^{28}Si Enrichment for Quantum Computers Using Ion Implantation and Layer Exchange

Ella Schneider

PhD student
Surrey Ion Beam Centre
Advanced Technology Institute
University of Surrey

www.uknibc.co.uk/e.b.schneider@surrey.ac.uk

Contents

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^{28}Si – Based Quantum Computers

- Spin qubits hosted in silicon are promising quantum computer architectures due to their
  - long coherence times and high fidelity gates
  - compatibility with CMOS/classical electronics for industrial manufacturing
  - scalability prospects – best chance for scaling up to million/billions qubits required for a practical quantum computer to run error correction protocols

- A cryogenically-cooled, defect-free ^{28}Si crystal acts a 'semiconductor vacuum'
  - ideal noise-free environment for qubits

^{nat}Si		
^{28}Si	^{29}Si	^{30}Si
92.2%	4.7%	3.1%

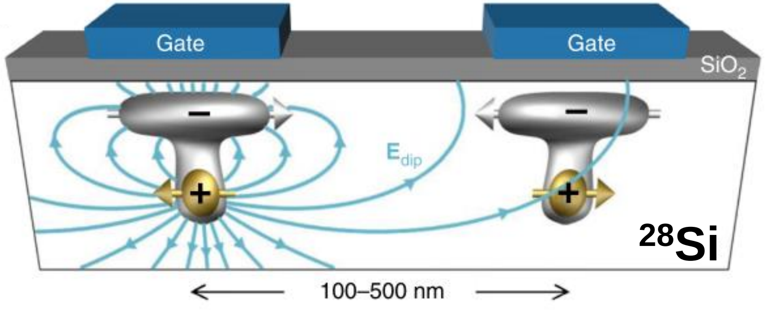
- ^{29}Si nuclear spin interacts with qubits degrading controllability and lifetime of states
- ^{30}Si should also be avoided as varying isotope-dependent bond lengths cause strain-induced magnetic fields which affects qubit controllability

- A readily available source of ^{28}Si is essential to quantum computer research and future production

^{28}Si – Based Quantum Computers (CMOS platforms)

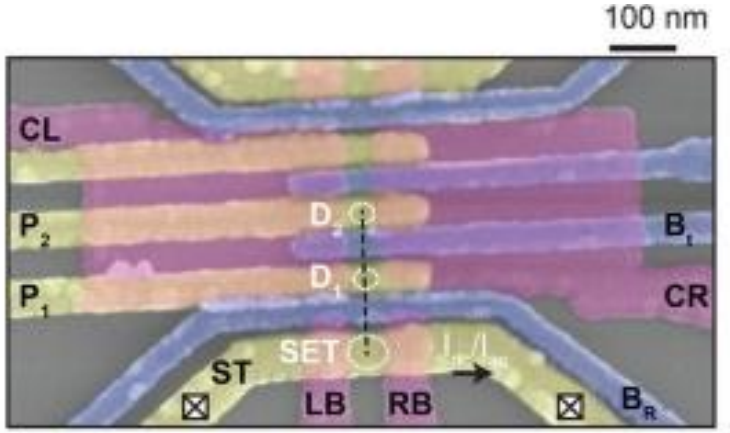
Architectures that use standard Si fabrication processes

UNSW’s Flip-Flop Qubit

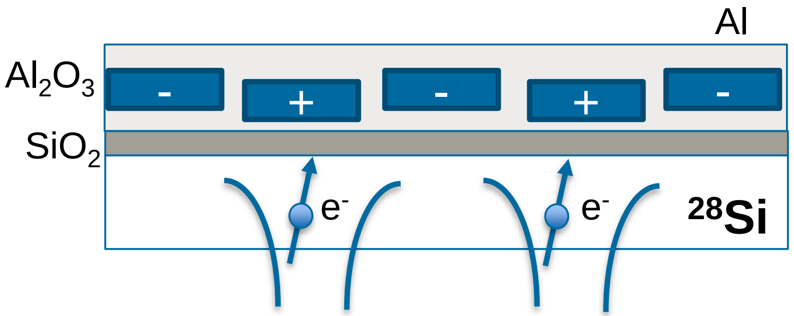


- P donor nucleus and valence electron hosted in ^{28}Si
- Gates on top control qubits
- Electric dipole interaction

Intel/Delft’s FinFET Quantum Dot Qubits



(Cross-section of dotted line)



- Electrode defined FinFET quantum dots ^{28}Si
- Potentials confine electrons that are used as qubits
- Barriers control qubit interactions

(Top) G.Tosi, et al. *Nat. Commun.* **8** 450 1–11 (2017)
(Bottom) H. G. J. Eenink, et al. *Nano Lett.* **12** 8653-8657 (2019)

Requirements of Quantum-Grade ^{28}Si

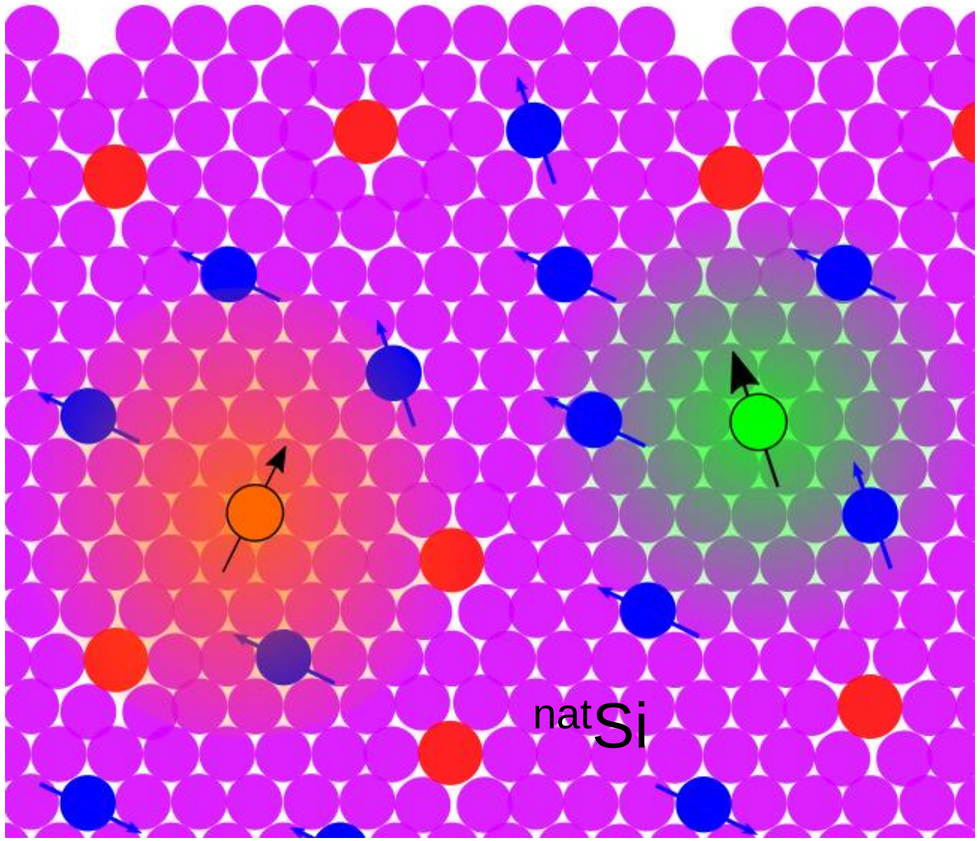
- Qubit coherence time > gate operation time
- Uniform qubit environment for reproducibility (controllability)

