Intro to QIS (and plasmas)

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Princeton Plasma Physics Laboratory (PPPL)

Presented to

PPPL School on Plasmas for Microelectronics and Quantum Information Science

July 28 - August 1, 2025

Quantum Information Science (QIS)

Compute

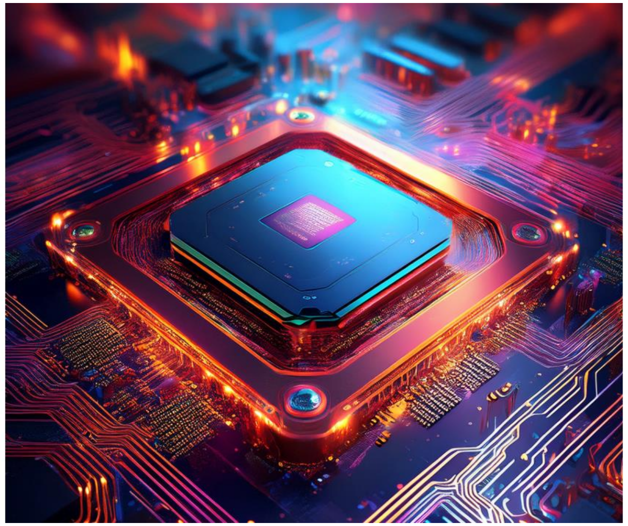
- Complexity
- Power
- Optimization

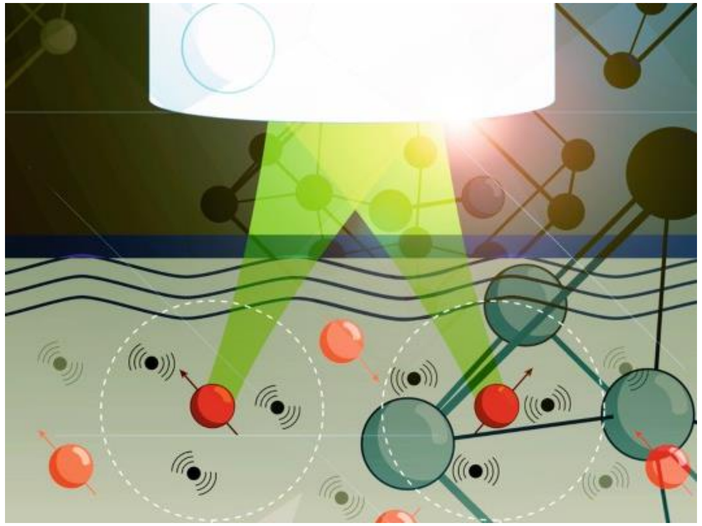
Communicate

- Transport quantum information
- Secure signals

Sense

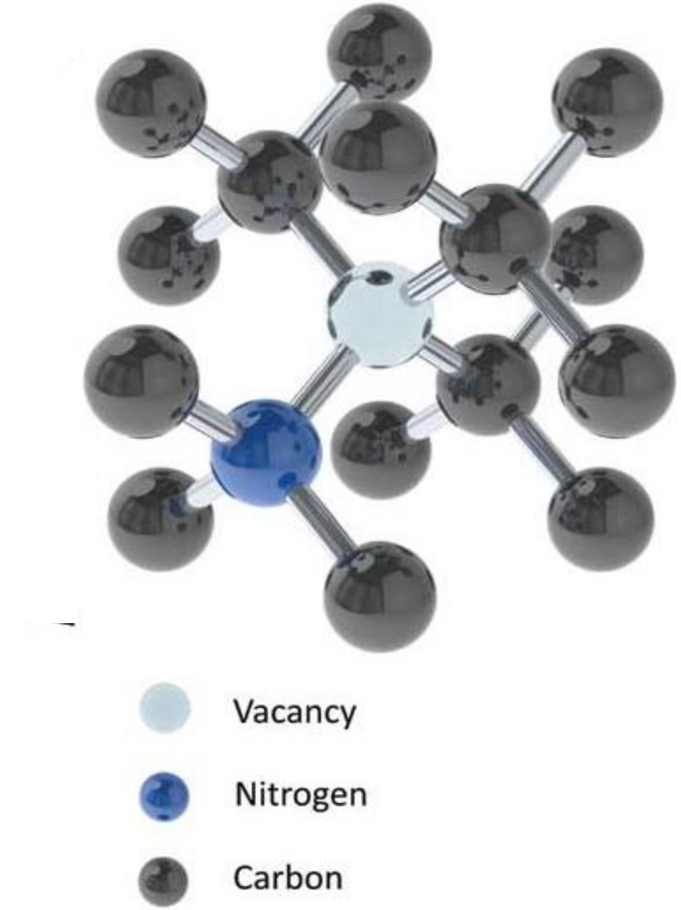
- Sensitivity
- Spatial resolution





Rovny et al, Nat Rev Phys 2024

QIS: Using Quantum States for Sensing



$$H = D(T)S_Z^2 \quad + \gamma_e \vec{B} \cdot \vec{S} \quad + d_\parallel [E_Z + \mathcal{E}_Z(\overleftrightarrow{\sigma})] S_Z^2$$
$$+ d_\perp [E_X + \mathcal{E}_X(\overleftrightarrow{\sigma})](S_X S_Y + S_Y S_X)$$
$$- d_\perp [E_Y + \mathcal{E}_Y(\overleftrightarrow{\sigma})](S_X^2 - S_Y^2)$$

