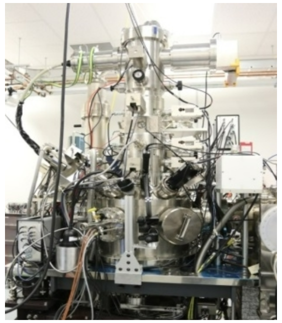
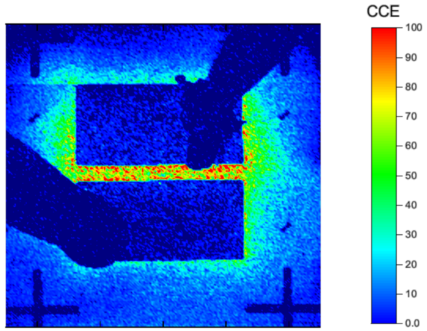
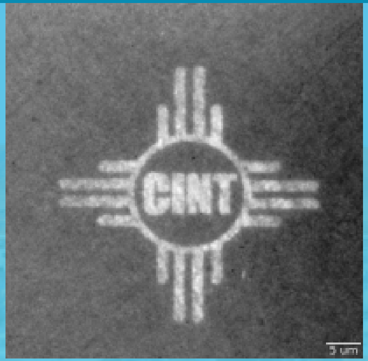
SAND2021-15320C

Sandia National Laboratories

Focused Ion Beam Implantation for the Fabrication of Single Defect Centers in Wide Bandgap Substrates using in-situ Counting and Photoluminescence







Edward Bielejec

Sandia National Laboratories, Albuquerque, NM 87185

U.S. DEPARTMENT OF ENERGY NNSA National Nuclear Security Administration

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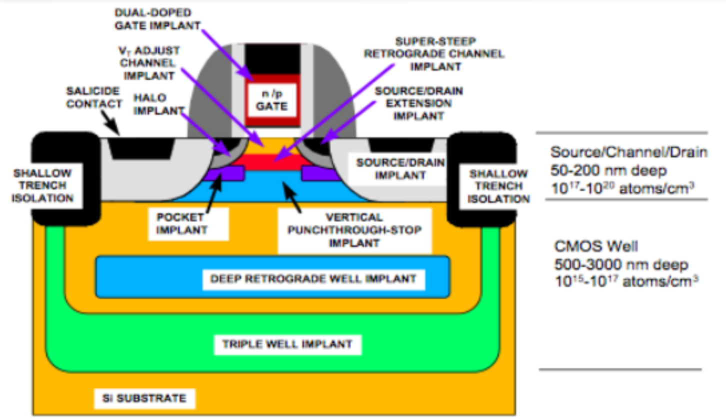
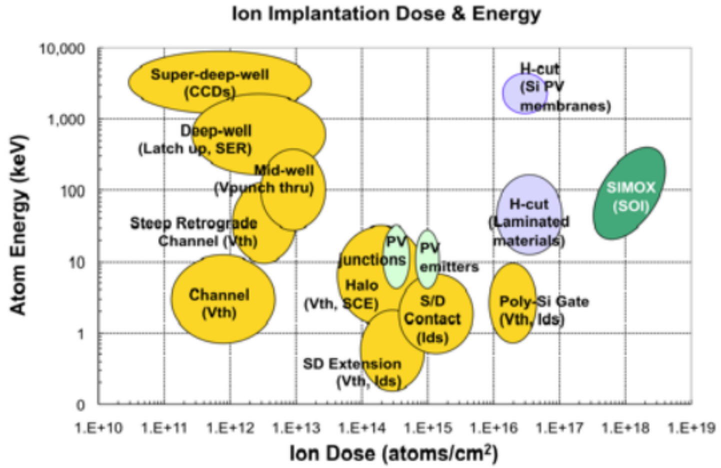
Ion Implantation and Irradiation for Device Fab

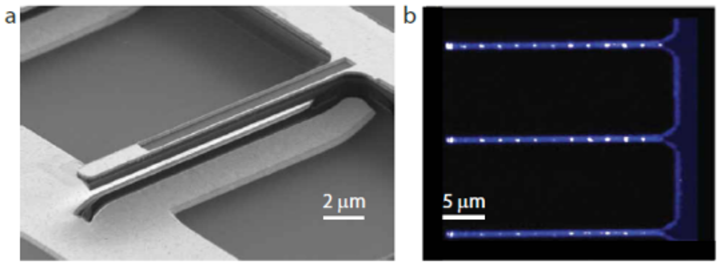
Ion Implantation has been a work-horse for the semiconductor industry



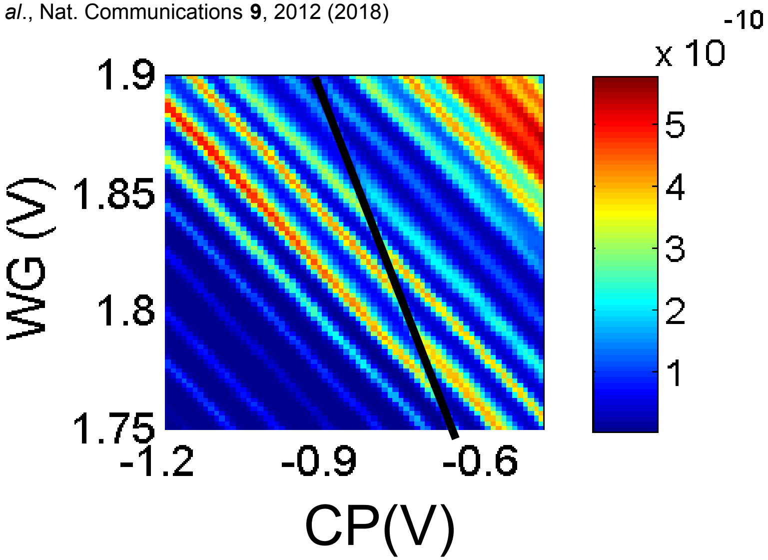


S. B. Felch, *et al*., Proceedings of PAC2013, Pasadena, CA (2013)

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



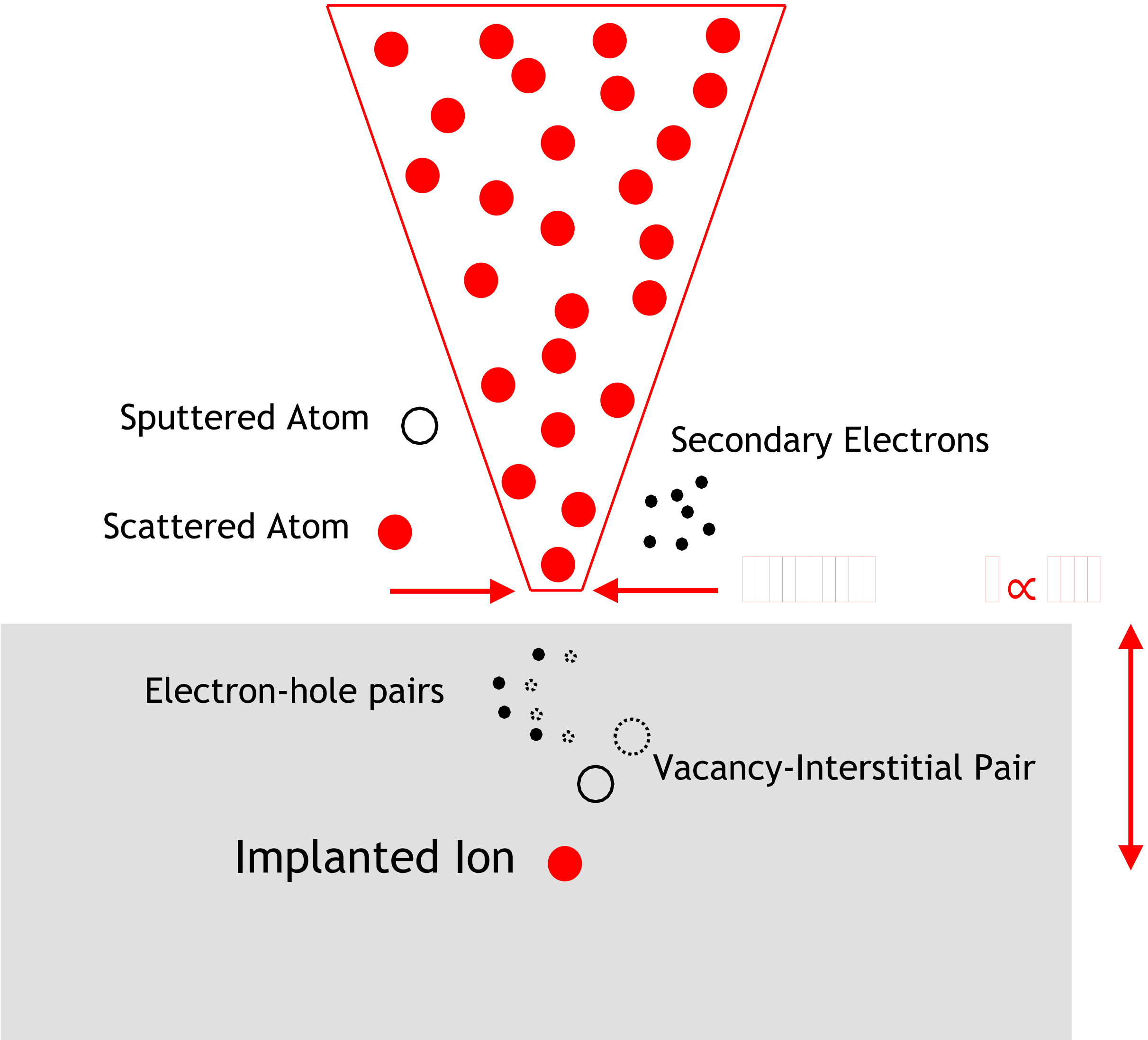
Y. Sohn, *et al*., Nat. Communications **9**, 2012 (2018)



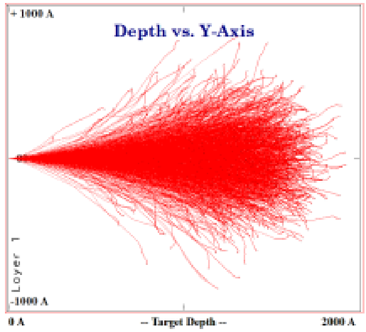
Singh et al. Appl. Phys. Lett. ***108****, 062101 (2016)*

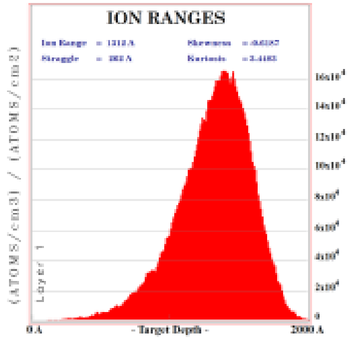
Focused Ion Beam implantation for fabrication of single atom devices and nanofabrication

Overview of the Focused Ion Beam (FIB) Implantation



SRIM simulation – XY Straggle





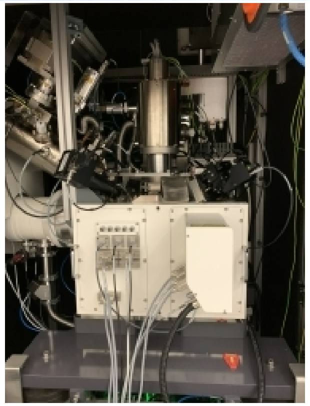
SRIM simulation

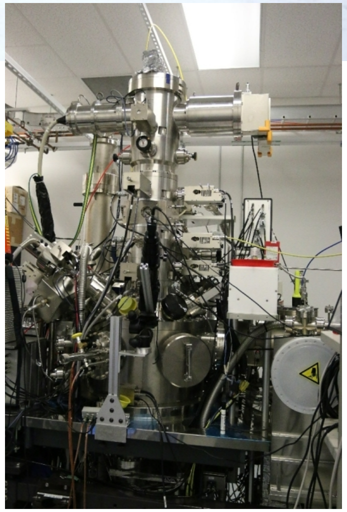
200 keV Si into diamond

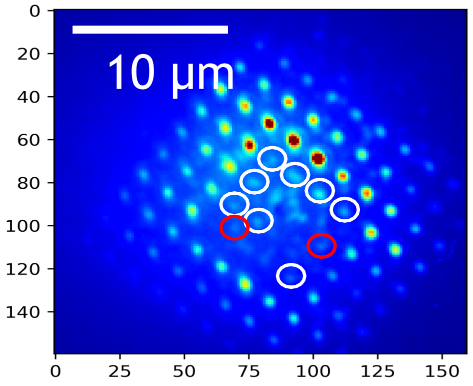
131 ± 26 nm

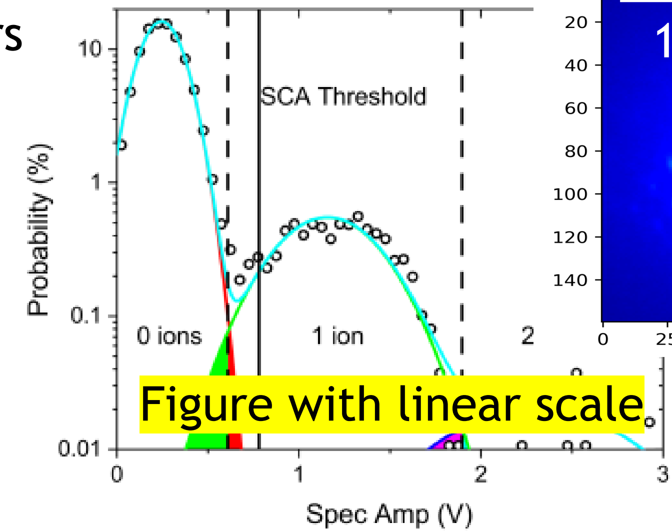
Outline

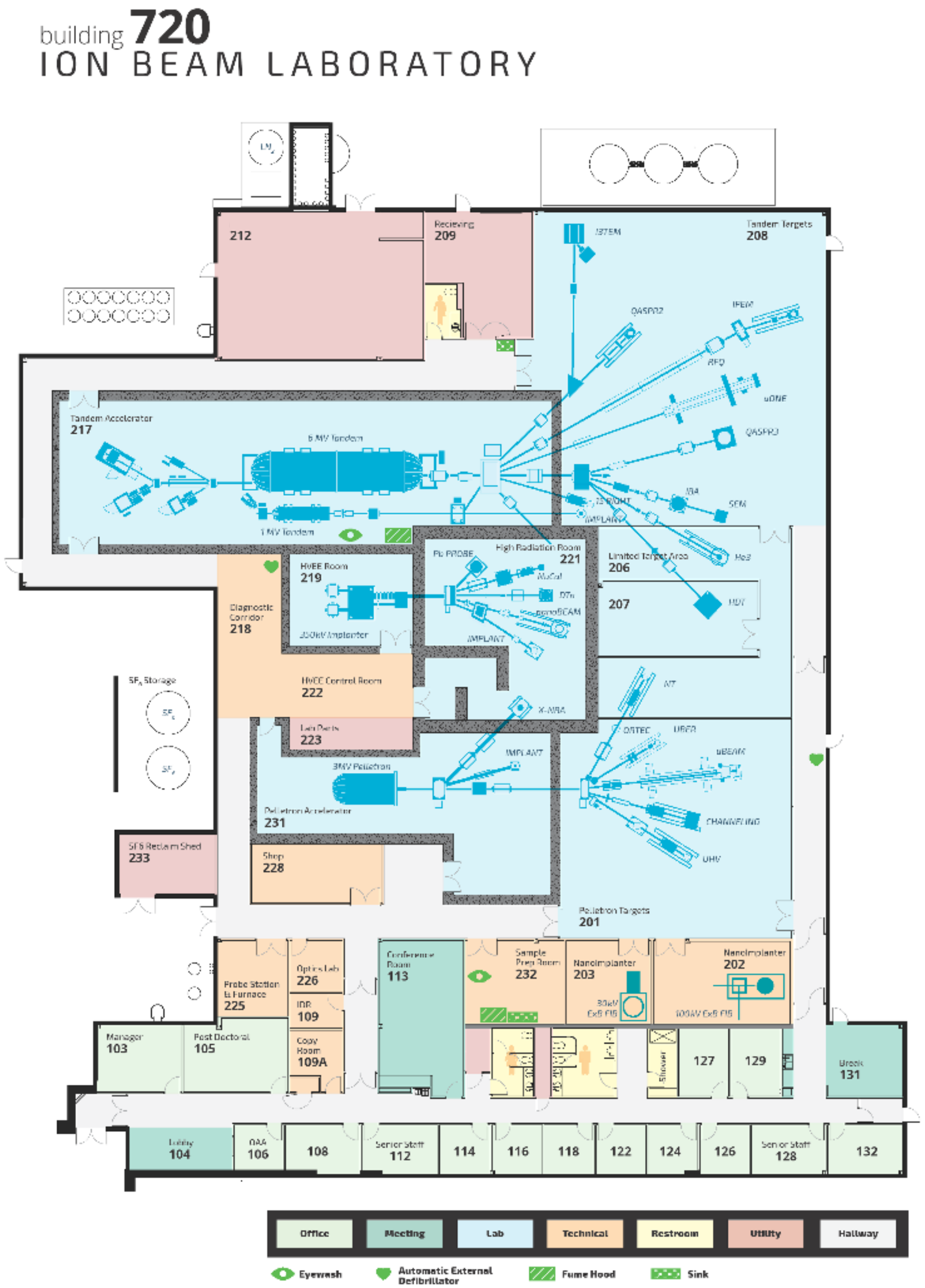
- Introduction to Sandia's Ion Beam Laboratory (IBL)
- Multi-species Focused Ion Beam (FIB)
  - Sandia's nanoImplanter and Raith Velion
- Single Defect Centers in Wide Bandgap Substrates
  - In-situ Counting – control the # of implanted ions
  - In-situ Photoluminescence – verify defect centers
- Summary











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 to mm's

(5) 100 kV ExB FIB nanoImplanter

(6) 35 kV ExB FIB Raith Velion

(7) 35 kV Zeiss HeIM

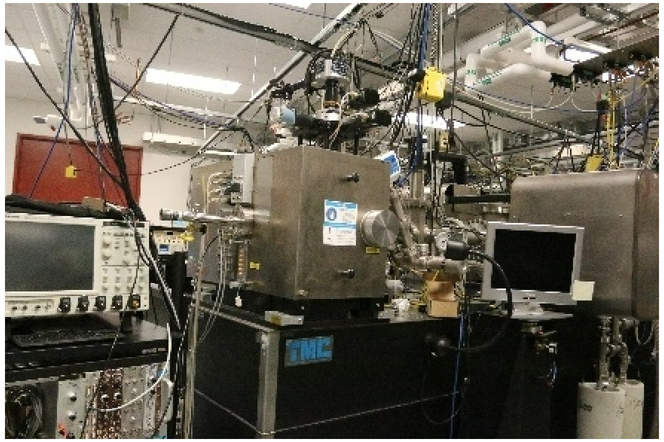
Installing

(8) 35 kV Plasma FIB

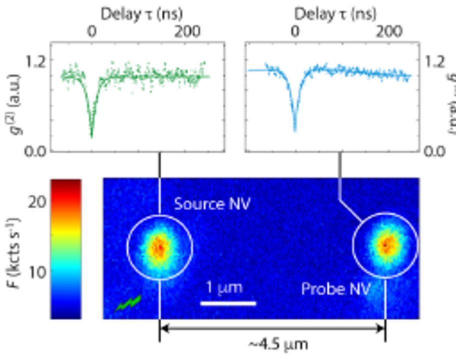
Low energy focused nanobeams <1 to 20 nm

High Energy Focused Ion Beams ~1 μm to mm’s

- 6 MV Tandem microbeam (microONE)

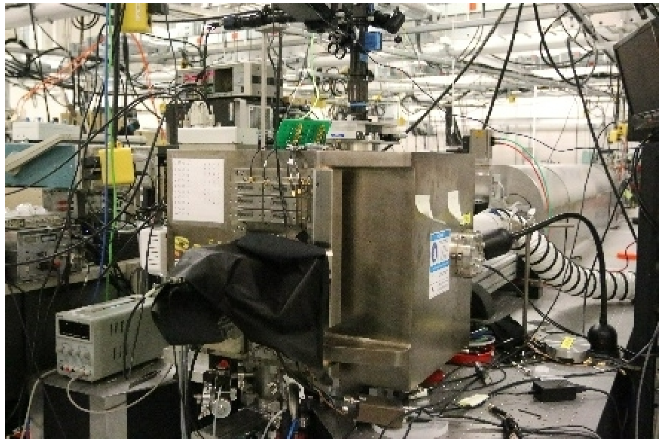


- High resolution laser stage
- Spot size <1 μm
- Energy 0.8 – 70 MeV
- H to Au

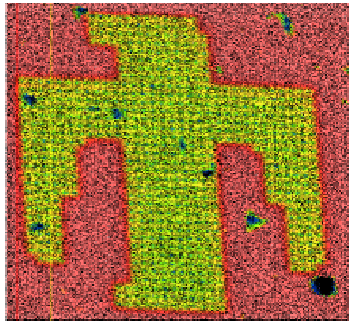


NV PL in diamond

- 3 MV Pelletron microbeam (Light Ion Microbeam)

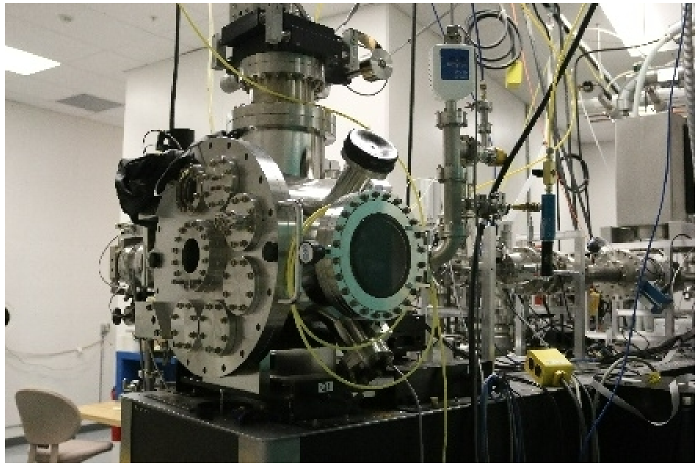


- High resolution laser stage
- Spot size <600 nm
- Energy 0.25 – 3 MeV
- H, He, N, Ar, Xe, …