^{nat}Si

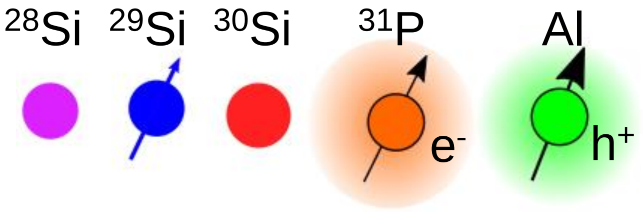
^{28}Si	^{29}Si	^{30}Si
92.2%	4.7%	3.1%

1. Enrichment	~99.9% ^{28}Si ~800ppm ^{29}Si[1]	• Remove enough ^{29}Si to eliminate noise created by uncontrolled nuclear spin interactions
2. Al content	<40 ppm Al Estimated from effective hole Bohr radius in Si	• Al hole can interact with qubit with more strength than the nuclear spin of ^{29}Si
3. Crystallinity	Perfect single crystal	• Any lattice defects are a source of decoherence - needs to be a uniform single crystal. ^{30}Si creates strain.
4. Contamination	<10 ppm C, N, O[2] Acceptable levels of CMOS wafers	• Presence inhibits epitaxial growth • Contaminants with spin will affect lifetime
5. Thickness	>50 nm [3]	• Spin atom/qubit needs to be away from noisy surface defects

Surface defects



Wavefunction radius/lattice constant to scale



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[1] K. M. Itoh and H. Watanabe, *MRS Commun*. **4**, 143 (2014)
[2] D Holmes, et al. *Phys. Rev. Materials* **5**, 014601 (2021)
[3] G. Tosi, et al. *Nat. Commun.* **8** 450 1–11 (2017)

^{28}Si Enrichment Techniques

Fundamental ^{28}Si separation technique	Method	Project/Institution	Achieved enrichment
Laser excitation	➢ Laser isotope separation. Selective ionisation of gases by means of precisely tuned lasers	SILEX, Silicon Quantum Computing Pty Ltd (SQC) and UNSW (Sydney)	-
Centrifugation of SiF$_4$	➢ Bulk Czochralski, chemical vapour deposition (CVD), float-zone crystallisation	Avogadro Project [1]	99.9993%
	➢ Plasma enhanced CVD with electron cyclotron resonance discharge	Institute of Chemistry of High-Purity Substances, Russia [3]	99.9986%
	➢ CVD	Isonics [4] Princeton University, USA [5]	99.924% 99.89%
	➢ Molecular beam epitaxy (also Ge enrichment)	Technical University of Munich, Germany[6]	99.9%
Ion beam magnetic filtering	➢ Hyperthermal ion beam deposition	National Institute of Standards and Technology (NIST), USA [6]	99.99987%
	➢ Small area (negative) ion implantation	University of Melbourne and UNSW [7]	99.97%
	➢ Layer exchange (conventional implanters or SIMPLE)	Surrey University Ion Beam Centre (IBC) [8]	99.7%
	➢ Conventional ion implantation	Surrey University IBC [9]	99.6%

Highest

↑

Lowest

Highest

↑

Lowest

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1 N. V. Abrosimov, et al., Metrologia, vol. 54, no. 4, pp. 599-609, 2017.

2 P. Becker, et al., Physica Status Solidi (A) Applications and Materials Science, vol. 207, no. 1, pp. 49-66, 2010.

3 J.Y. Li, C.T. Huang, L.P. Rokhinson, and J.C. Sturm, Appl. Phys. Lett. 103, 162105 (2013)

4 K. Itoh and H. Watanabe, MRS Communications, vol. 4, no. 2, pp. 143-157, 2014.

5 J. Sailer, et al., Phys. Status Solidi – Rapid Res. Lett. 3, 61 (2009)

6 K Tang et al 2020 J. Phys. Commun. 4 035006

7 D. Holmes et al. Phys. Rev. Materials 5, 014601 (2021) .

8 J. England, D. Cox, N. Cassidy, B. Mirkhaydarov, and A. Perez-Fadon, *Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms*, vol. 461, no. September, pp. 30–36, 2019, doi: 10.1016/j.nimb.2019.09.013

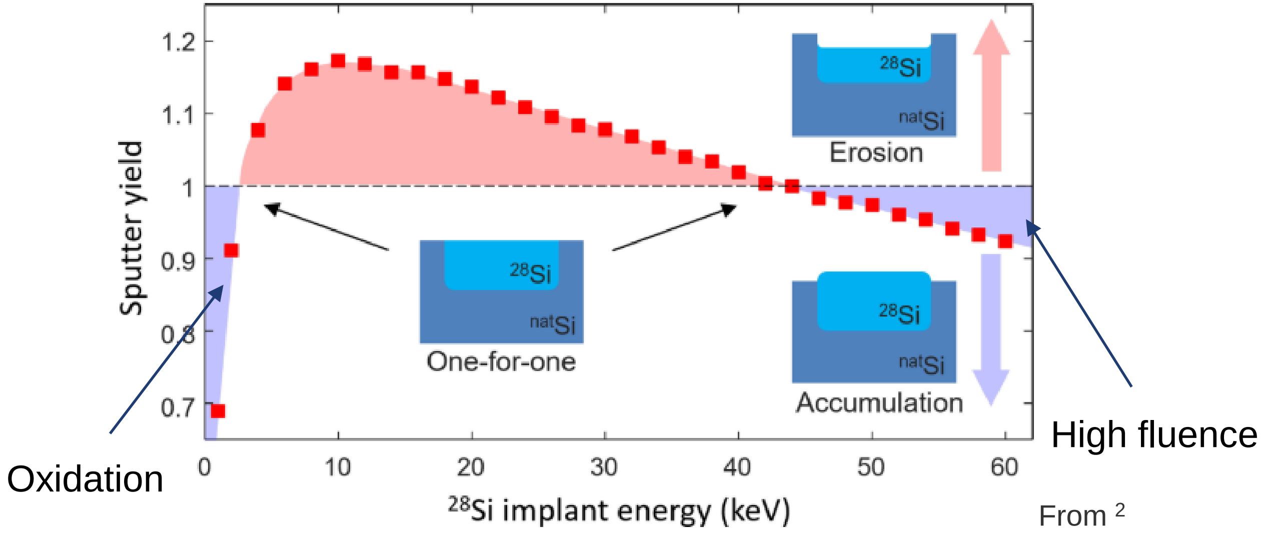
9 Ella B Schneider *et al* 2021 *J. Phys. D: Appl. Phys.* **54** 355105