Zero-field splitting (T)

Zeeman effect ($\vec{B}$)

Stark effect ($\vec{E}, \overleftrightarrow{\sigma}$)

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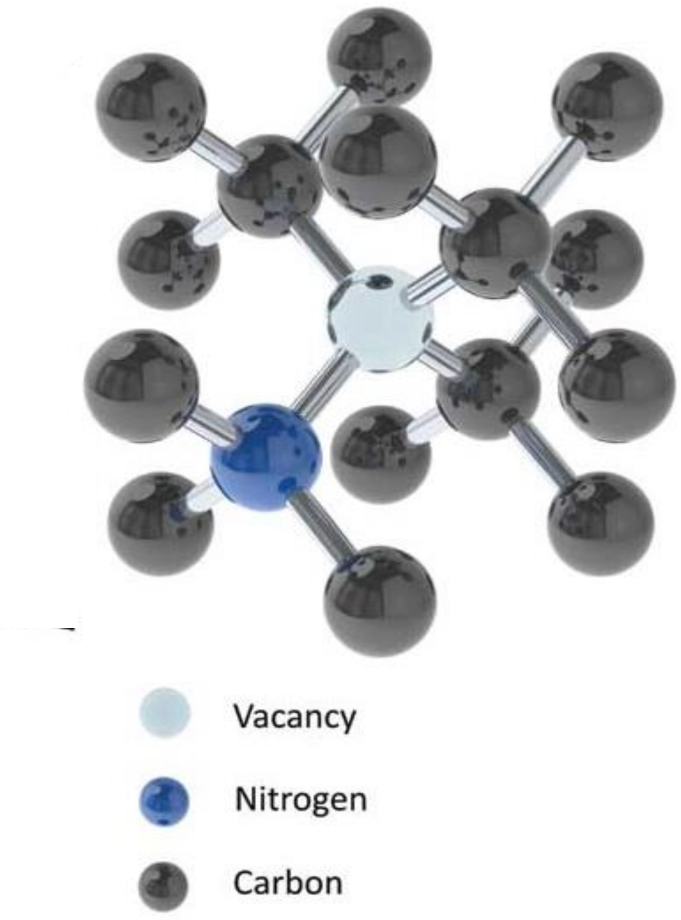

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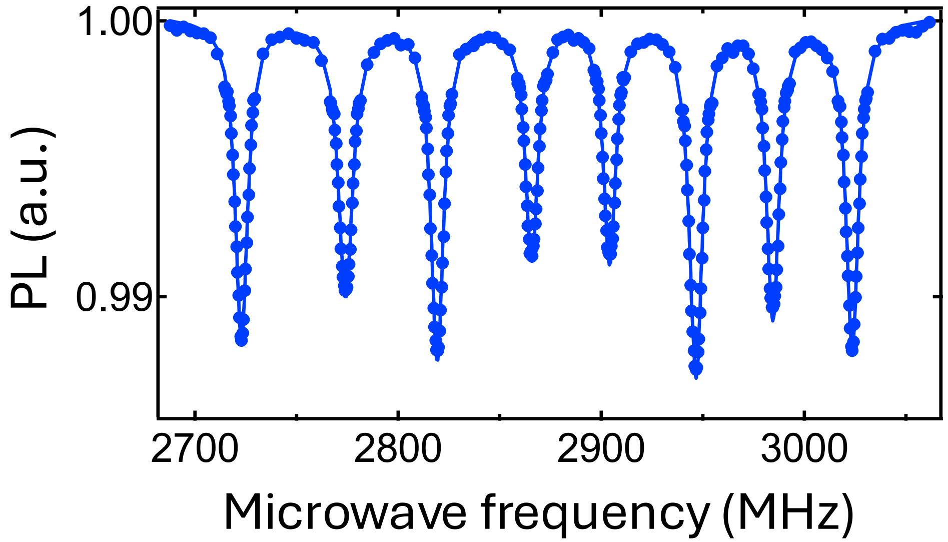
Optically-detected magnetic resonance (ODMR)