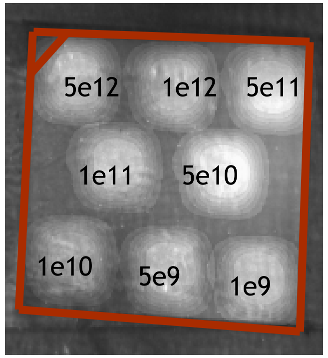


IBIC on PIN diode

- 350 kV HVEE microbeam (NanoBeamLine)



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

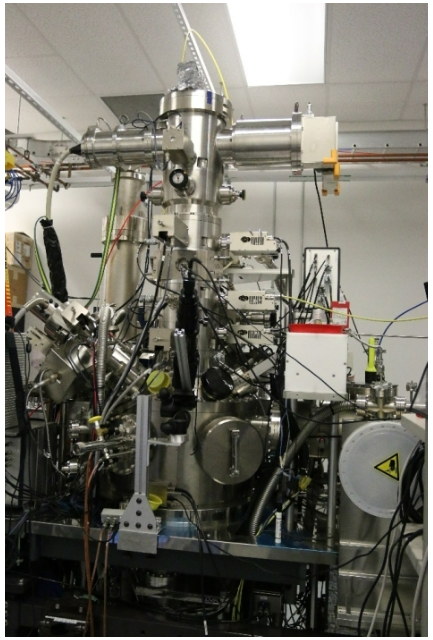


Ni into diamond

A. Lozovoi, *et al.*, Accepted for Publication in *Science Advances* (2021)

Low Energy Focused Ion Beams <1 to 20 nm

- 100 kV A&D FIB100NI (nanoImplanter)

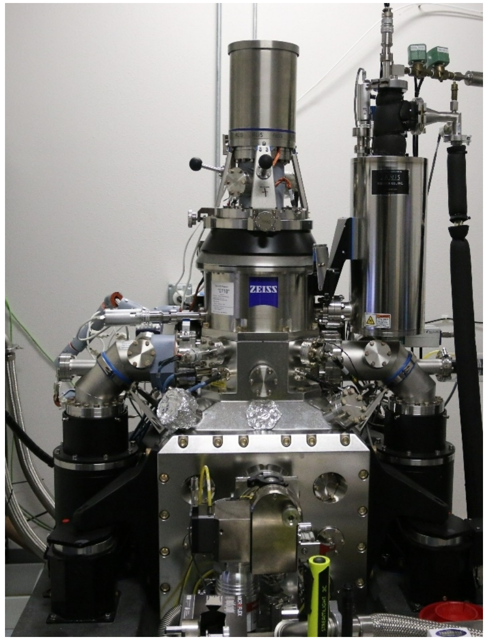


- High resolution laser stage
- Spot size <10 nm (Ga)
- Energy 10 – 200 keV
- 1/3 periodic table

- 35 kV Raith Velion (Velion)

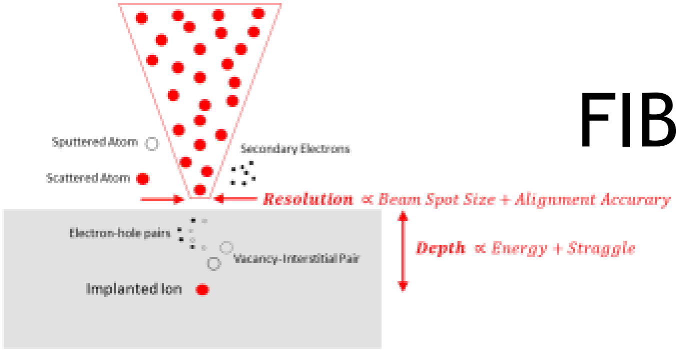
- High resolution laser stage
- Spot size <6 nm (Ga)
- Energy 5 – 70 keV
- 1/3 periodic table

- 35 kV Zeiss Orion Plus (HeIM)



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

All equipped with 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

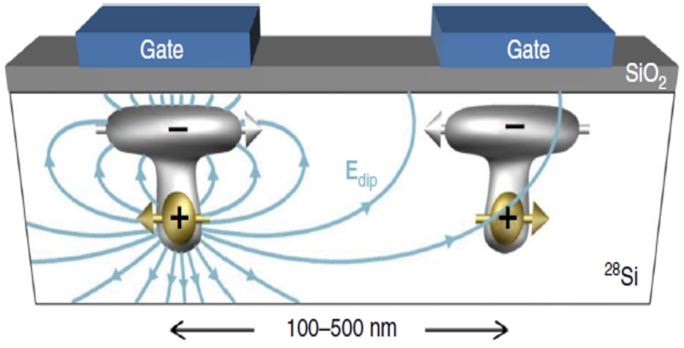
Alignment accuracy ~ 10's nm

Overall resolution ~ 20-50 nm

What is the needed resolution?

Silicon quantum processor with robust long-distance qubit couplings

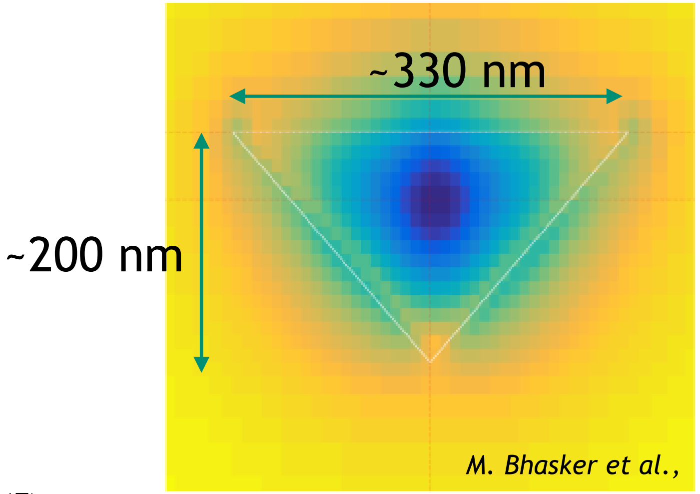
Guilherme Tosi[1], Fahd A. Mohiyaddin[1,3], Vivien Schmitt[1], Stefanie Tenberg[1], Rajib Rahman[2], Gerhard Klimeck[2] & Andrea Morello[1]



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

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

Nanophotonic Applications



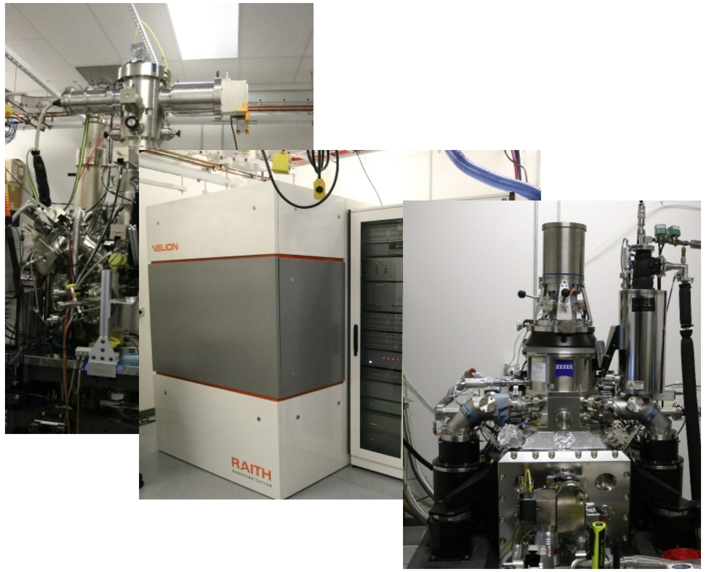
Center of mode is ~ 55 nm below the surface of the waveguide ☑

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

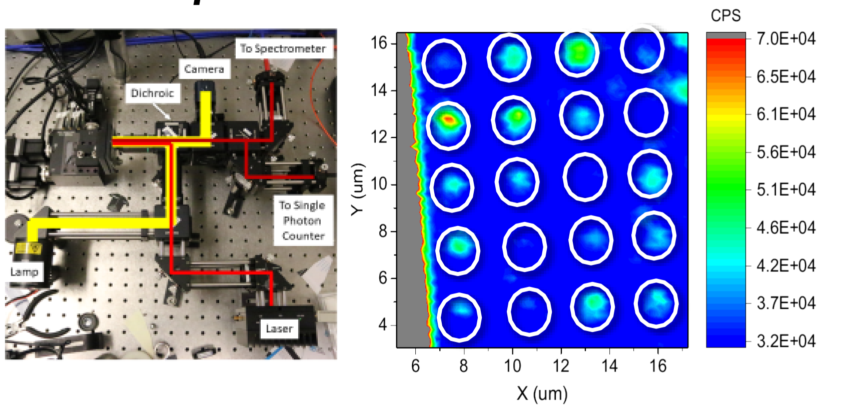
Practical Example of FIB Implantation

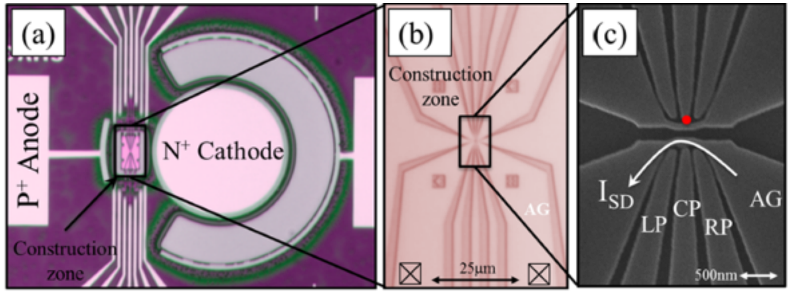
Conversation of what is needed?

FIB Implantation/Irradiation

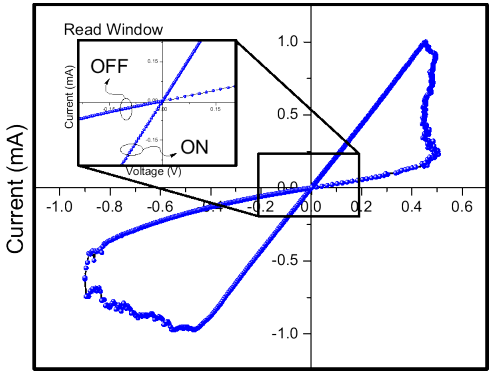


Post Implantation Characterization



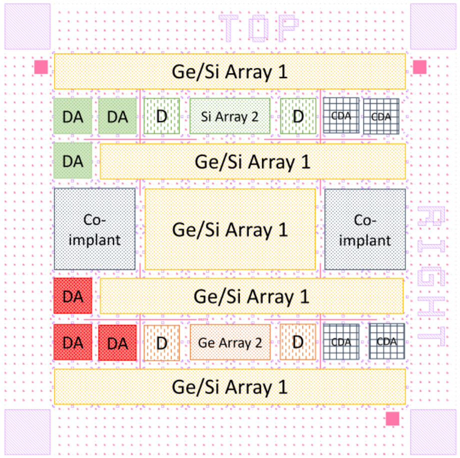


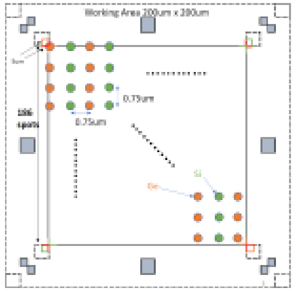
J. Pachaco, *et al*., *Rev. of Sci. Instr.* (2017)



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

Design and layout of sample



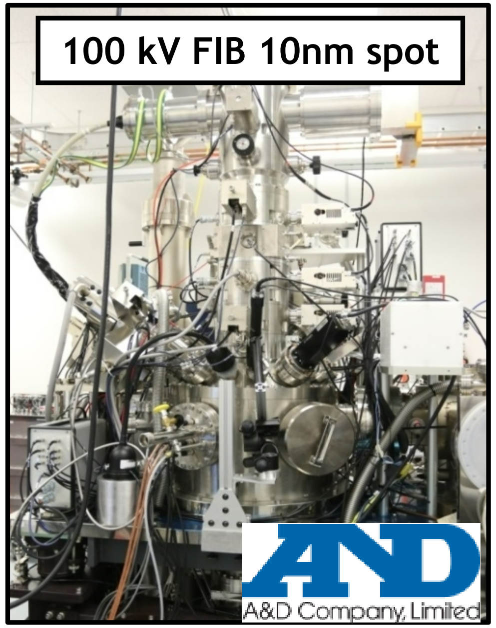


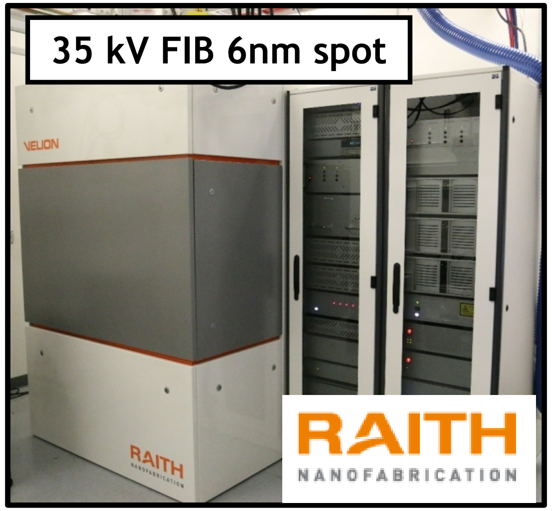
Post Implantation Sample Prep

https://cint.lanl.gov/

Capabilities accessible through CINT User Proposals

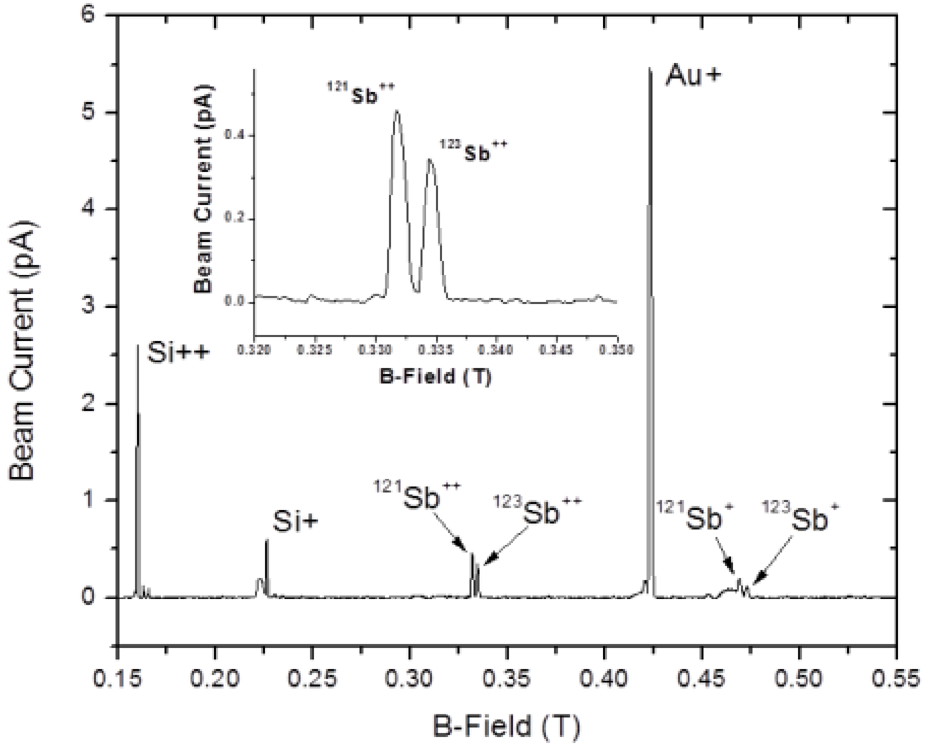
Focused Ion Beam Implantation - nanoImplanter (nI) and Velion





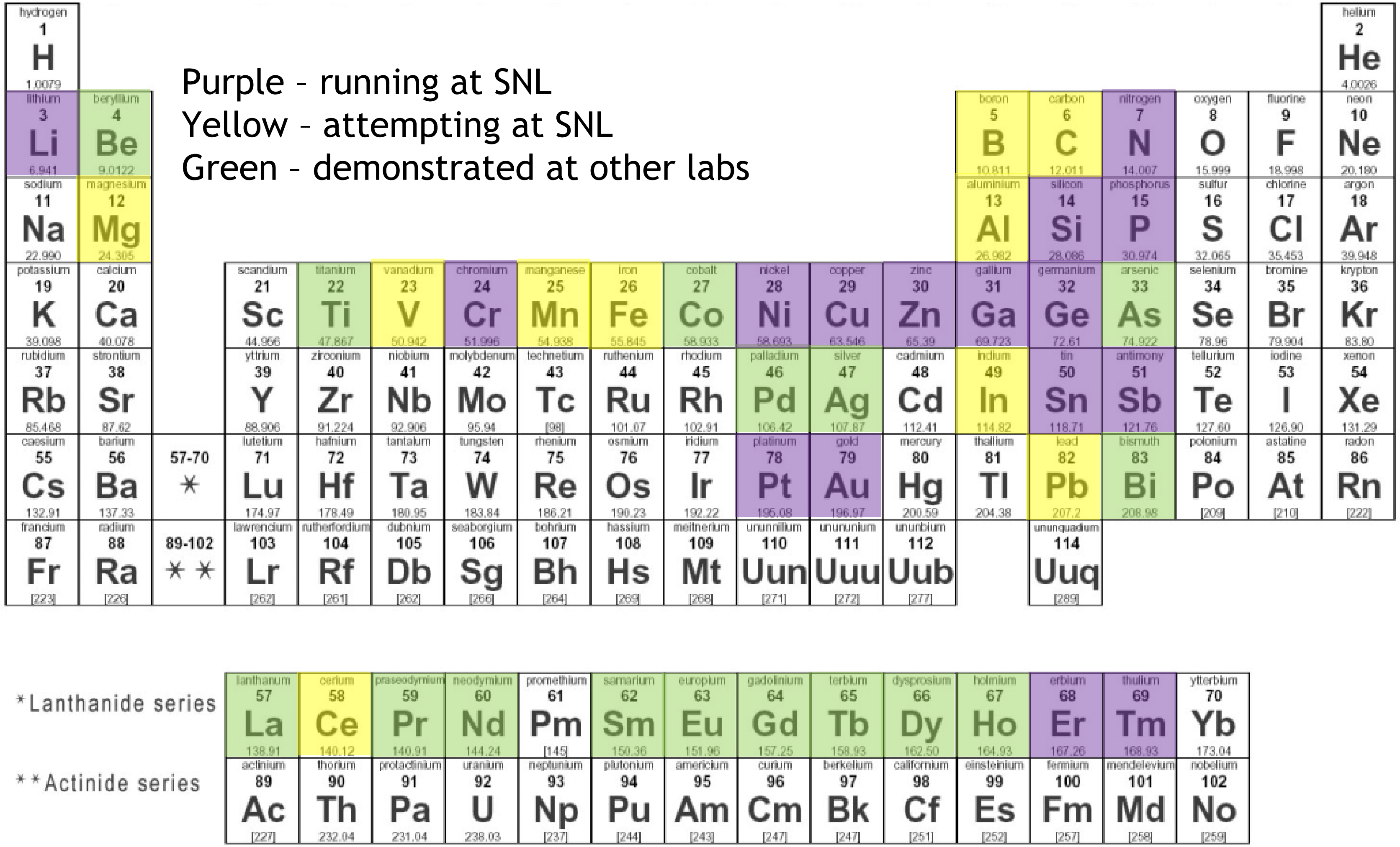
- Focused ion beam system (FIB)
  → nm beam spot size on target
- ExB Filter (Wien Filter)
  → Multiple ion species
    e.g., Li, Si, P, Sb, etc… (separating out ^{28}Si, ^{29}Si, etc…)
- Fast blanking and chopping
  → Single ion implantation
- Direct-write lithography
  → nm targeting accuracy
- Low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