Direct ^{28}Si Implantation into ^{nat}Si Substrates for Enrichment

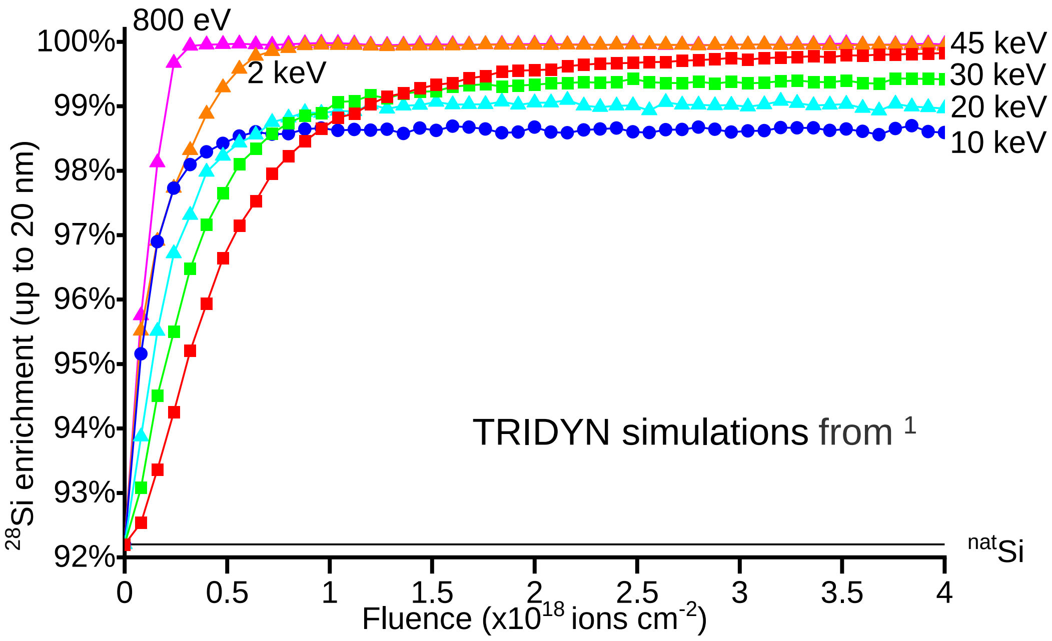
- During implantation, ^{28}Si concentration in ^{nat}Si builds up
  - Aim is to reach ~100% ^{28}Si
- Depending on the energy, ^{28}Si ions will either sputter away the implanted ^{28}Si (erosion) or continue to build up (accumulation)

- Erosion – ^{28}Si enrichment limited
- Accumulation – can either be achieved with
  - Ultra low energies <3 keV (‘deposition’)
    - High enrichment in low fluences
    - Very high oxidation in conventional implanters[1]
  - Low energies >45 keV[2]
    - 100% enrichment can be achieved and UHV not required
    - Very high fluences required to deplete ^{29}Si and ^{30}Si

- Isobaric contamination (N_2, CO) implanted
  - Can be avoided with negative ions[2] – reduces beam current



From [2]



TRIDYN simulations from [1]

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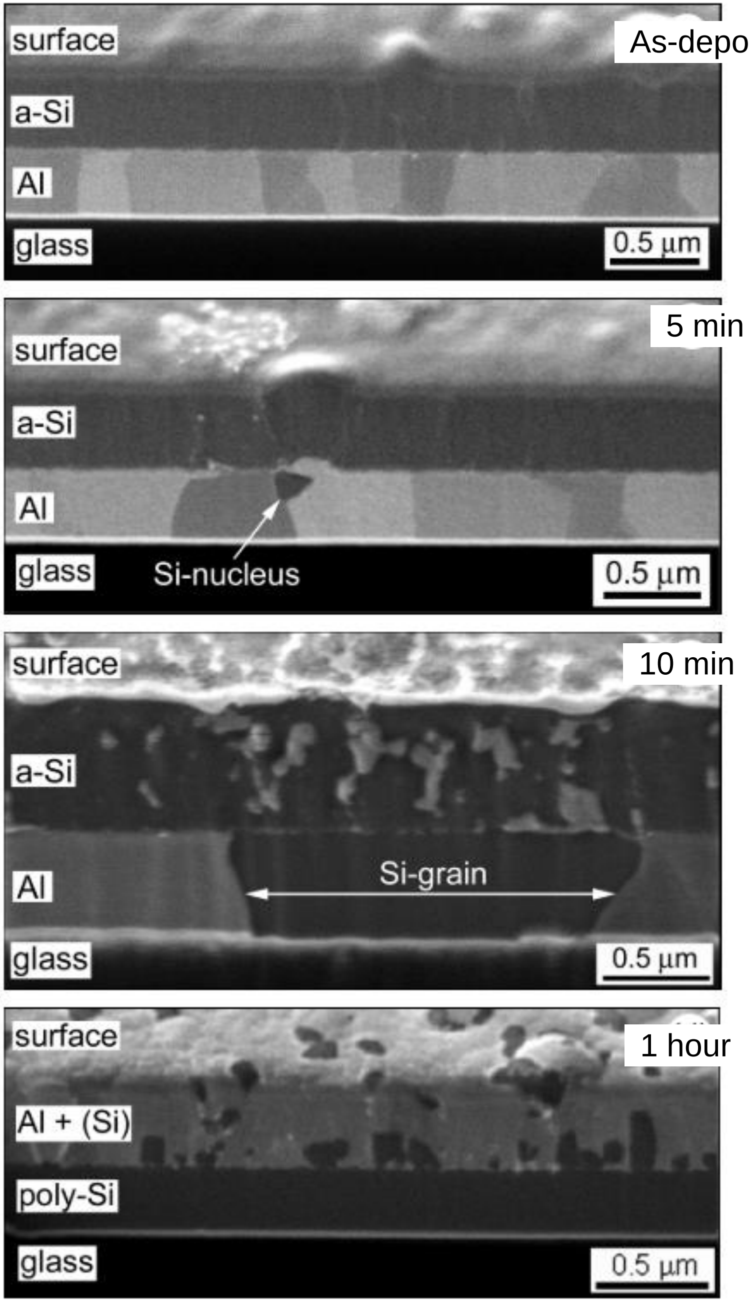
[1] Ella B Schneider *et al* 2021 *J. Phys. D: Appl. Phys.* **54** 355105

[2] D Holmes *et al* 2021 *Phys. Rev. Mater.* **5** 014601

Conventional Layer Exchange (Deposition Only)

- Commercial process used by CSG Solar[1]:
  1. Deposit Al onto glass
  2. Deposit a-Si onto Al
  3. Heat (~500°C / 1hr):
     1. a-Si dissolves into Al
     2. Si diffuses through Al
     3. Si precipitates out as c-Si
- Process driven because c-Si has lower E_{Gibbs} than a-Si
- Heterogeneous nucleation on Al grain boundaries
- Ostwald ripening produces continuous poly-Si layer
- Oxide interface between deposited Al and Si important

