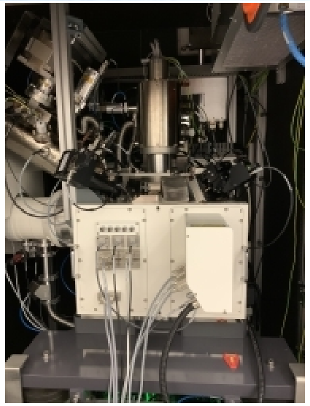
Outline

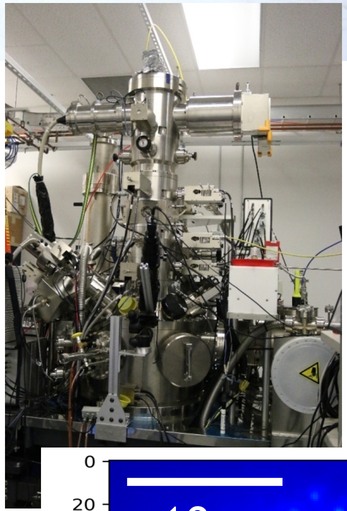
- Introduction to Focused Ion Beam Capabilities at Sandia's Ion Beam Laboratory (IBL)

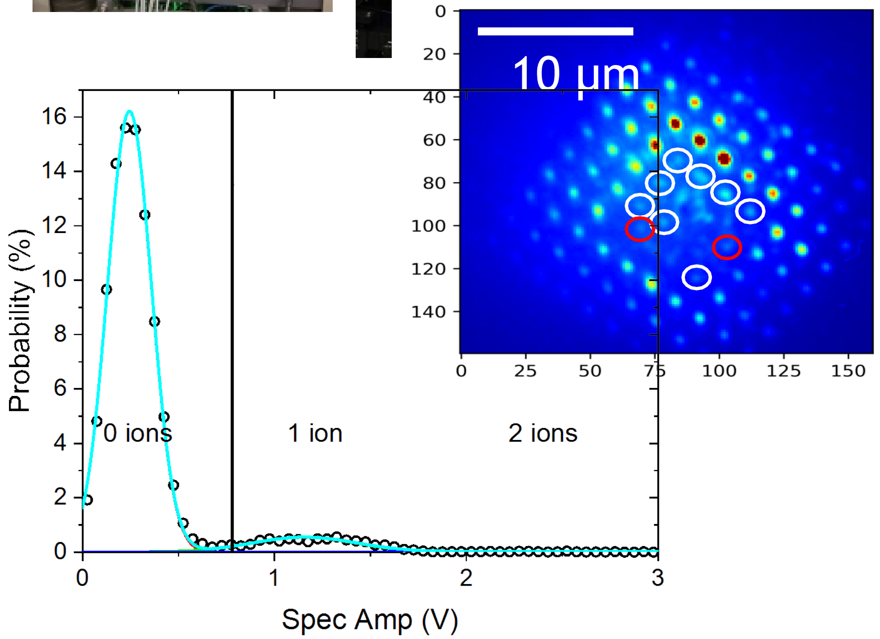
- Multi-species Focused Ion Beam (FIB)
  - Sandia's nanoImplanter and Raith Velion
  - New Liquid Metal Alloy Ion Source Development

- Single Defect Centers in Wide Bandgap Substrates
  - In-situ Counting – control the # of implanted ions
  - In-situ Photoluminescence – verify defect centers

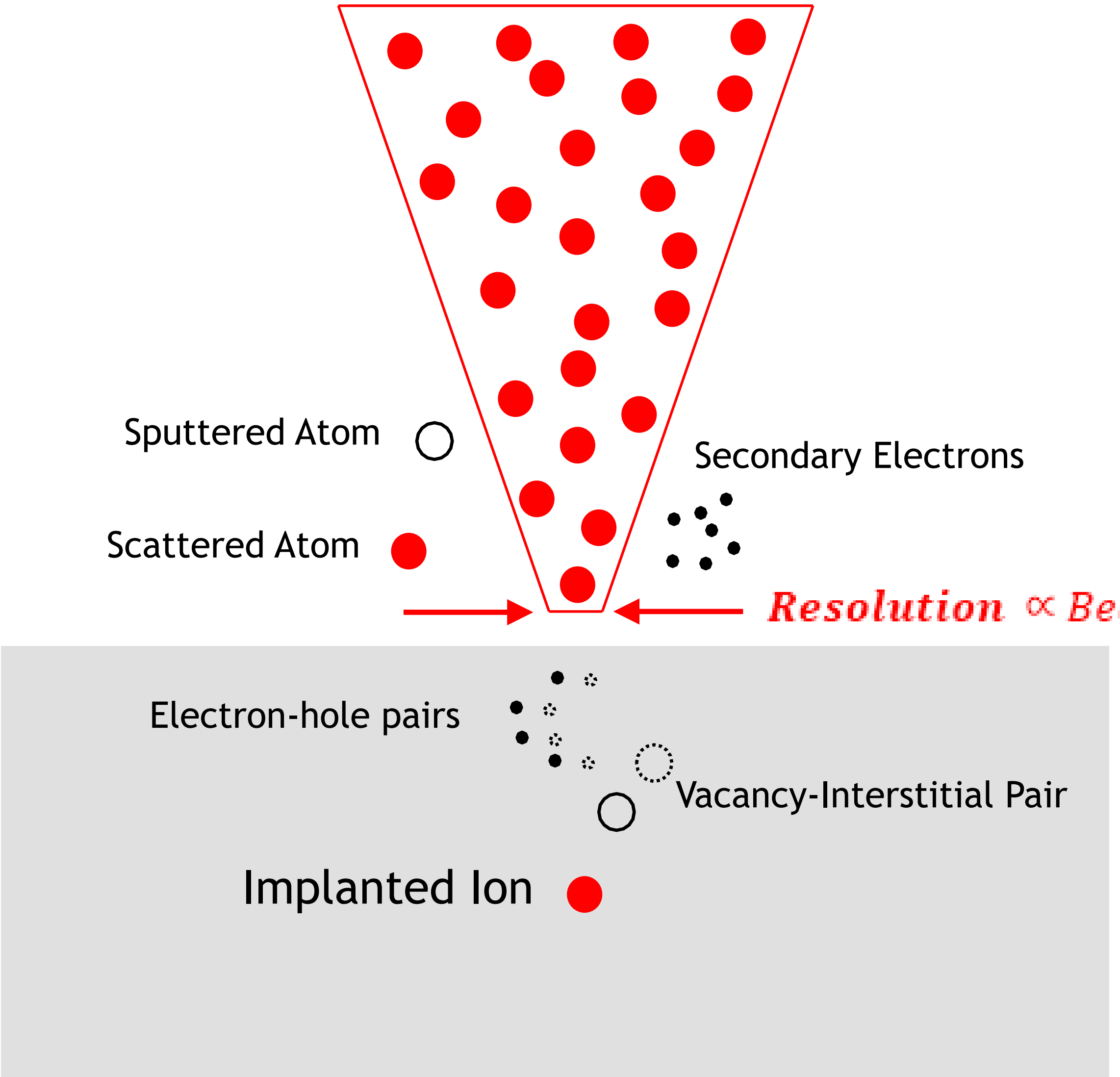
- Summary and Outlook for FIB Implantation







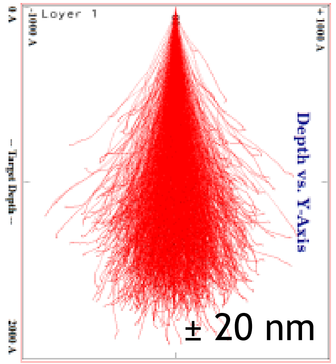
Nanofabrication using Focused Ion Beam (FIB) Implantation



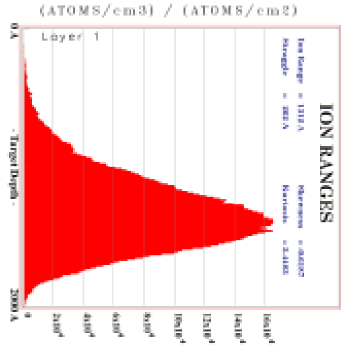
Resolution $\propto$ *Beam Spot Size + Alignment Accurary + XY Straggle*

SRIM simulation - XY straggle

200 keV Si into diamond



Depth $\propto$ *Energy + Z Straggle*



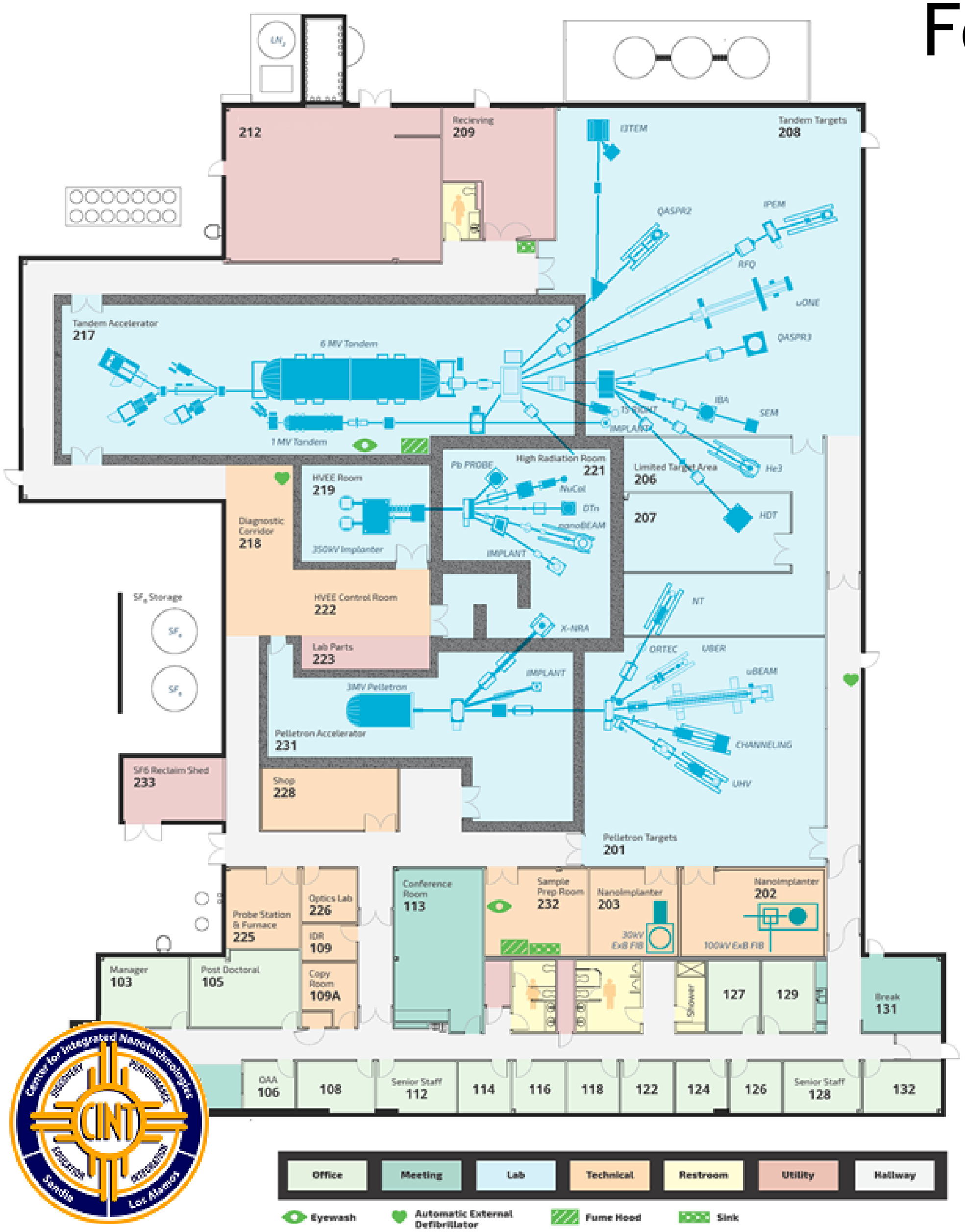
SRIM simulation - Z straggle

200 keV Si into diamond

131 ± 26 nm

building 720
ION BEAM LABORATORY

Overview of Sandia's Ion Beam Laboratory (IBL) Focused Ion Beam (FIB) Capabilities



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

Installing

(8) 35 kV Plasma FIB

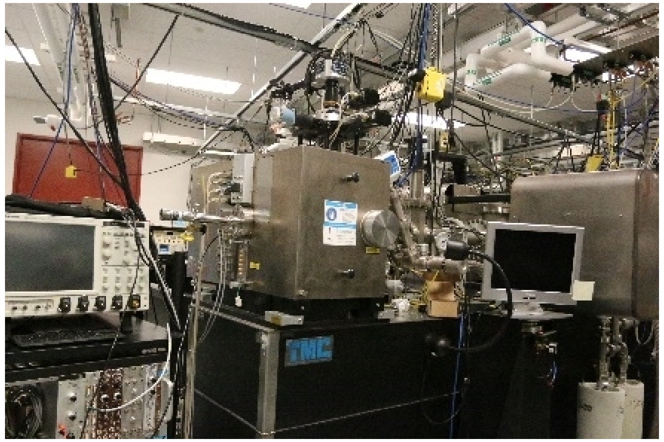
Low energy focused nanobeams <1 to 20 nm

25 end-stations including *in-situ* DLTS, PL, TEM, SEM, 1200°C heating, etc…

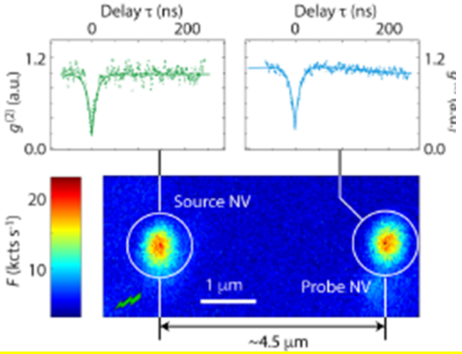
High Energy Focused Ion Beams ~1 μm

Michael Titze, Gyorgy Vizkelethy

- 6 MV Tandem microbeam (microONE)

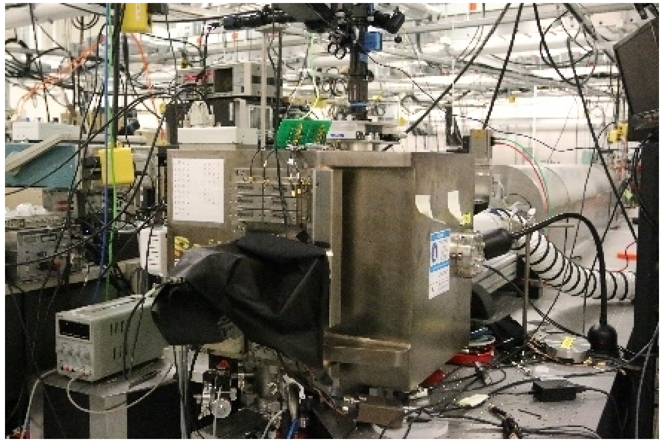


- High resolution laser stage
- Spot size <1 μm
- Energy 0.8 – 70 MeV
- H to Au

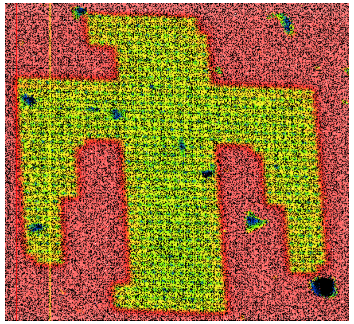


NV PL in diamond

- 3 MV Pelletron microbeam (Light Ion Microbeam)

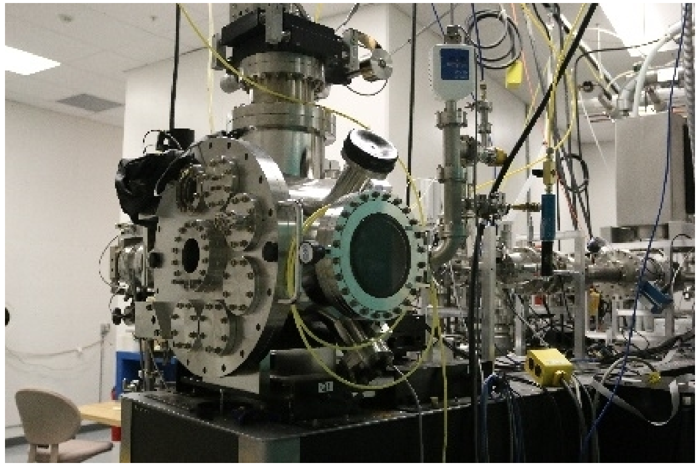


- High resolution laser stage
- Spot size <600 nm
- Energy 0.25 – 3 MeV
- H, He, N, Ar, Xe, …

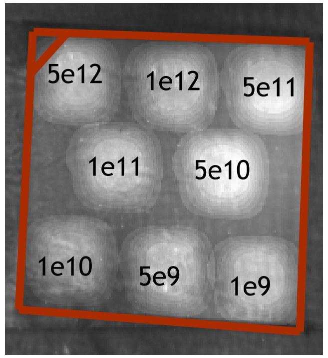


IBIC on PIN diode

- 350 kV HVEE microbeam (NanoBeamLine)



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



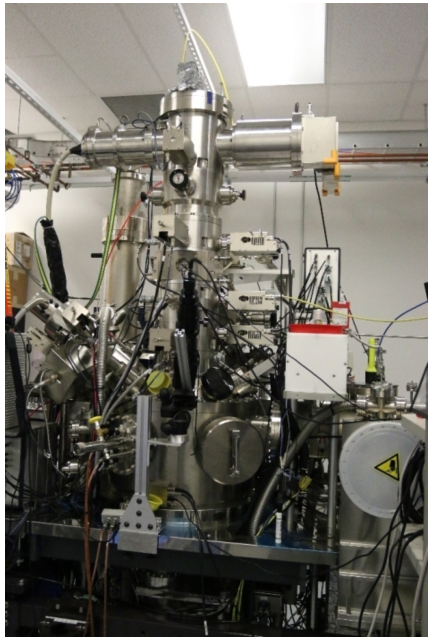
Ni into diamond

A. Lozovoi, *et al.*, Nature Electronics **4**, 171-742 (2021) and Science Advances **8**, 1 (2022)

Low Energy Focused Ion Beams <1 to 20 nm

Michael Titze, Alex Belianinov

- 100 kV A&D FIB100NI (nanoImplanter)

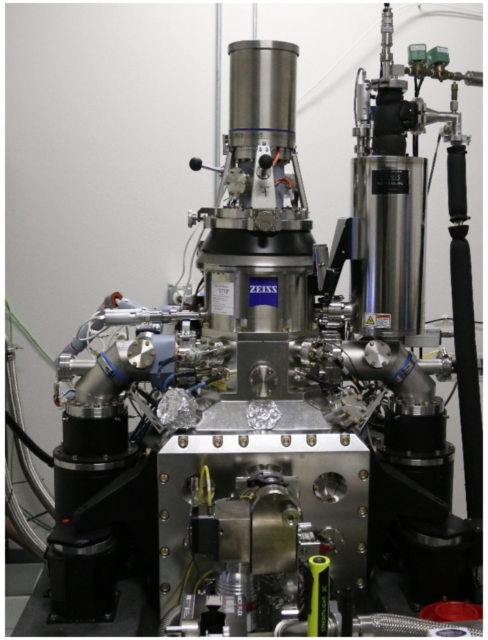


- High resolution laser stage
- Spot size <10 nm (Ga)
- Energy 10 – 200 keV
- 1/3 periodic table

- 35 kV Raith Velion (Velion)

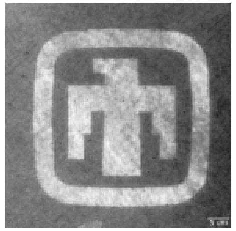
- High resolution laser stage
- Spot size <6 nm (Ga)
- Energy 5 – 70 keV
- 1/3 periodic table

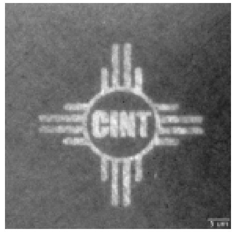
- 35 kV Zeiss Orion Plus (HeIM)

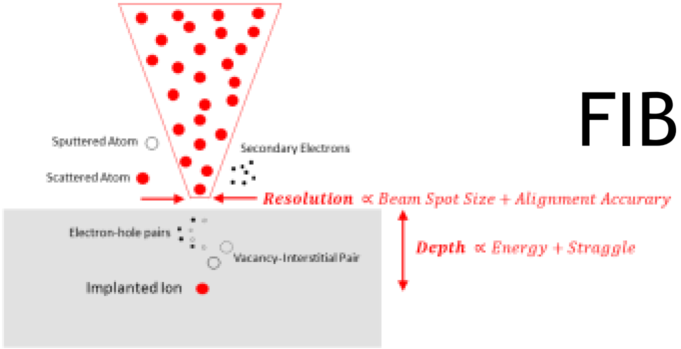


- Piezo stage
- Spot size <1 nm
- Energy 10 – 35 keV
- He

All three equipped with Raith lithography software for patterning







FIB Implantation Resolution

What our resolution?