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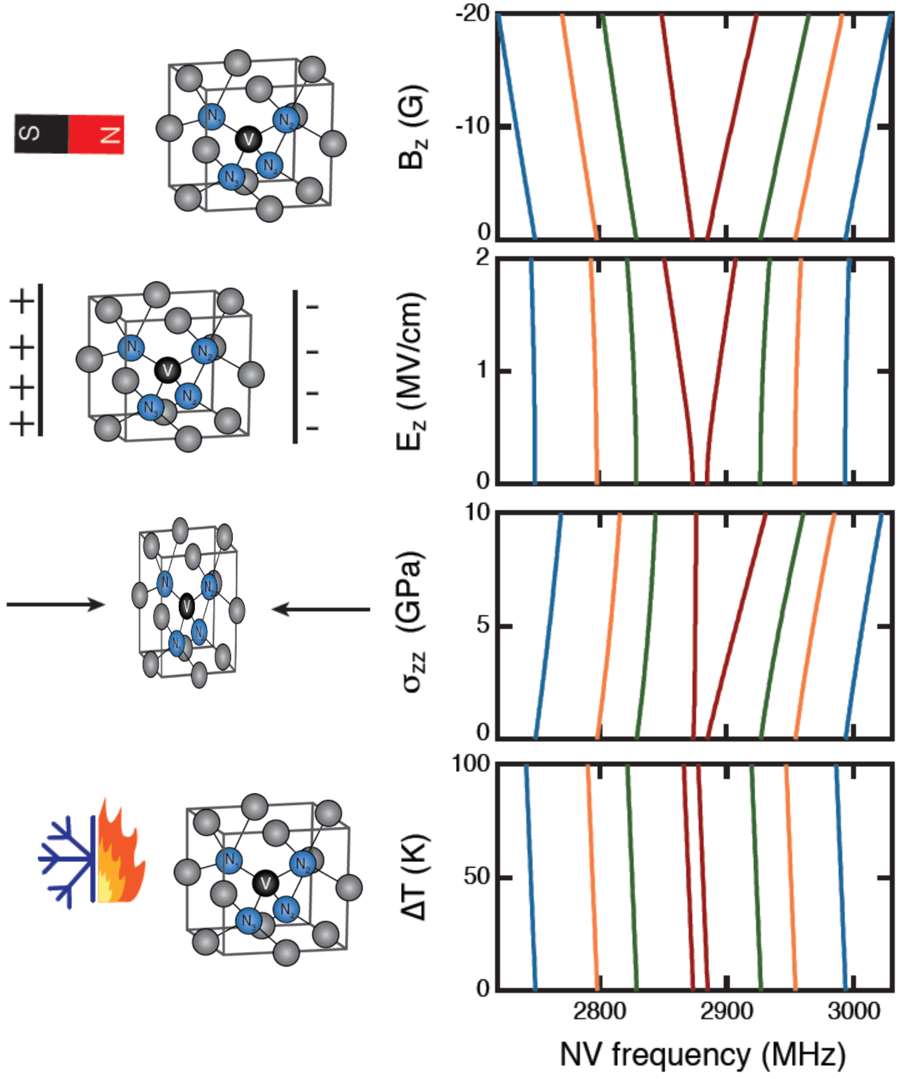

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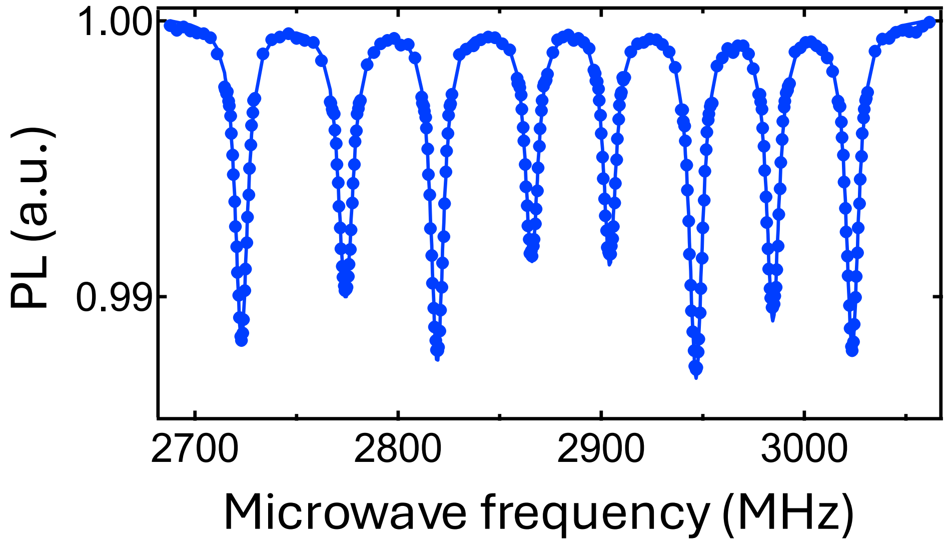
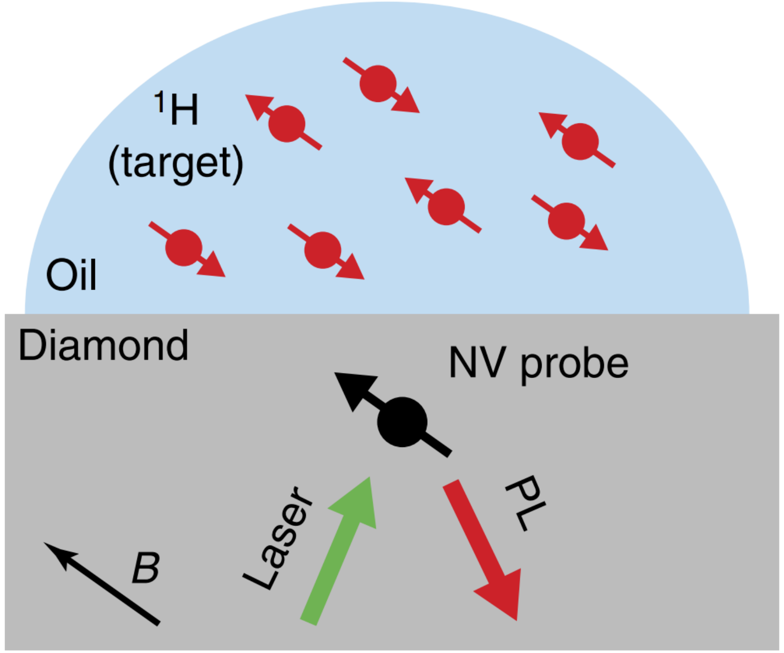