- Continuous c-Si layers have been grown onto Si wafers (Majni, Ottaviani 1977)



Tilted X-SEM of layer exchange on glass annealed at 500°C[2] >

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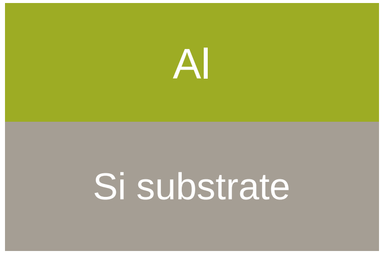
[1] Schneider, J and Evans, R, Proc. 21st EUPVSEC, Dresden (2006), 1032

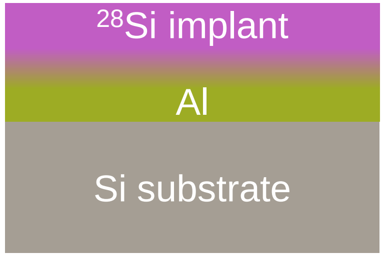
[2] Nast, O. "The aluminium-induced layer exchange forming polycrystalline silicon on glass for thin-film solar cells", Ph.D. thesis, University of Marburg (2000)

Layer Exchange with Ion Implantation for ^{28}Si Enrichment

1. Deposit Al onto native oxide-free Si substrate



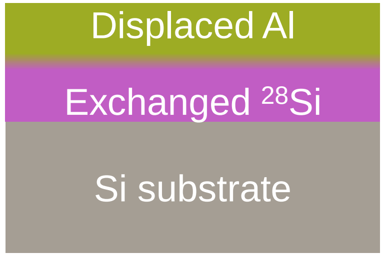
2. Implant ^{28}Si into Al



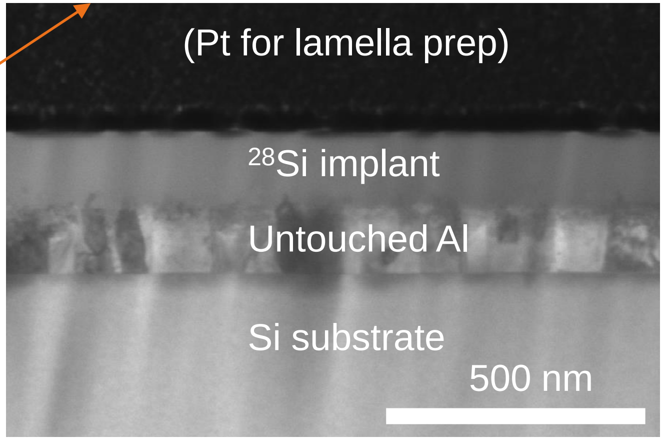
30keV/6.6E17cm^{-2} using IBC DF implanter

Implanted ^{28}Si diffuses into Al: epitaxially grows on substrate or nucleates at grain boundary

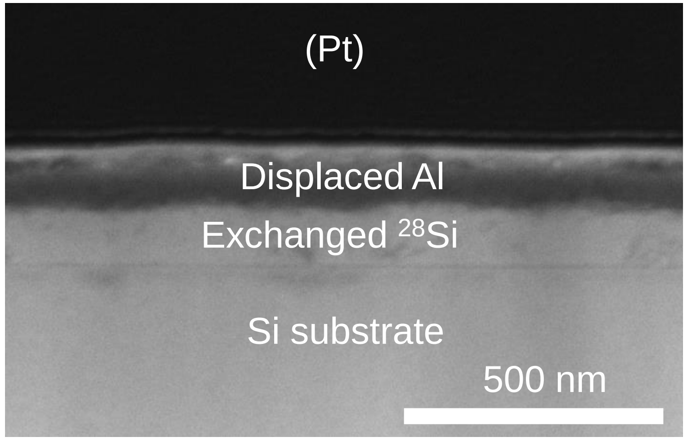
3. Anneal to initiate layer exchange



After 2x 30s anneals @ 500°C



10x lower fluence than direct implantation



Fabrication conditions
1. 250 nm deposited Al
2. ^{28}Si/30keV/6.6E17cm^{-2}
3. 2x 30s anneals @ 500°C in thermocouple RTA

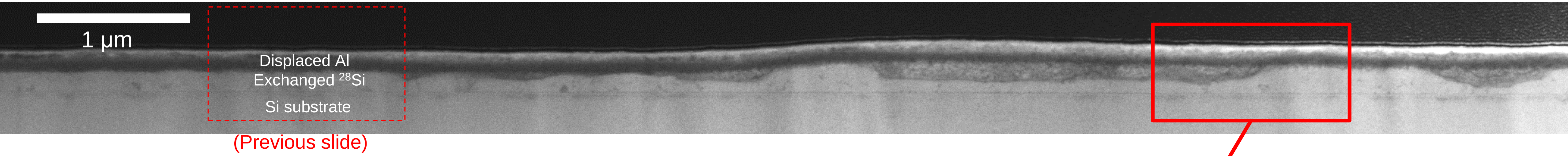
Bright field STEM

(Lamellas were prepared and imaged using the Surrey ATI Tescan FERA3 Plasma FIB. An FEI Ga FIB was used for lamella thinning)

Does potential energy remaining from implant process help drive layer exchange?

Implant Layer Exchange: Experimental Results

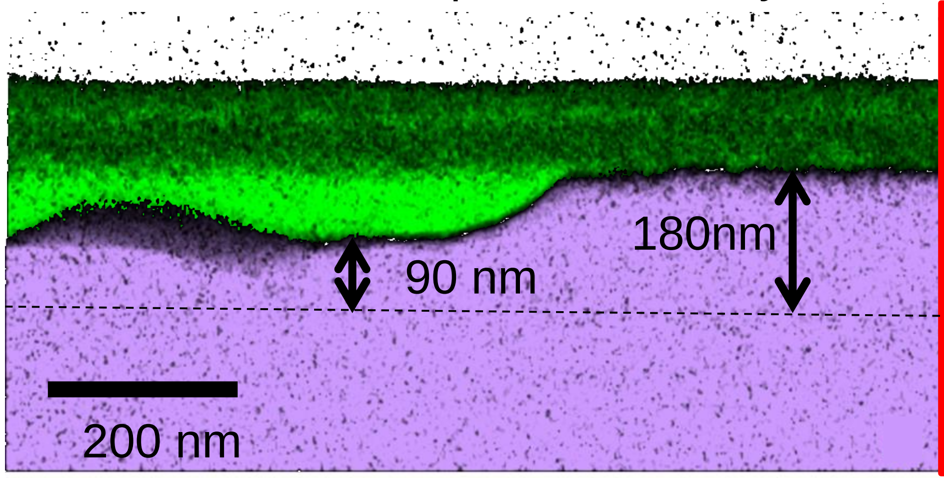
After 2x 30s anneals @ 500°C (across 10 µm)