<u>High Energy FIB</u>

Spot Size ~ 1 μm

Alignment accuracy ~ 1 μm

Overall resolution ~ 1-2 μm

<u>Low Energy FIB</u>

Spot Size ~ few nm

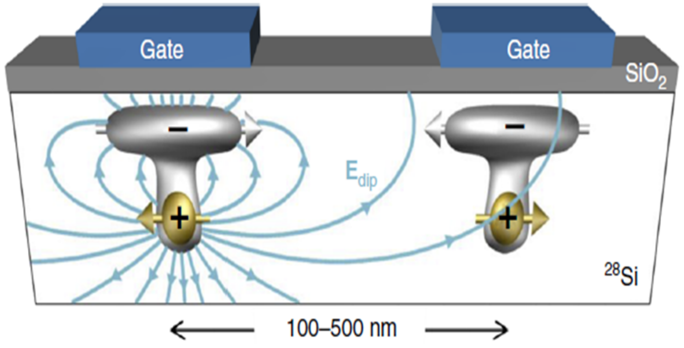
Alignment accuracy ~ 10's nm

Overall resolution ~ 20-50 nm

What is the needed resolution?

Silicon quantum processor with robust long-distance qubit couplings

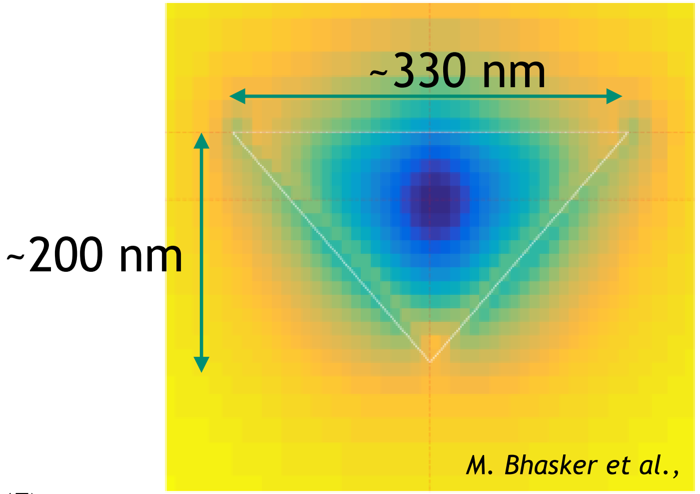
Guilherme Tosi[1], Fahd A. Mohiyaddin[1,3], Vivien Schmitt[1], Stefanie Tenberg[1], Rajib Rahman[2], Gerhard Klimeck[2] & Andrea Morello[1]



G. Tosi et al., Nat. Comm. ***8****, 450 (2017)*

- Depth z_d = 15-20 nm ☑
- Separation of 100-500 nm ☑

Nanophotonic Applications



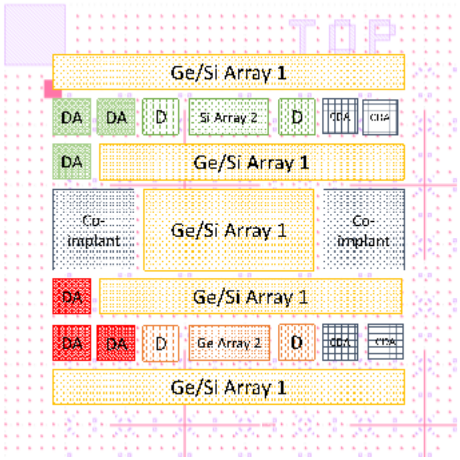
Center of mode is ~ 55 nm below the surface of the waveguide ☑

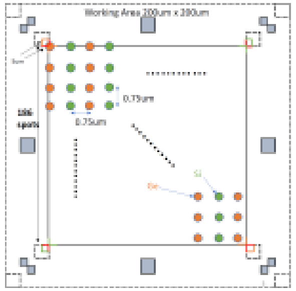
Low Energy Implantation? Lateral Positioning – OK, Target Depth – OK!

Practical Example of FIB Implantation

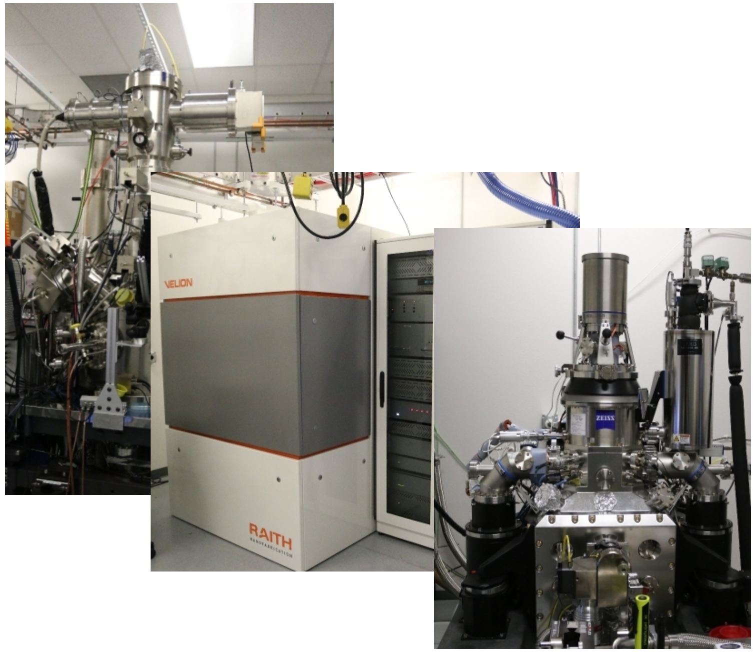
Conversation of what is needed?

Design and layout of sample

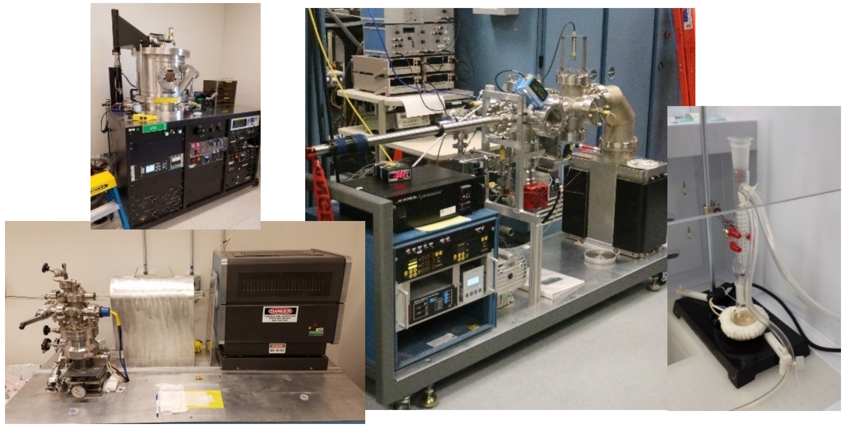




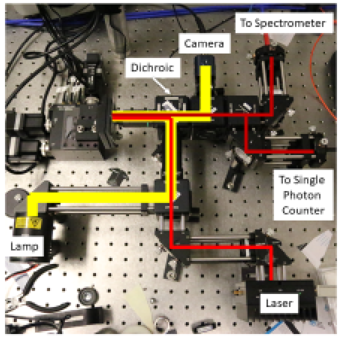
FIB Implantation/Irradiation

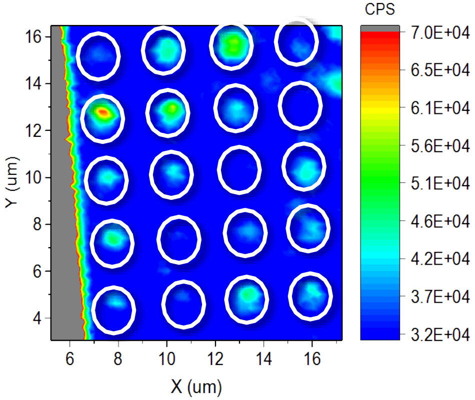


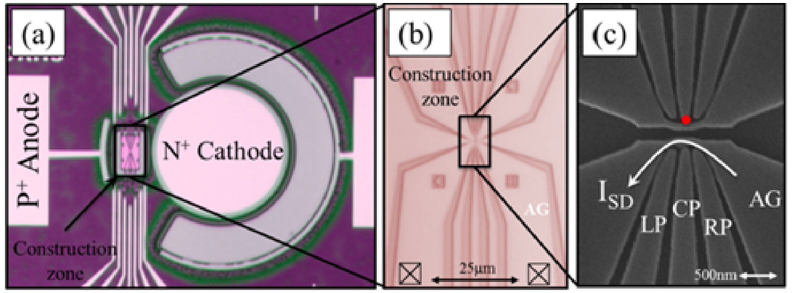
Post Implantation Sample Prep



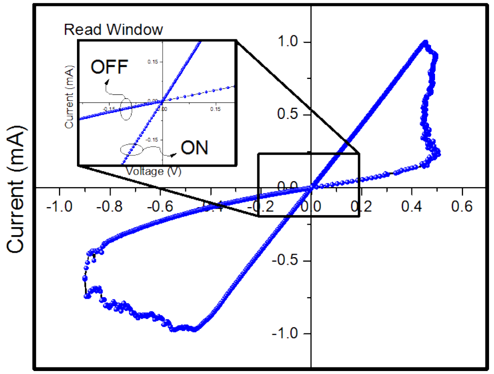
Post Implantation Characterization





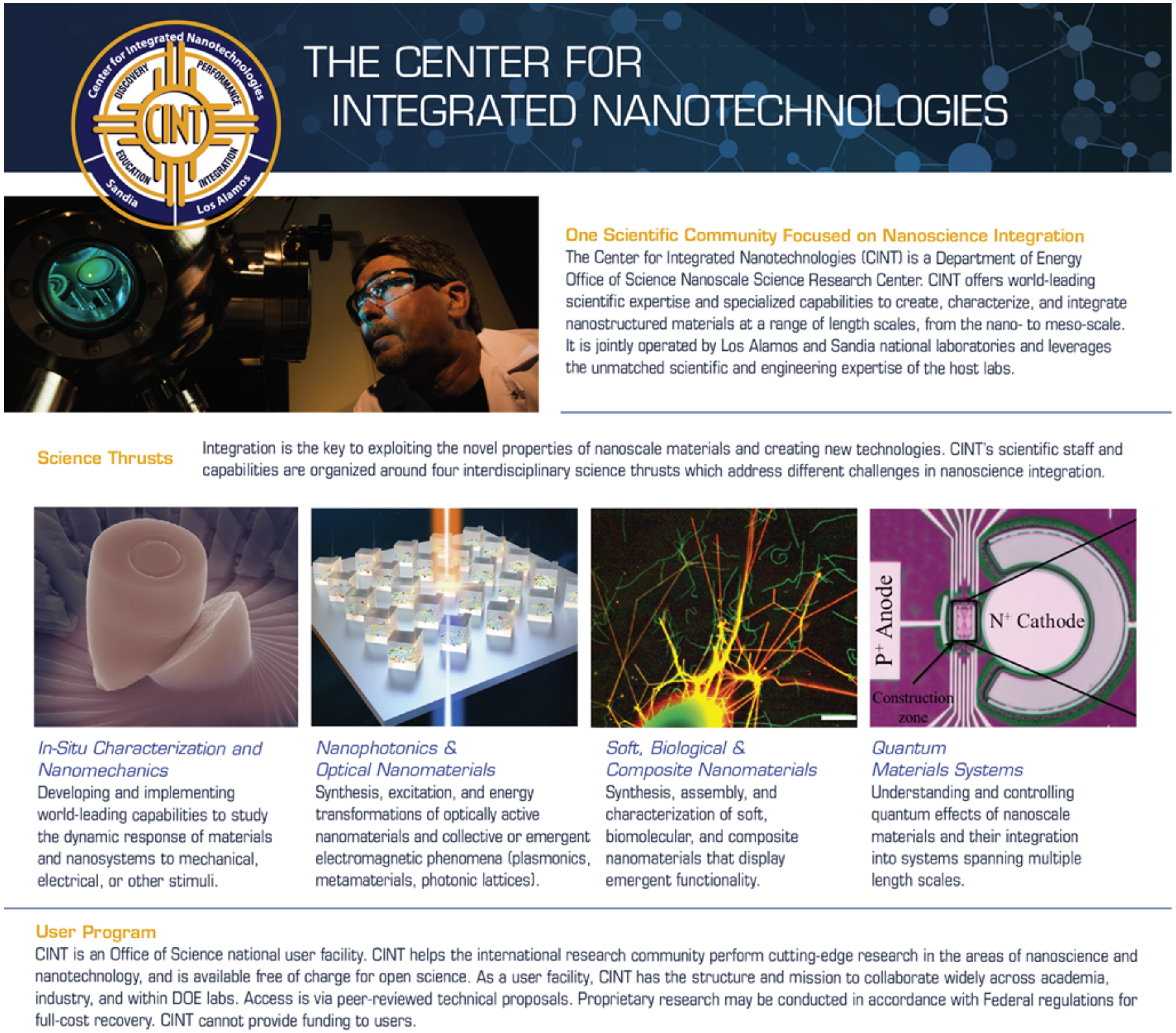


J. Pachaco, *et al*., *Rev. of Sci. Instr.* (2017)



J. Pachaco, et al. Appl. Phys. A **124** 626 (2018)

How can you access these capabilities? CINT User Proposals



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.

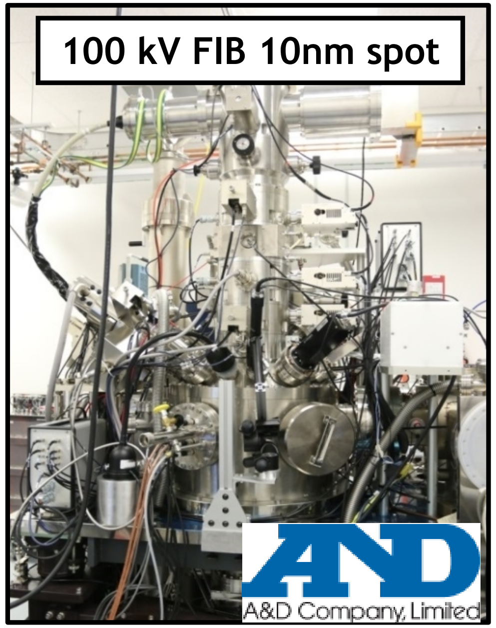
User Program

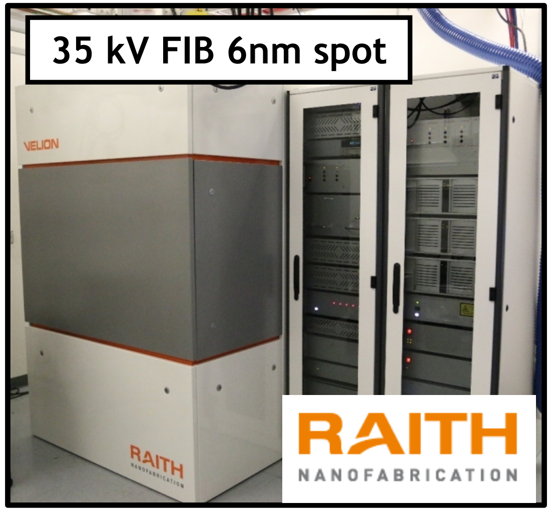
CINT is an Office of Science national user facility. CINT helps the international research community perform cutting-edge research in the areas of nanoscience and nanotechnology, and is available free of charge for open science. As a user facility, CINT has the structure and mission to collaborate widely 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) has both regular Fall and Spring call for proposals and Rapid Access available