Sb Source: Mass Spectrum



Liquid Metal Alloy Ion Sources (LMAIS) for FIB Implantation

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

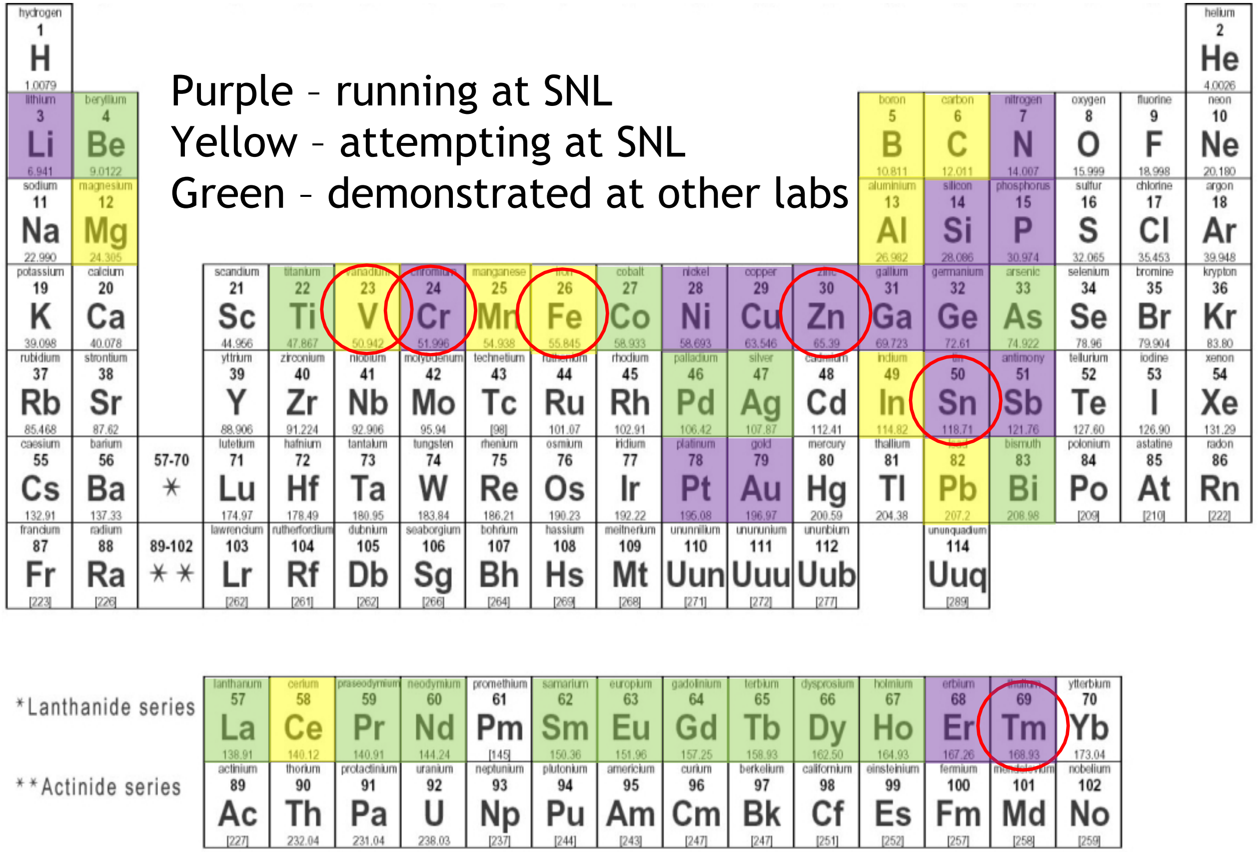


L. Bischoff, *et al.*, Applied Physics Reviews **3**, 021101 (2016)

After A. Weick University of Bochum

Wide Variety of Ion Species Available

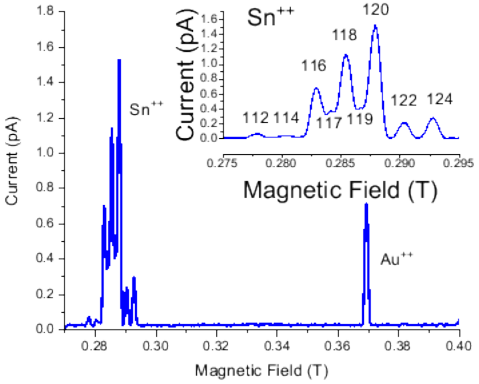
New Sources – V, Cr, Fe, Zn, Sn, Tm (easy-ish)



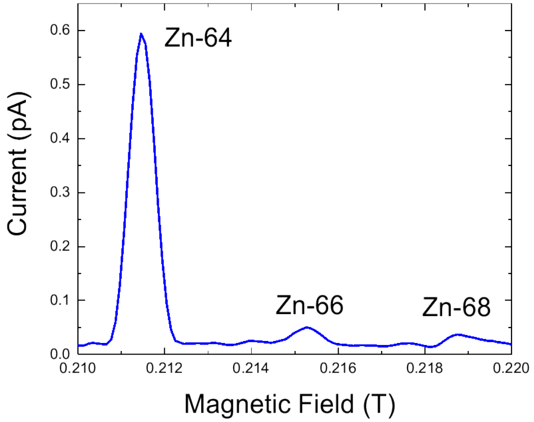
Purple – running at SNL
Yellow – attempting at SNL
Green – demonstrated at other labs

- Based on AuSiX or AuGeX alloys
- Easy to wet the tip and easy to run

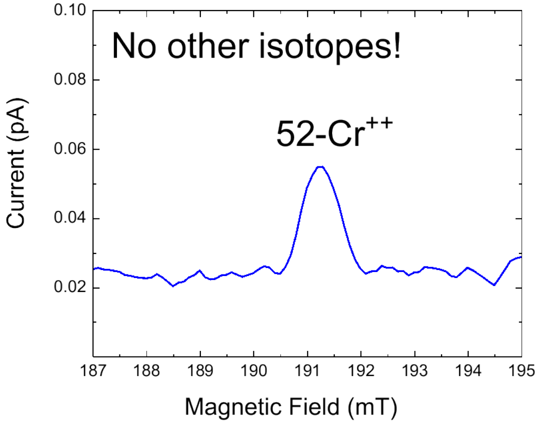
AuSn



AuSiZn

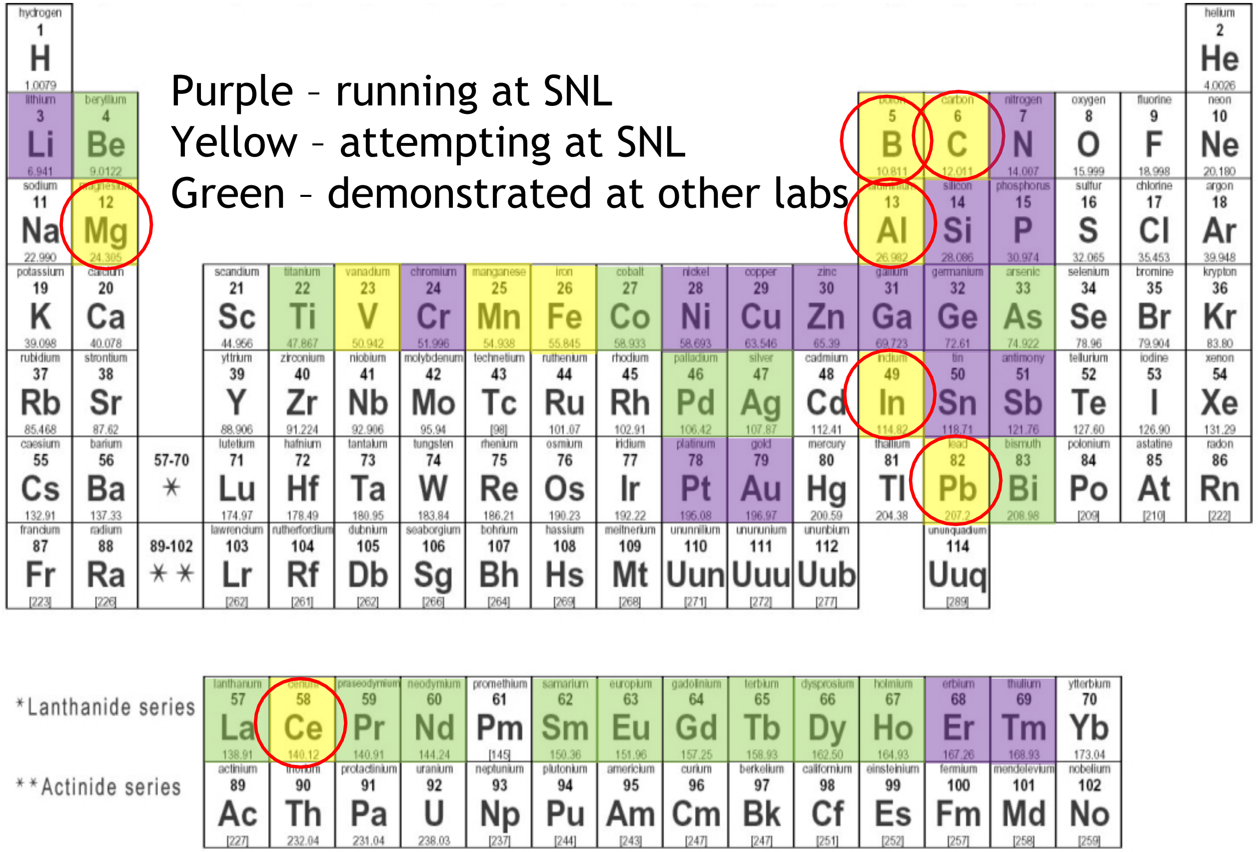


AuGeCr



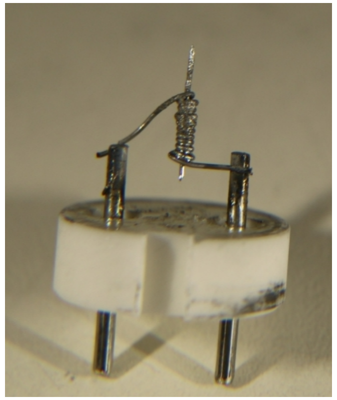
New Sources – B, C, Al, Ce, Mg, In, Pb (Hard)

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



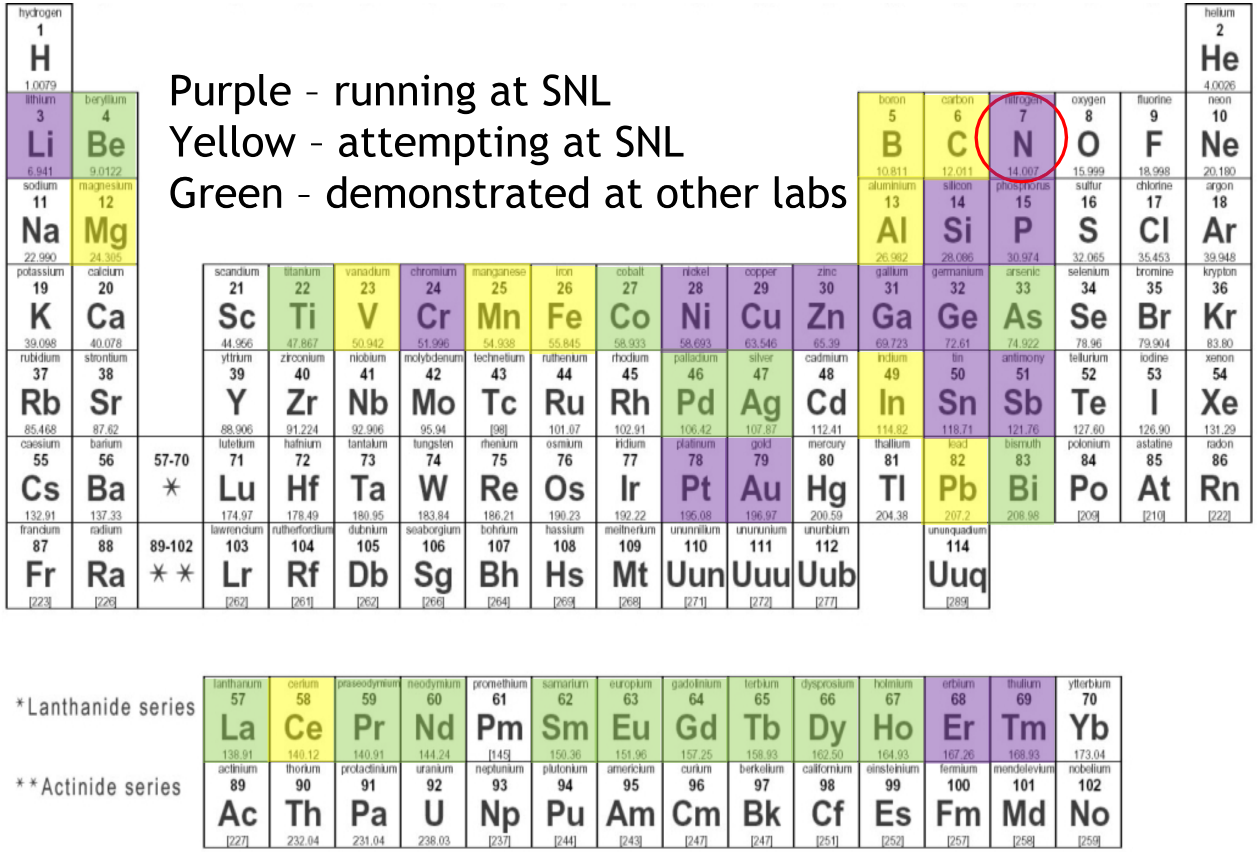
- B
  - → Hard to load into the alloy BAuGeNi
- C, Al, Ce
  - → AlCeC alloy oxides while running, killing beam
- Mg
  - → GaMg alloy oxides in the boat
- Pb
  - → PbSn alloys with W tip, working on new materials

SNL SnPb tip
on Kovar wire



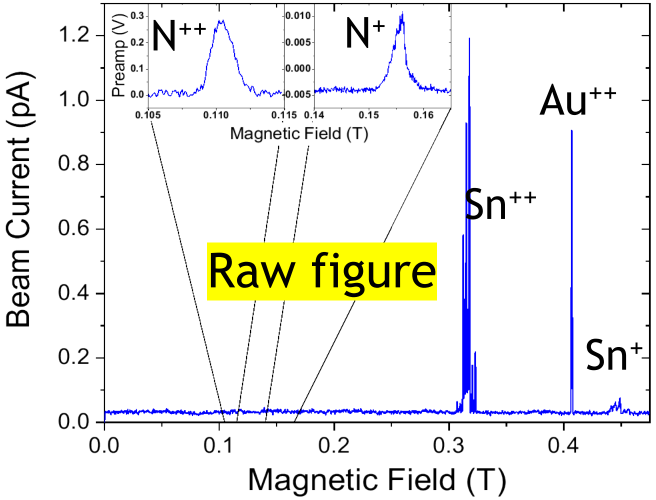
New Sources – N (How to do???)

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

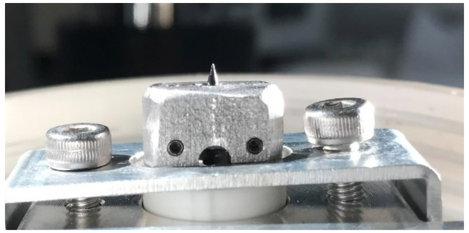


(1) Liquid metal alloys with implanted N

i.e., AuSn+N, In+N

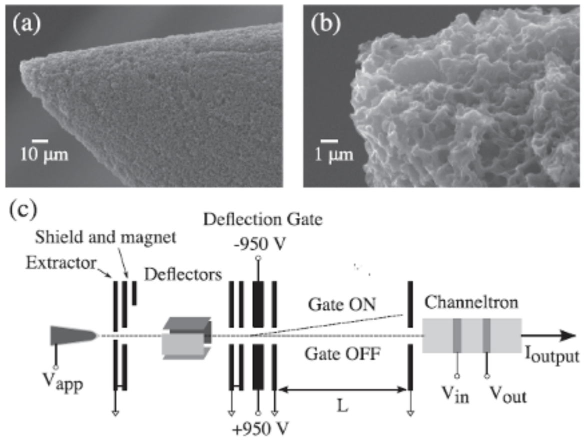


(2) Ionic Liquid Ion Source (with MIT)



(At SNL for testing)

Carbon Xerogel Microtip with ILIS



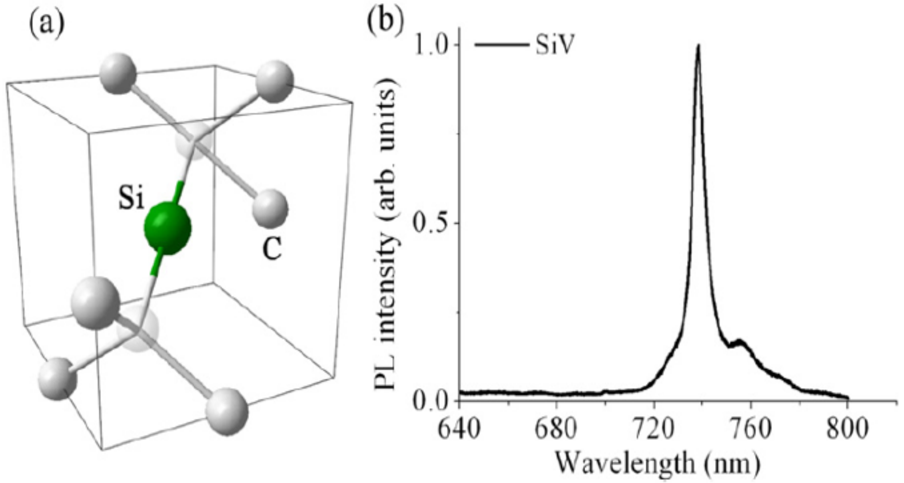
C. S. Perez-Martinez, *et al.*, *Appl. Phys. Lett.* **107**, 043501 (2015)

Single Defect Centers in Wide Bandgap Substrates

- Defect centers in wide bandgap substrates have applications from metrology to quantum computation

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)

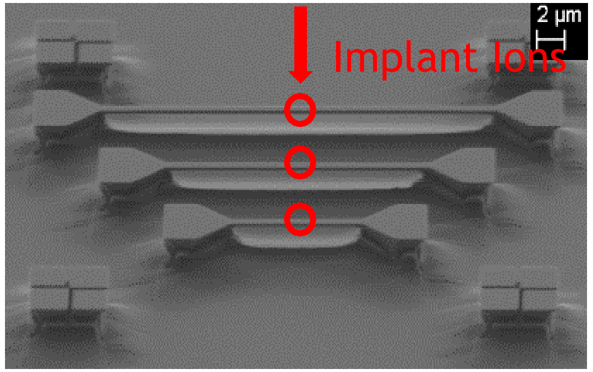
- How to produce a single defect center where you want it?

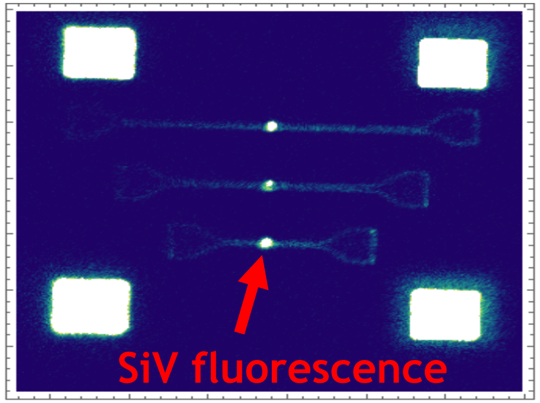
1.) Location – focused ion beam implantation to control location

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

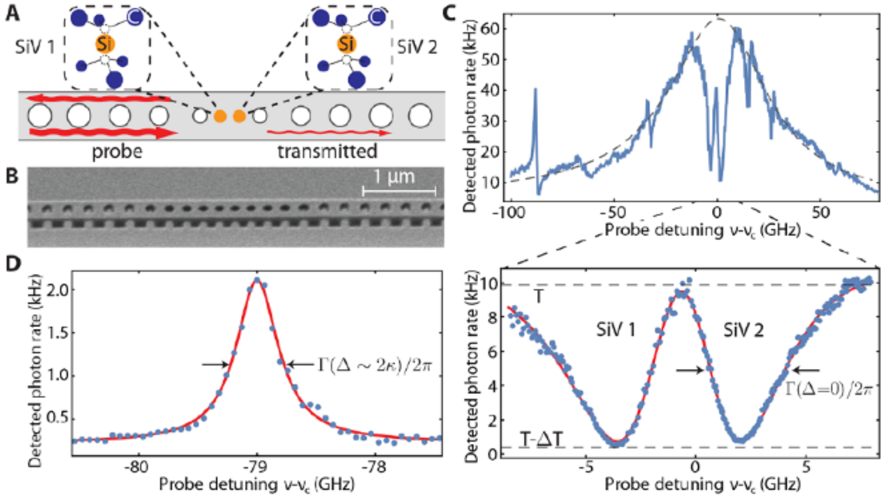
Use FIB implantation to control the spatial location

Diamond Nanobeams (with Harvard)



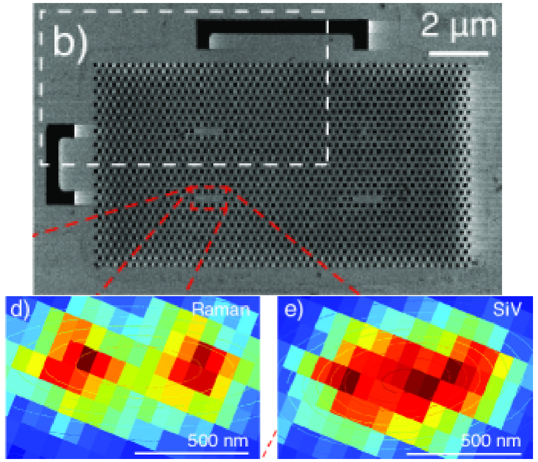


A. Sipahigil, *et al.*, *Science* (2016)



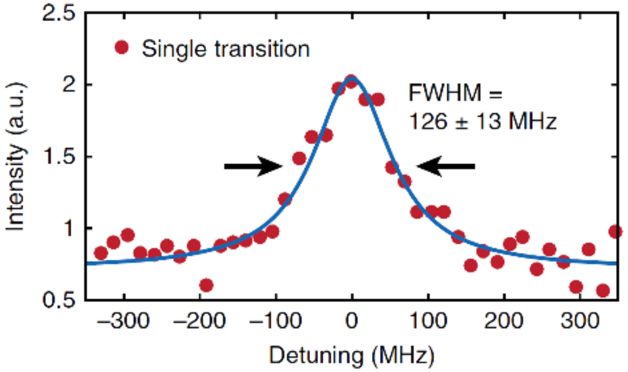
R. E. Evans, *et al.*, *Science* (2018)

2D photonic Crystals (with MIT)