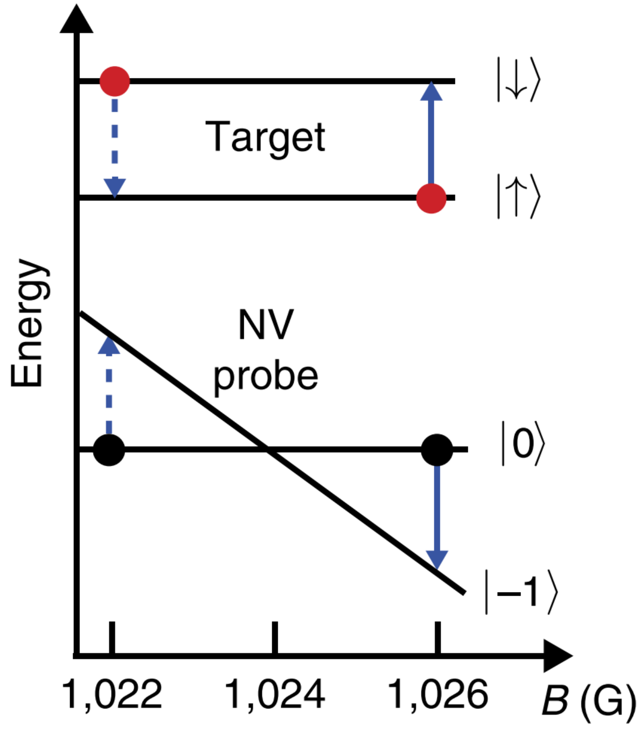
QIS: Using Quantum States

1. **Create a controllable quantum object**
   → e.g. 'NV defect' in diamond



2. **Control quantum object**
   → e.g. Laser control of quantum state

3. **Use quantum object**
   → e.g. Transfer polarization to nuclear spins



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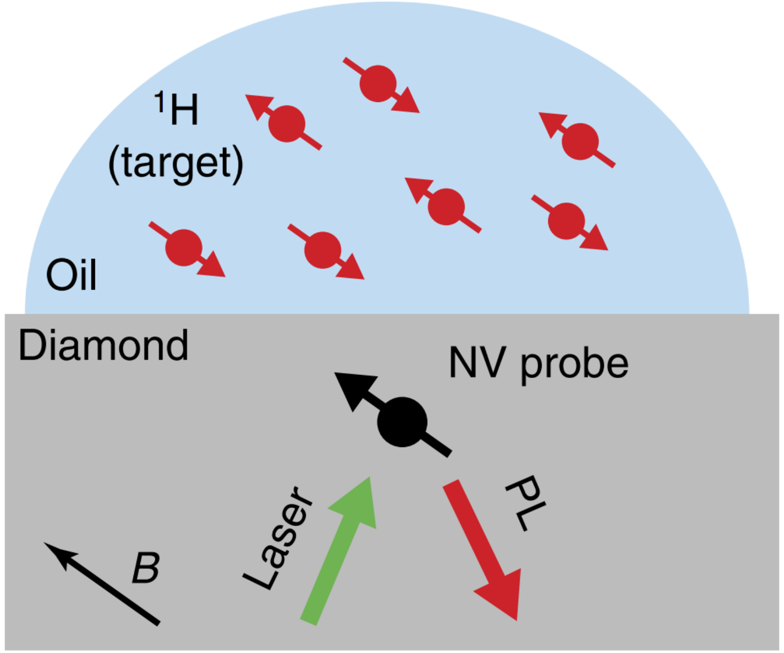

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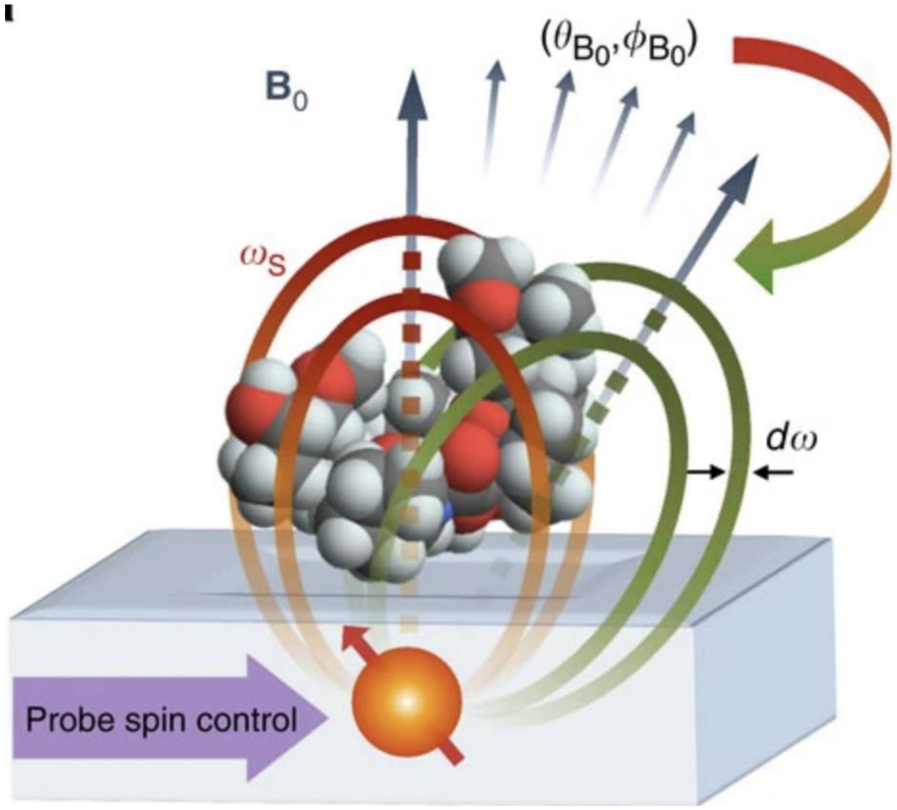
3. **Use quantum object**
   → **e.g. Single molecule magnetic resonance imaging (MRI)**