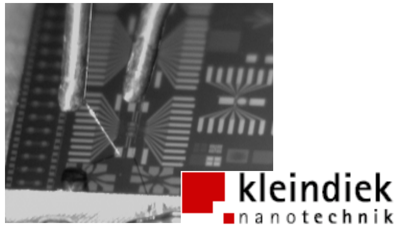
https://cint.lanl.gov/

FIB Implantation using the nanoImplanter and Velion

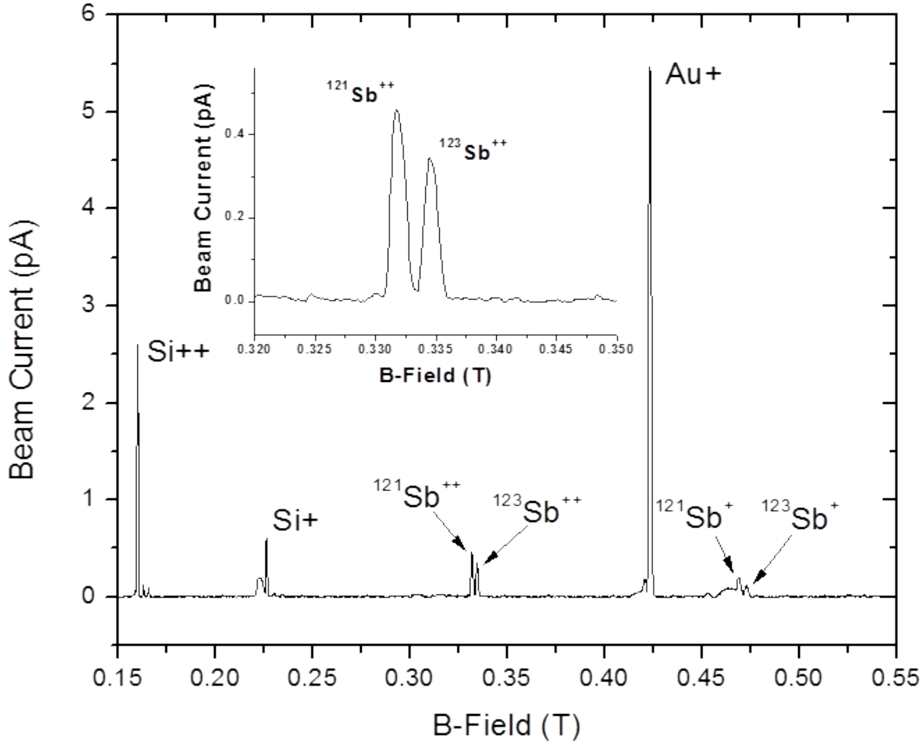




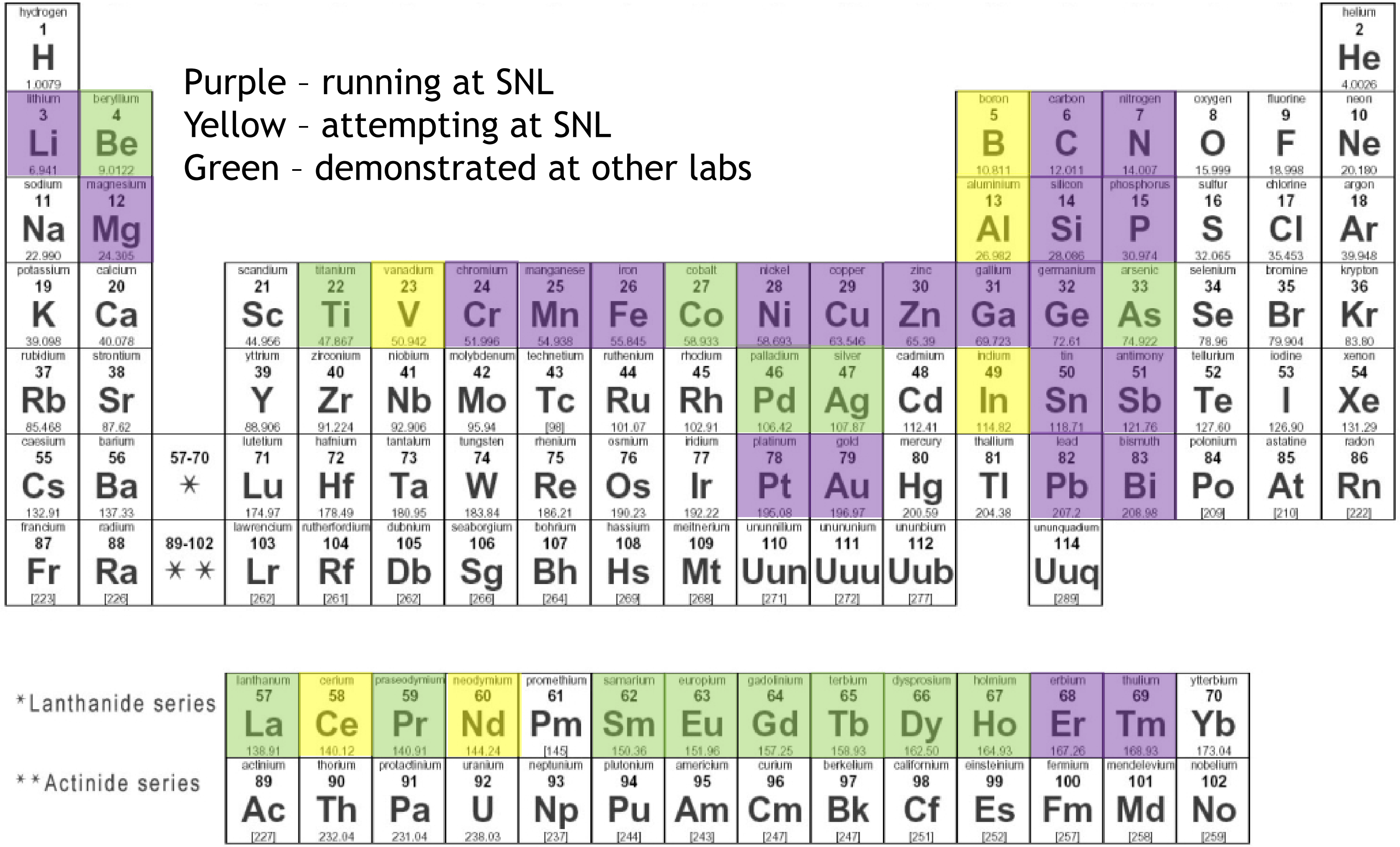
- Focused ion beam system (FIB)
  → nm beam spot size on target
- ExB Filter (Wien Filter)
  → Multiple ion species
    e.g., Li, Si, P, Sb, etc… (separating out ^{28}Si, ^{29}Si, etc…)
- Fast blanking and chopping
  → Single ion implantation
- Direct-write lithography
  → nm targeting accuracy
- Low temperature stage
- In-situ electrical probes



Sb Source: Mass Spectrum



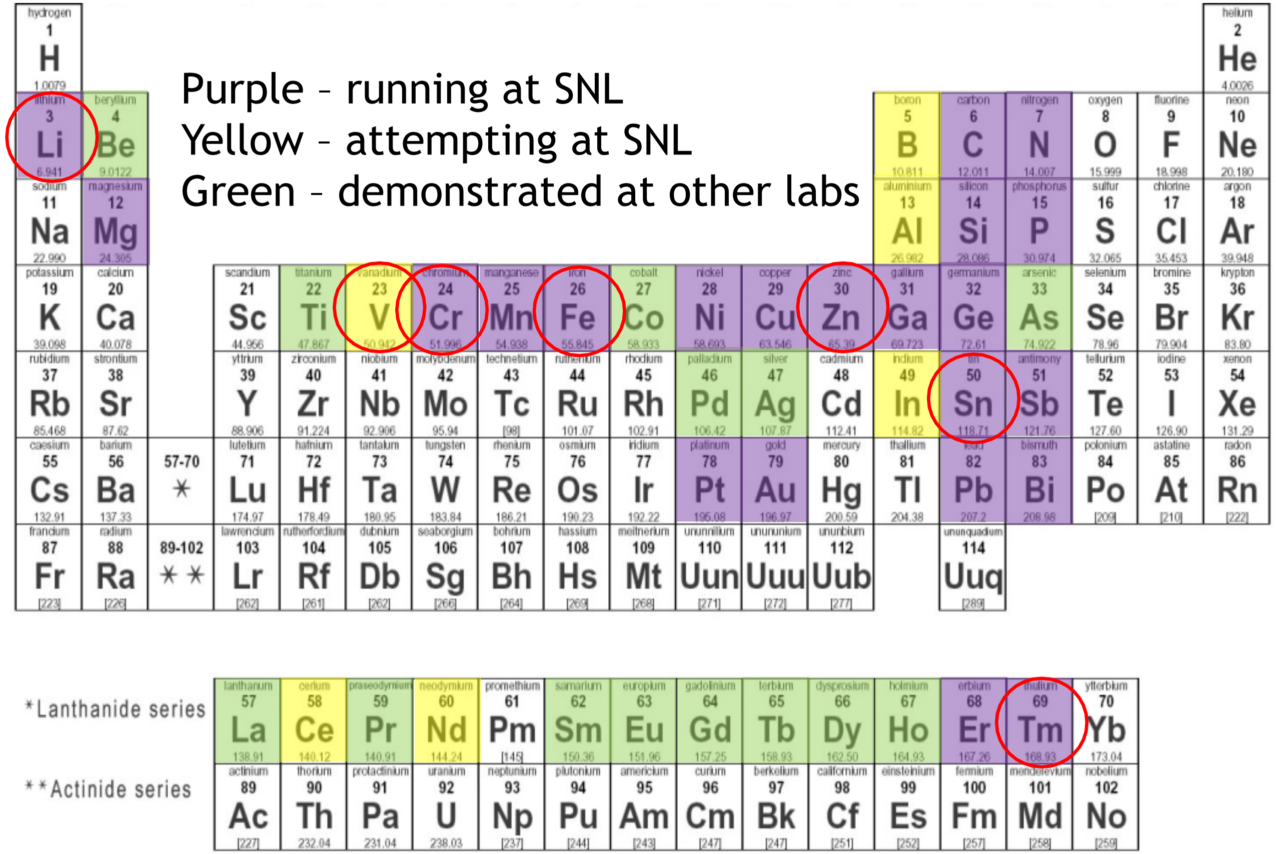
Available Liquid Metal Alloy Ion Sources (LMAIS) for FIB Implantation



After A. Weick University of Bochum
L. Bischoff, *et al.*, Applied Physics Reviews **3**, 021101 (2016)

Wide Variety of Ion Species Available and Developing More!

Examples of New Sources – Li, V, Cr, Fe, Zn, Sn, Tm (easy-ish)

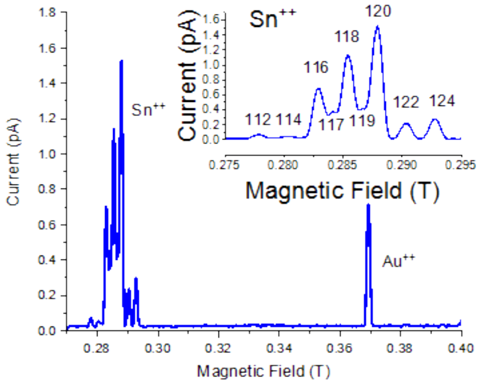


Purple – running at SNL
Yellow – attempting at SNL
Green – demonstrated at other labs

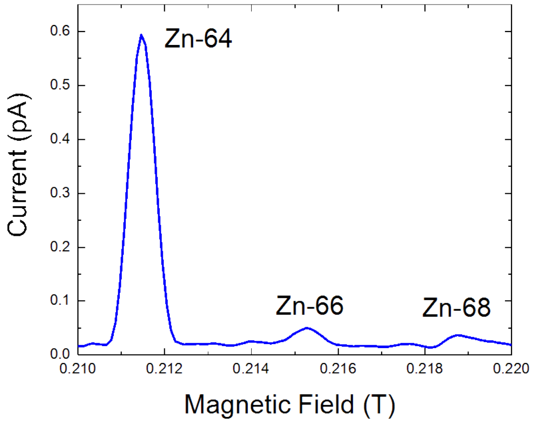
- Based on AuSiX or AuGeX alloys
- Easy to wet the tip and easy to run
- For example, AuSiLi

M. Titze *et al.*, JVST B, **39**, 012802 (2021)

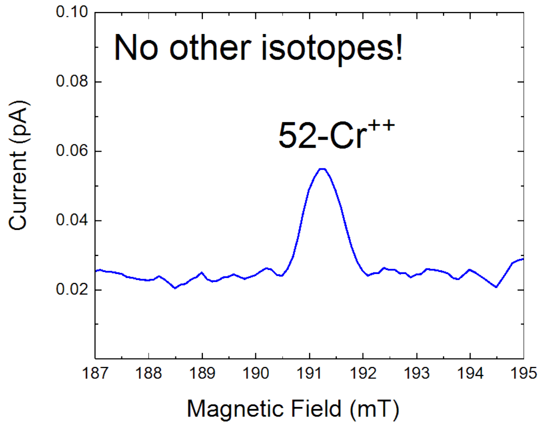
AuSn



AuSiZn



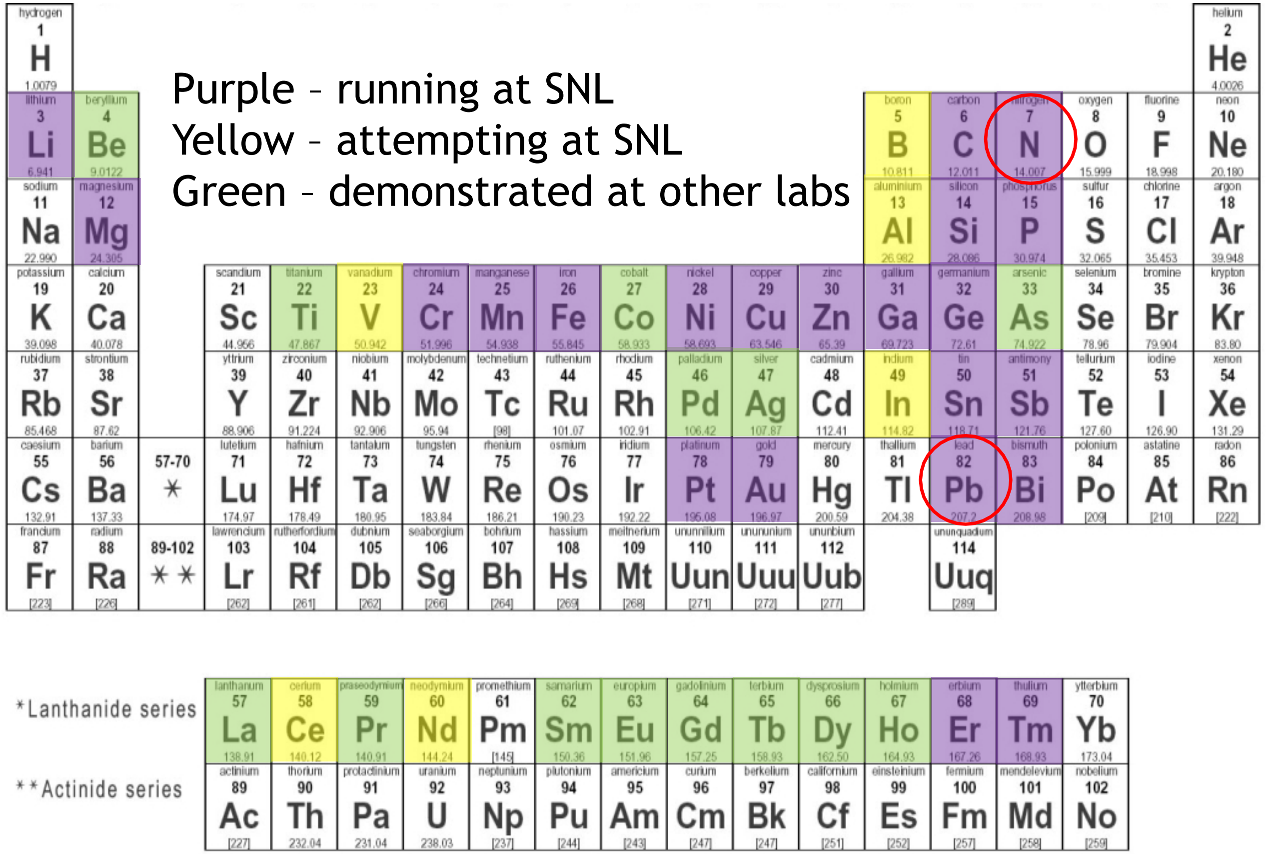
AuGeCr



Aaron Katzenmeyer, Michael Titze, Alex Belianinov

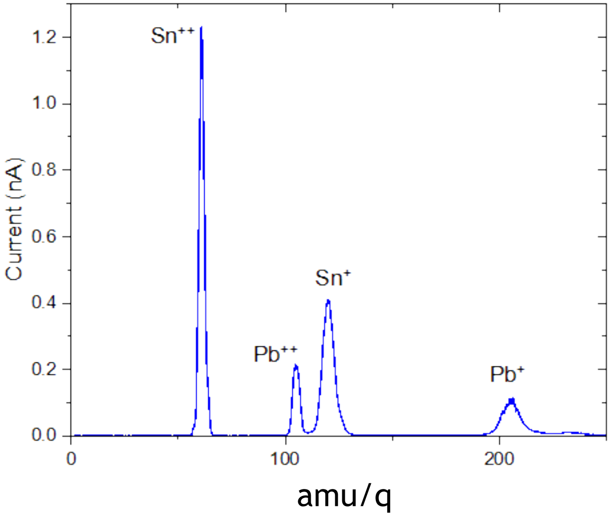
Examples of ***HARD*** New Sources – Pb and N

Purple – running at SNL
Yellow – attempting at SNL
Green – demonstrated at other labs



- Pb

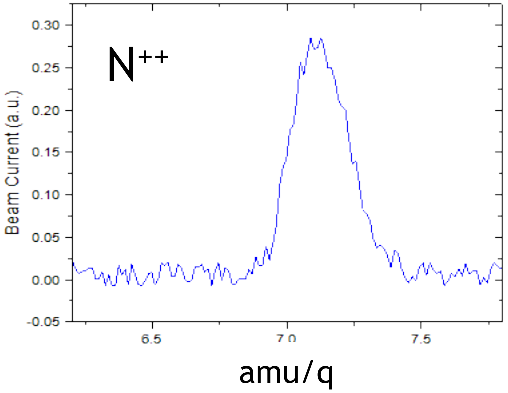
→ PbSn alloys with W tip, use Kovar tips!

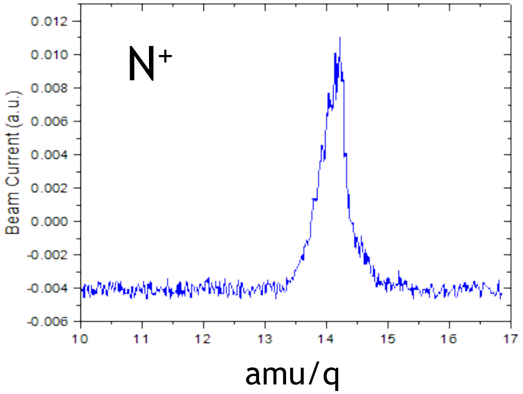


- N

→ Liquid metal alloys with implanted N

i.e., AuSn+N

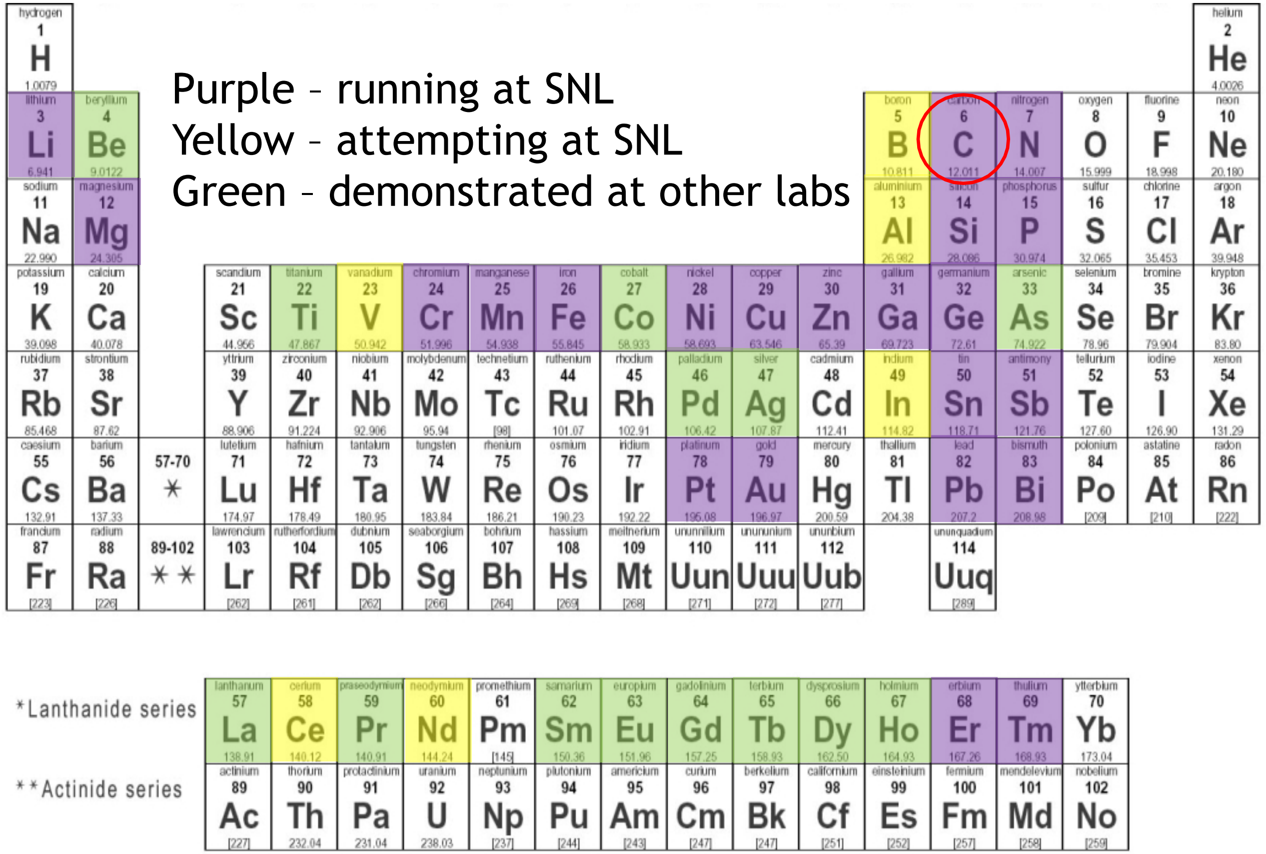




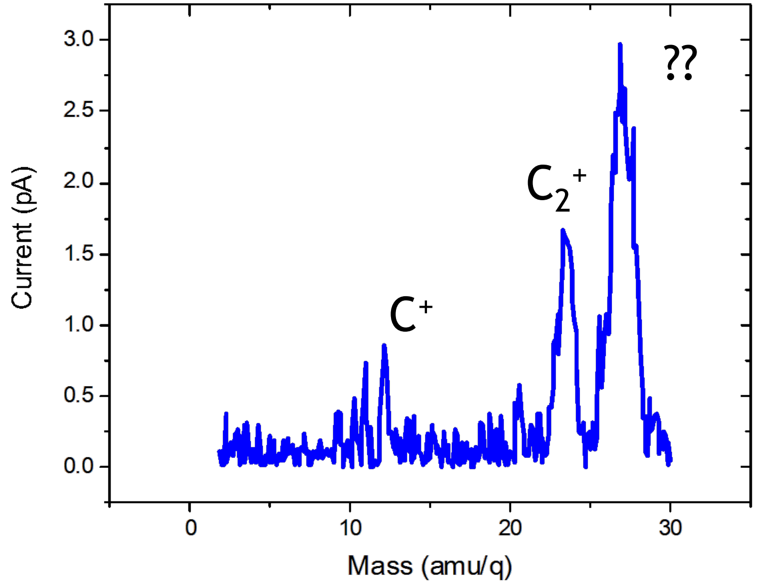
Michael Titze, Alex Belianinov

Examples of ***HARD*** New Sources – C

Purple – running at SNL
Yellow – attempting at SNL
Green – demonstrated at other labs



- C
  → CeC alloy with



(1) Source fabricated at Helmholtz-Zentrum Dresden-Rossendorf

(2) Source tested at Raith

(3) Source used at SNL to implant XXX

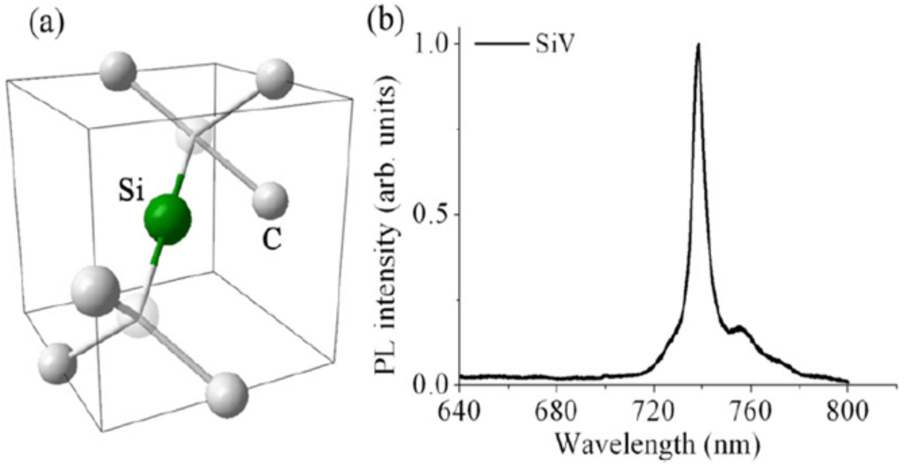
In collaboration with Nico Klinger, Gregor Hlawacek and Paul Mazarov

Creation of Single Defect Centers in Wide Bandgap Substrates

- Defect centers in wide bandgap substrates have applications ranging from metrology, quantum computing, quantum sensing to biological

Ex. Silicon Vacancy Centers in diamond

The ion beam implantation and detection techniques are mainly material agnostic!