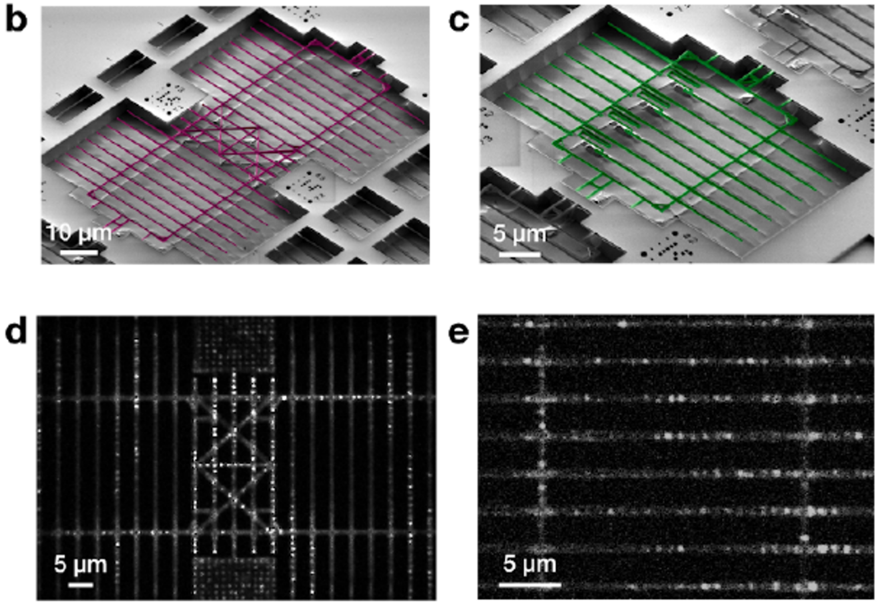
Accuracy is <50 nm

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



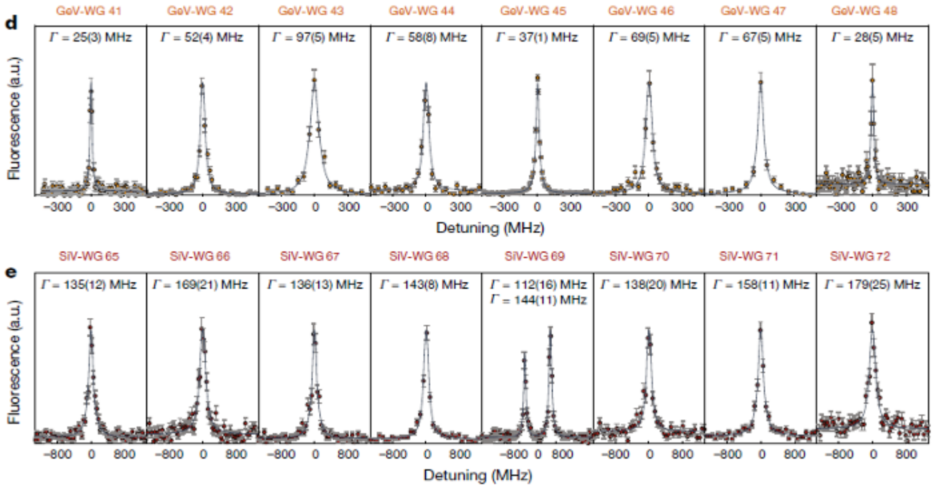
Use FIB implantation to control the spatial location

Diamond waveguides with AlN photonics (with MIT)

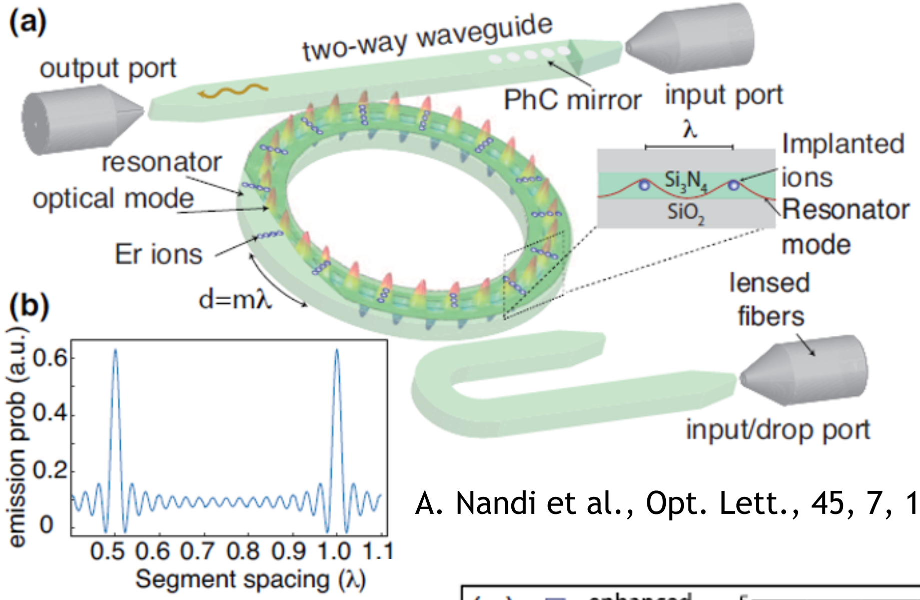


16 and 8 channel "quantum micro-chiplets"

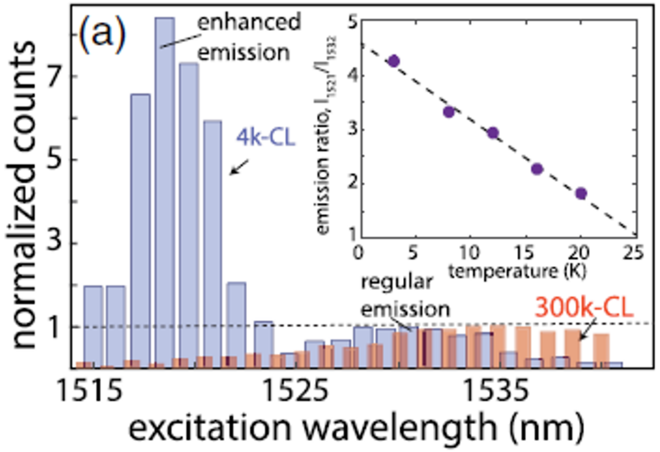
Noel H. Wan et al., Nature, 583, 226–231(2020)



Silicon Photonics (with Purdue)



A. Nandi et al., Opt. Lett., 45, 7, 1631 (2020)



Use Counted Implantation and Photoluminescence to Better Understand Yield

Yield = # measured SiV / # implanted Si

<u>In-situ Photoluminescence</u>

- 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

See EQ01.03.03 V. Chandrasekaran for more details

<u>In-situ Counted Implantation</u>

- 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

See EQ01.09.03 M. Titze for more details

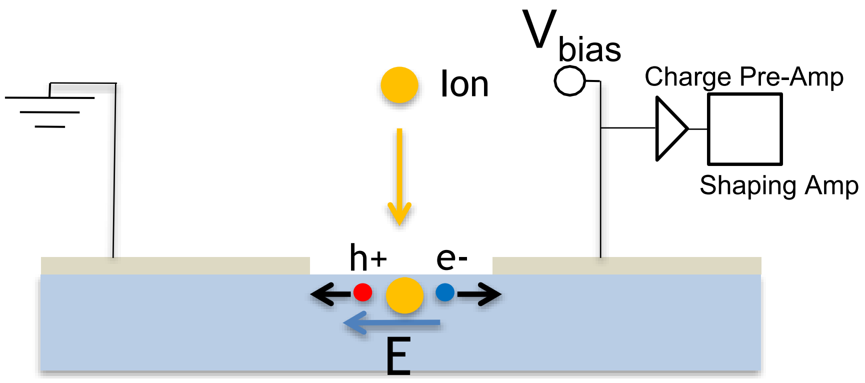
In-situ Counted Ion Implantation

Yield = # measured SiV / # implanted Si

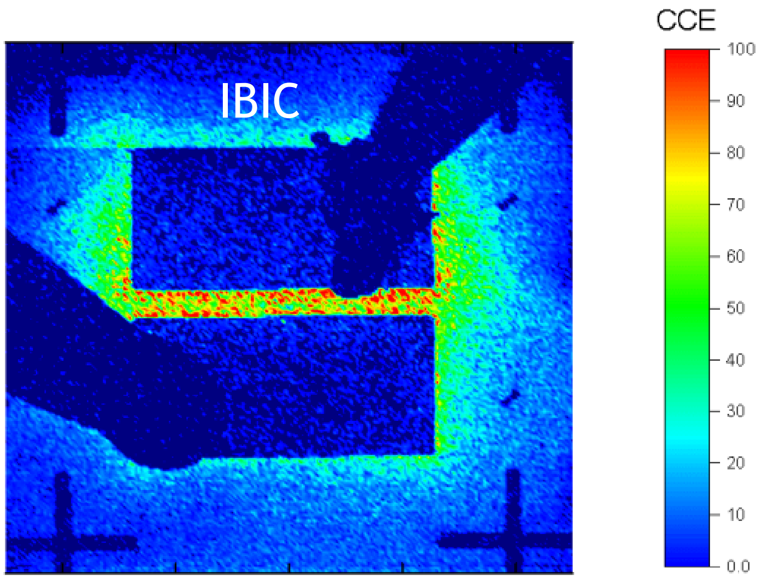
Poisson Statistics

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

Ion Beam Induced Charge (IBIC)



Side View

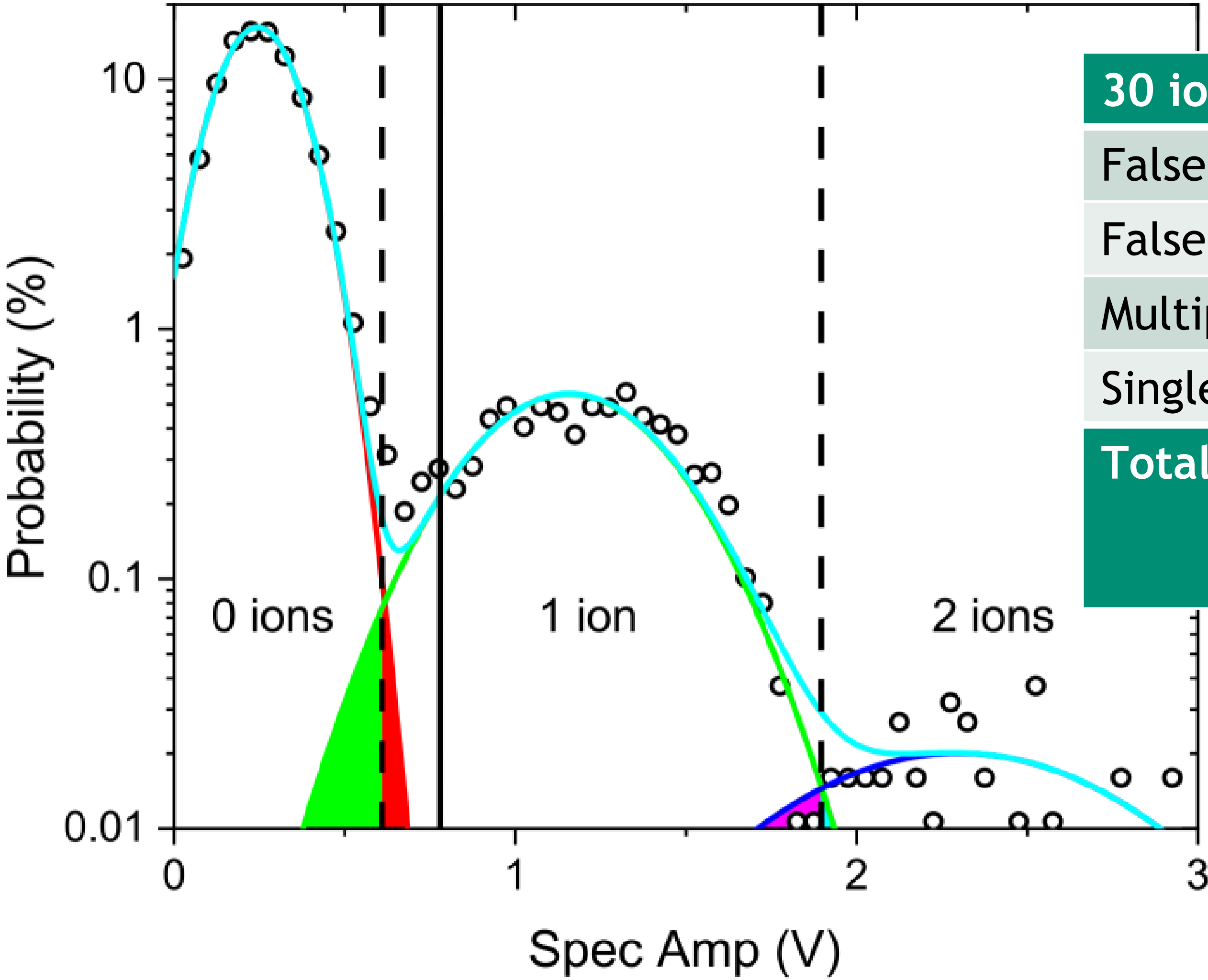


$$CCE = \frac{Charge\ Collected}{Charge\ Depositied} x\ 100$$

IBIC/detection demonstrated for low energy heavy ions

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

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

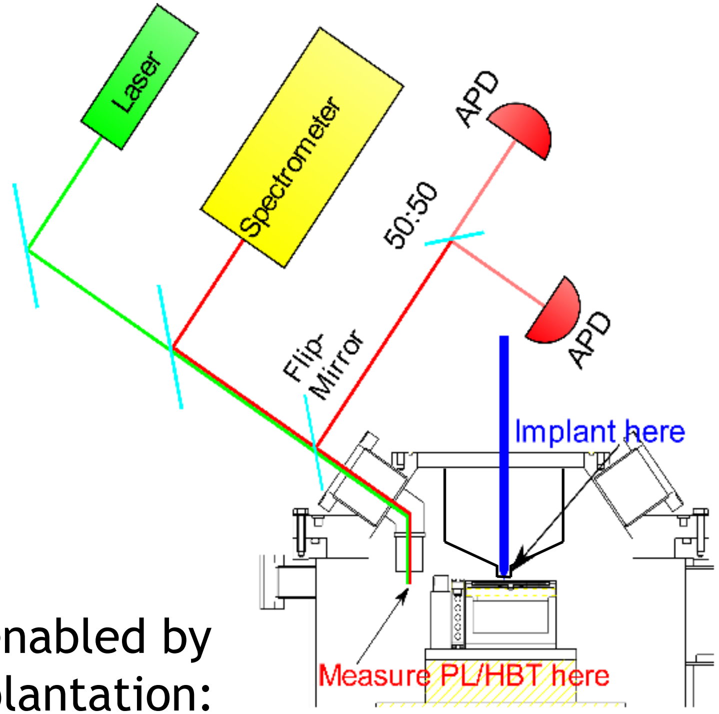
See EQ01.09.03 M. Titze for more details

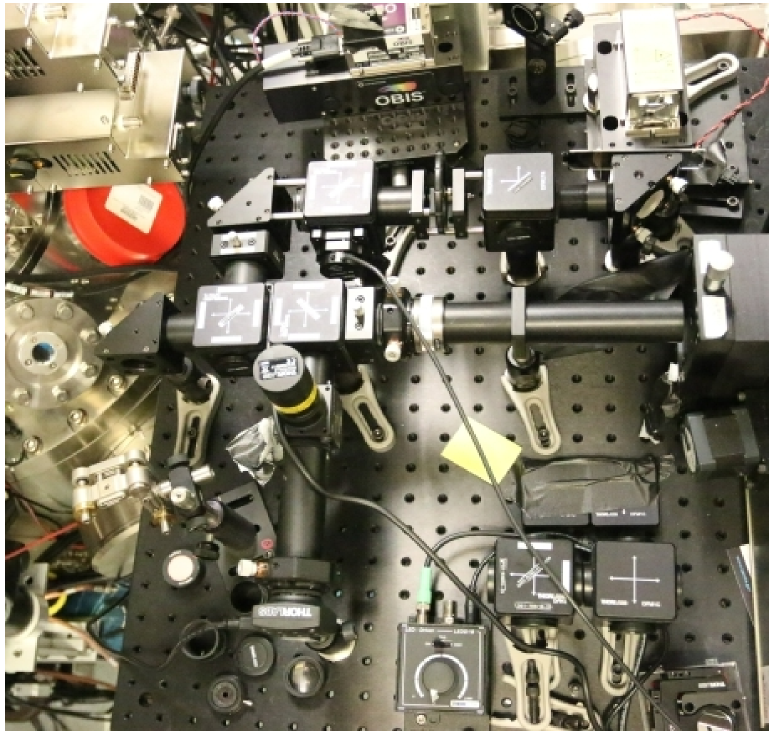
In-situ counting to reduce the error in the number of implanted ions

In-situ Photoluminescence

Yield = # measured Vsi / # implanted Si

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





Replace with picture of in-situ setup on nI

Two-step process enabled by high resolution implantation:

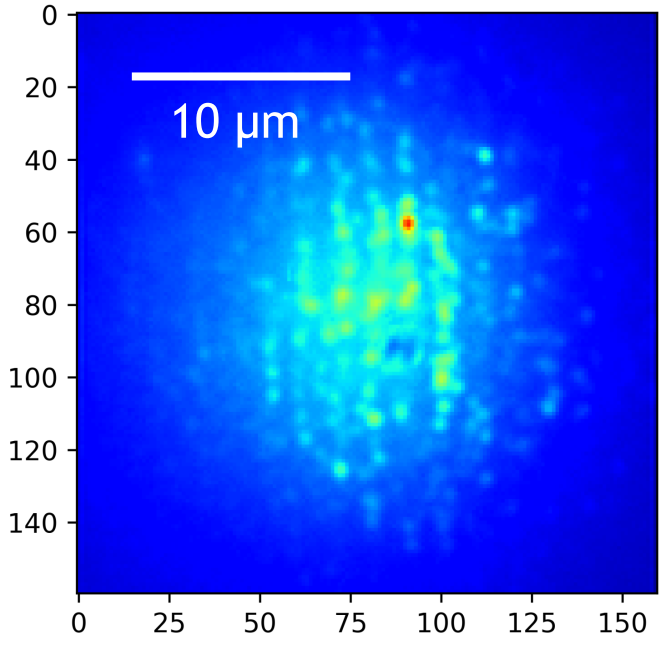
(1) Aligned implantation, <40 nm

(2) Detect PL

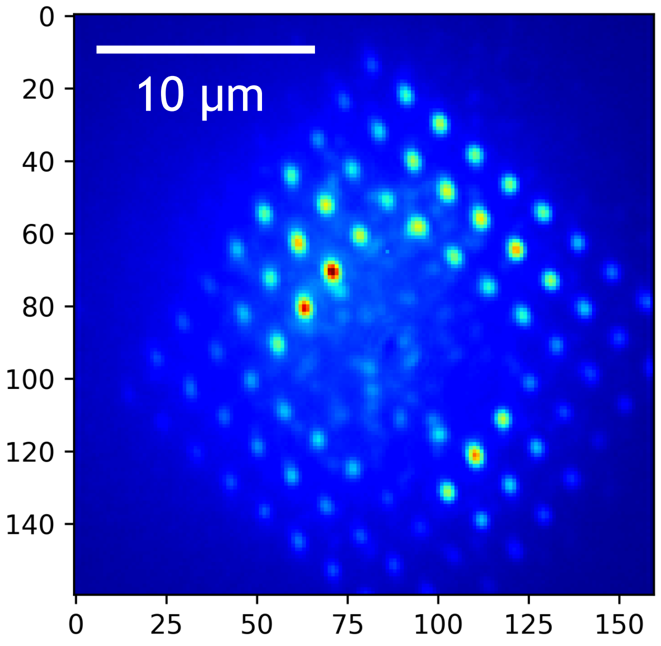
In-situ PL to confirm the optically emission from the defect centers

In-situ Photoluminescence

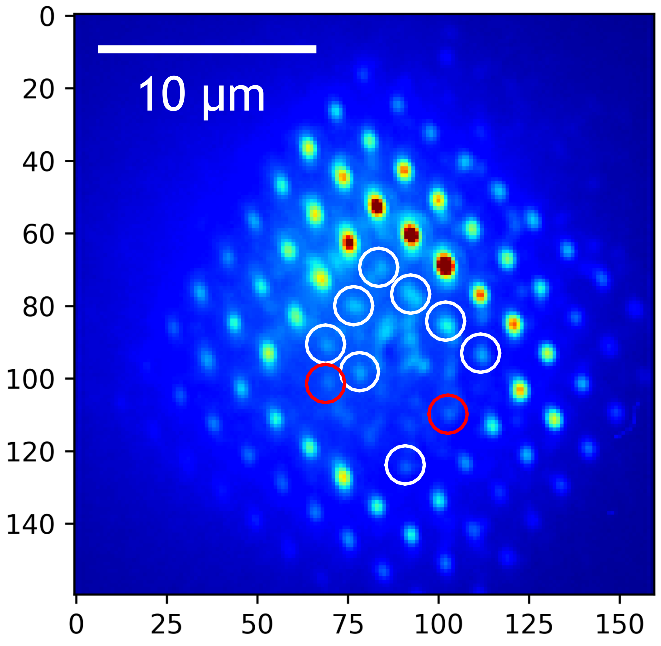
Background PL



Implant Alignment Grid



Implant/PL Repeat to fill Array



See EQ01.03.03 V. Chandrasekaran for more details

Replace with figure with constant color scale

Preliminary results suggest this works, BUT limited by high background counts

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, Jacoby Henshaw, L. Basso, H. Byeon, A. Mounce, P. Keyayias, M. Lilly (SNL)

V. Chandrasekaran, Han Htoon (LANL)

V. Costa (UNM)

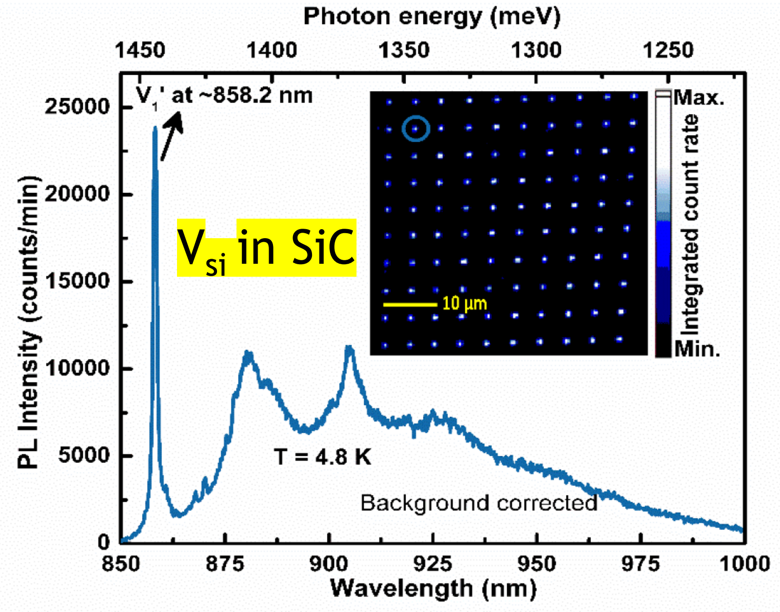
And we have continued to support a wide range of user groups through CINT

Summary

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

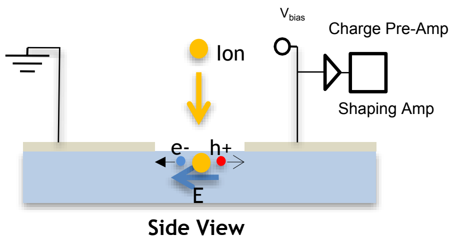
→ Viable solution for prototyping – fast and easy!