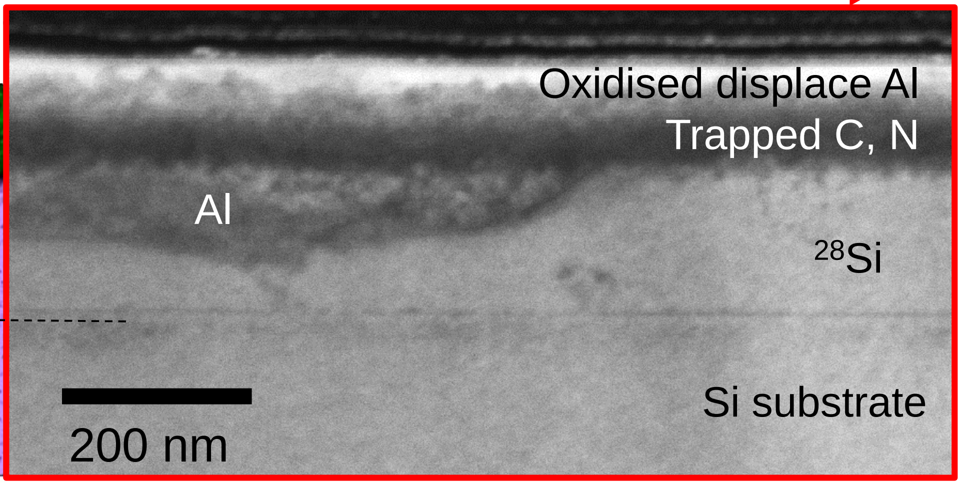
- Layer exchange occurring across large range but uneven across sample

Fabrication conditions
1. 250 nm deposited Al
2. ^{28}Si/30keV/6.6E17cm^{-2}
3. 2x 30s anneals @ 500°C in thermocouple RTA

EDX false-colour map, Si & Al X-rays

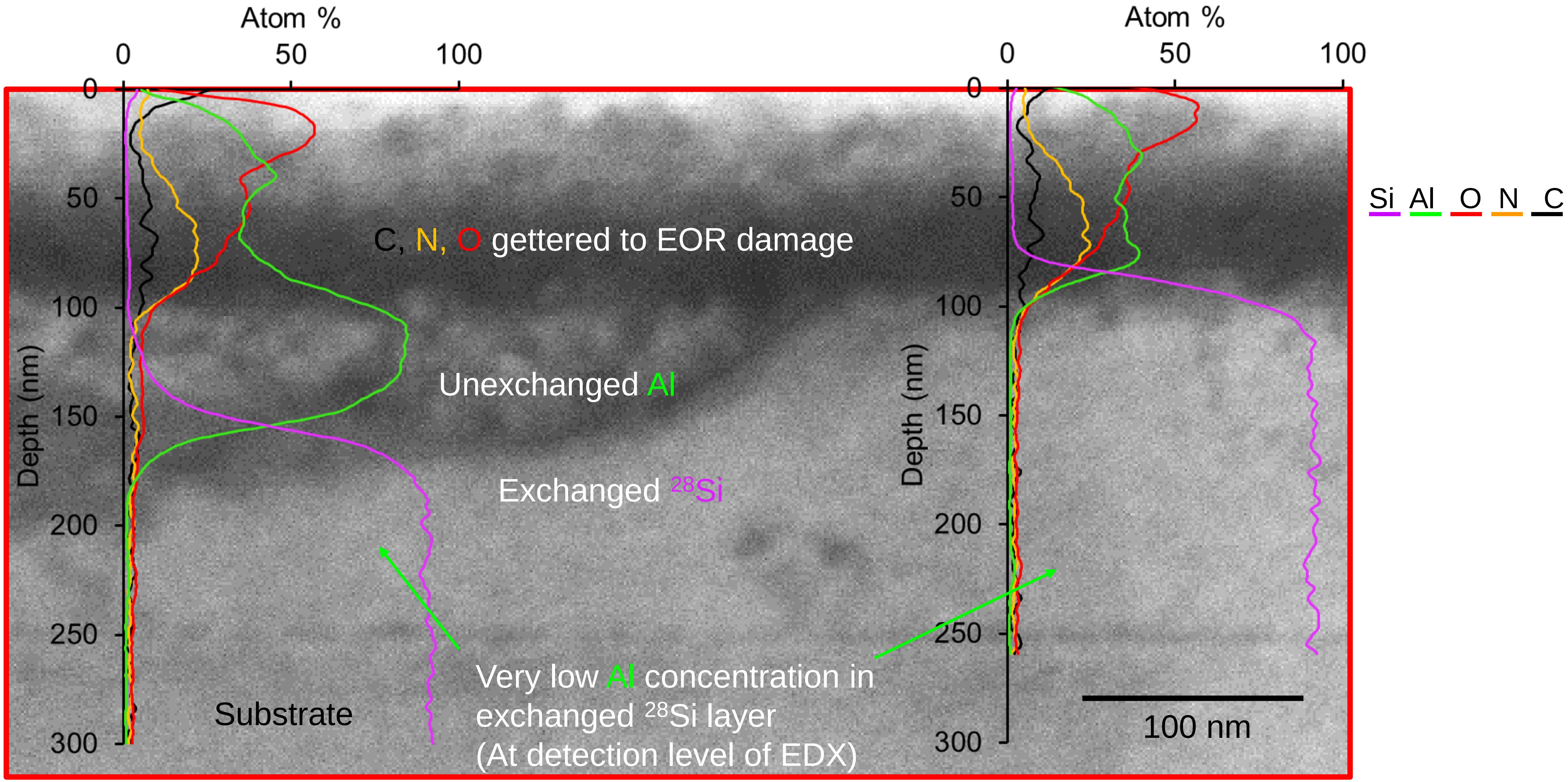


TEM image



- EDX map shows layer exchange of implanted ^{28}Si is occurring - not always completely (left)
- Thickness of ^{28}Si layer varies from 90 nm – 180 nm

Si Purity: TEM-EDX line profile in region of interest



C, N, O gettered to EOR damage

Unexchanged Al

Exchanged ^{28}Si

Substrate

Very low Al concentration in exchanged ^{28}Si layer
(At detection level of EDX)