I. Aharonovich *et al.*, Rep. Prog. Phys. **74**,076501 (2011)

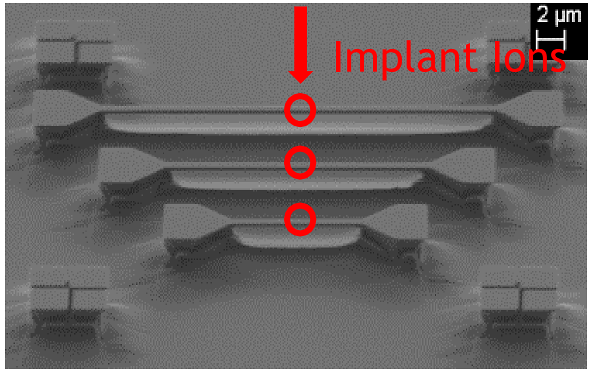
- How to produce a single defect center where you want it?

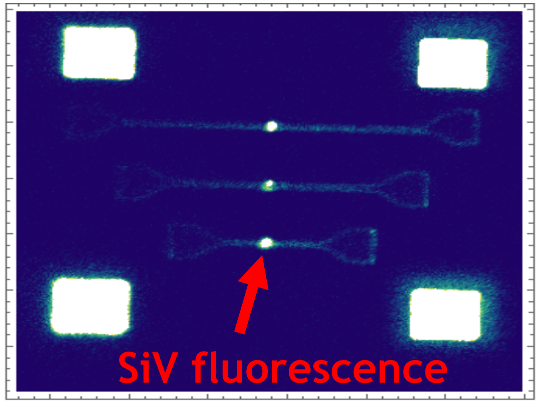
1.) Location – focused ion beam implantation to control location

2.) Yield – counted implantation to control the number of ions and in-situ PL to confirm an optically active defect center

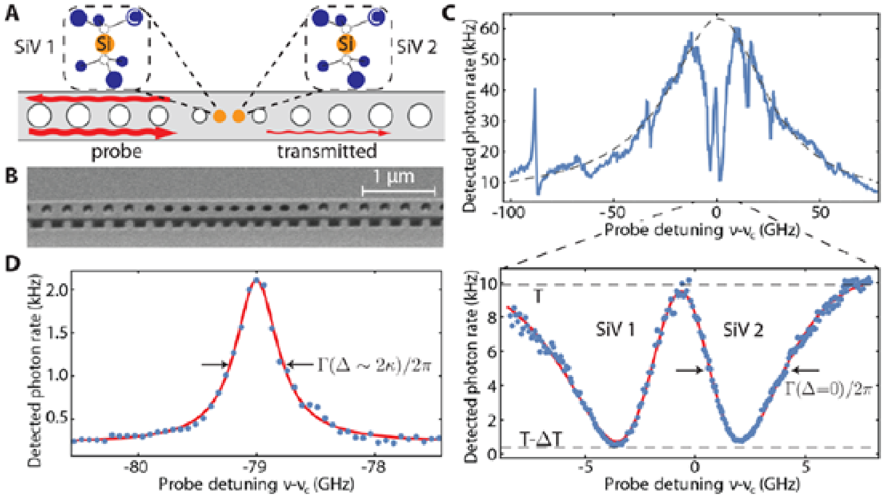
Location solved*, use FIB implantation to control the spatial location

Diamond Nanobeams (with Harvard)



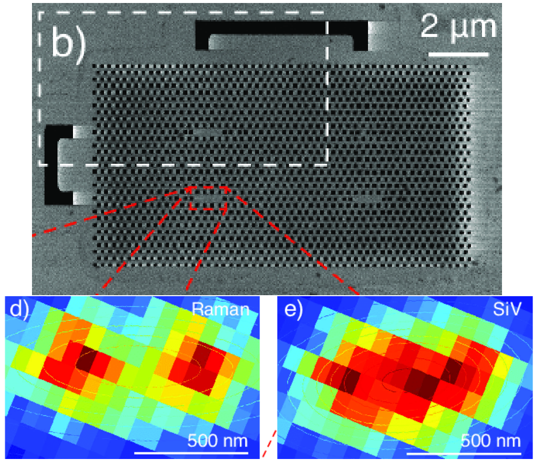


A. Sipahigil, *et al.*, *Science* (2016)

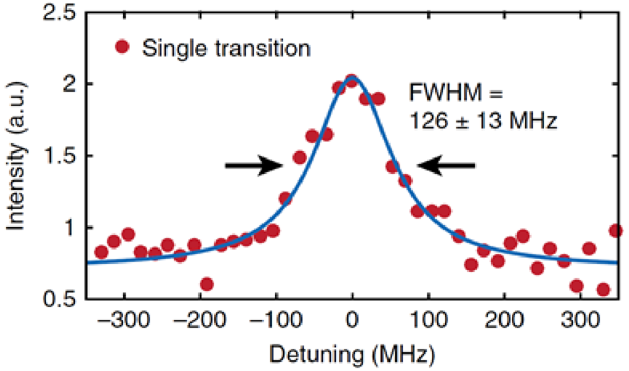


R. E. Evans, *et al.*, *Science* (2018)

2D Photonic Crystals (with MIT)



T. Schroder, *et al.*, Nature Communications (2017)

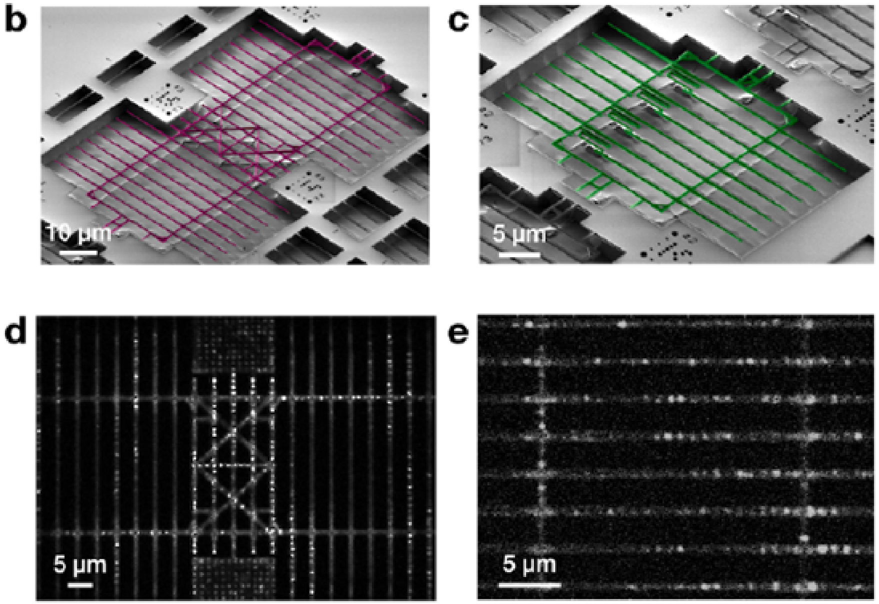


Spatial Accuracy is <50 nm

* Application dependent

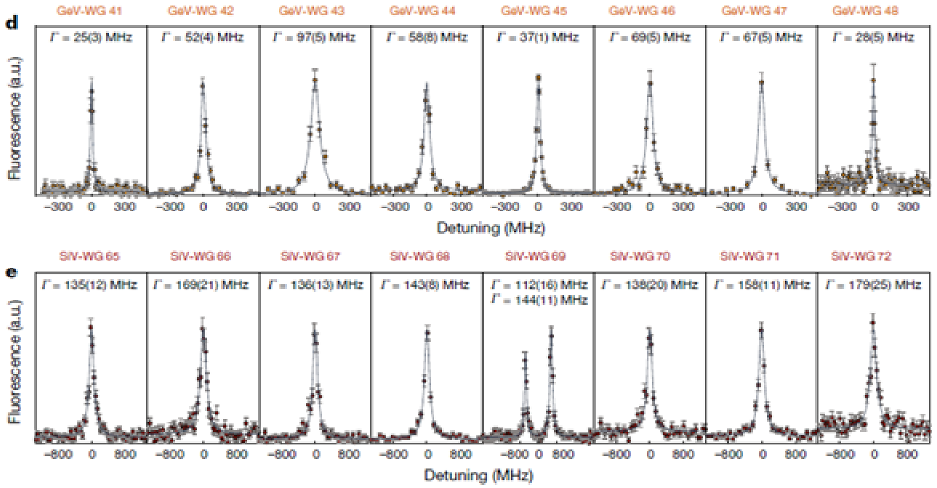
Another set of CINT user projects using FIB Implantation

Diamond waveguides with AlN photonics (with MIT)

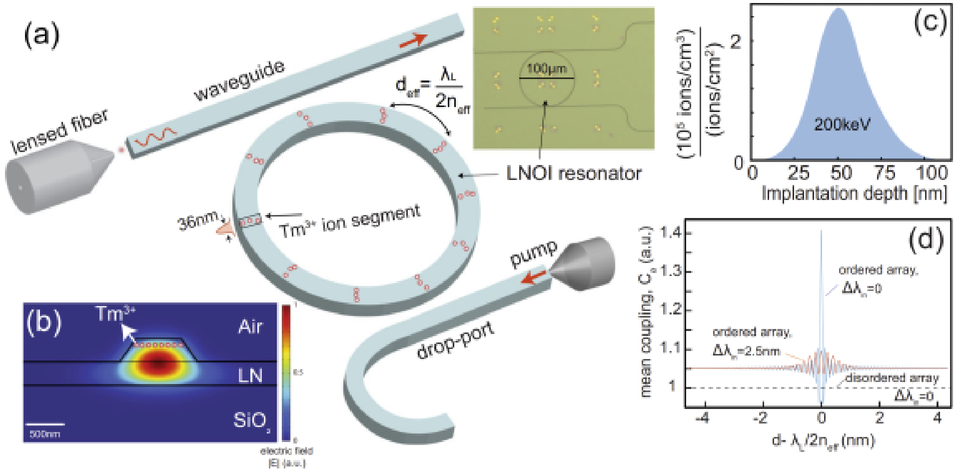


16 and 8 channel "quantum micro-chiplets"

Noel H. Wan *et al.*, Nature, **583**, 226-231(2020)

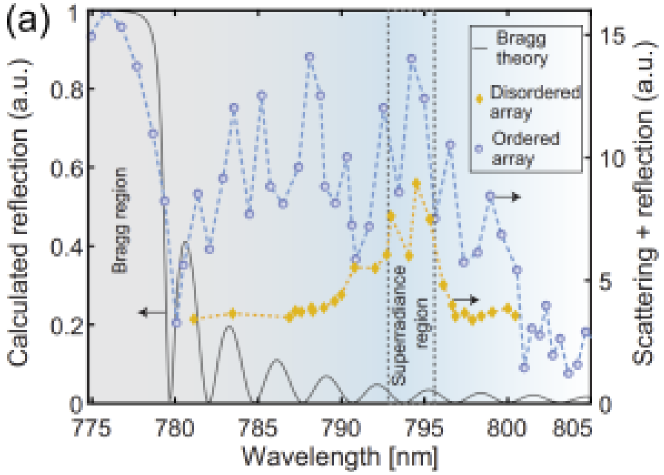


Lithium Niobate Photonics (with Purdue)



D. Pak *et al.*, Comm. Physics, **5**, 89 (2022)

FIB implantation works for a wide range of substrates



We Use Counted Implantation and Photoluminescence to Better Understand the Yield

Yield = # measured SiV / # implanted Si

In-situ Photoluminescence

- Low activation yield limits our ability to make high yield arrays

(Yield numbers are typically 3-10%)

- In-situ photoluminescence can reduce the error in the number of defect centers

In-situ Counted Implantation

- Timed Implantation dominated by Poisson statistics for small numbers

(Uncertainty in number of ion is $\sqrt{N}$)

- In-situ counting can reduce the error in the number of implanted ions

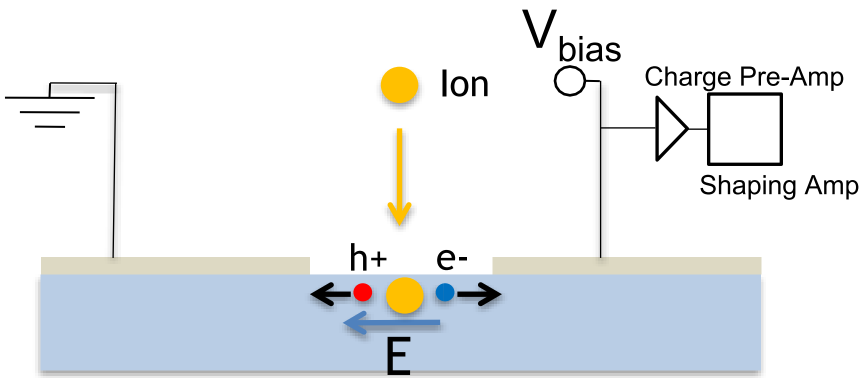
In-situ Counted Ion Implantation to control the number of implanted ion

Yield = # measured SiV / # implanted Si

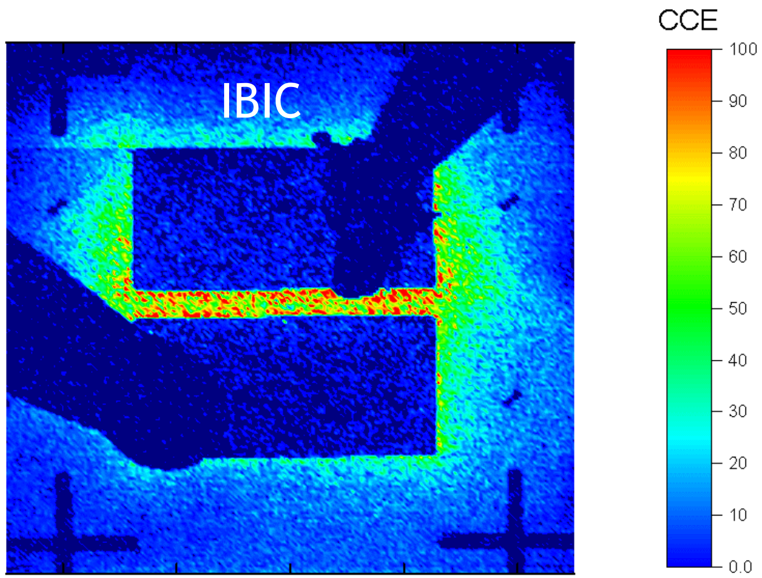
Poisson Statistics

(Uncertainty in number of ion is $\sqrt{N}$)

Ion Beam Induced Charge (IBIC)



Side View



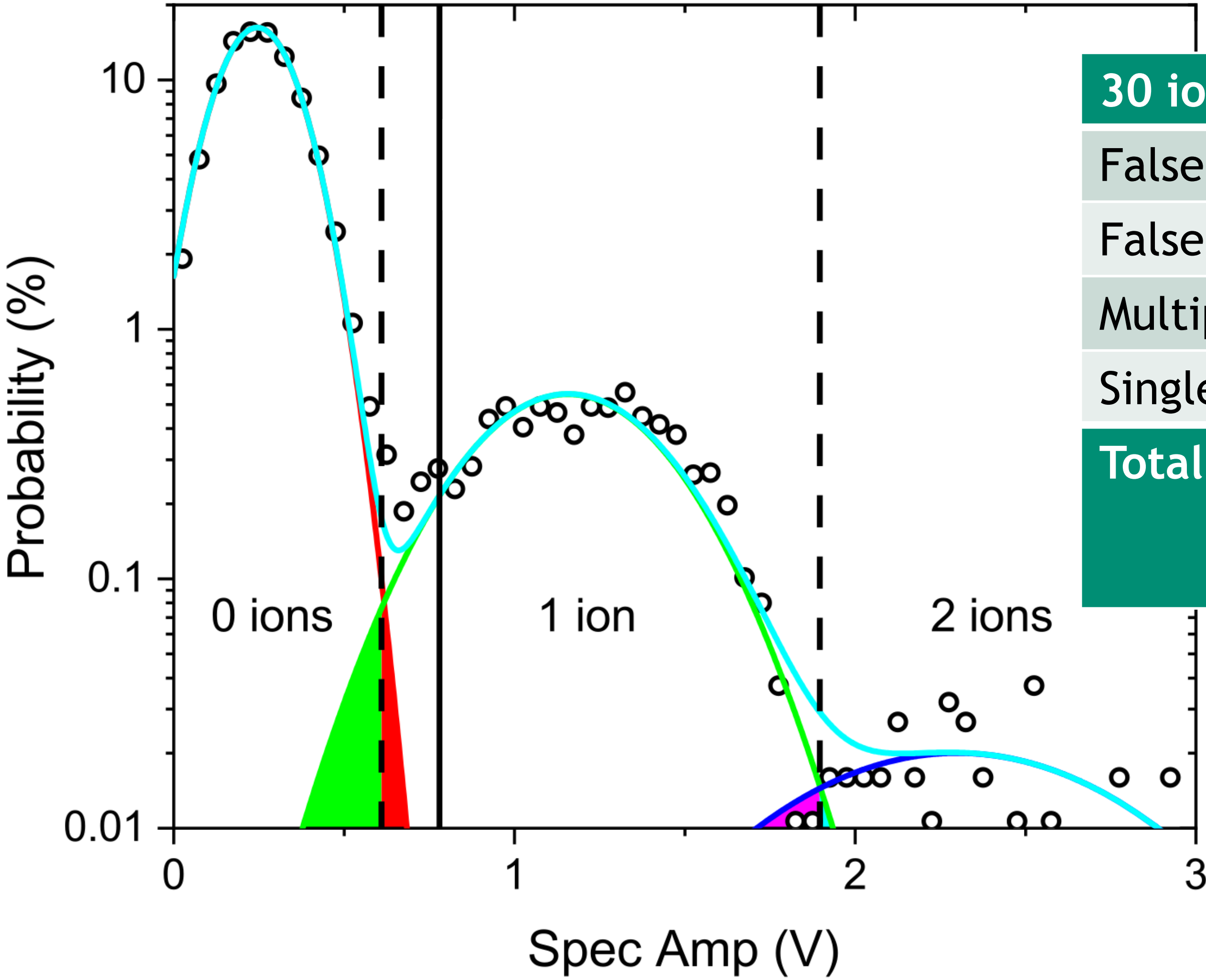
$$CCE = \frac{Charge\ Collected}{Charge\ Depositied} \ x\ 100$$