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



S. Pavunny et al., Scientific Report **11**, 3561 (2021)

- Path Towards Deterministic Defect Centers in Wide Bandgap Materials



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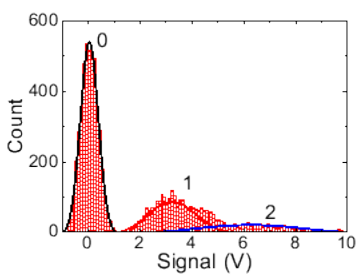
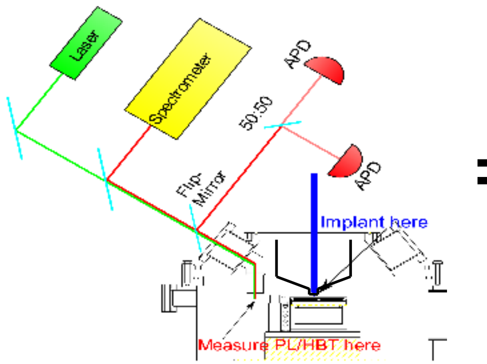


Abraham *et al*, APL **109**, 063502 (2016)

+



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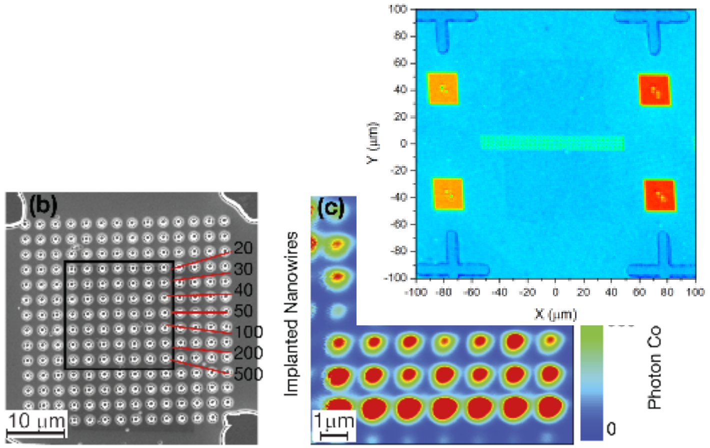


L. Marseglia, *et al.*, *Optics Express* (2018)

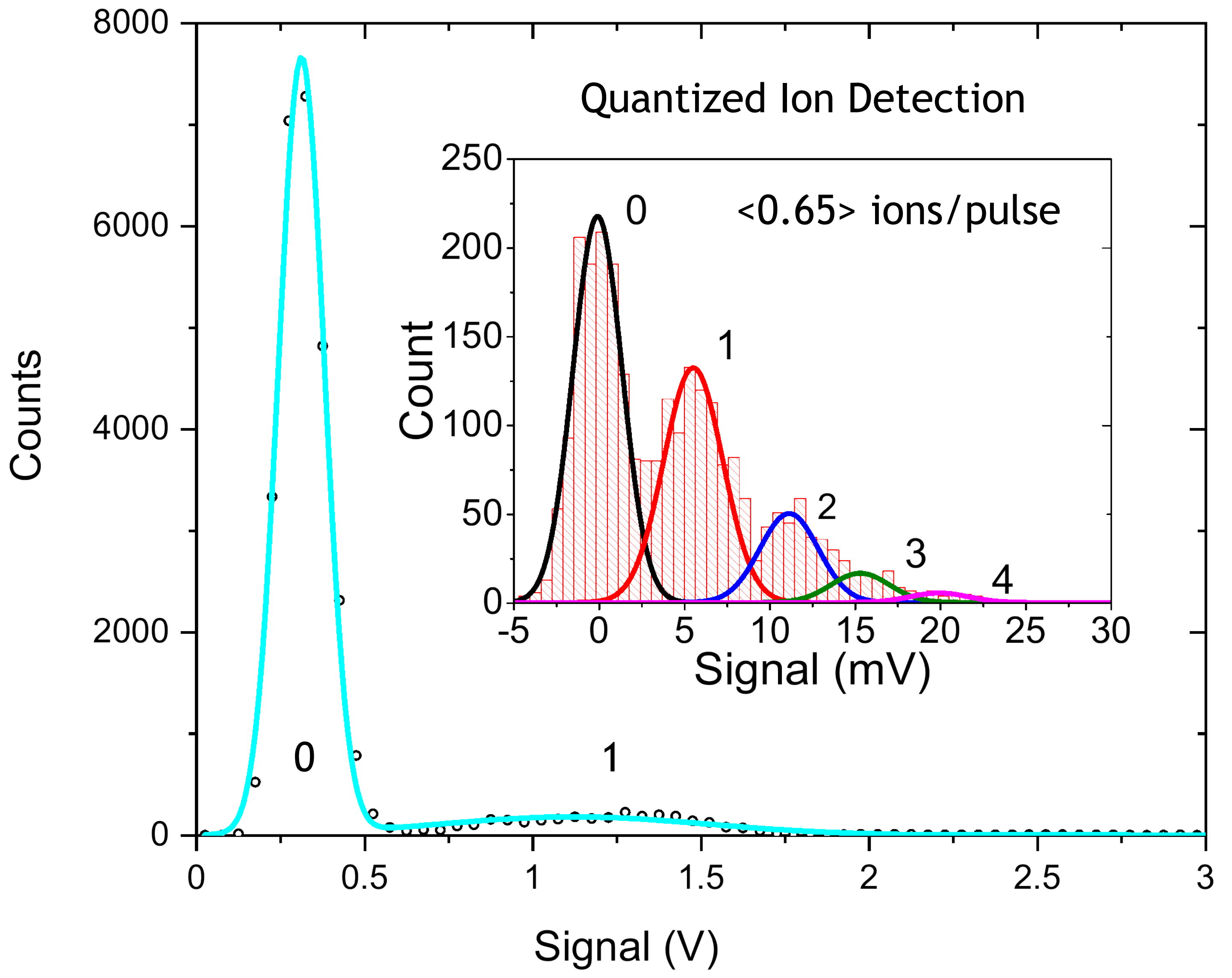
Control the number of ions

Confirm Optically Active Defect Centers

https://cint.lanl.gov/

Extra Slides

Single Ion Diamond Detection at <0.1> ions/pulse



Signal-to-noise ratio

$$SNR = \frac{\mu_{signal}}{\mu_{noise} + \sigma_{noise}}$$

SNR = 4.7

Good distinction between 0 and ≥1 ion

See EQ01.09.03 M. Titze at 2:00 pm for more details

Overview of High Resolution Implantation Techniques

Resolution

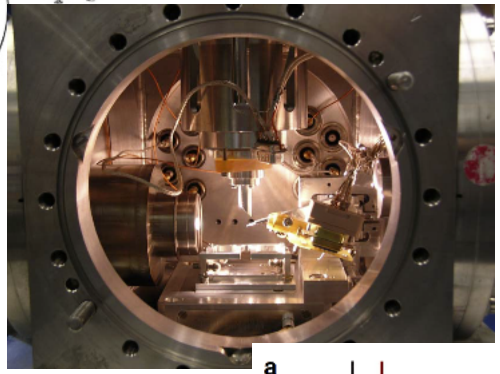
Speed

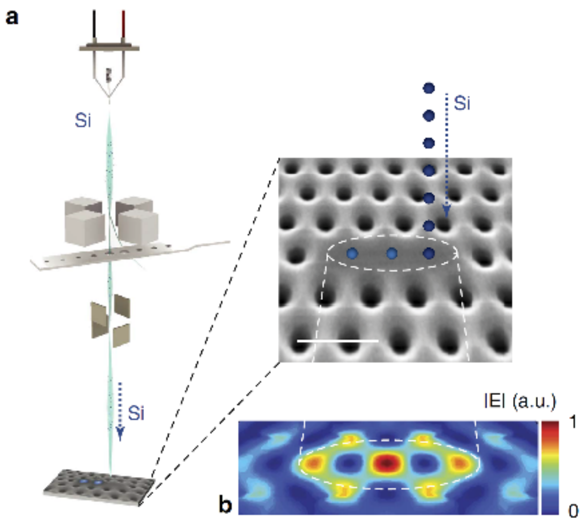
(1) Hydrogen Lithography

(2) Probe Based Implantation

(3) Focused Ion Beam (FIB) Implantation

(4) Masked Implantation

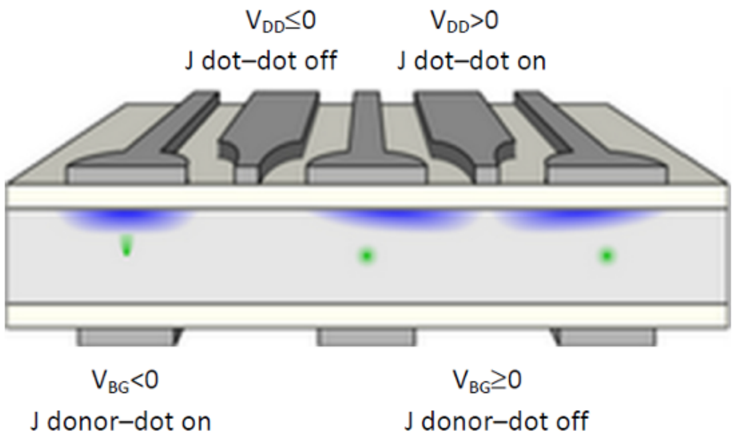




Needed Resolution for Si Qubit Applications

A spin quantum bit architecture with coupled donors and quantum dots in silicon

T. Schenkel[1], C. C. Lo[1], C. D. Weis[1], J. Bokor[2], A. M. Tyryshkin[3], and S. A. Lyon[3]
[1]Ion Beam Technology Group, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
[2]Department of Electrical Engineering and Computer Sciences, University of California, Berkeley, CA 94720, USA
[3]Electrical Engineering Department, Princeton University, Princeton, NJ, USA
Contact-email: T_Schenkel@LBL.gov
(October 10, 2011)

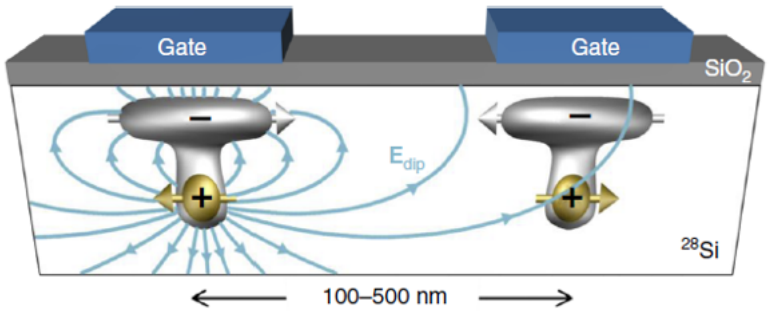


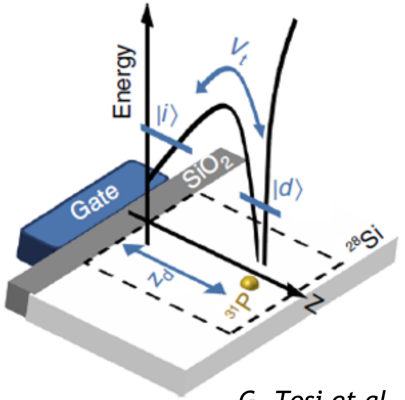
T. Schenkel et al., US 8,816,325 B2 (2014)

- Depth ~20-50 nm
- Donor separation ~100 nm

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]





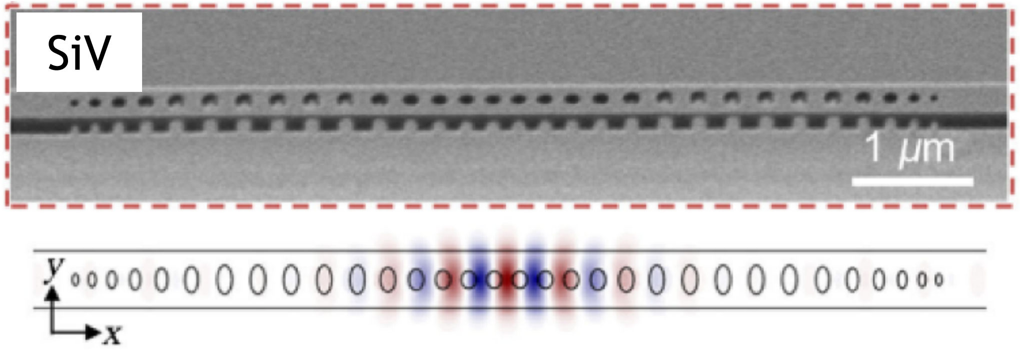
- Depth z_d = 15 nm or larger
- Separation of 100-500 nm

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

FIB Implantation? Lateral Resolution – OK, but Depth Resolution requires low energies!

Needed Resolution for Nanophotonics Applications

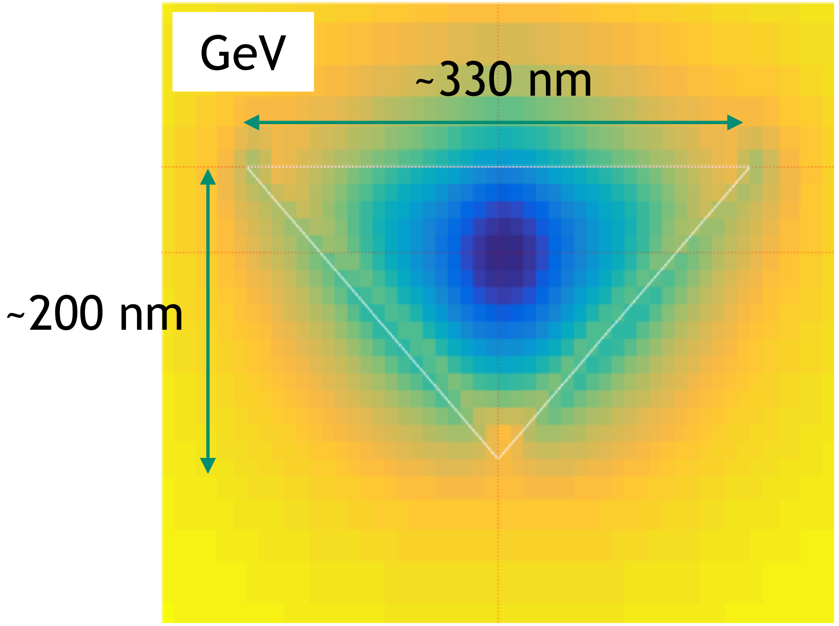
Coupling Diamond defect center to cavity



M. J. Burek et al., Phys. Rev. Applied ***8****, 024026 (2017)*

- Design the cavity around ion straggle
- Linear fall off in coupling strength

Cross-sectional energy profile of cavity mode



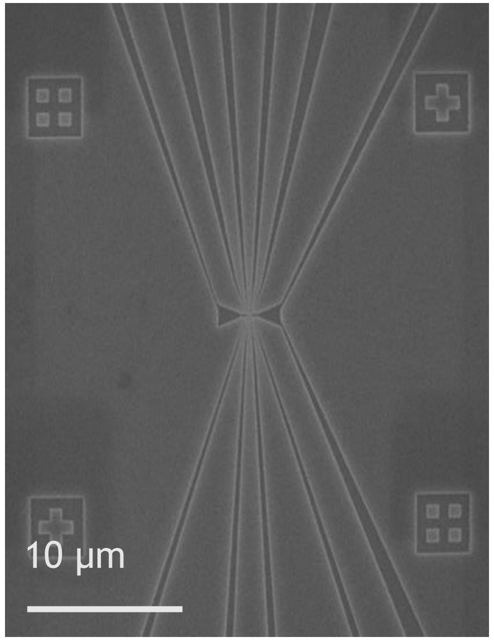
Center of mode is ~ 55 nm below the surface of the waveguide

M. Bhasker et al.,

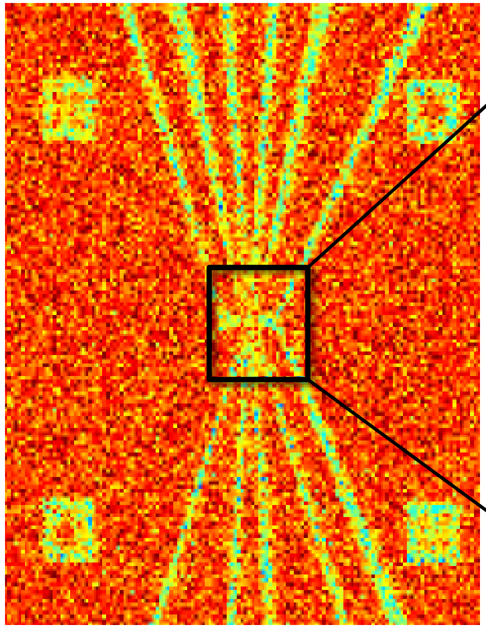
FIB Implantation? Lateral Resolution – OK, Depth Resolution – OK!

Targeting Resolution for FIB Implantation

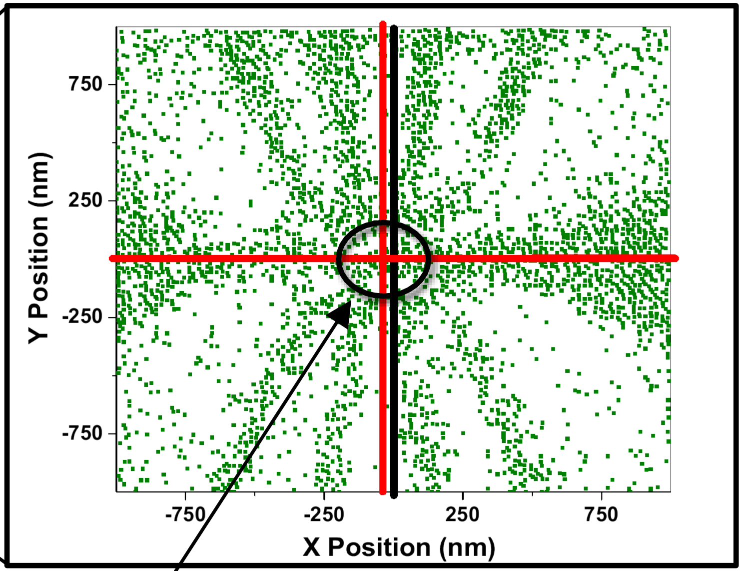
SEM of nanostructure



Ion Beam Induced Charge Collection (IBIC) of nanostructure

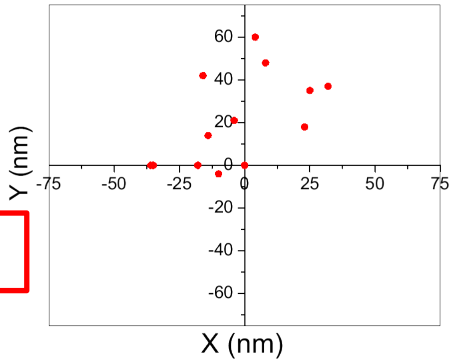


High resolution IBIC for targeting



Δx ~35 nm
Δy ~0 nm

Targeting Acuracy

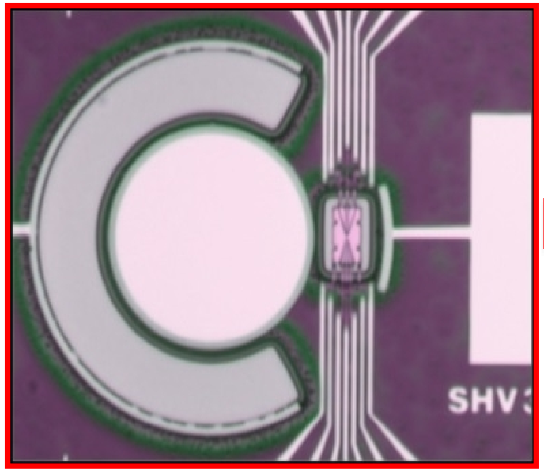


Targeting resolution <40 nm, new Velion should improve this number

How to address the yield question?