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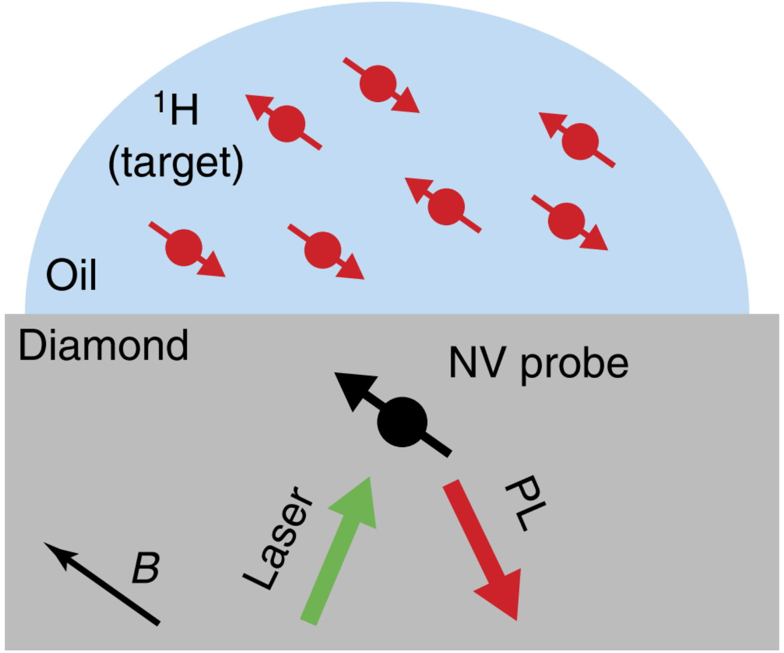

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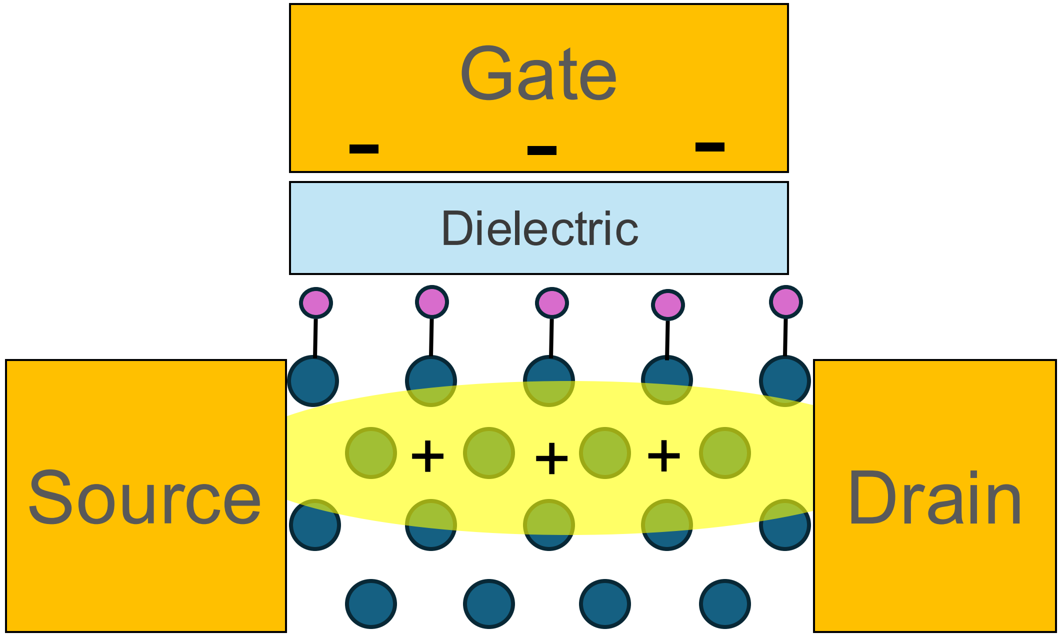
Material / Device Requirements:

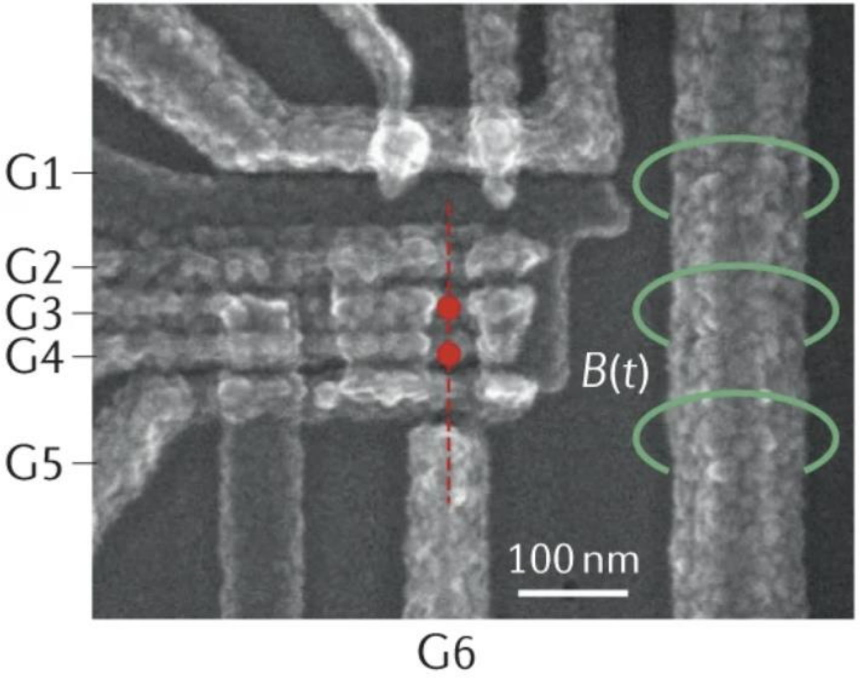
A. Quantum compatible material
B. Engineered quantum objects
   - Position & Function
C. Engineered quantum control structures
D. Maintain quantum properties

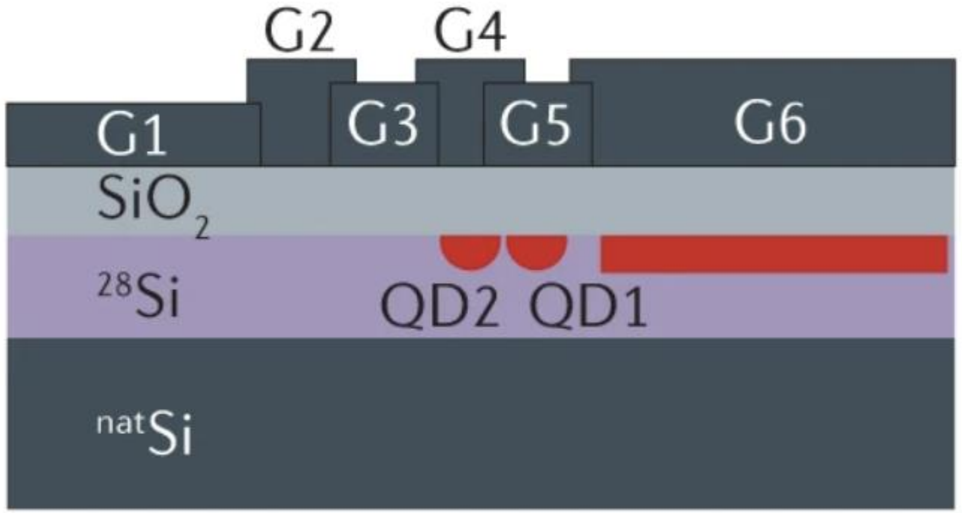
Classic Microelectronics



- Charges
- Capacitances
- Currents
- Electric potential
- Sensitive to stray charges