IBIC/detection demonstrated for low energy heavy ions

Abraham *et al*, APL **109**, 063502 (2016)

Michael Titze

Demonstrated Single Ion Diamond Detection at <0.1> ions/pulse



30 ions /<1> SiV	Timed	In-Situ	Post-Analysis
False Positives	-	< –1 ppb	2.3 %
False Negatives	-	8.6 %	–0.9 %
Multiple Ions	-	5.8 %	1.7 %
Single as Double	-	-	–0.2 %
Total	**+18.3 / –18.3 %**	**+14.4 / –0 %**	**+4.0 / –1.1 %**

- 7x improvement in the error in implanted ions as compared to timed implantation

Titze *et al*, Nano Letters **22**, 3212-3218 (2022)

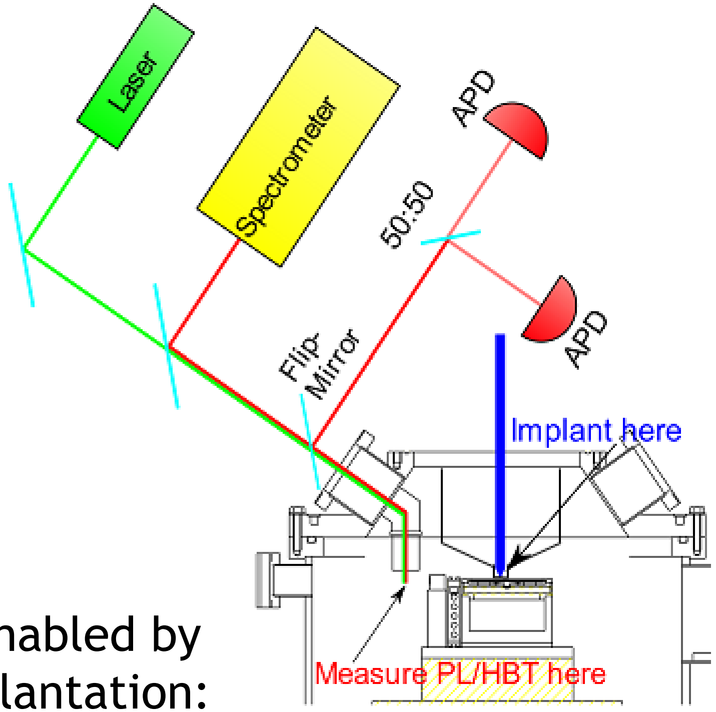
- Aside, <0.1> ions/pulse using a 1 μs pulse length needs ~30 fA of beam current or ~100k ions/s

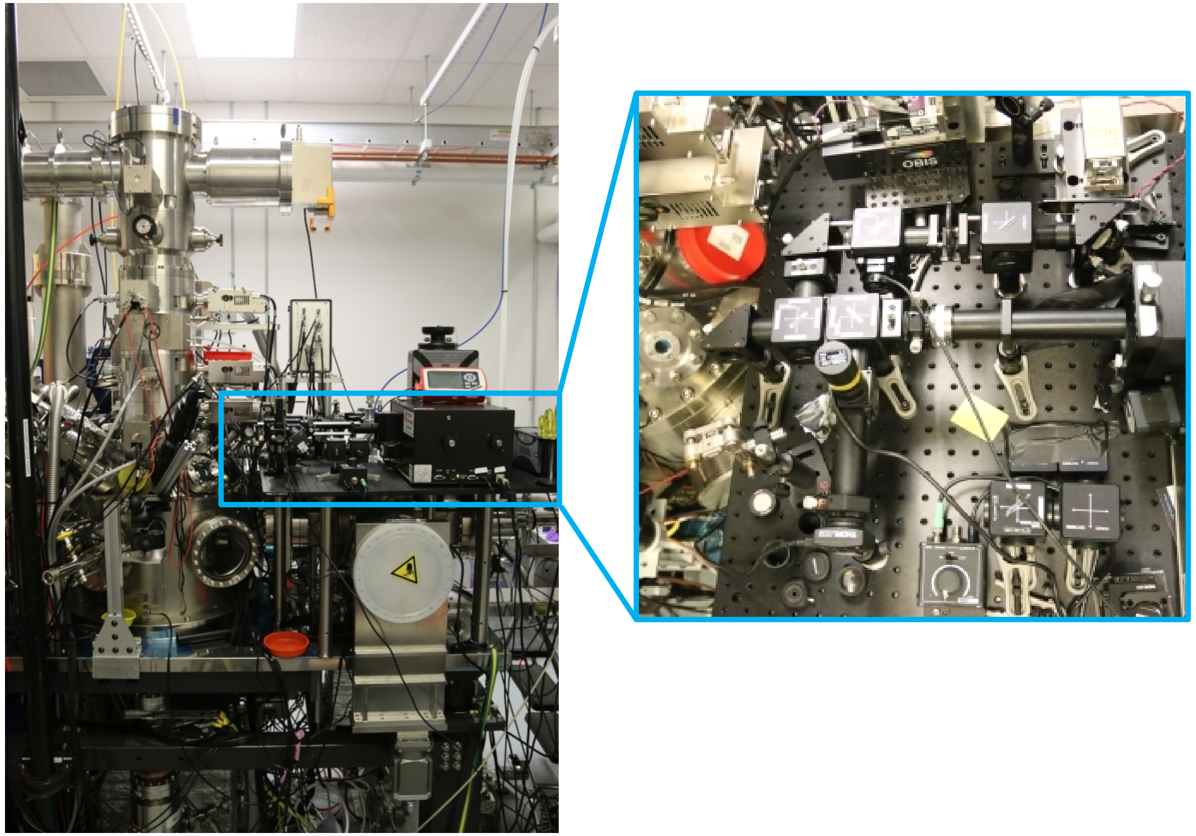
In-situ counting to reduce the error in the number of implanted ions

In-situ Photoluminescence to confirm optical defect center

Yield = # measured Vsi / # implanted Si

We switched from SiV in diamond to Vsi in SiC as can measure as implanted samples without annealing





Two-step process enabled by high resolution implantation:

(1) Aligned implantation, <40 nm

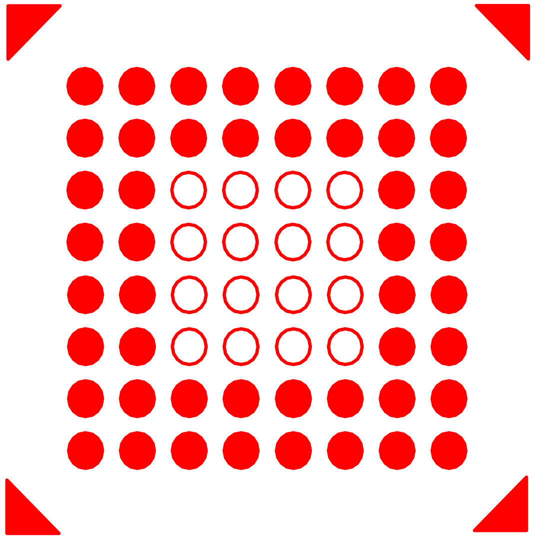
(2) Detect PL

In-situ PL to confirm the optically emission from the defect centers

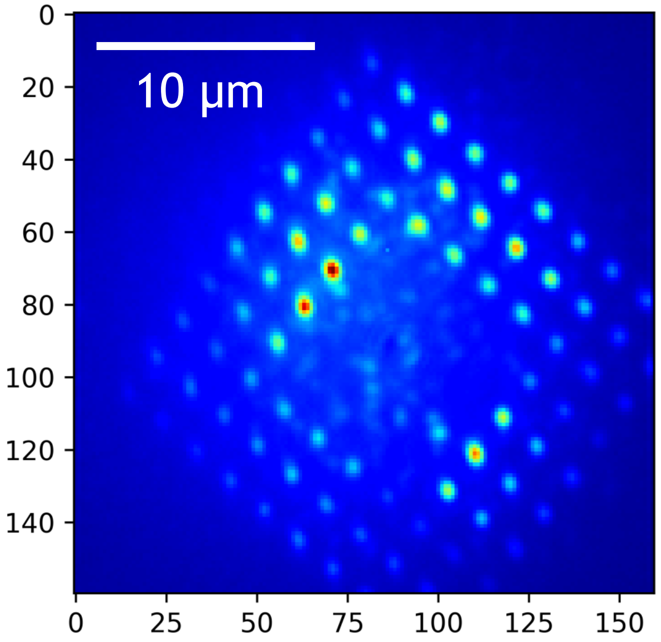
V. Chandrasekaran, Han Htoon, Michael Titze

Demonstration of In-situ Photoluminescence in SiC

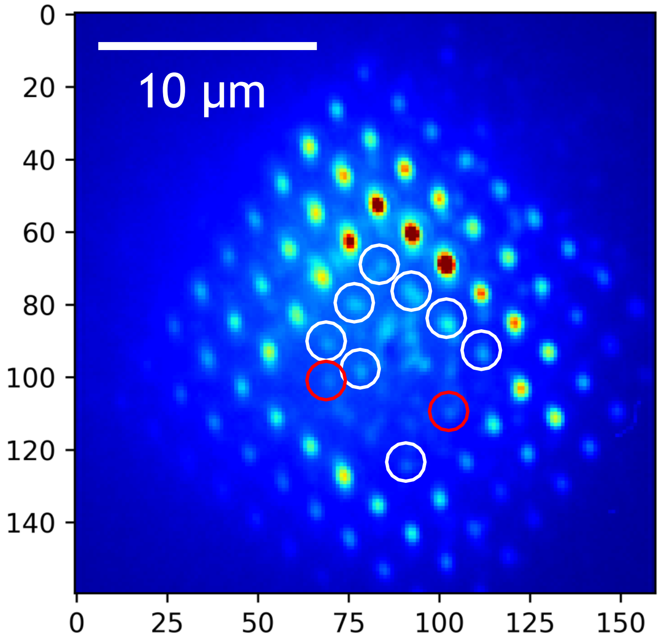
Design of array



Implant Alignment Grid



Implant/PL Repeat to fill Array



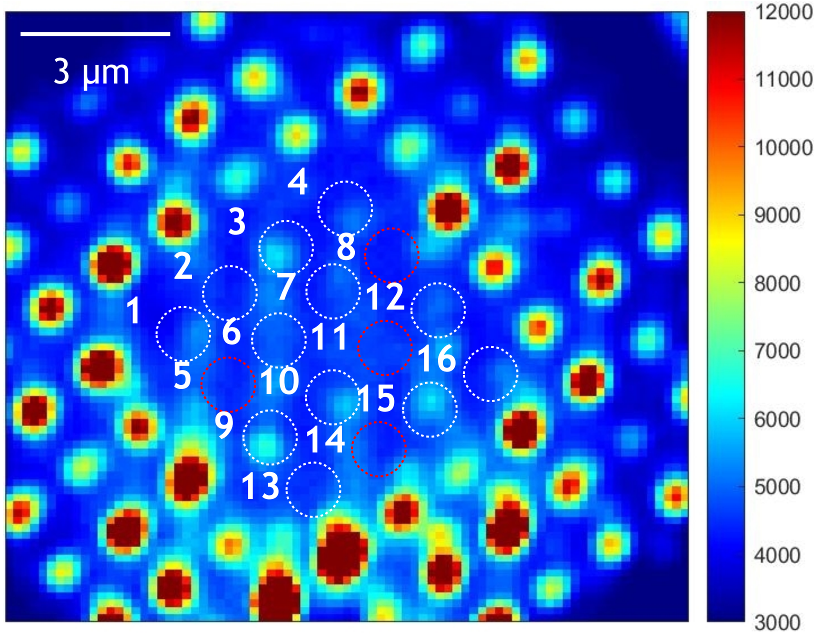
- Alignment markers
- <500> Ions/spot implanted once that acts as landmark to find in-situ arrays during PL check
- In-situ array points to be filled

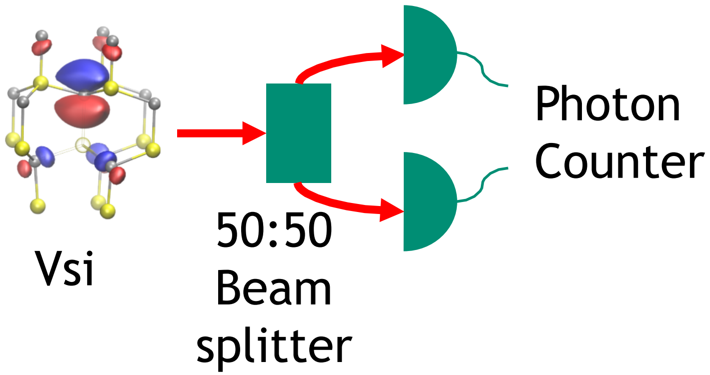
Chandrasekaran *et al*, Adv. Sci., 2300190 (2023)

Preliminary results suggest the in-situ PL works, BUT is limited by high background counts

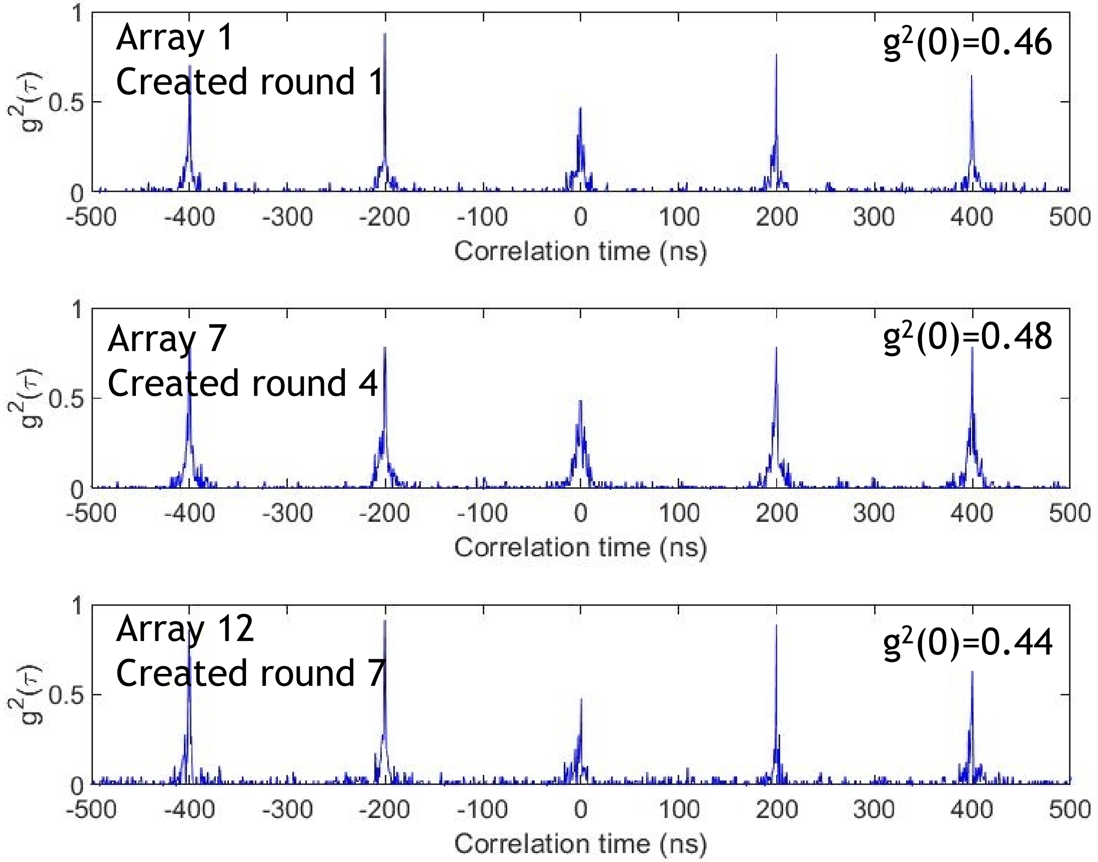
V. Chandrasekaran, Han Htoon

Ex-situ $g^2(\tau)$ check on <3> ions/spot arrays shows single defects





3 representative examples (okay, good examples ☺!)



**No background correction*

Chandrasekaran *et al*, Adv. Sci., 2300190 (2023)

9/16 (56%) of filled array points showed $g^2(0)<0.5$ without background correction → single defects

Acknowledgements

Sandia has developed strong internal ion beam implantation and optical groups

- **M. Titze**, **W. Hardy**, **J. L. Pacheco**, **J. B. S. Abraham**, G. Burns, A. Flores, G. Vizkelethy (SNL)
- **M. Zaibari**, **Jacob Henshaw**, **L. Basso**, **H. Byeon**, A. Mounce, P. Kehayias, M. Lilly (SNL)
- **V. Chandrasekaran**, Han Htoon (LANL)
- V. Acosta (UNM)

And we have continued to support a wide range of user groups through CINT

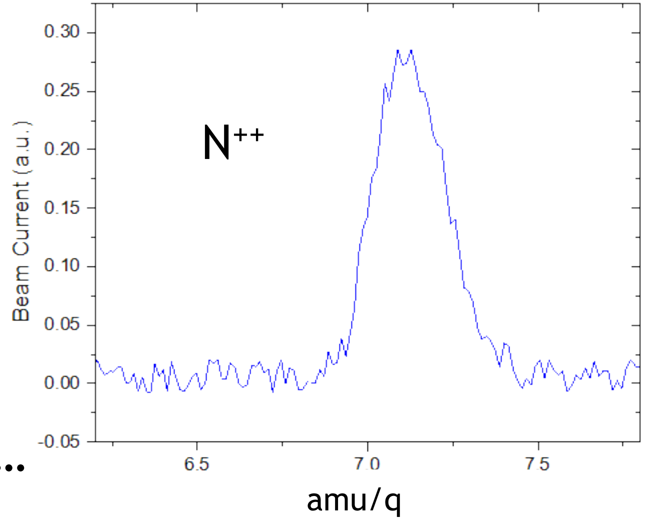
Summary

- We have demonstrated focused ion implantation for fabrication of single atom devices and nanofabrication

→ Viable solution for prototyping – fast and easy!