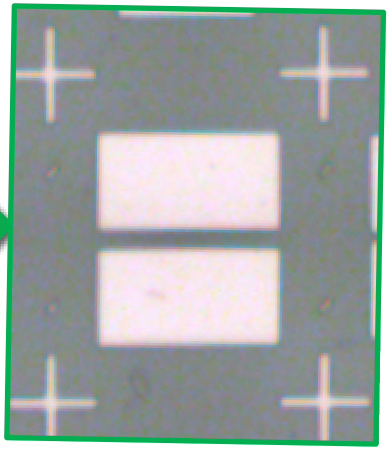
Our Approach is to use single ion detectors to better understand the yield

Silicon



Translating single ion detection to diamond

Diamond



J. Seamons, *et. al*. Appl. Phys. Lett, 93, 043124 (2008)

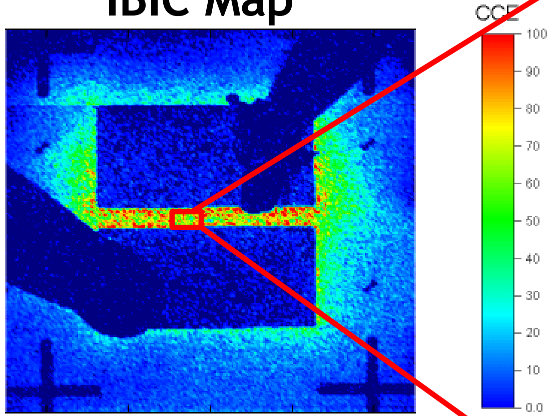
E. Bielejec, *et. al*. Nanotechnology 21, 085201 (2010)

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

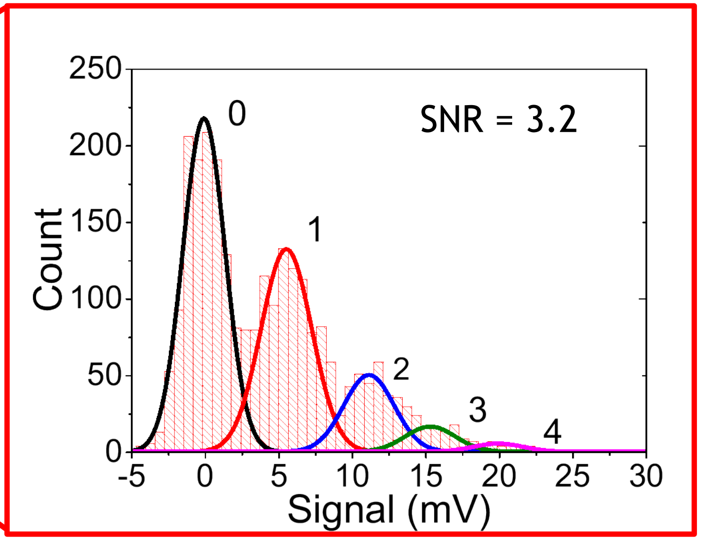
Allows us to improve understanding of the yield by directly counting the number of implanted ions

Single Ion Diamond Detectors

IBIC Map

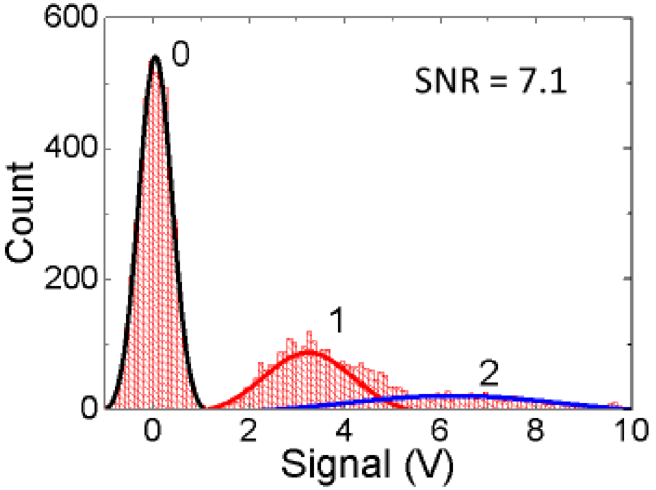


Quantized Detection <0.65> ion



Optimizing gain for single ion detection

Single Ion Counting <0.2> ion



$$SNR = \frac{\mu_{signal}}{\mu_{noise} + \sigma_{noise}}$$

Signal amplitudes match Poisson statistics to 4%

Single Ion Detection in Diamond with high SNR

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

Failure Modes for Single Ion Implantation

Three failure modes:

(1) ***Implant Multiple Ions***

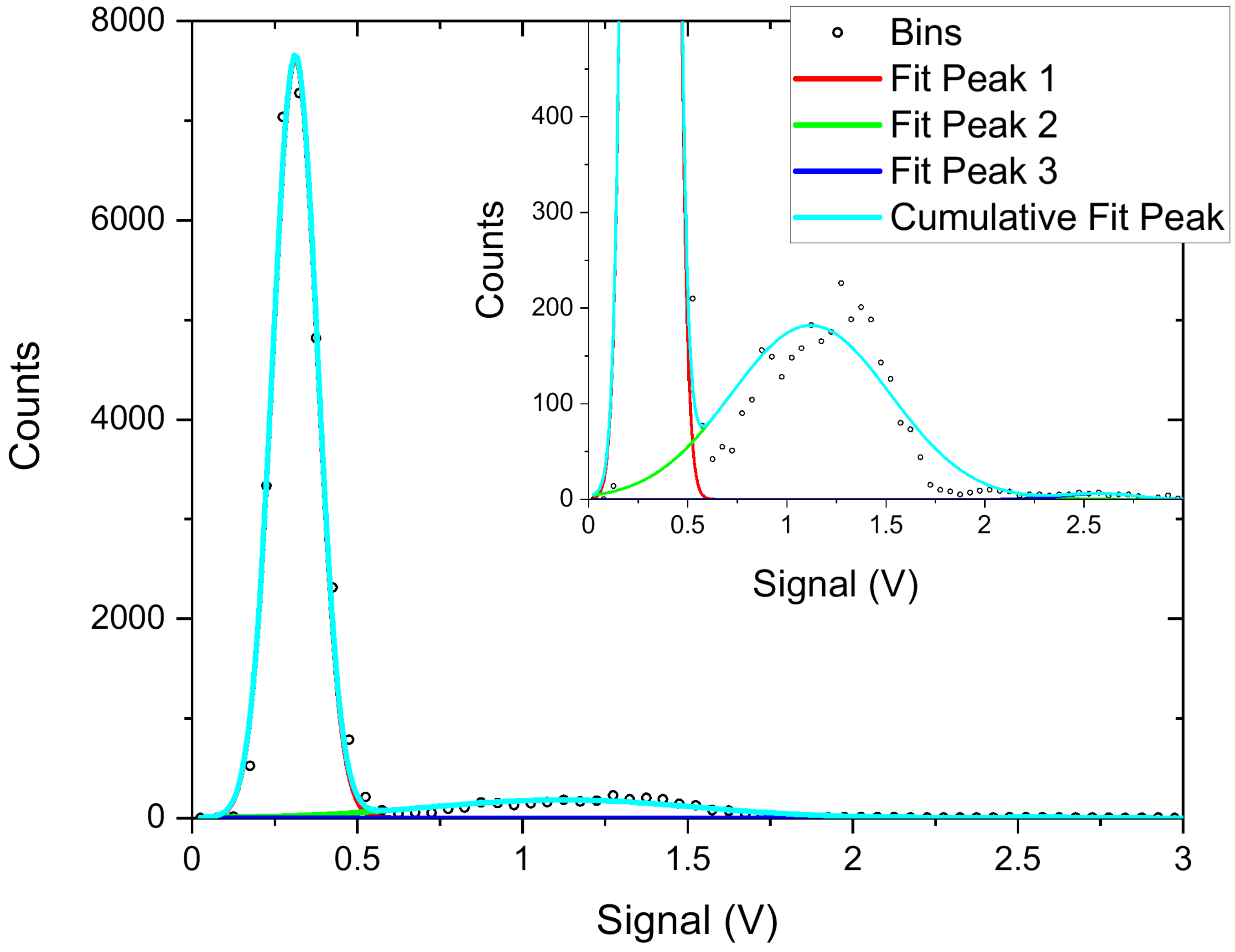
Implant >1, but count as 1

(2) ***False Positive***

Implant 0 ions, but count as 1

(3) ***False Negative***

Implant 1 ion, but count as 0



Failure Mode #1: Implanting Multiple Ions

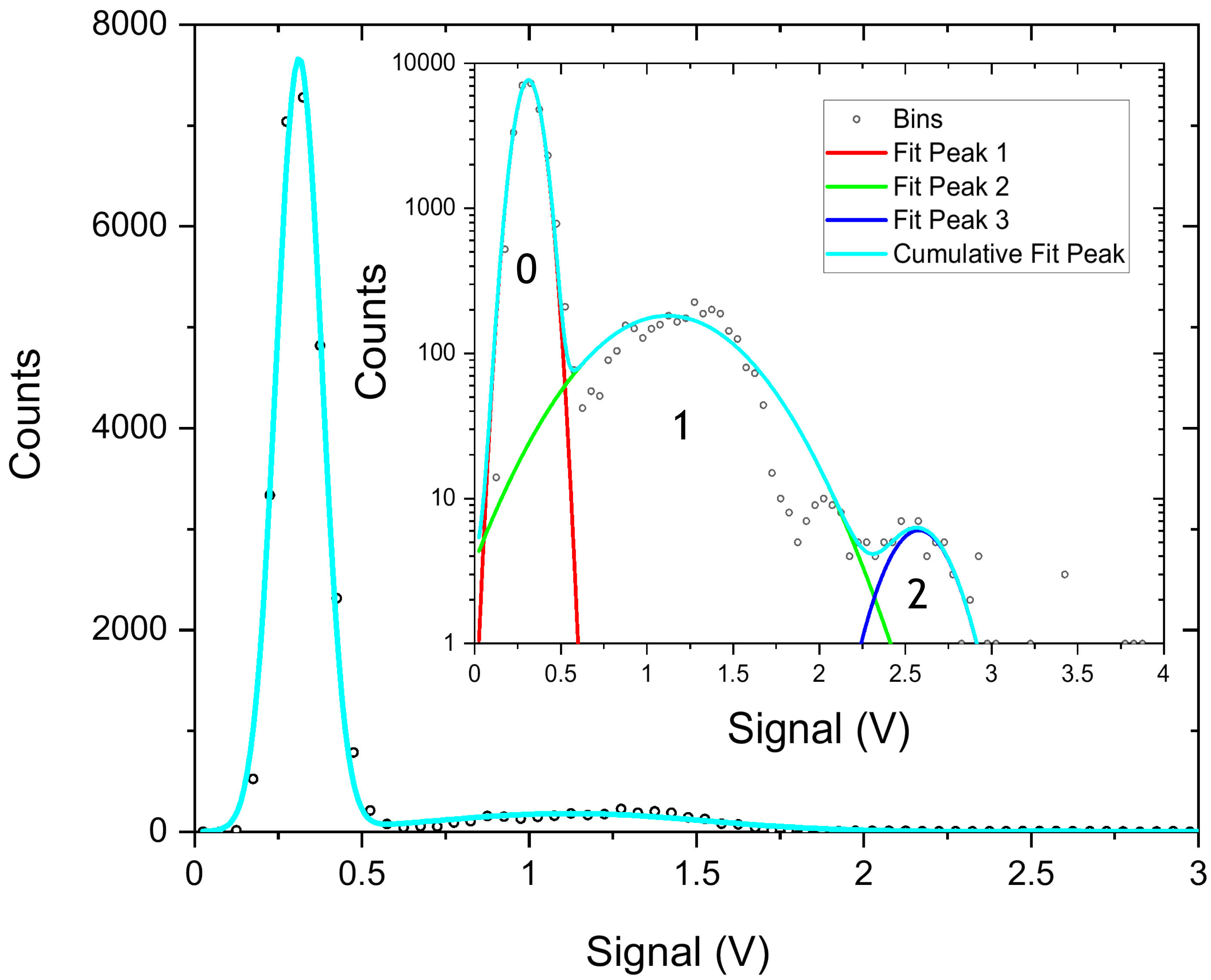
(1) ***Implant Multiple Ions***

Implant >1, but count as 1

Ion implantation follows a Poisson distribution

<0.1> ions/pulse: 90% 0's, 9% 1's and 0.45% 2 or more

Effectively 5% error in # of ions, for example count in 20 singles and get 21 ions



Failure Mode #2: False Positives

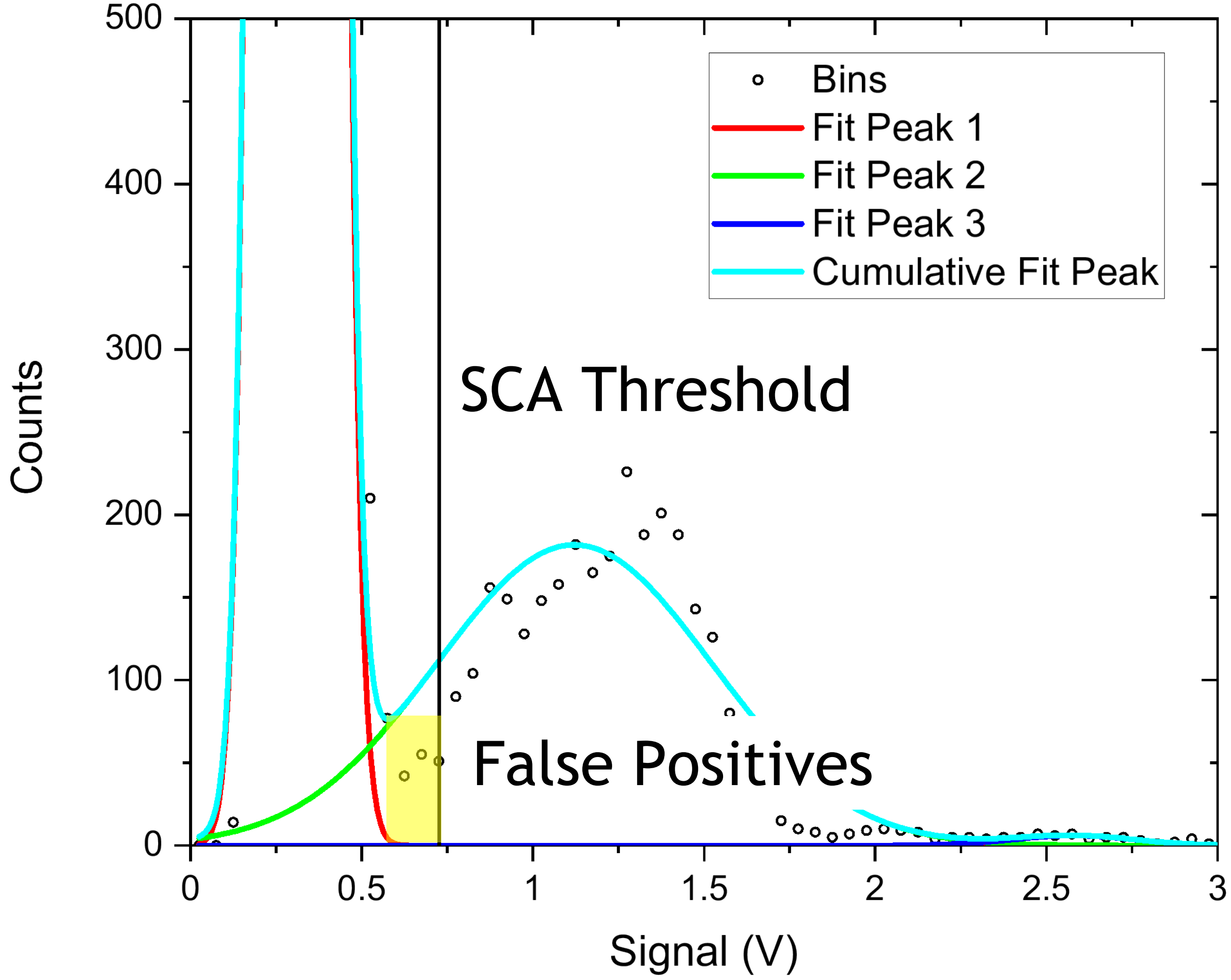
(2) ***False Positive***

Implant 0 ions, but count as 1

In case on right, we have a SNR of 4.7 between 0 and 1

We adjust the SCA threshold to avoid false positives

Leads to practically no (~1 ppb) false positives



Failure Mode #3: False Negatives

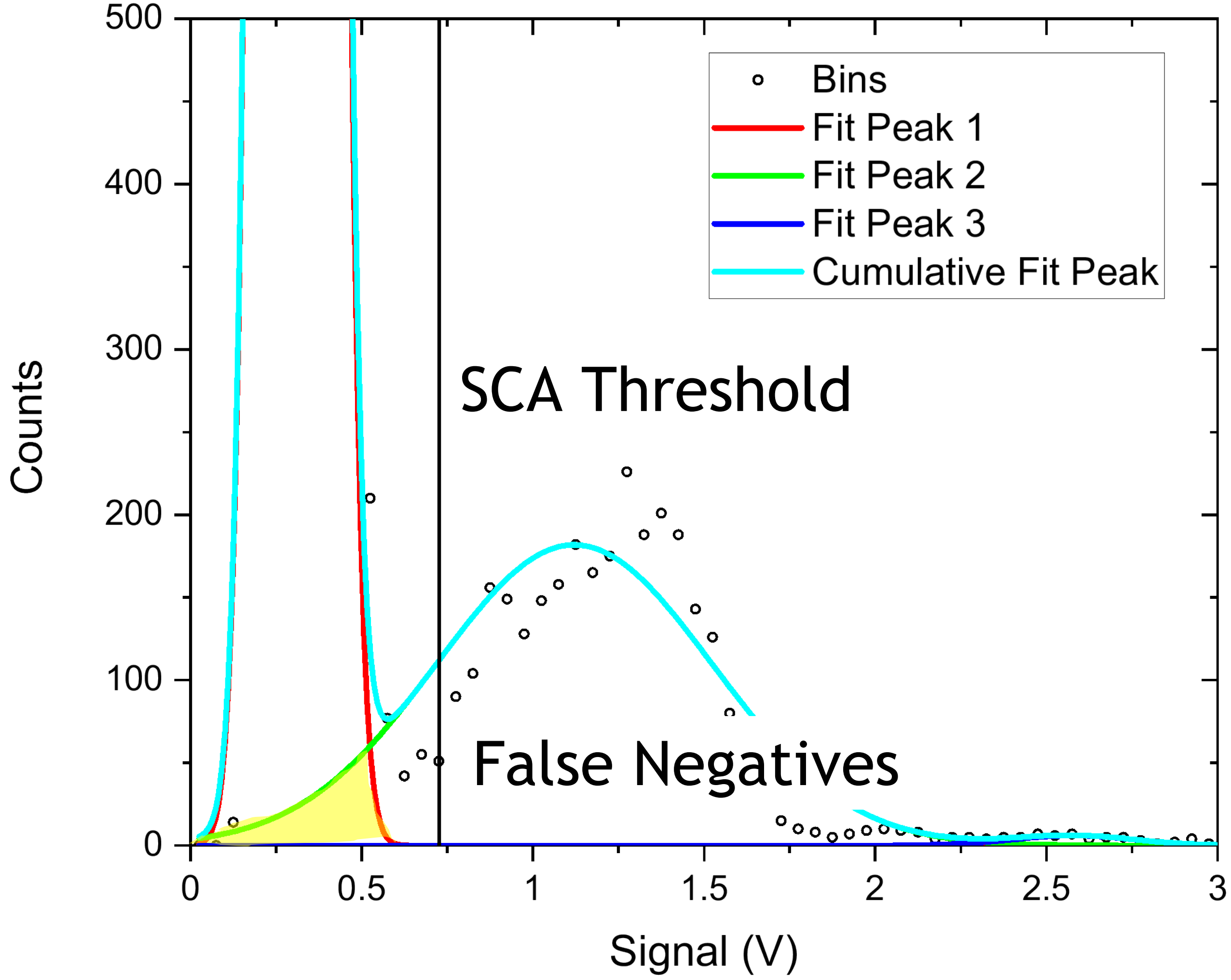
(3) ***False Negative***

Implant 1 ion, but count as 0

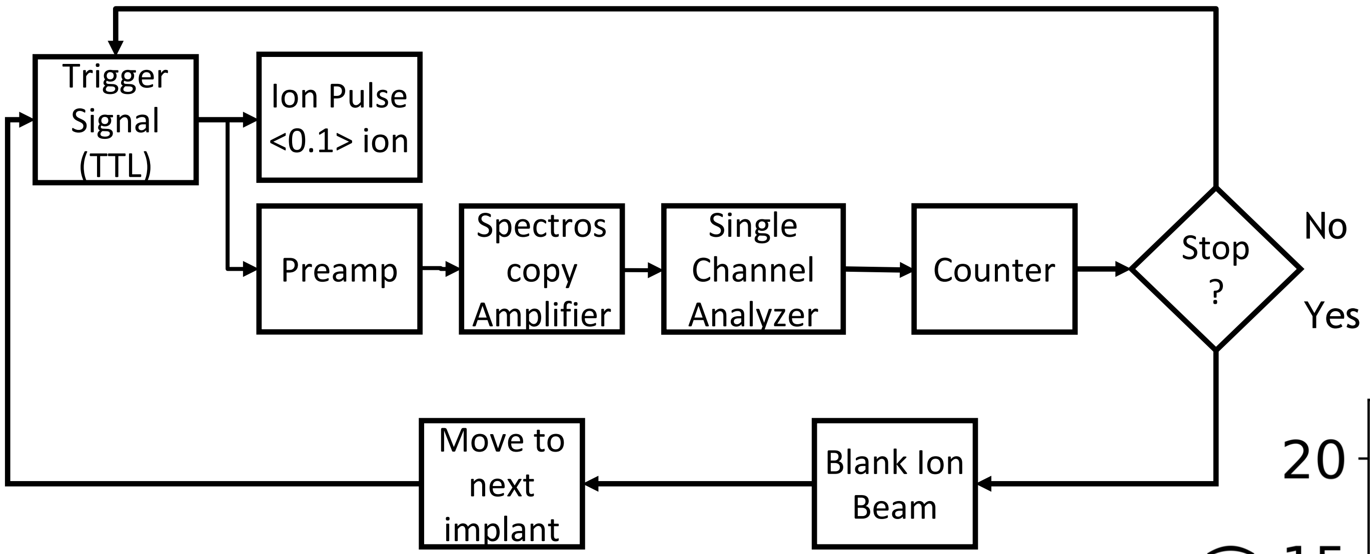
Adjust the SCA threshold to avoid false positives