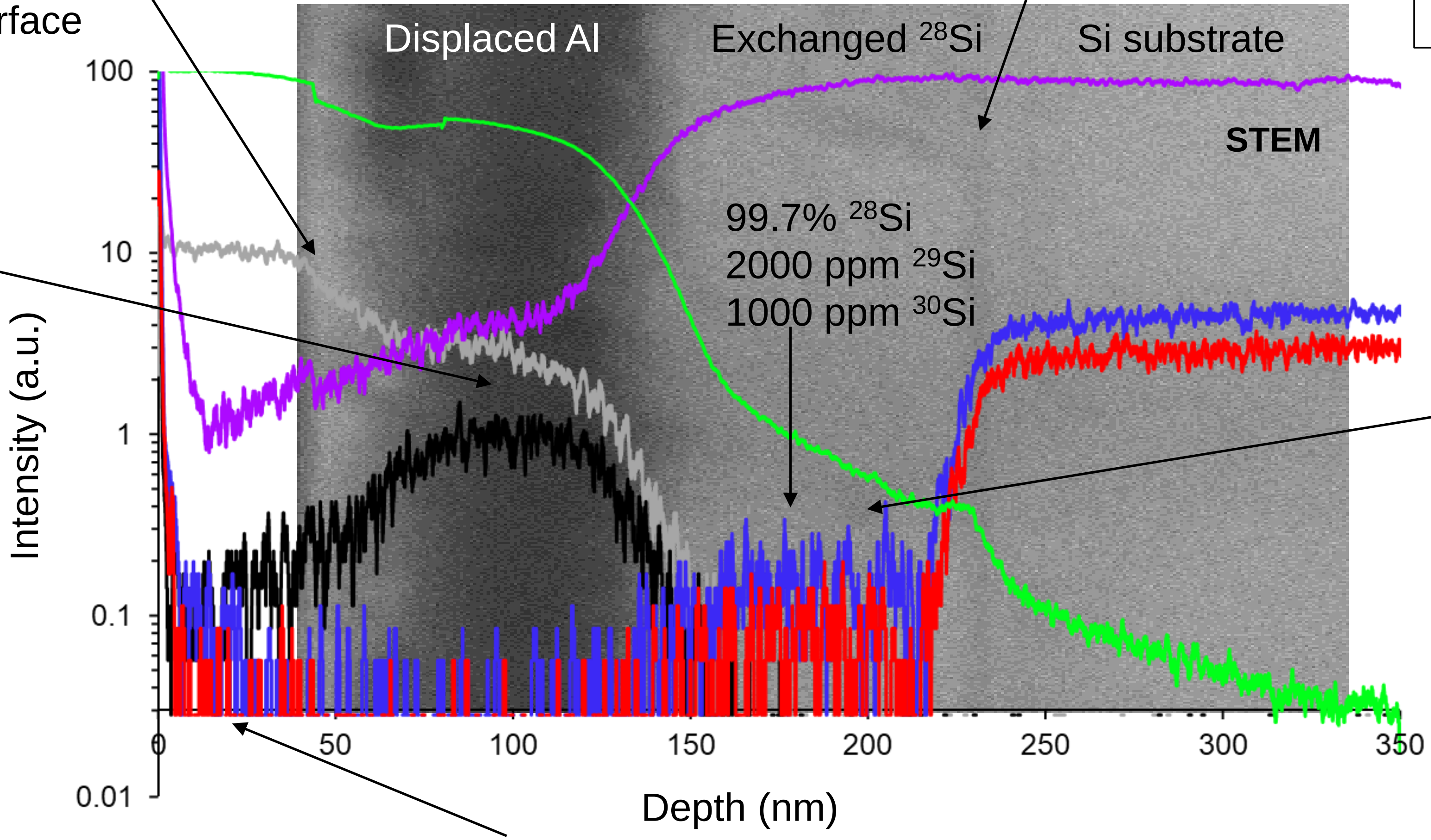
Isotopic Enrichment: ToF-SIMS

(Surrey Mech. Eng.) 25kV Bi probe and 3kV Cs etch over 100x100 µm^2 area
Intensity of Al vs Si is not proportional to concentration! (Due to secondary ionisation yields and matrix effects)

Al oxidised from surface

Substrate interface

Isobaric contamination gettered in Al



99.7% ^{28}Si
2000 ppm ^{29}Si
1000 ppm ^{30}Si

^{29}Si and ^{30}Si intensity lower in exchanged ^{28}Si layer compared to natural levels in substrate

^{29}Si:^{30}Si ratio > ^{nat}Si

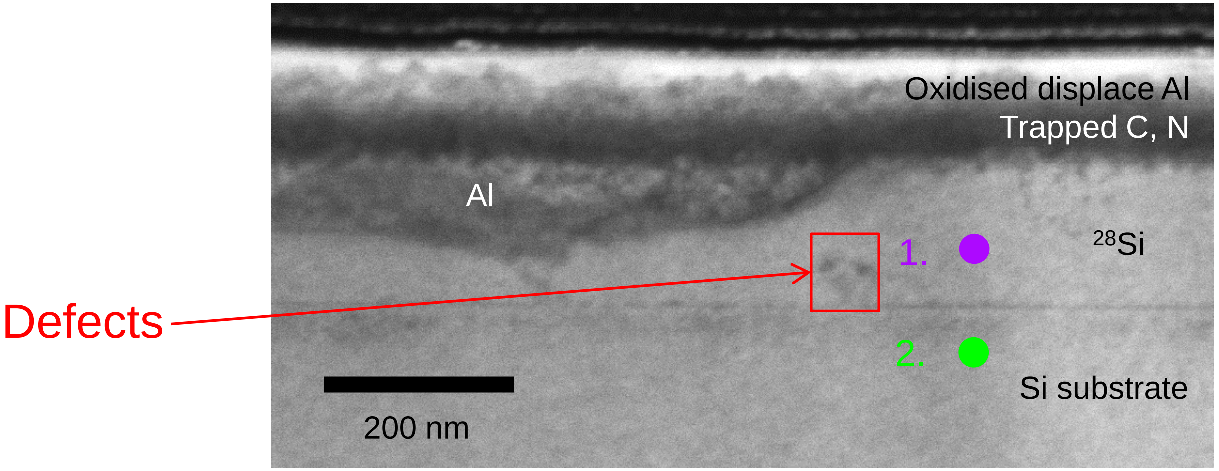
No C or O in exchanged ^{28}Si layer

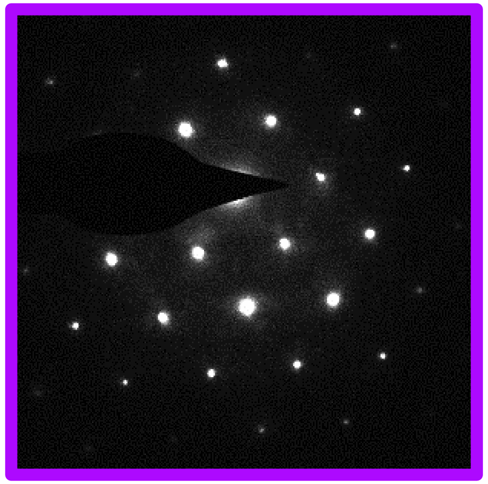
Need standard for quantitative measurement of Al in exchanged ^{28}Si layer

SIMS sputter rate non-linear due to surface oxidation

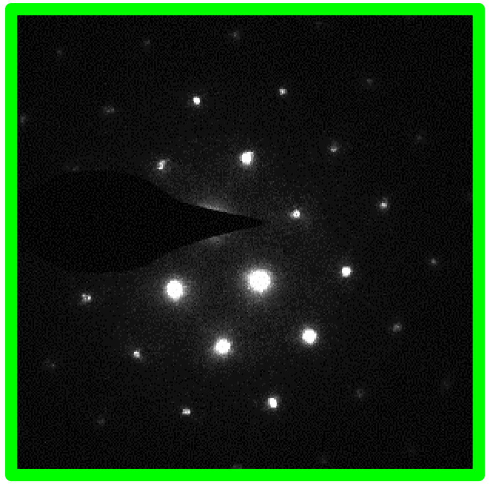
Evidence of Epitaxial Growth: TEM Nanobeam Diffraction