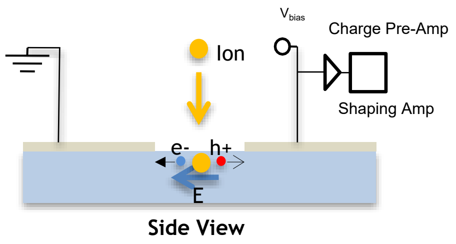
→ On-going work in diamond, SiN, SiC, hBN, GaN, AlN, GaOx, etc…

→ LMAIS for 42/114 elements and development still on-going



Multiple sources run, with ~1.5 days of runtime

- Pathway Towards Deterministic Defect Centers in Wide Bandgap Materials using FIB Implantation



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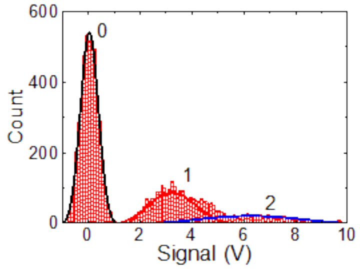
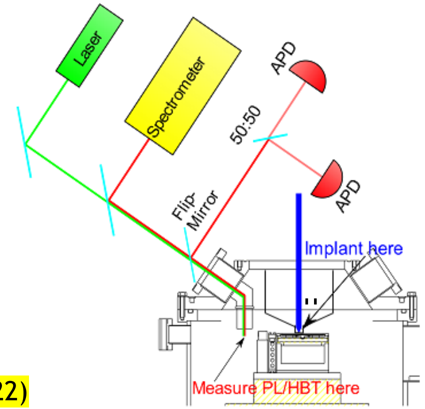


Abraham *et al*, APL **109**, 063502 (2016)

Titze et al, Nano Letters 22, 3212-3218 (2022)

+



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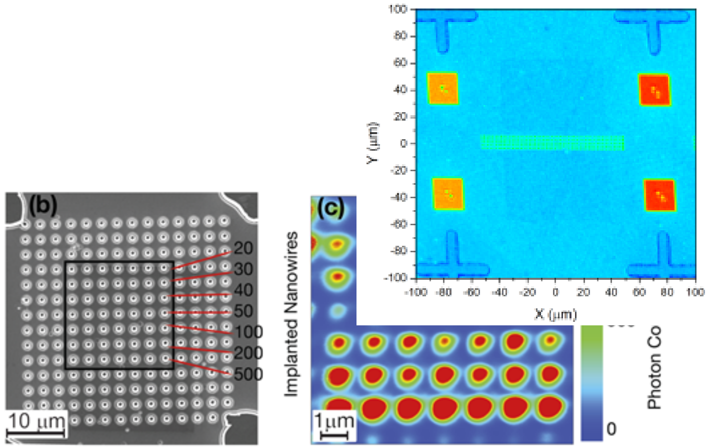


L. Marseglia, *et al.*, *Optics Express* (2018)

Control the number of ions

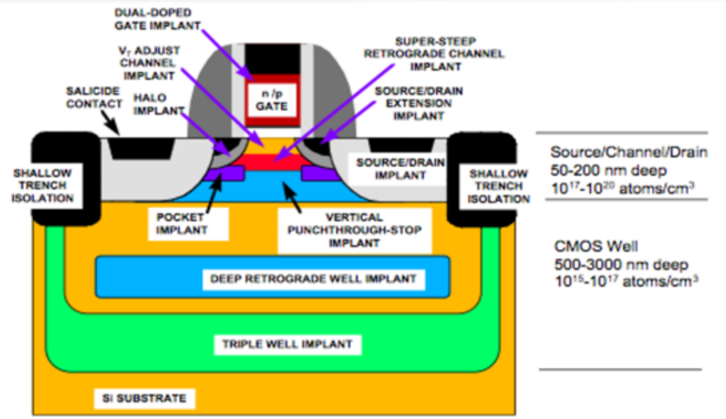
Confirm Optically Active Defect Centers

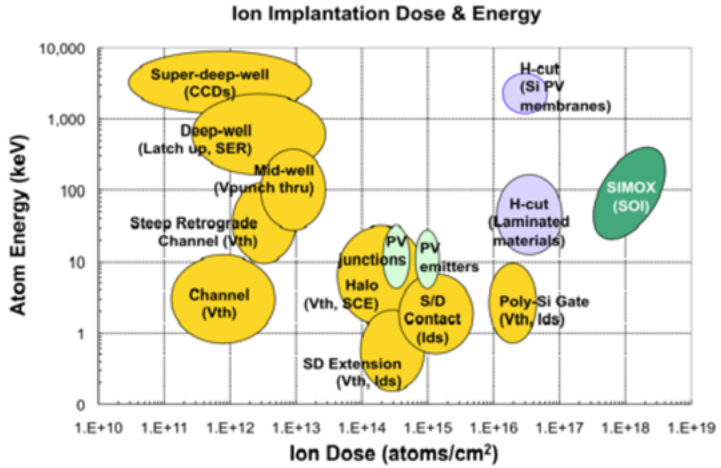
https://cint.lanl.gov/

Extra Slides

Ion Implantation and Irradiation for Device Fab

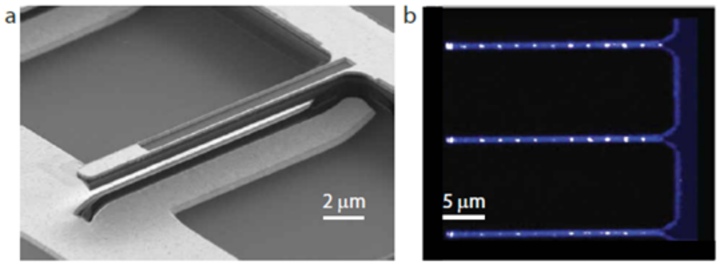
Ion Implantation has been a work-horse for the semiconductor industry



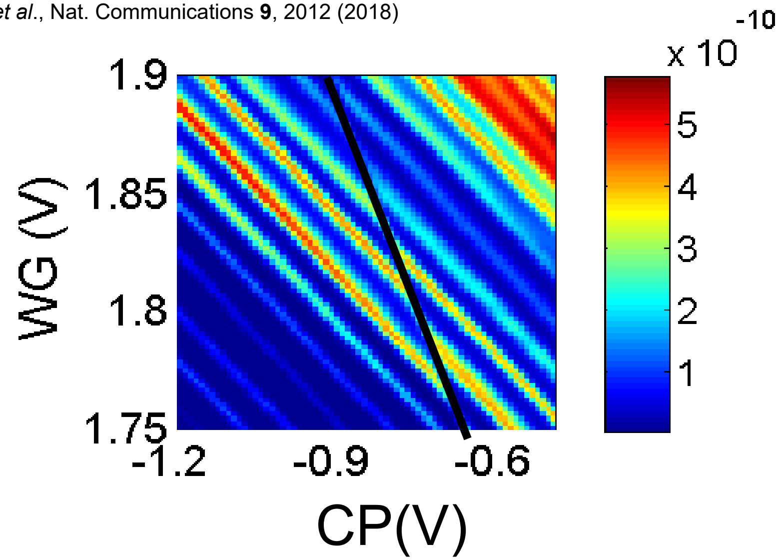


S. B. Felch, *et al.*, Proceedings of PAC2013, Pasadena, CA (2013)

Our work is centered on localized implantation and fabrication based on a deterministic number of implanted ions



Y. Sohn, *et al.*, Nat. Communications **9**, 2012 (2018)

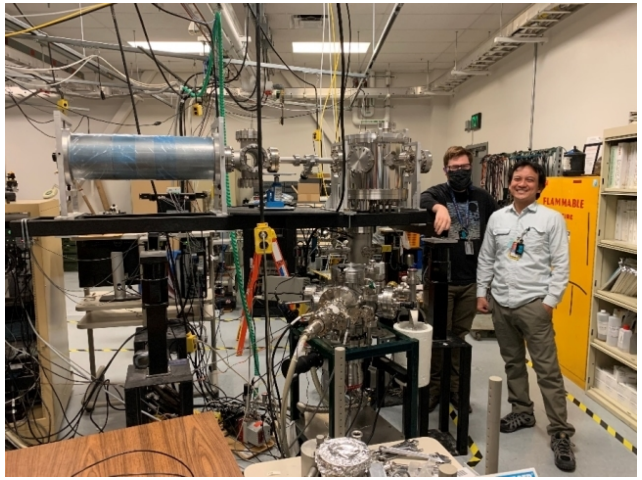


Singh et al. Appl. Phys. Lett. ***108****, 062101 (2016)*

Focused Ion Beam implantation for fabrication of single atom devices and nanofabrication

Capability Currently Under Development

Pulsed Power Electron Gun

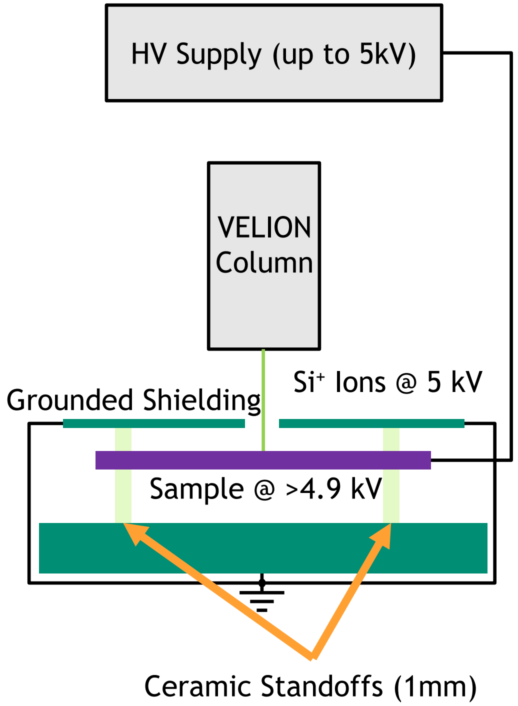


Specifications

- 300 keV
- >>10 A of beam current
- <25 ns pulse lengths

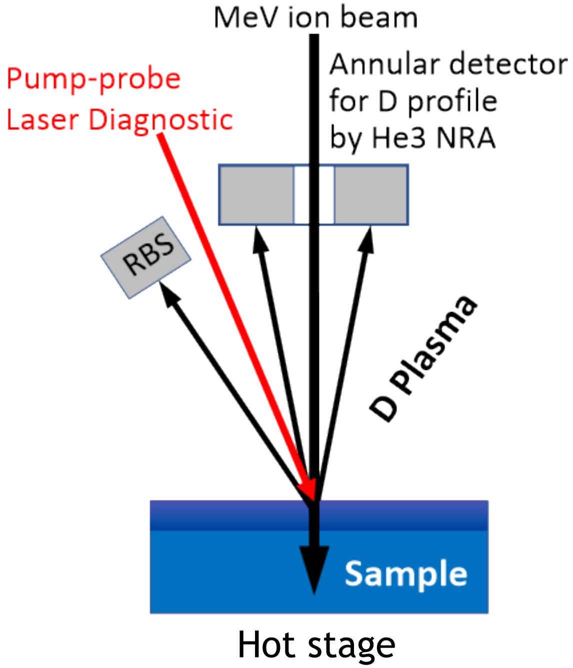
Enables improved yield for wide bandgap defect centers

Low Energy Implantation Stage



Enables lower energy implantation, albeit with a loss of spatial resolution

Combined Ion Irradiation + Ion Beam Analysis + Plasma Exposure



Enables exploration of real-time evolution in sample properties including erosion and redeposition

Aaron Katzenmeyer

Pb Source based on SnPb alloy

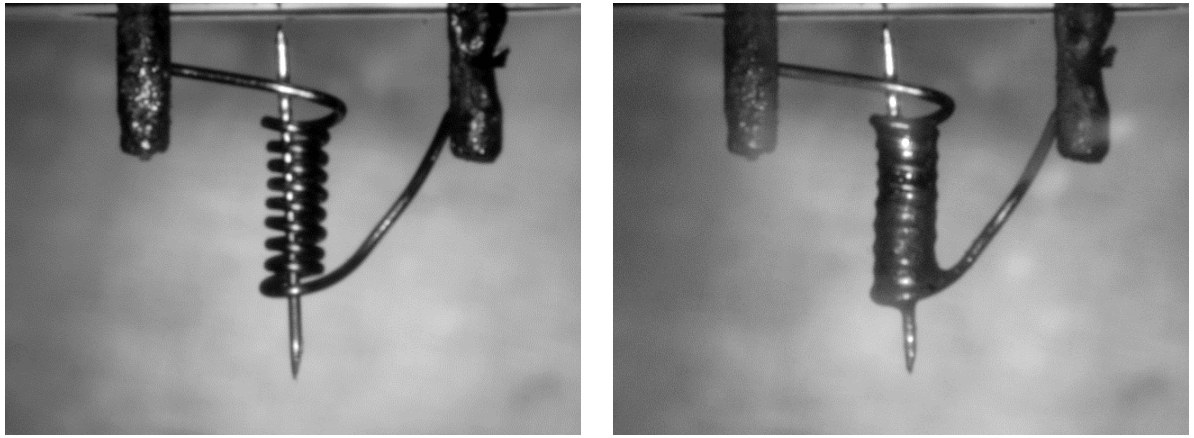
- Solder (SnPb) does not wet W, but Solder does wet a soldering ***iron***

- We attempted multiple homemade sources made from Fe, Monel, Kanthal, Kovar. Of these ***Kovar (Fe-Ni-Co)*** gave the best performance

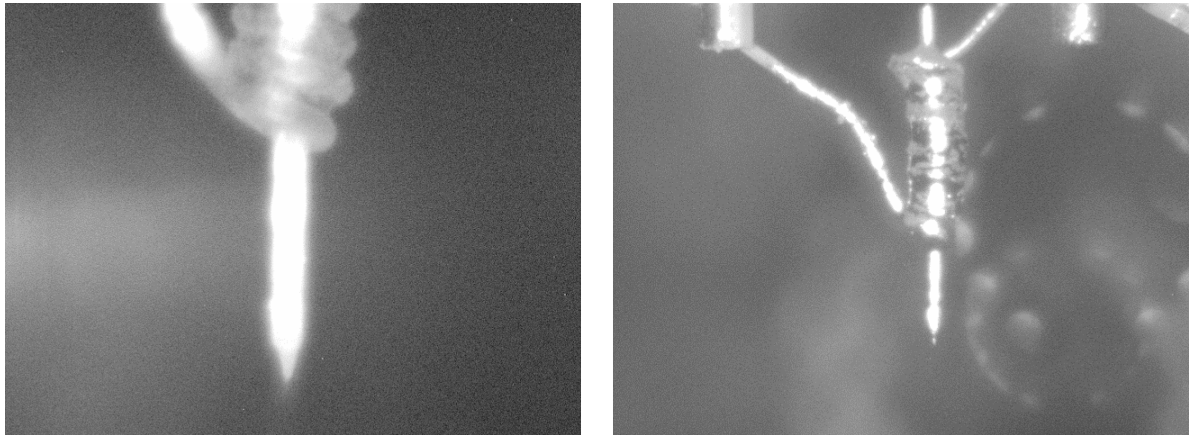
- We tested both mechanically cut and electrochemically etched tips welded to the reservoir to improve alloy flow and heating

As tested in the Raith Velion these tips achieve lifetimes of <2000 μA*hr and spot sizes of <40 nm

Mechanically cut tips



Electrochemically etched tips using Iron Chloride



Andy Mounce

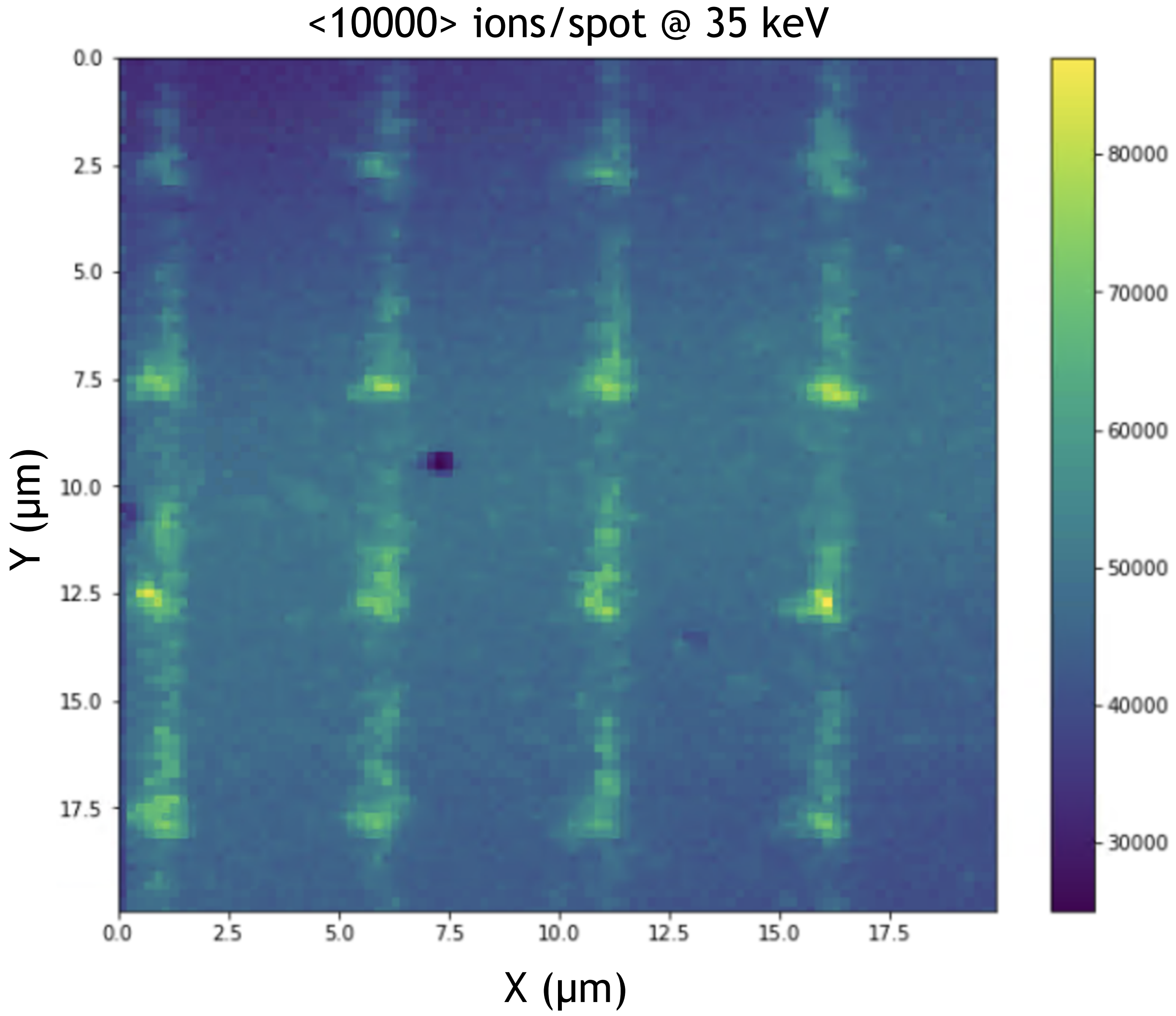
Preliminary Example of PbV in Diamond Substrate

- Observed PbV spectral signature in diamond substrates using FIB implantation
- Vertical stripes in the datasets?

  - Different Pb isotopes?

    ^{208}Pb (≈52%)
    ^{206}Pb (≈24%)
    ^{207}Pb (≈23%)
    ^{204}Pb (≈1%)

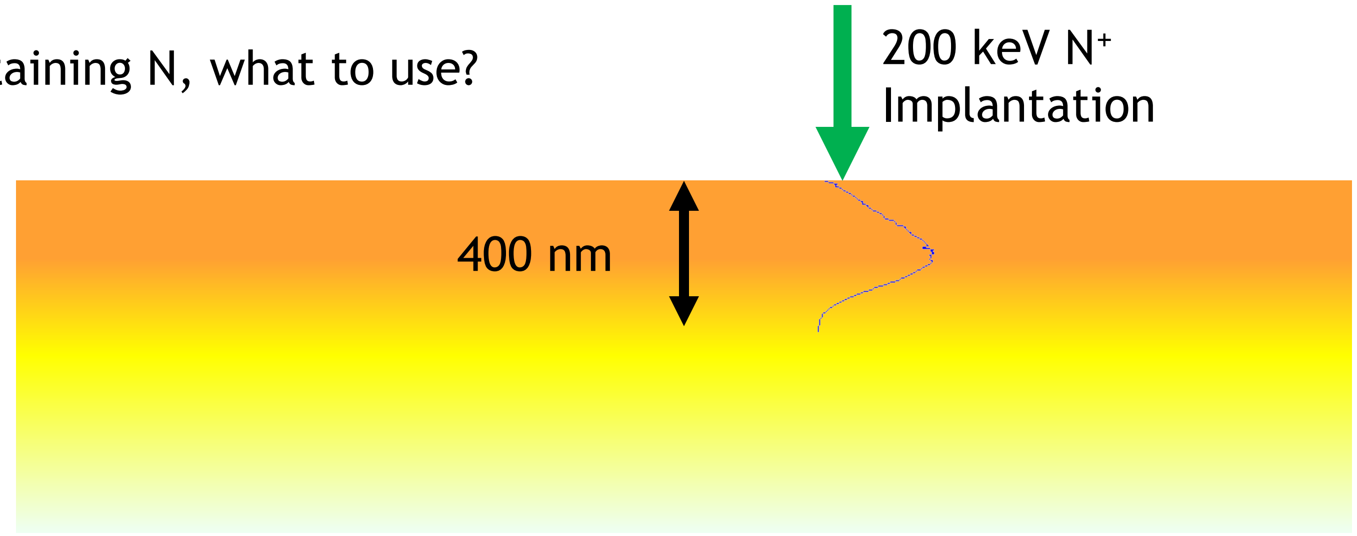
  - In the Raith Velion our m/Δm is not large enough to isolate the individual isotopes



Barney Doyle, Yong Wang

Fabrication of a Nitrogen Liquid Metal Alloy Ion Source

Need a low melting point (200-500C) alloy containing N, what to use?