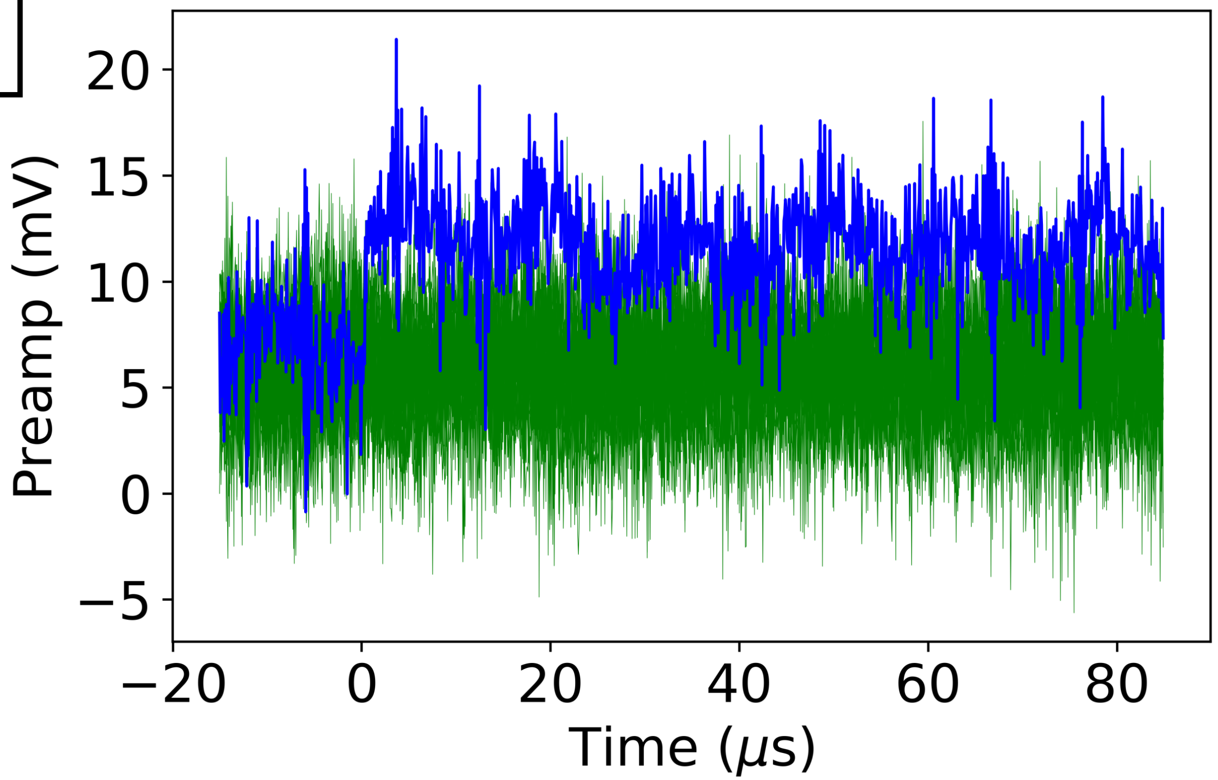
SCA optimization leads to ~10% false negatives

→ Largest source of error in our process



In-situ Counting Experiment

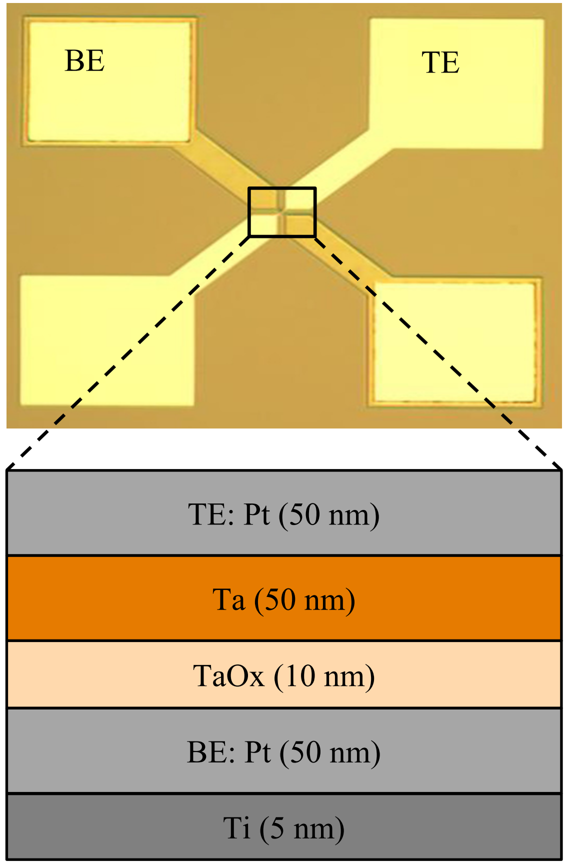


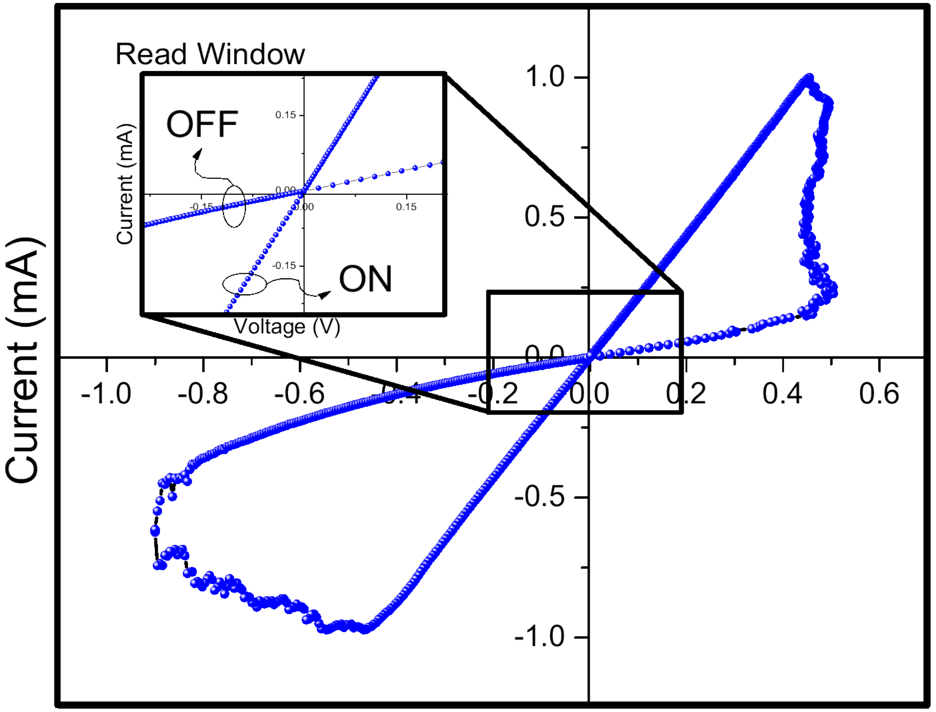


Fully automated process, but still very difficult and slow!!!!

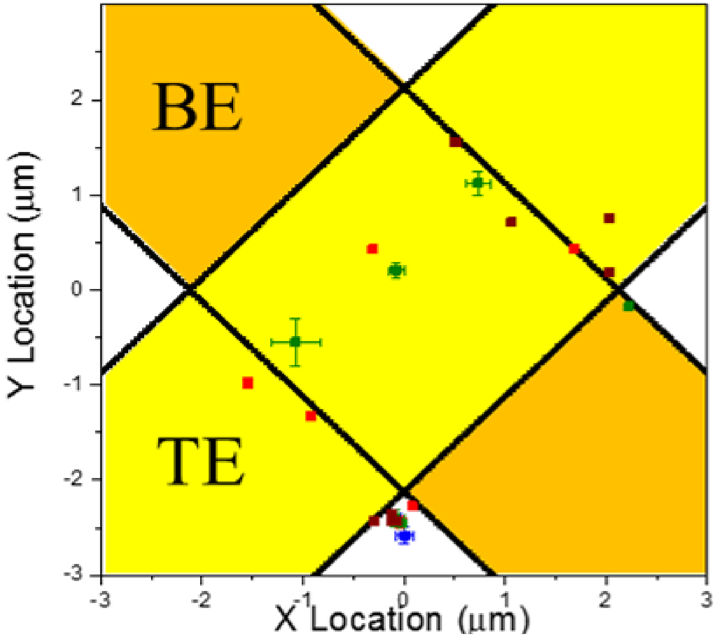
Example – Ion Beam Seeding of Memristors

- Memristor based memories are promising solid-state memories with high speed, low power, high density and are naturally radiation hardened



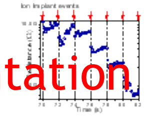


Filament Formation

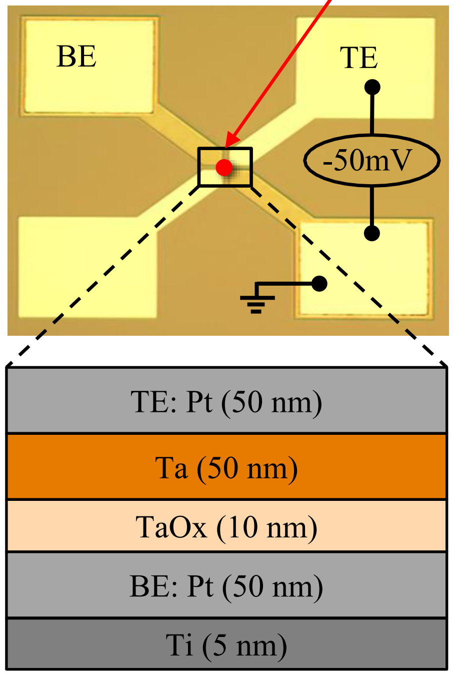


- Can we use focused ion beam implantation to act as seeding sites for the conductive filament formation bypassing electroformation?

Modification of the TaOx Film Resistance using Ion Irradiation

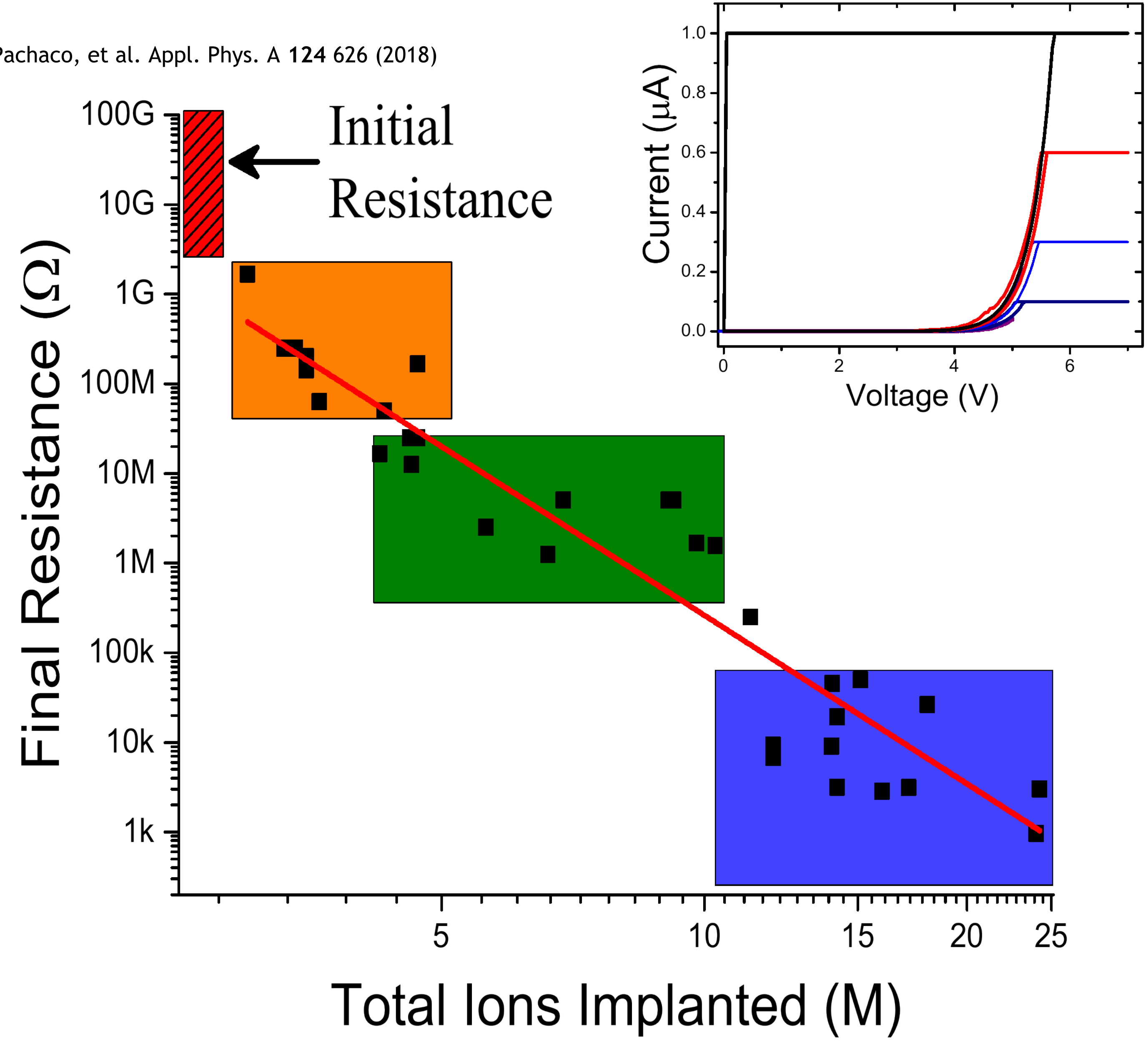


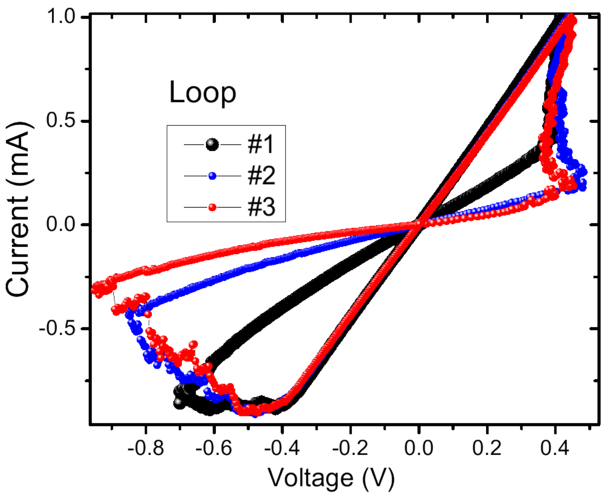
200 keV Si Implantation

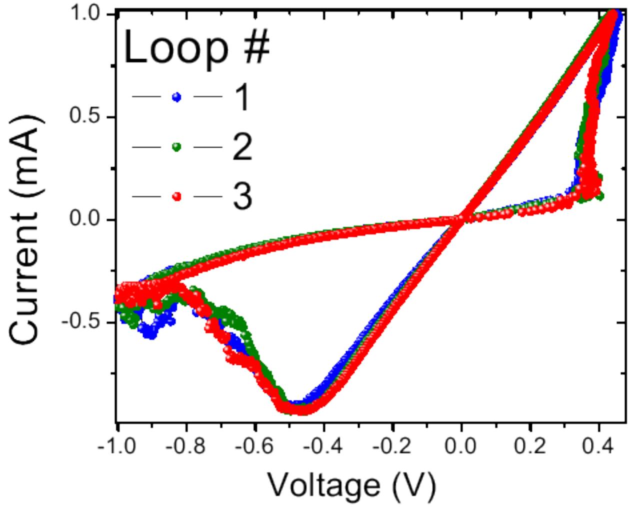


Ion Beam Seeding of Memristor Filament

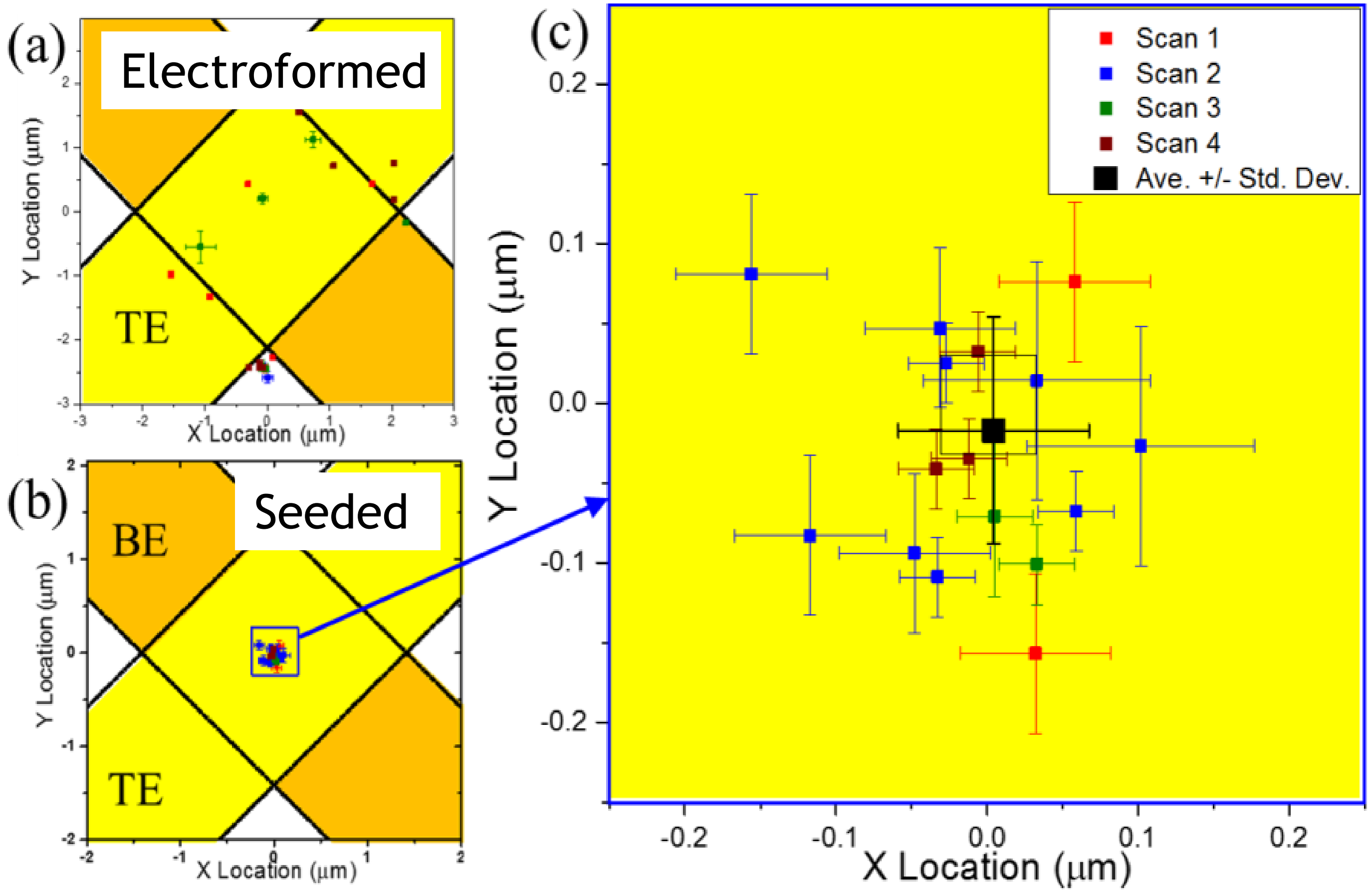
J. Pachaco, et al. Appl. Phys. A **124** 626 (2018)







Control over Filament Location



Results:

- Control over filament location and resistivity
- No Electroforming is necessary

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

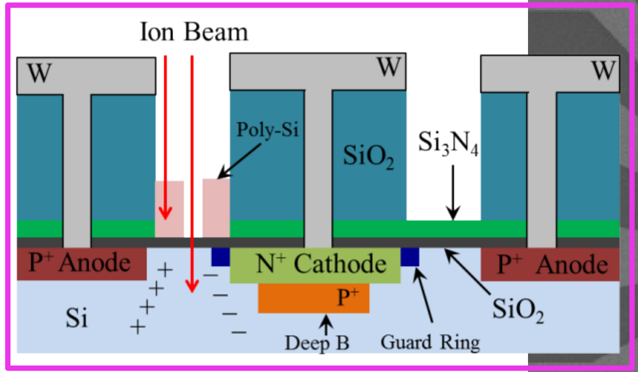
Single Donor Devices for Si Qubits

Si-MOS platform integrated with:

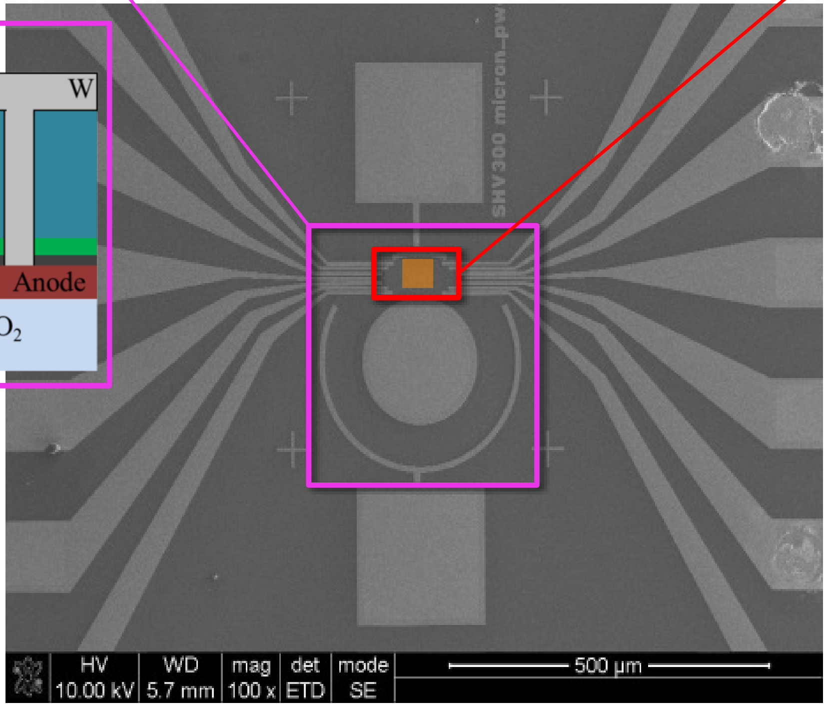
- Single Ion Detector
- Nanostructure for Qubit operation

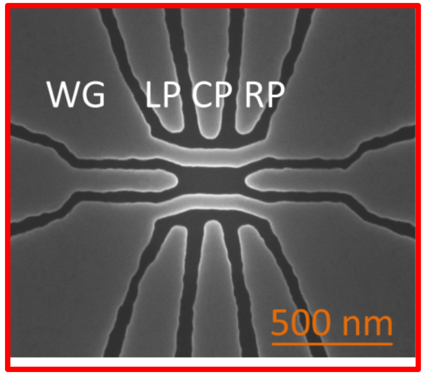
Single Ion Detector

Nanostructure

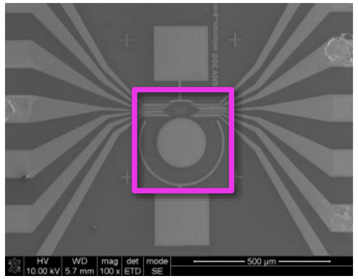


Ion Beam Induced Charge (IBIC)