TEM image





Exchanged ^{28}Si layer



Substrate

- Diffraction pattern in ^{28}Si layer matches substrate
- Implies epitaxial ^{28}Si crystallisation

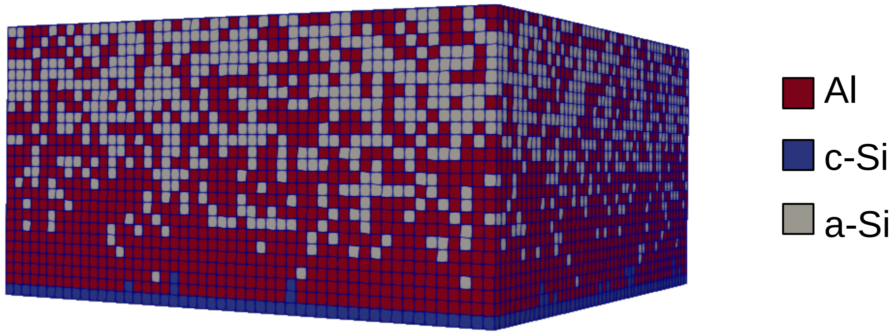
Summary of Results

	Requirement	**Achievement**
1. Enrichment	~99.9% ^{28}Si ~800 ppm ^{29}Si	99.7% ^{28}Si 2000 ppm ^{29}Si
2. Al concentration	<40 ppm Al	~1% (10 000 ppm)
3. Crystallinity	Perfect single crystal	Epitaxial growth with some defects
4. Contamination	<10 ppm C, N, O	< detection limit
5. Thickness and uniformity	>50 nm	90-180 nm Need improvement

Challenges: Enrichment, Al concentration, Uniformity

Future Work

- Optimise Si-Al interface and Al quality to improve layer exchange uniformity
  - Used Tyndall facilities through European access scheme ASCENT+ with Brenda Long (UCC) and Nikolay Petkov (MTU)
- Optimise implant conditions
  - Improve enrichment with better implanter mass resolution
  - ^{28}Si implant using SIMPLE at Surrey Ion Beam Centre through RADIATE
- Optimise anneal
- Reduce residual Al concentration in exchanged ^{28}Si
  - Does better epitaxy minimise inclusion in exchanged layer?
  - Remove by chemistry or gettering
- Modelling (kMC SPPARKS)
- Seeking a partner for spin lifetime measurements



Questions

www.uknibc.co.uk/e.b.schneider@surrey.ac.uk

Conclusions: Why ion implanted Al-induced layer exchange?

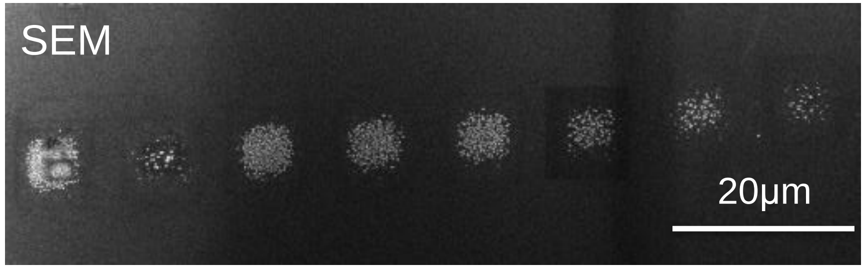
1. CMOS fabrication techniques
    - Use standard SiF_4 feedstock and an industrial implanter
2. Implant fluence 10x lower than what is required for direct implantation
    - Industrial implanter could process single wafer in 3 hours (high but plausible)
3. Less sputtering of implanted Si
4. Implanted layer at higher energy than equilibrium state due to amorphisation and implant damage – can drive process
    - Very short annealing processes
5. Getter isobaric interference

- Main issue: residual Al concentration – plan to getter or remove chemically

Advantages of SIMPLE

(Single Ion Multispecies Positioning at Low Energy)

- Although SIMPLE is a single ion implanter it can be used in high fluence mode to implant islands of 20x20 μm^2
- High doses over small areas (~μm^2) in short times (~10 minutes)
  - Compared to a Danfysik broad area implant ~10^2 hrs
- Different doses on same sample
- Wien filter – ^{28}Si mass selection
- Ultra High Vacuum – oxidation minimised
- Liquid Metal Ion Sources made in-house (Au-Si)