Implantation

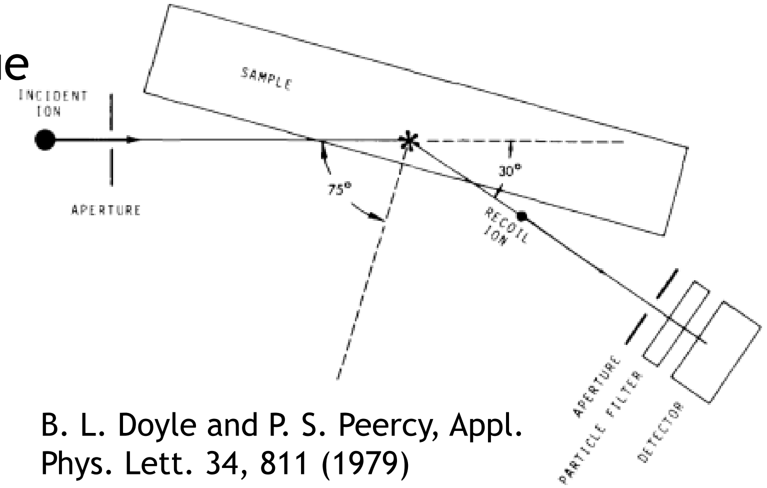
Implanted a 200 µm thick $Au_{80}Sn_{20}$ foil with Nitrogen to $5x10^{17}$ ions/cm2

→ Implantation level chosen based on solid-solubility limit for H

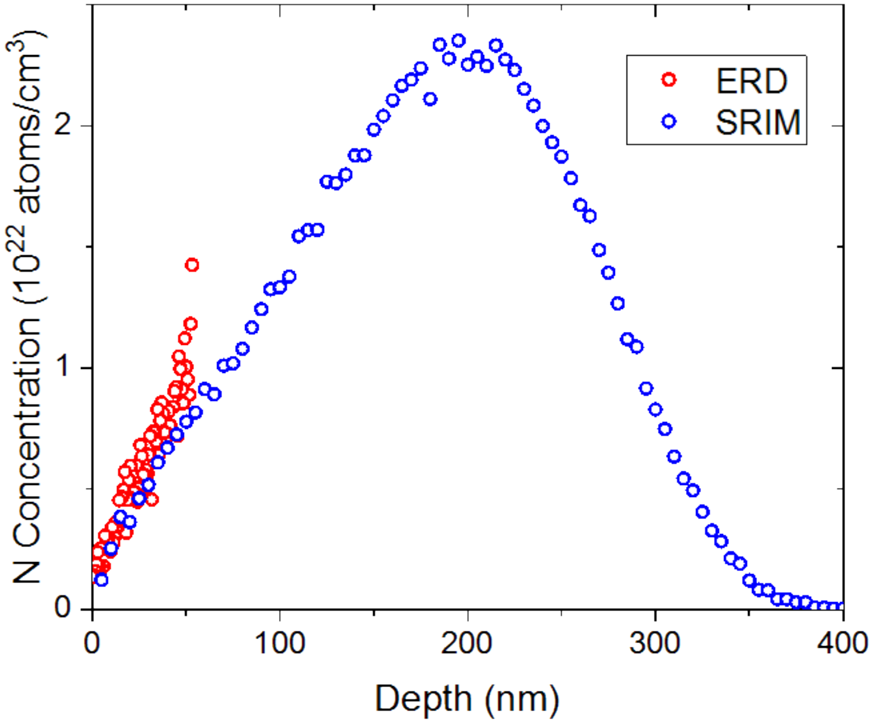
Characterization

Elastic Recoil Detection (ERD) is used to probe the first 100 nm of the implanted foil

→ Limited interrogate depth due to multiple scattering of ions



B. L. Doyle and P. S. Peercy, Appl. Phys. Lett. 34, 811 (1979)



We can produce the material, ***but*** can we make a usable LMAIS?

Aaron Katzenmeyer

Tip fabrication from implanted AuSnN foil

(1) Heat W tip to remove surface contaminants

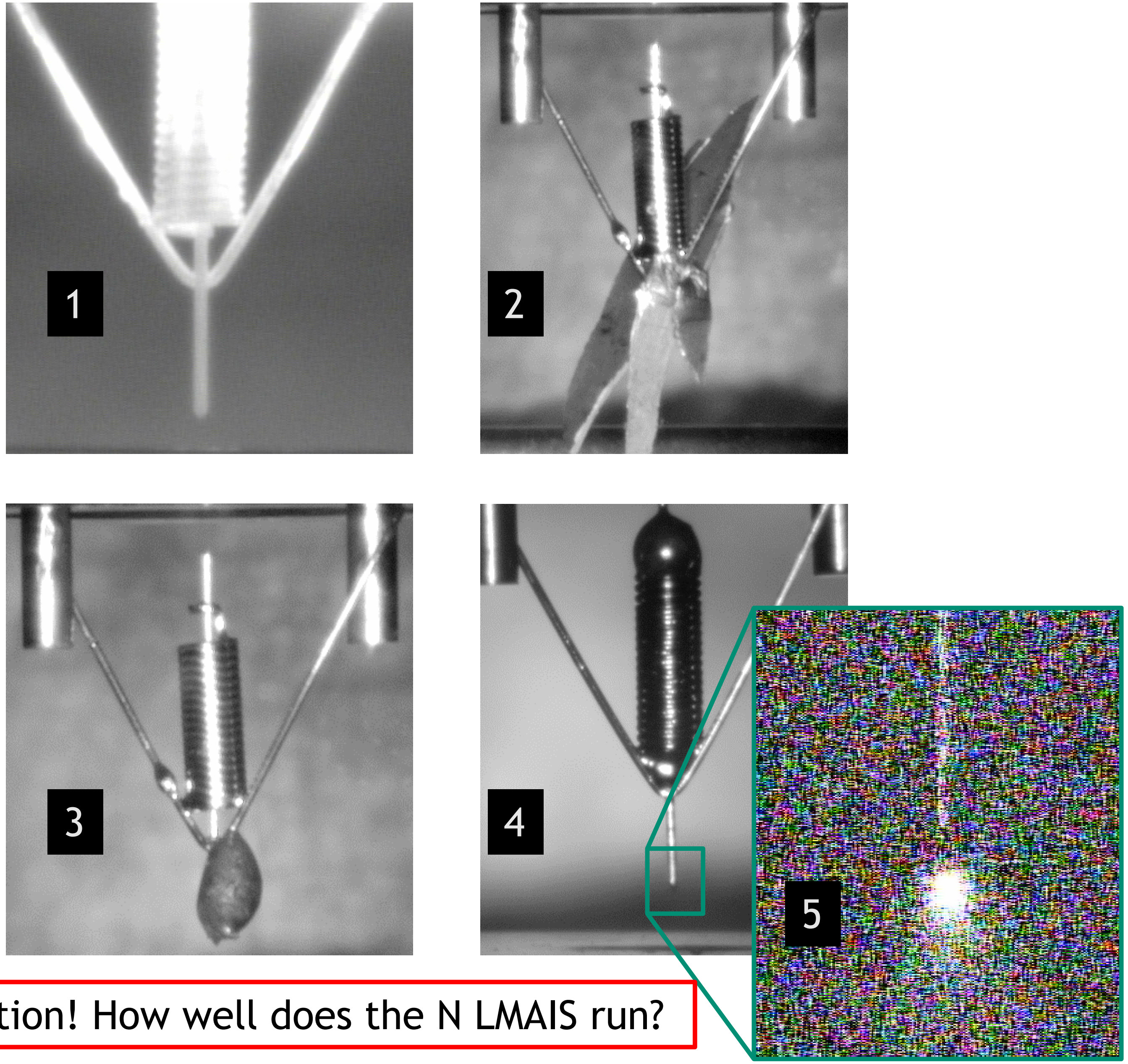
(2) Tack weld on implanted foil to the tip

(3) Heat tip to melt the foil onto the tip

(4) Repeat steps (2) and (3) until tip and the reservoir is filled

(5) Test tip for emission

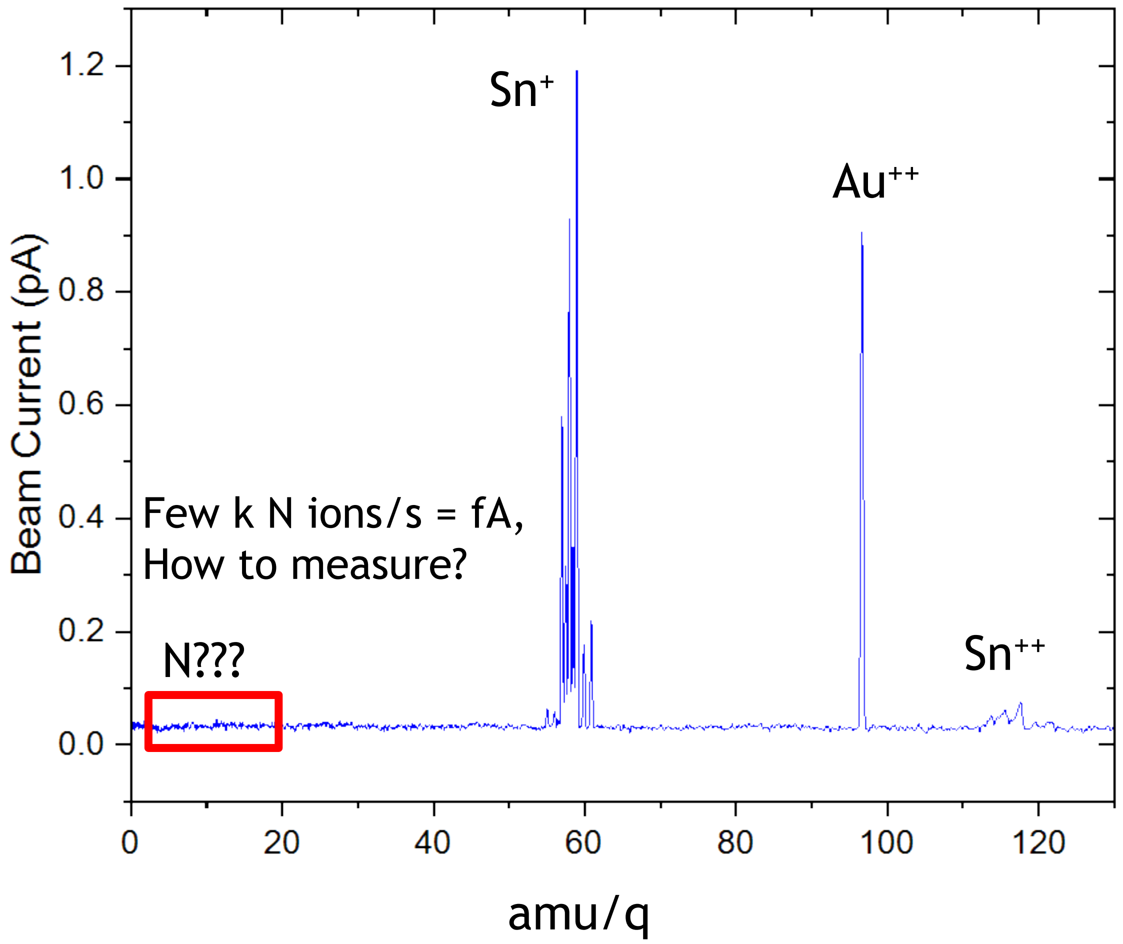
(6) Install tip into nanoImplanter



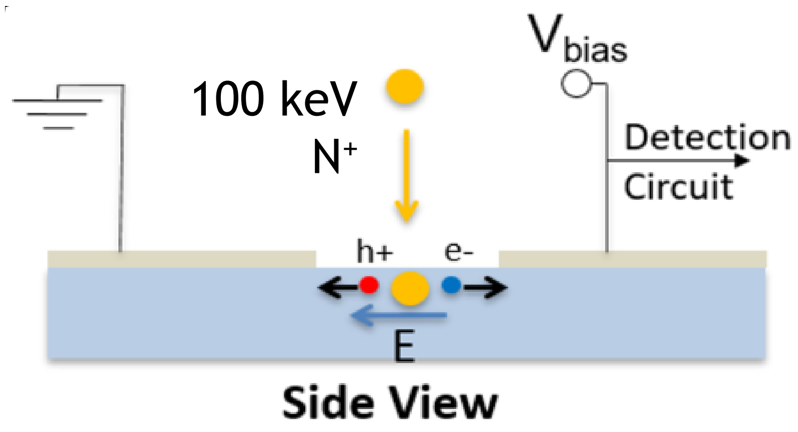
Very challenging source preparation! How well does the N LMAIS run?

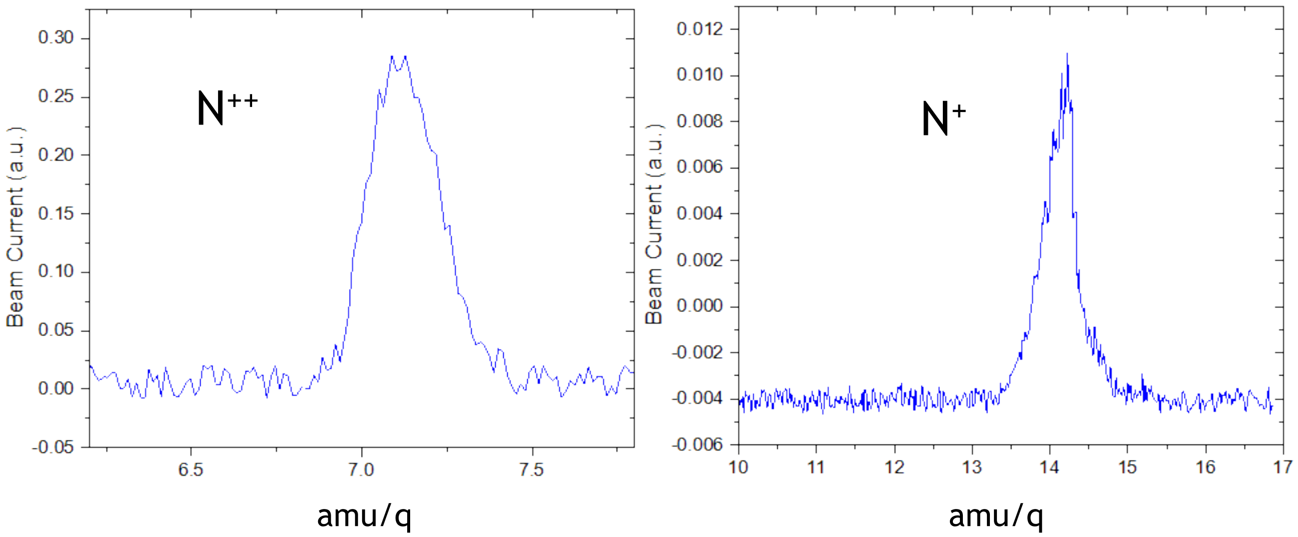
Michael Titze

N Source Characterization – Mass Spectrum via IBIC



Ion Beam Induced Charge (IBIC) to measure N beam

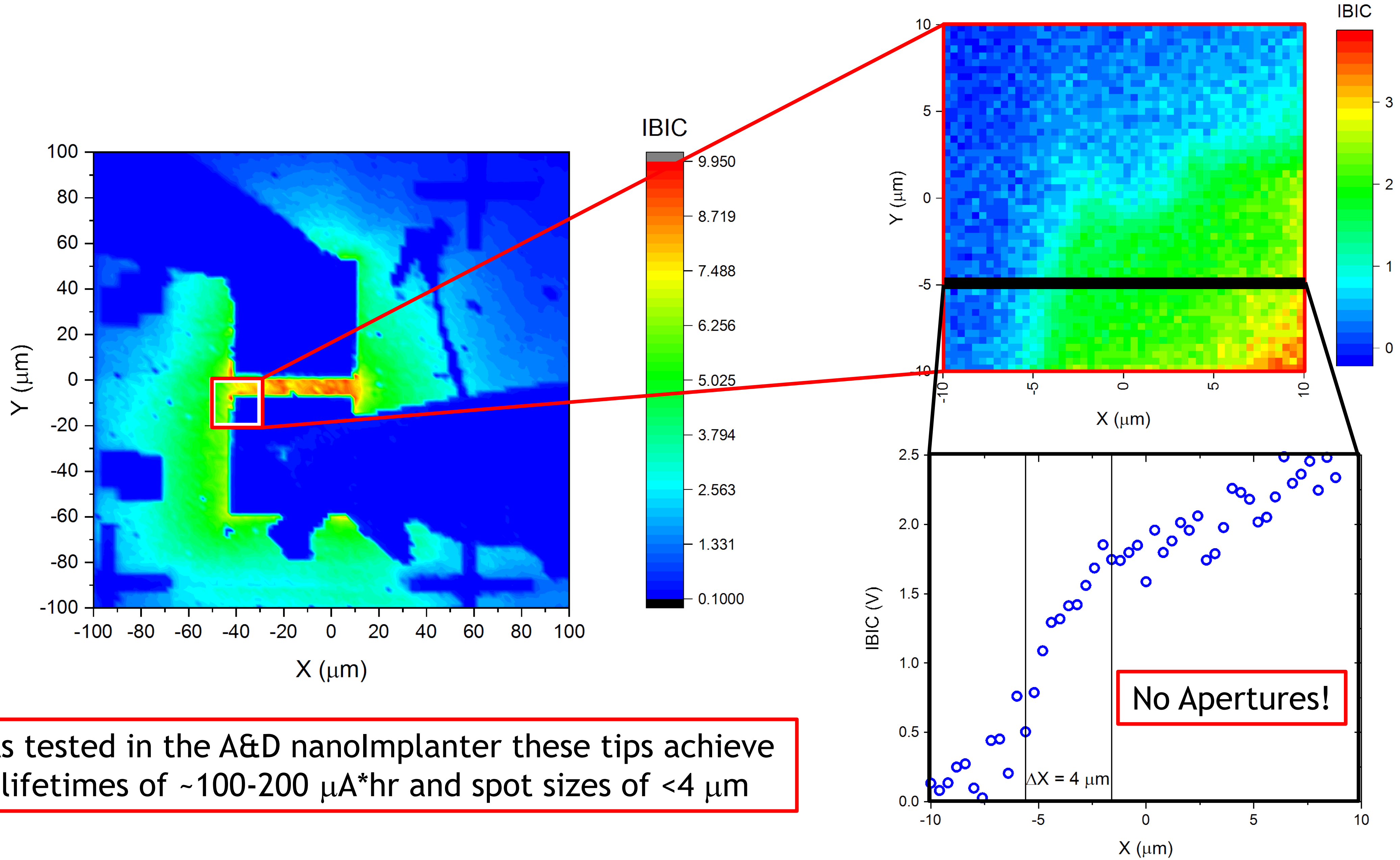




Observe Atomic nitrogen single and doubly charged ions in the source at ~20k ions/s