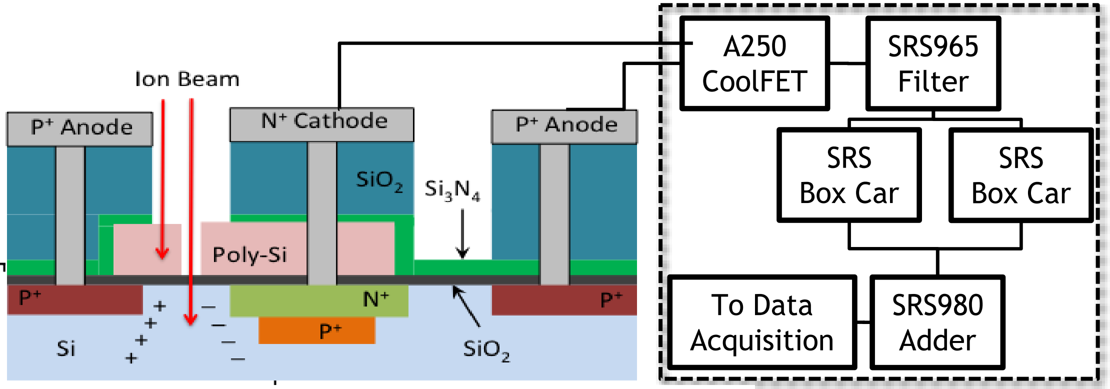


Combined Single Ion Detector and Nanostructures Demonstrated

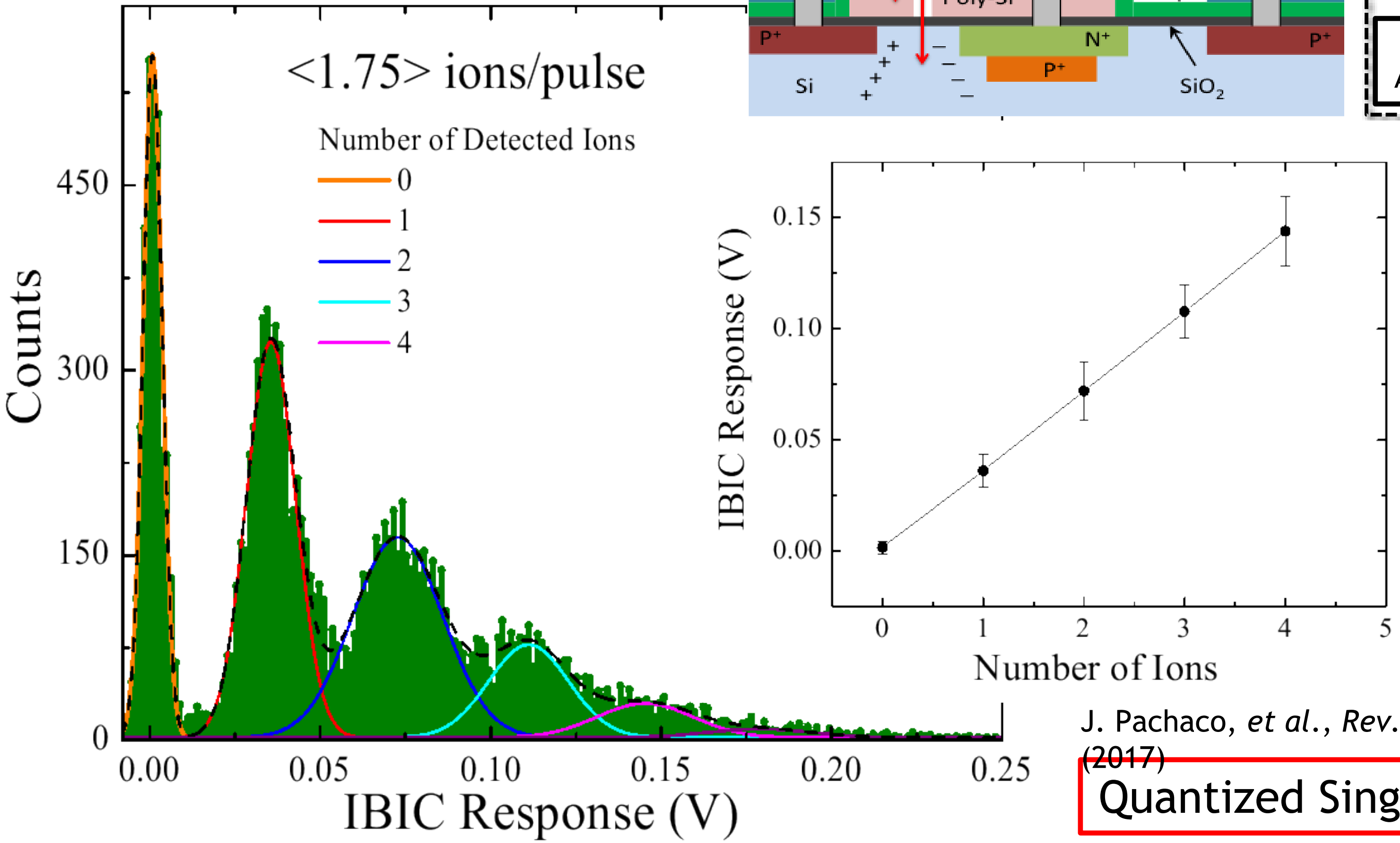


Integrated Single Ion Detectors

Ion Beam Induced Charge Collection (IBIC)

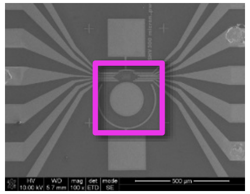


200 keV Si Detection



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

Quantized Single Ion Detection



Lowest SNL Detected Single Ion Implantation

Ion	Energy (keV)	SiO_2 thickness (nm)	Range[a] (±straggle) (nm)	e-h pairs (k)[a]	SNR	Error rate (%)	Detection efficiency (%)
Si	200	7	273 (±76)	39	21.2	≪1	100
Sb	120	35	25 (±17.5)	8.5	5.2	...	...
Sb	50	7	25 (±9)	5.0	4.4	...	...
Sb	20	7	11 (±5)	1.4	2.5	15	87.6

[a]Range and e-h pairs calculated from the SiO_2/Si interface into the Si substrate.

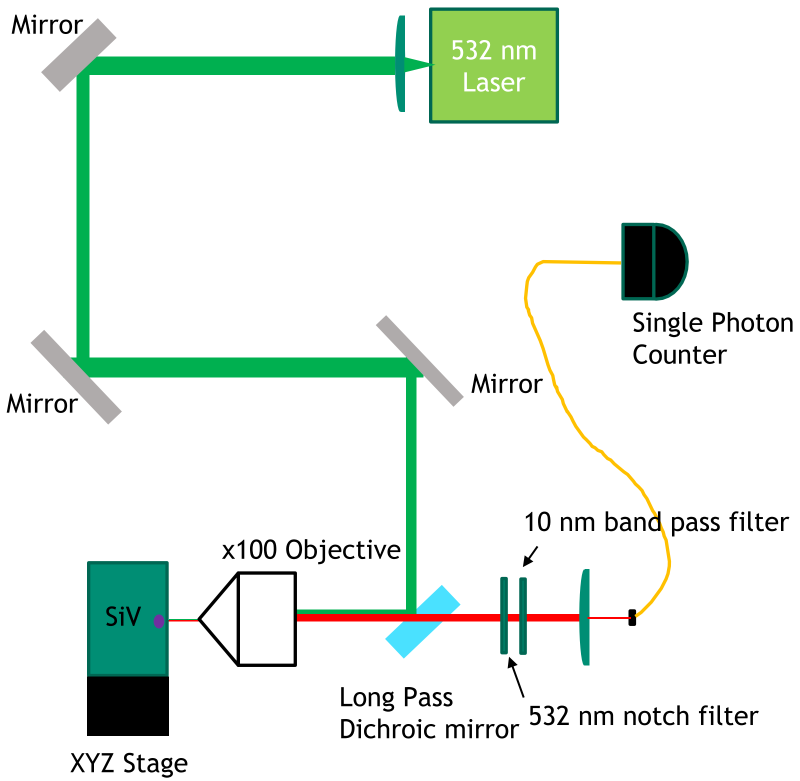
$$SNR = \frac{\mu_S}{\mu_N + \sigma_N} = 2.5$$

How to further improve SNR?

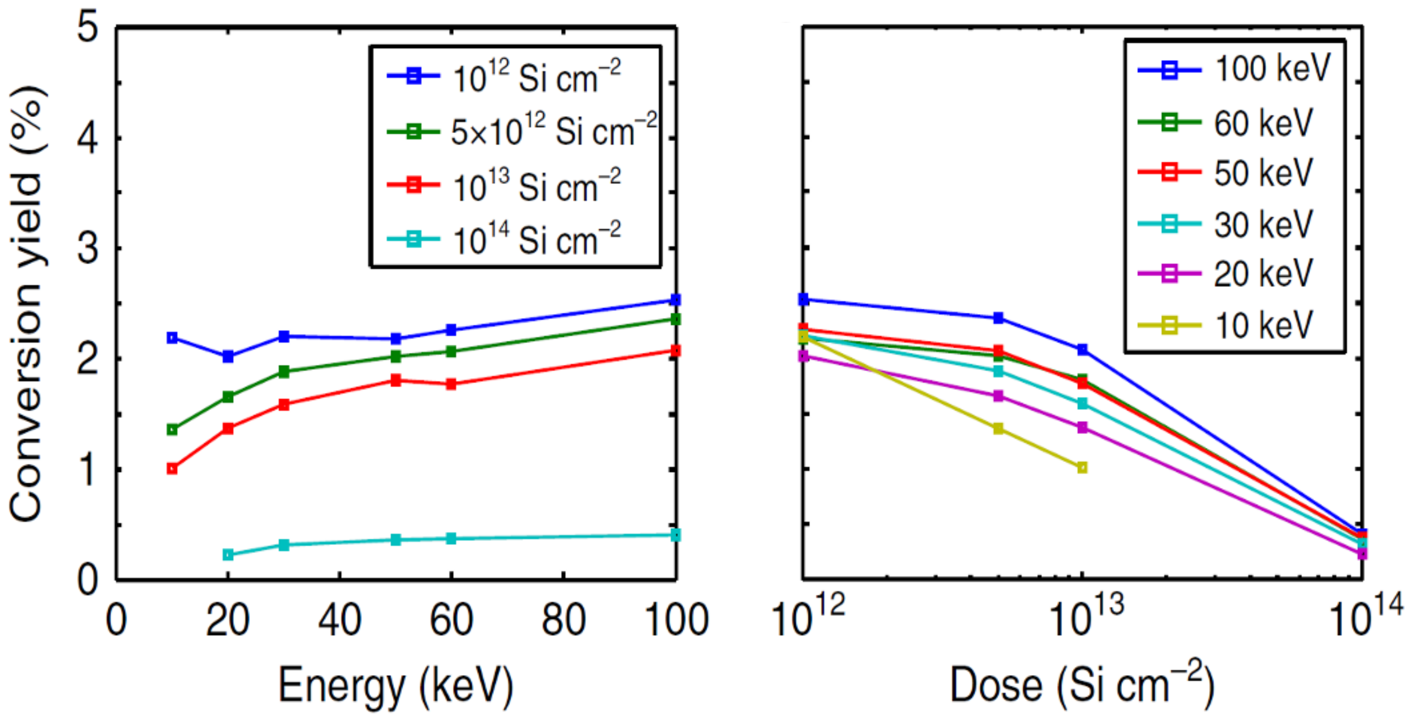
- In-vacuum preamp ☑
- Cool detector and FET ☑

Path forward to low energy <10 nm implantation and detector

Yield: Mapping SiV Photoluminescence



Yield = # measured SiV / # implanted Si



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

Similar yield to

S. Sangtawesin et. al. Appl. Phys. Lett. 105, 063107 (2014)

S. Tamura et. al. Appl. Phys. Express 7, 115201 (2014)

Yield is low and dominated by Poisson Statistics!

Yield Improvements Efforts

(1) Post electron irradiation

(2) Post ion irradiation

(3) Sequential ion irradiation

(1)–(3): Create excess vacancies

(4) Hot ion irradiation — Enable vacancy migration

B. Diller *et al.*, arXiv:1909.08778

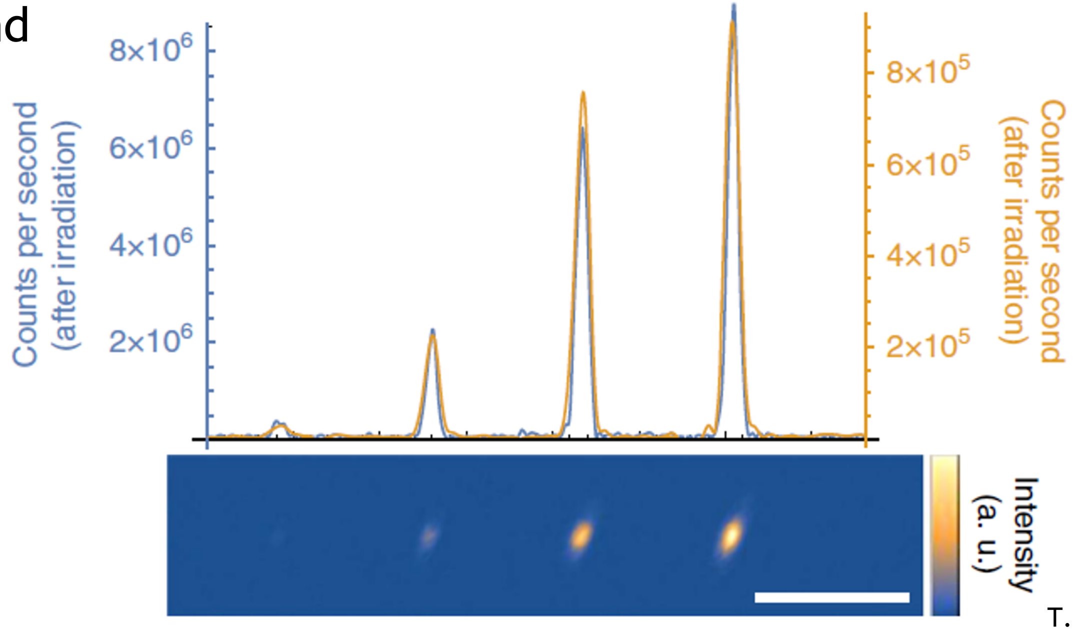
(5) Substrate doping — Enable preferential defect formation?

B. Rose *et al.*, Science **361**, 60-63 (2018)

T. Luhmann *et al.*, Nature Comm. **10**, 4956 (2019)

After electron irradiation and anneal

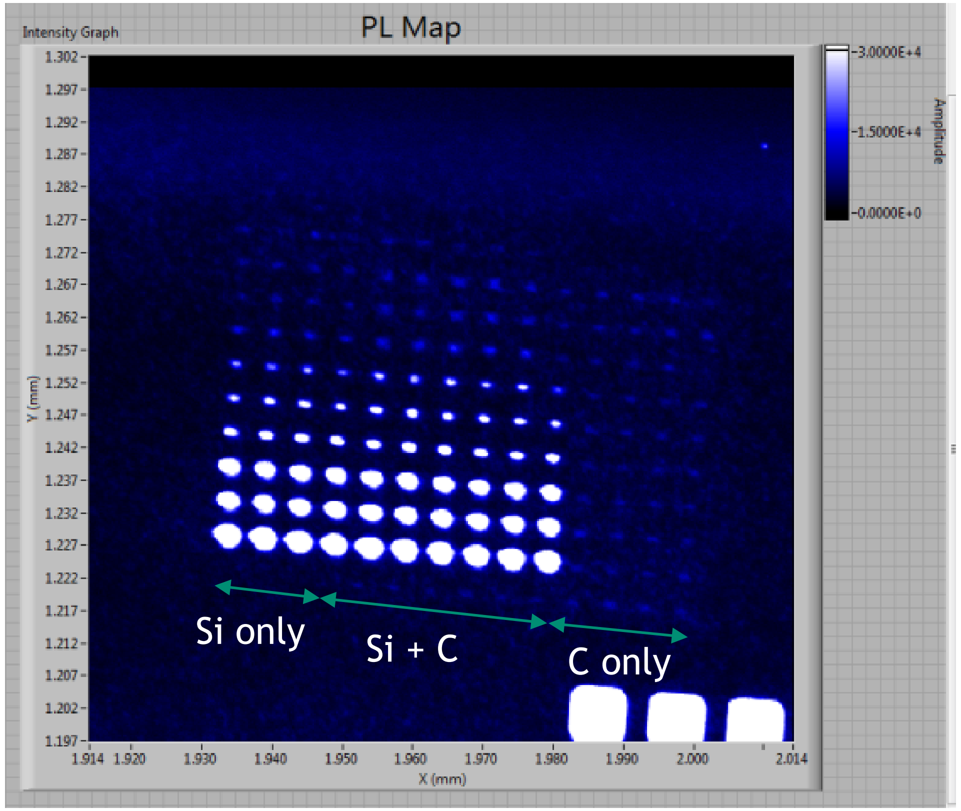
10x improvement

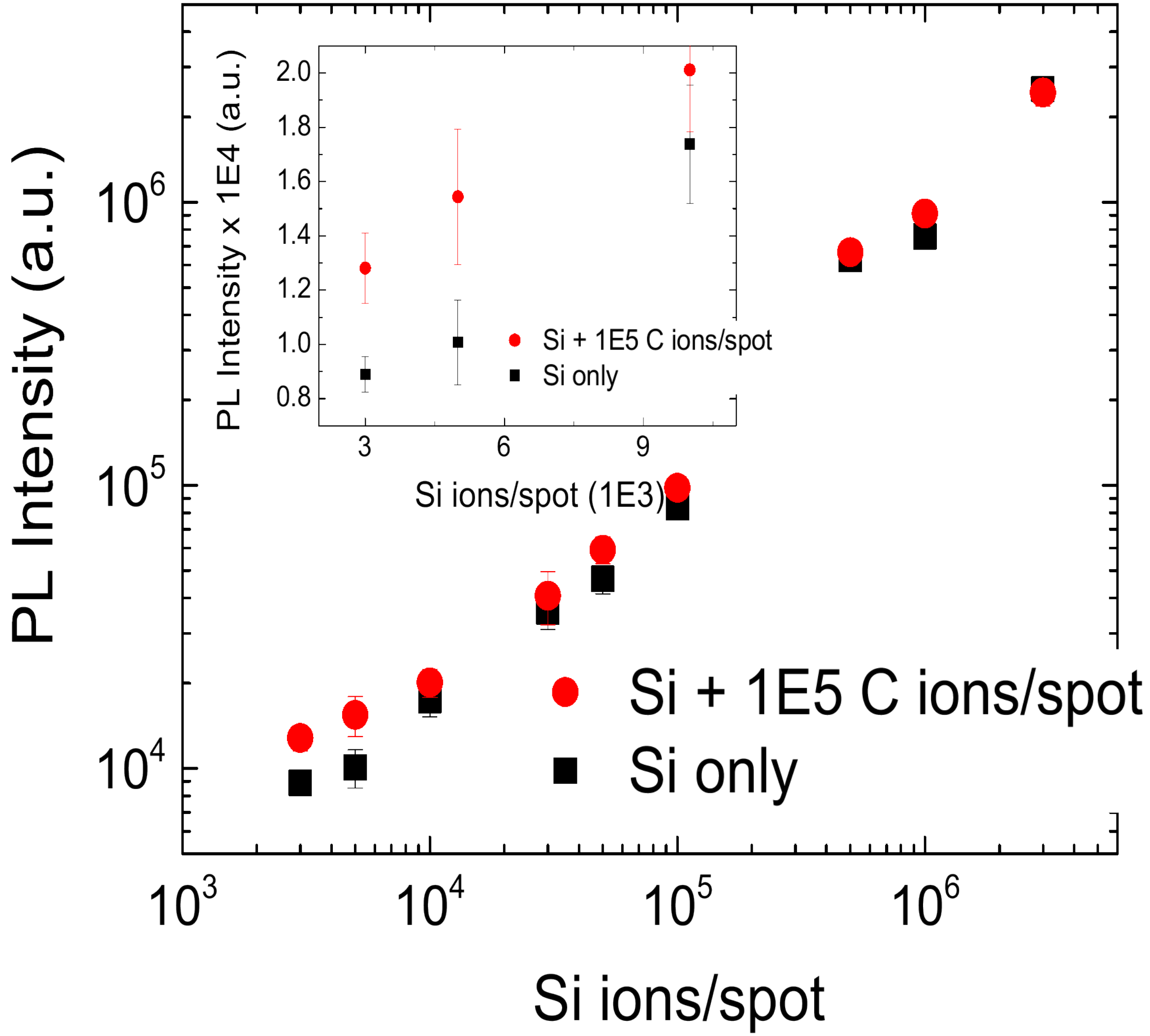


After ion implant and anneal

T. Schroder *et al.*, Nature Communications **8**, 15376 (2017)

Sequential Implantation – μm sized areas





Units

1E5 C ions/spot → ~1E13 ions/cm²

3E3 Si ions/spot → ~4E11 ions/cm²

→ Below the damage threshold

Si+C PL is ~1.4x larger than Si PL → Yield improvement observed in μm spots

Progress on Nitrogen Source Development

Approaches for building a nitrogen source for the nanoImplanter:

(1) Implant N into a known working source

(2) Use ionic liquid sources as demonstrated in the literature

Start with a working LMAIS – AuSn, In, AiSi, etc… and implant N

(1) Can we implant N into the LMAIS?

- N implanted into AuSn and In alloys at 200 keV N^+ to fluences of 5E17 ion/cm^2

(2) Does N stay in the alloy?

- N concentration measured by 28 MeV Si^{+5} ERD

- AuSn+N → N stays in place at least in the very near surface (figure on right)

- In+N → N concentration is less than calculated:
    - N diffused deeper into the In
    - Or it may have left the sample

(3) Can we form a N containing tip?

- On-going! But, at most we expect ~4e3 N/s