Quantum Microelectronics

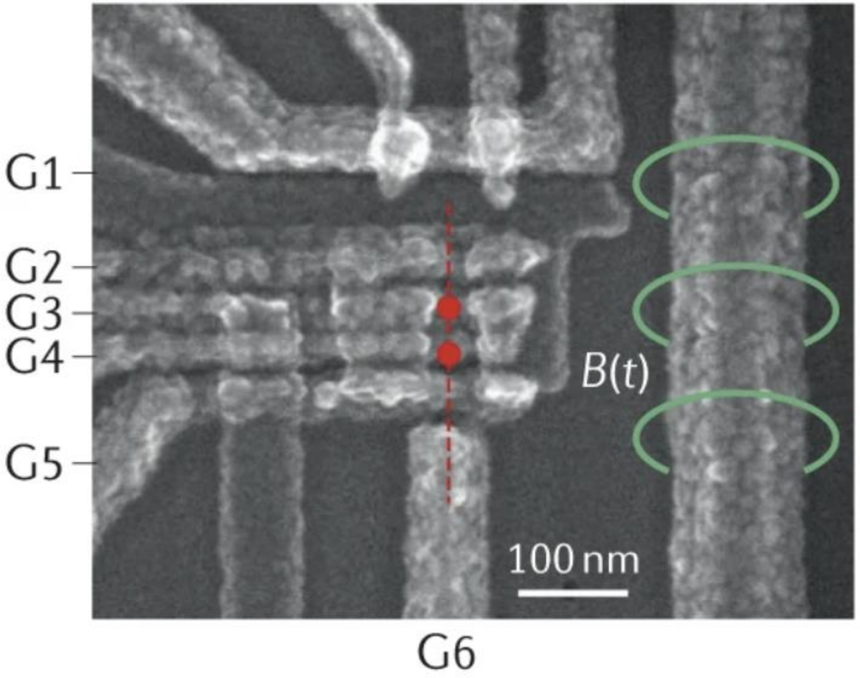


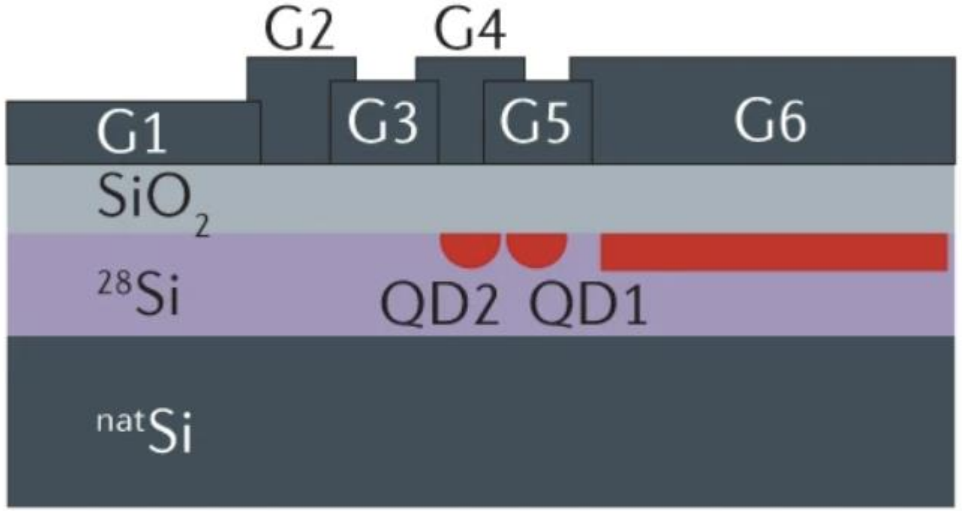


Quantum Microelectronics

Bit = 0 or 1

Qubit = superposition of 0 *and* 1

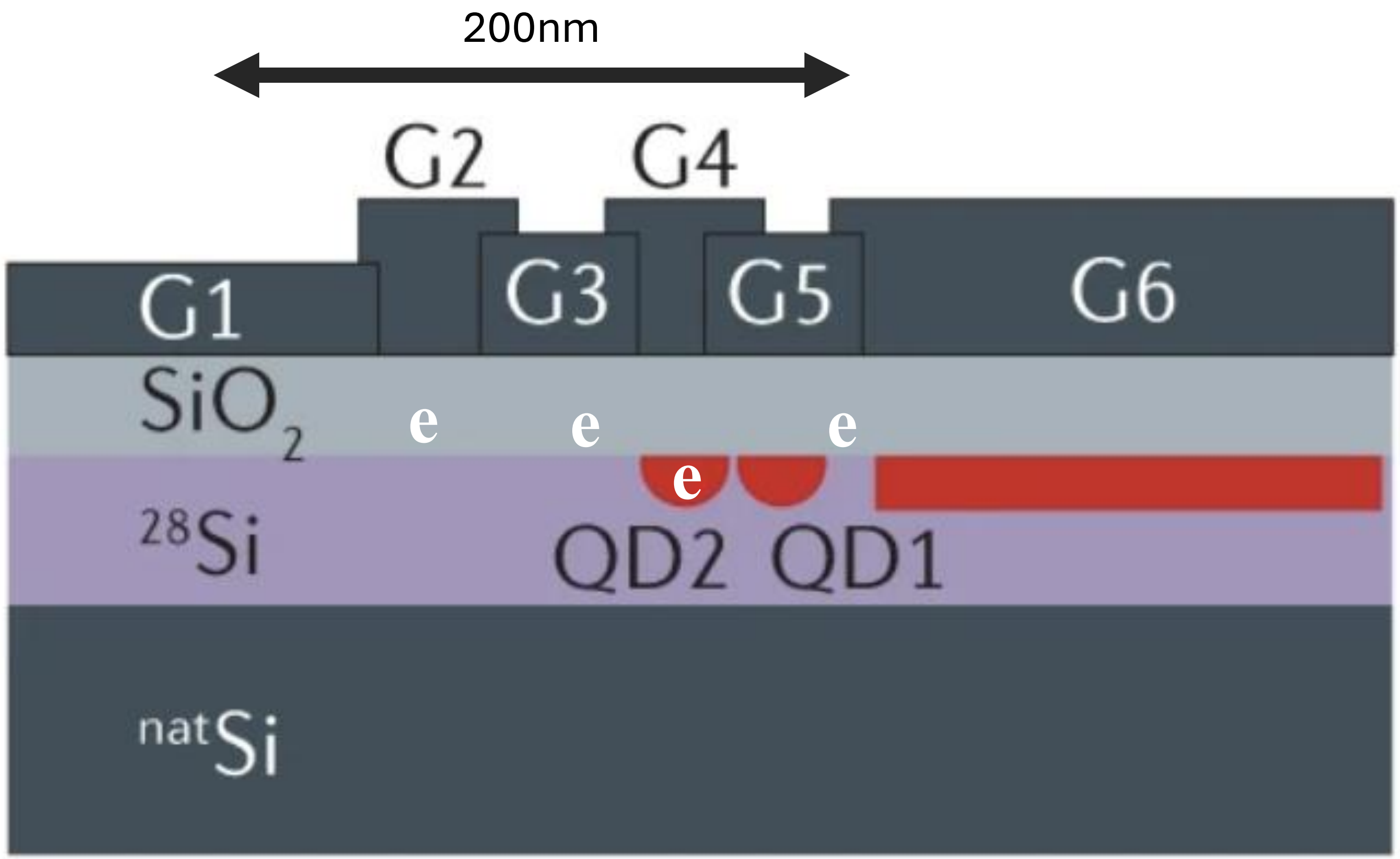




- Quantized electron position and electron flow
- Smaller number of electrons
- **VERY sensitive to stray charges**

https://doi.org/10.1002/adfm.202105488 https://doi.org/10.1038/s42254-021-00283-9