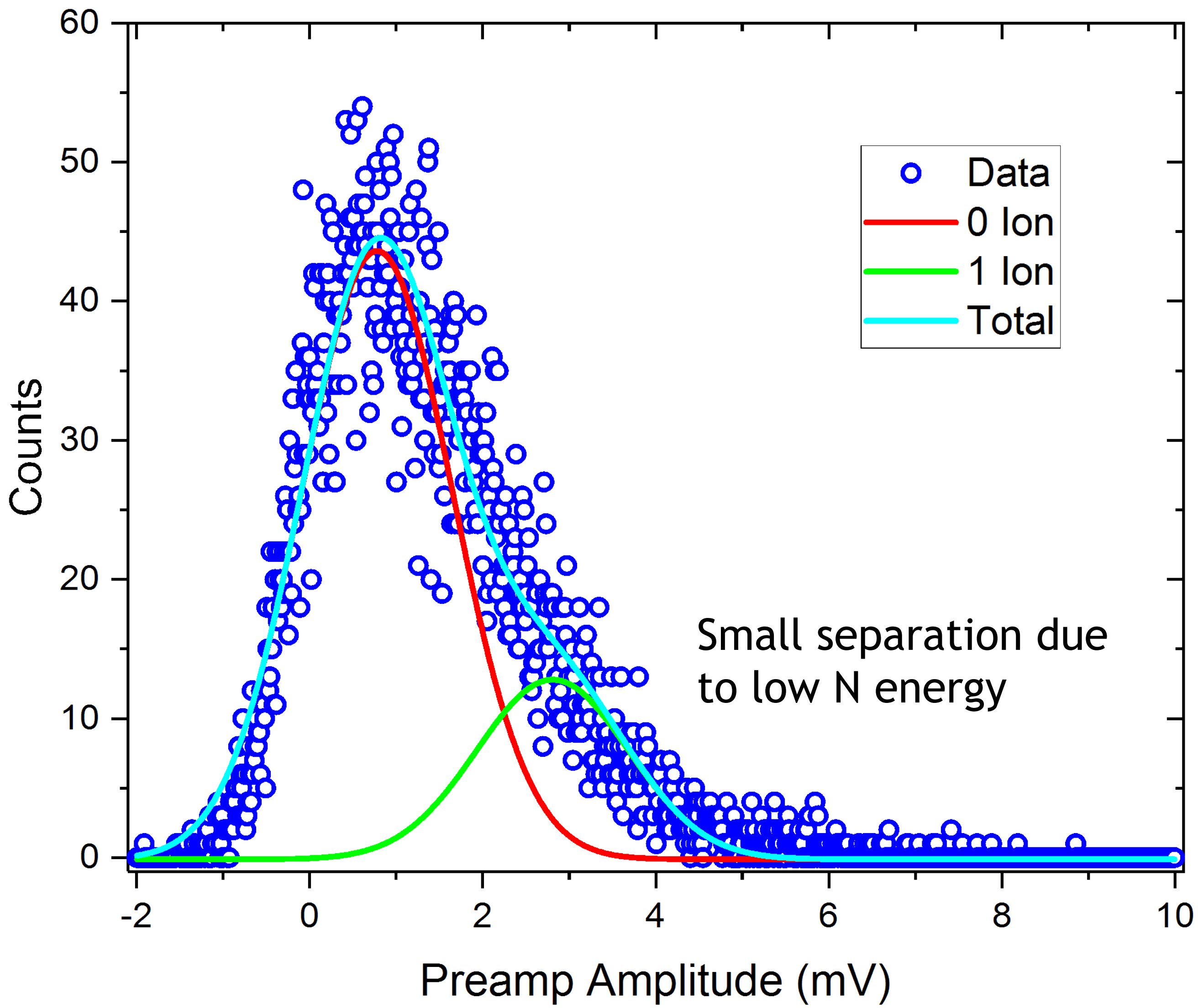
Michael Titze

N Source Characterization – Spot Size via IBIC



As tested in the A&D nanoImplanter these tips achieve lifetimes of ~100-200 μA*hr and spot sizes of <4 μm

Characterization – Beam Current measured via IBIC



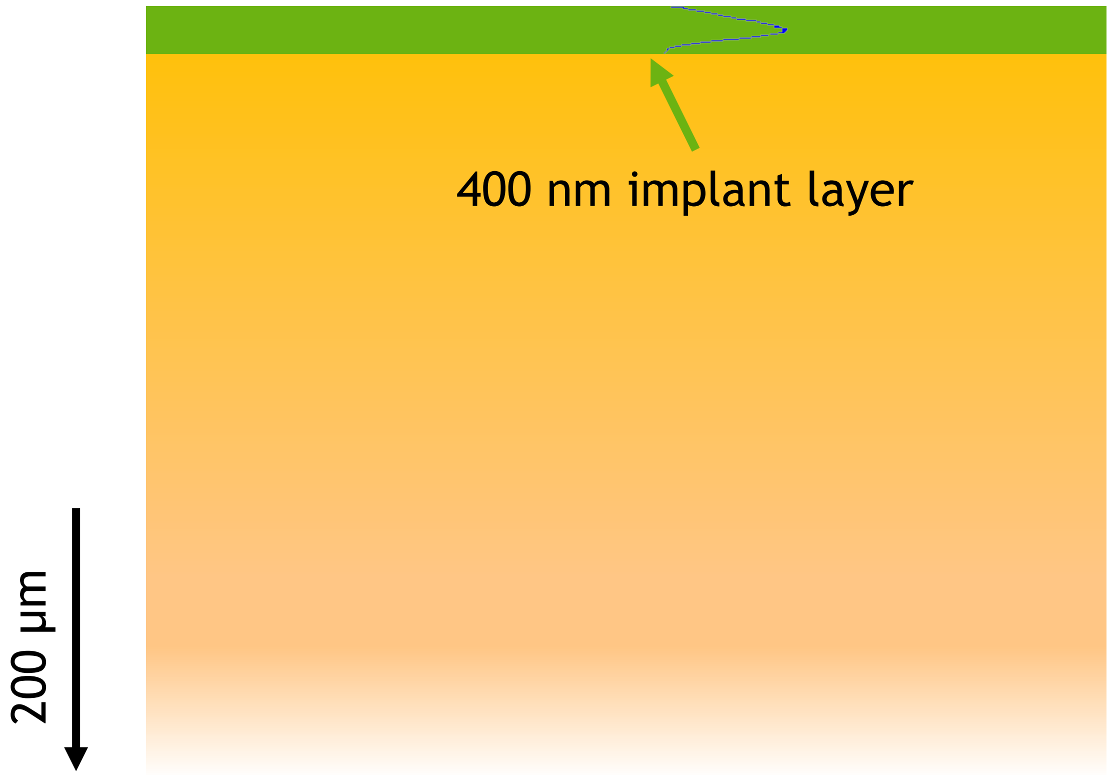
1. Fit peak to Gaussians
2. Area under peak corresponds to likelihood of event

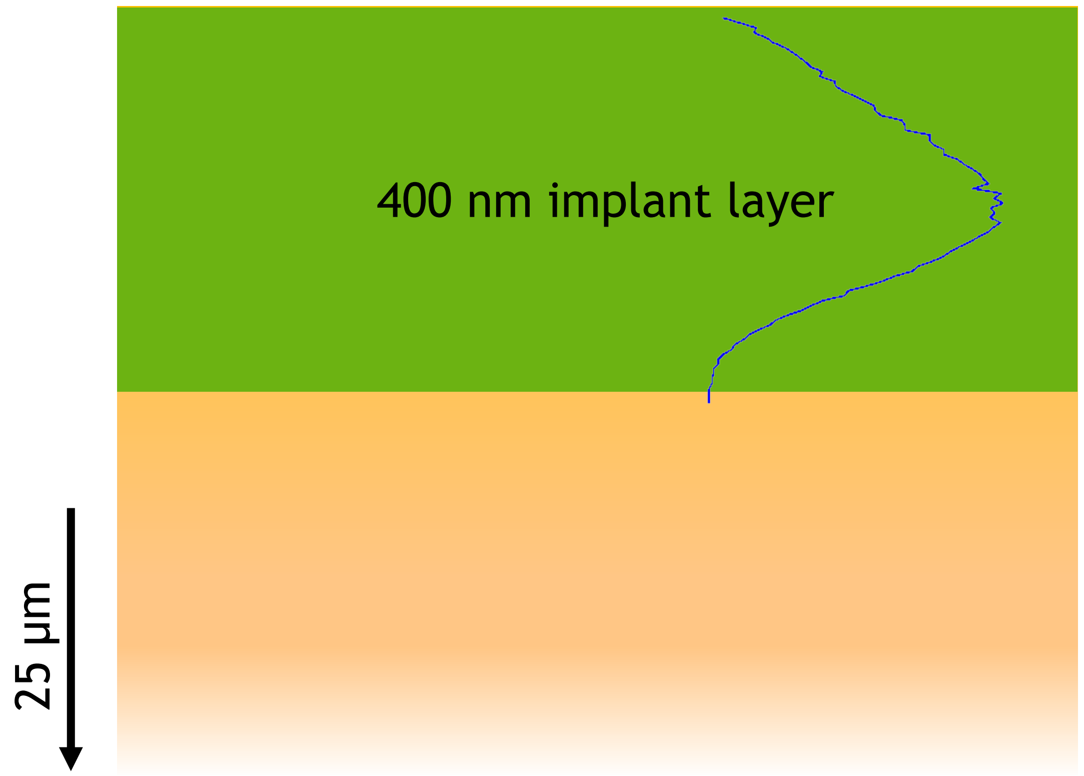
<1> ion every 50 μs ≡ 20 k ions/s

N Source – Improvements

Higher beam current

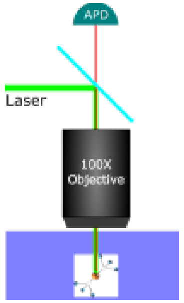
- Thinner foil to same fluence → higher overall concentration
- Fabrication in N atmosphere → additional N absorption

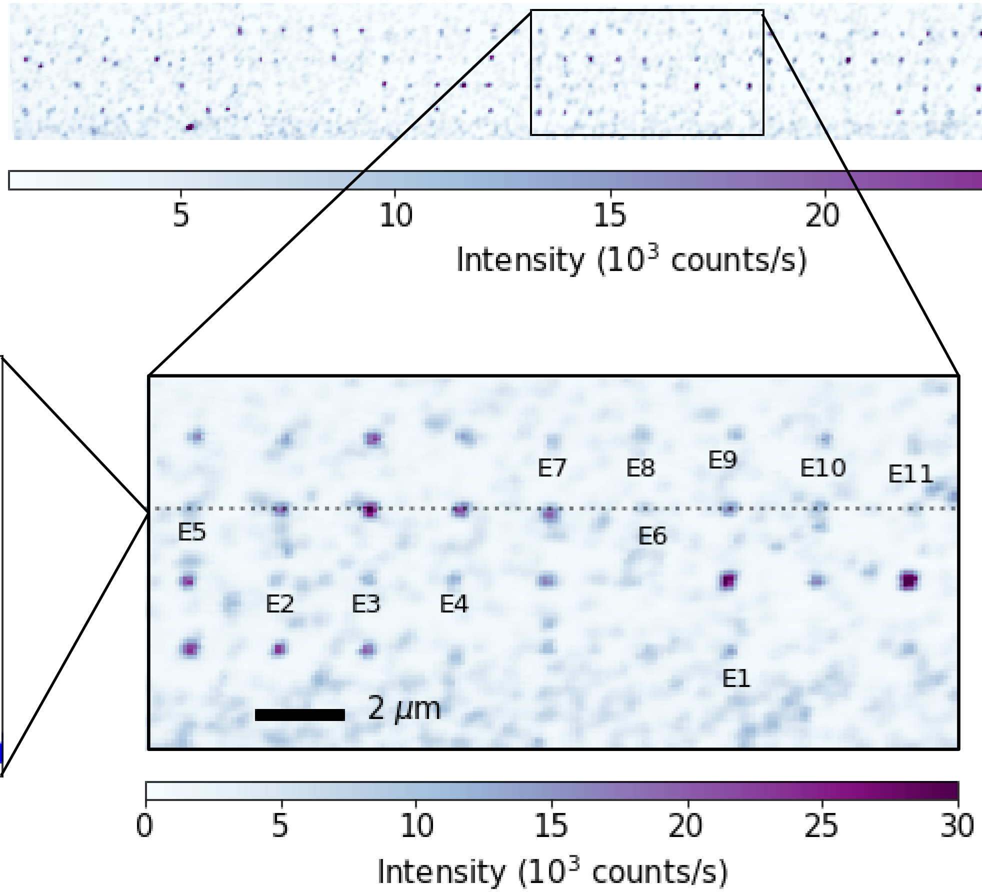


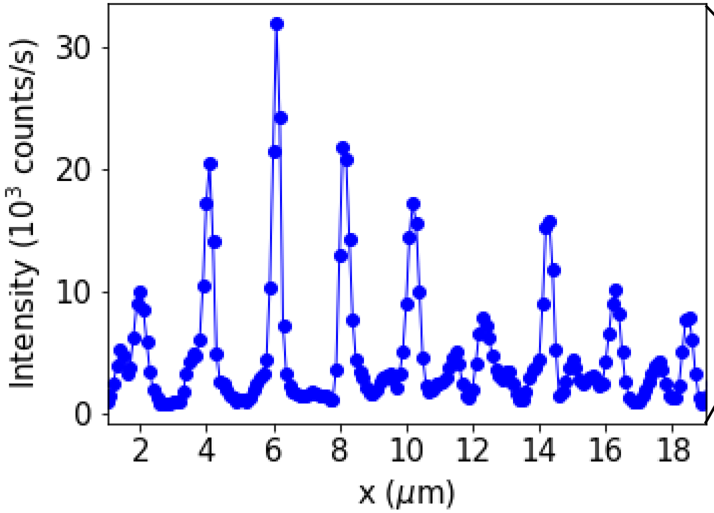


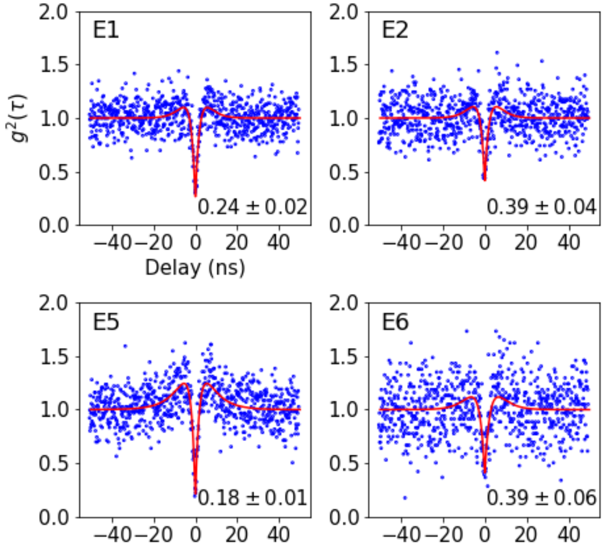
Photoluminescence of the counted sample shows single defects

PL map of the counted ion sample





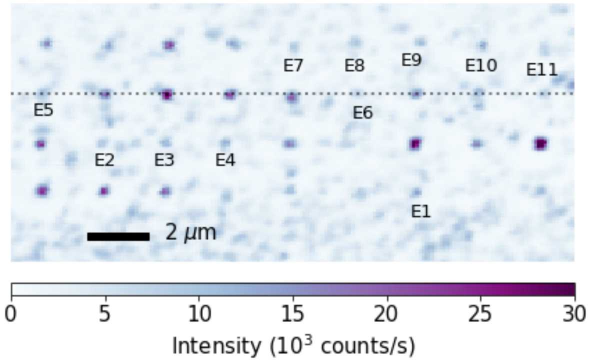




Heejun Byeon and Andrew Mounce at CINT

High confidence in the number of implanted Si ions → confirmed by $g^2(\tau)$

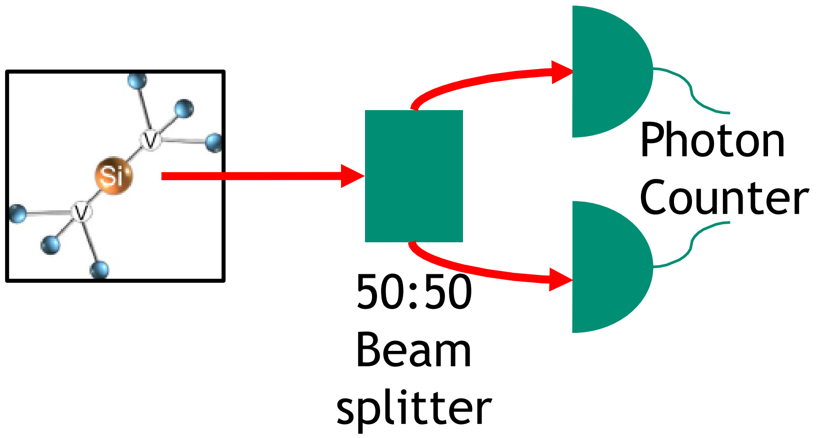
Hanbury-Brown-Twiss Interferometry

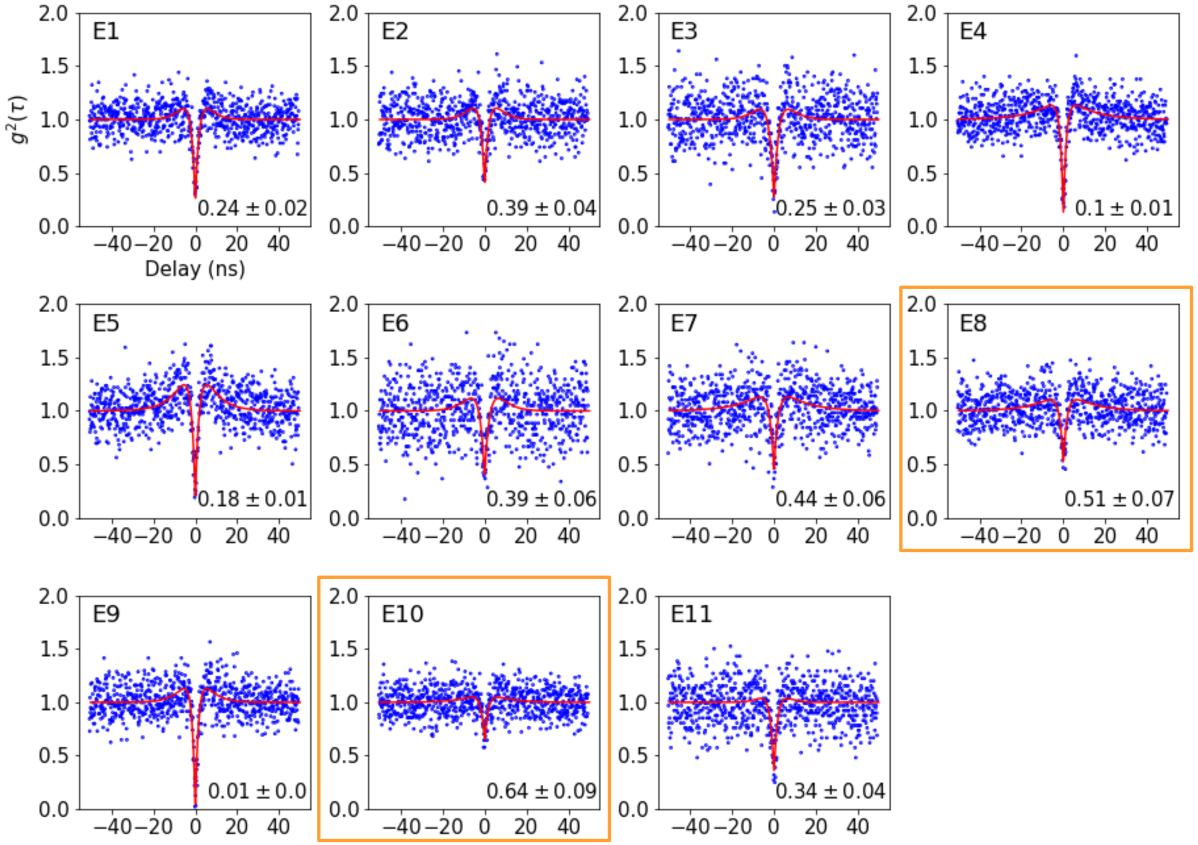


Single emitter will emit only 1 photon at a time

2nd order autocorrelation

Only 1 photon at each counter at any time





Not all locations identified as single emitters from PL are single emitters