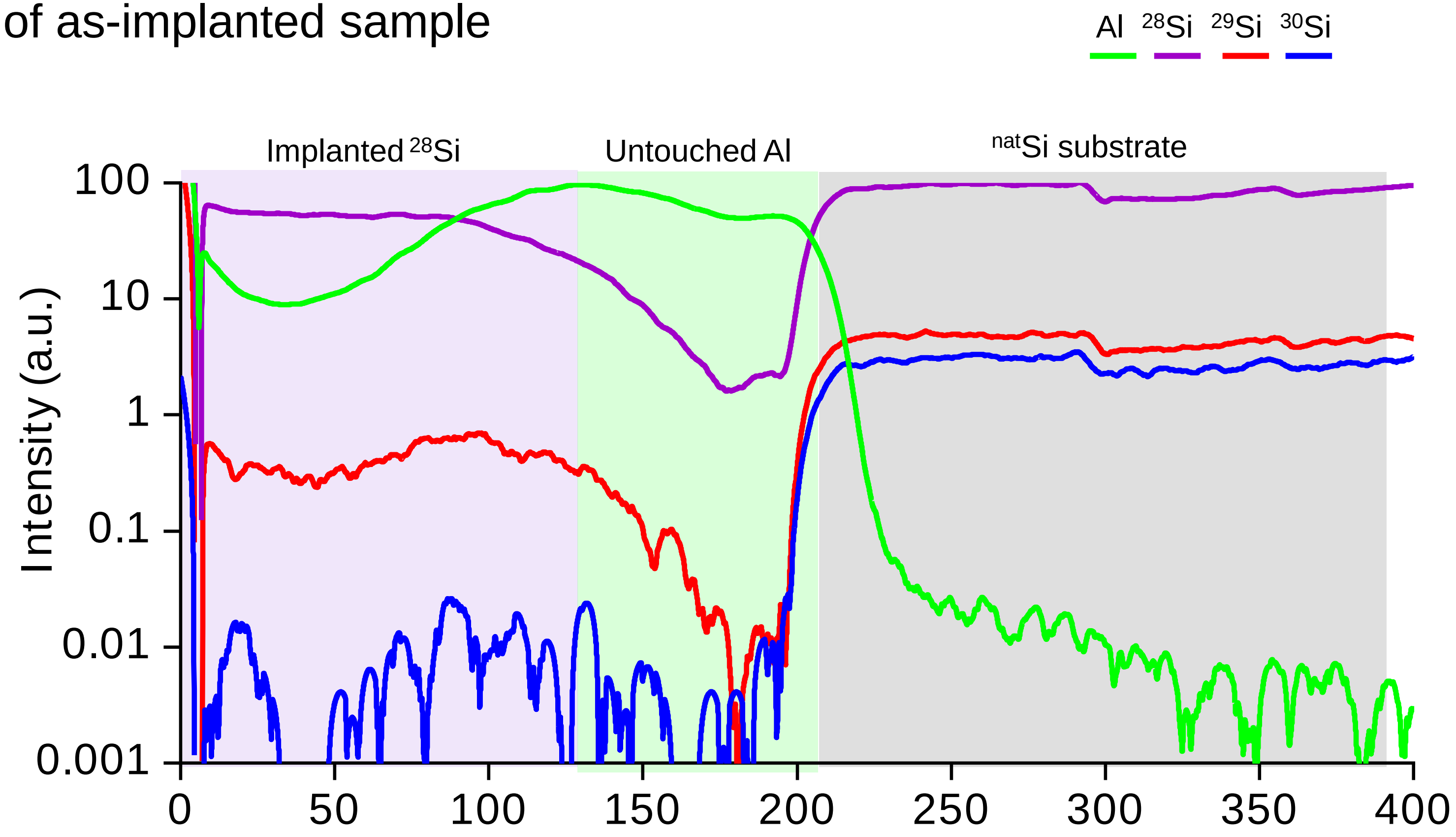


Islands formed by layer exchange of SIMPLE-implanted Ge

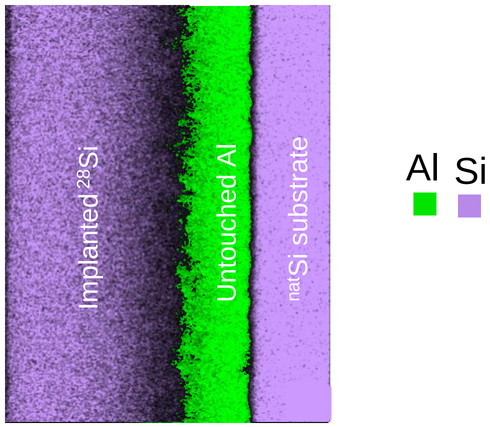
J. England, D. Cox, N. Cassidy, B. Mirkhaydarov, and A. Perez-Fadon, Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms, vol. 461, pp. 30–36, 2019, doi: 10.1016/j.nimb.2019.09.013.

Implanted ^{29}Si and ^{30}Si Contamination

ToF SIMS of as-implanted sample



TEM-EDX false-colour maps of Si and Al X-rays

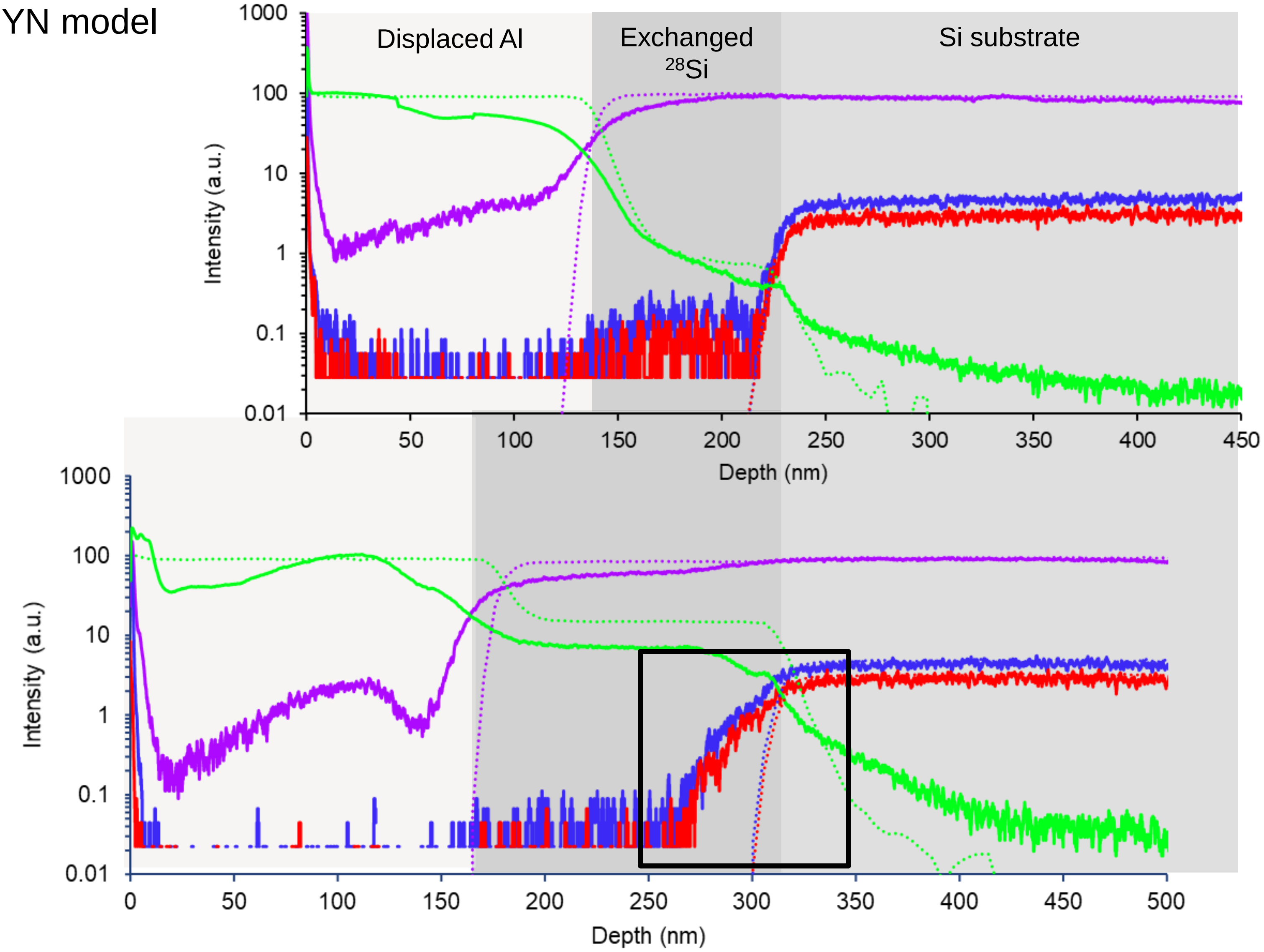


Implant Layer Exchange: Substrate Diffusion

Fabrication conditions:
1. 250 nm and 325 nm deposited Al
2. ^{28}Si/30keV/6.6E17cm^{-2}
3. 500°C

ToF-SIMS and TRIDYN model

250 nm Al
1 minute @ 500°C



325 nm Al
1 hour @ 500°C

Experiment modelled by sharp interface – negligible diffusion of substrate over 1 minute

Diffusion of substrate ^{29}Si and ^{30}Si into enriched ^{28}Si layer over 1 hour