Quantum Microelectronics



SiO2 trap density O(10^{10}/cm^2)

https://doi.org/10.1016/j.matpr.2020.03.630

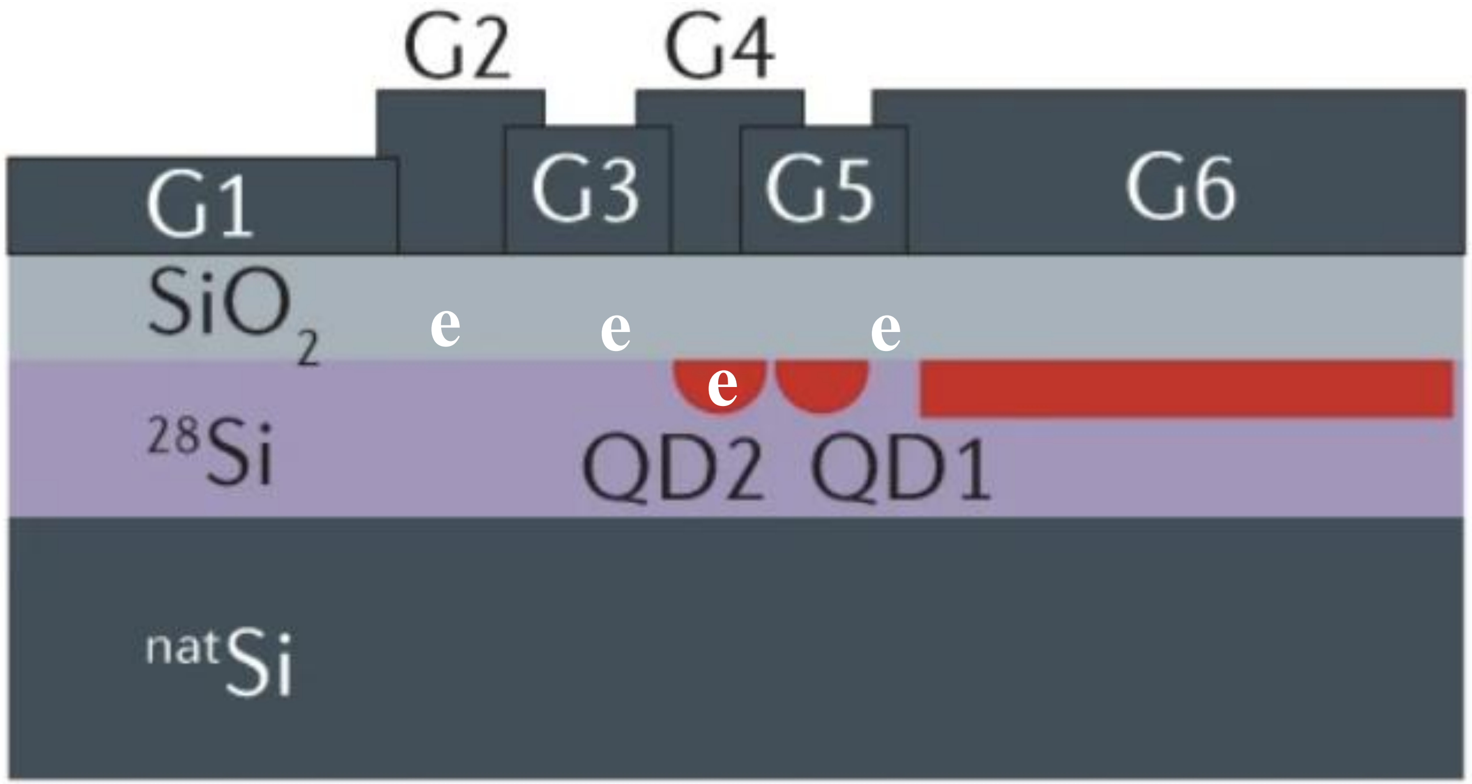
Quantum ‘relaxation time’

≈

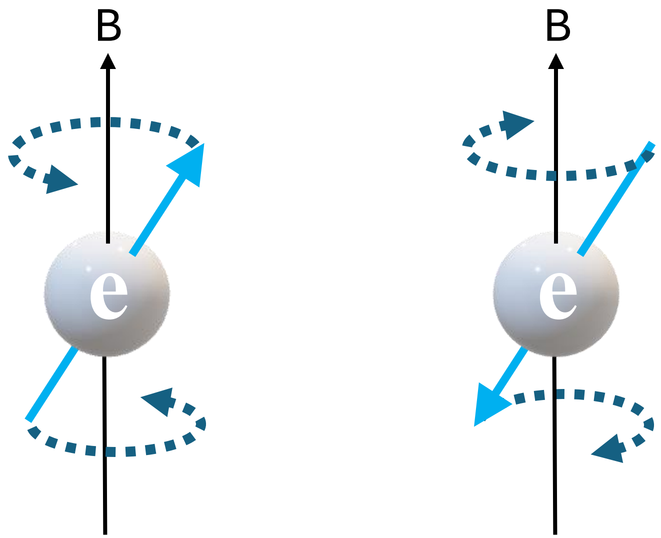
Bit flip error 0 ↔ 1

Sensitive to electric fields

Quantum Microelectronics – Magnetic Spins



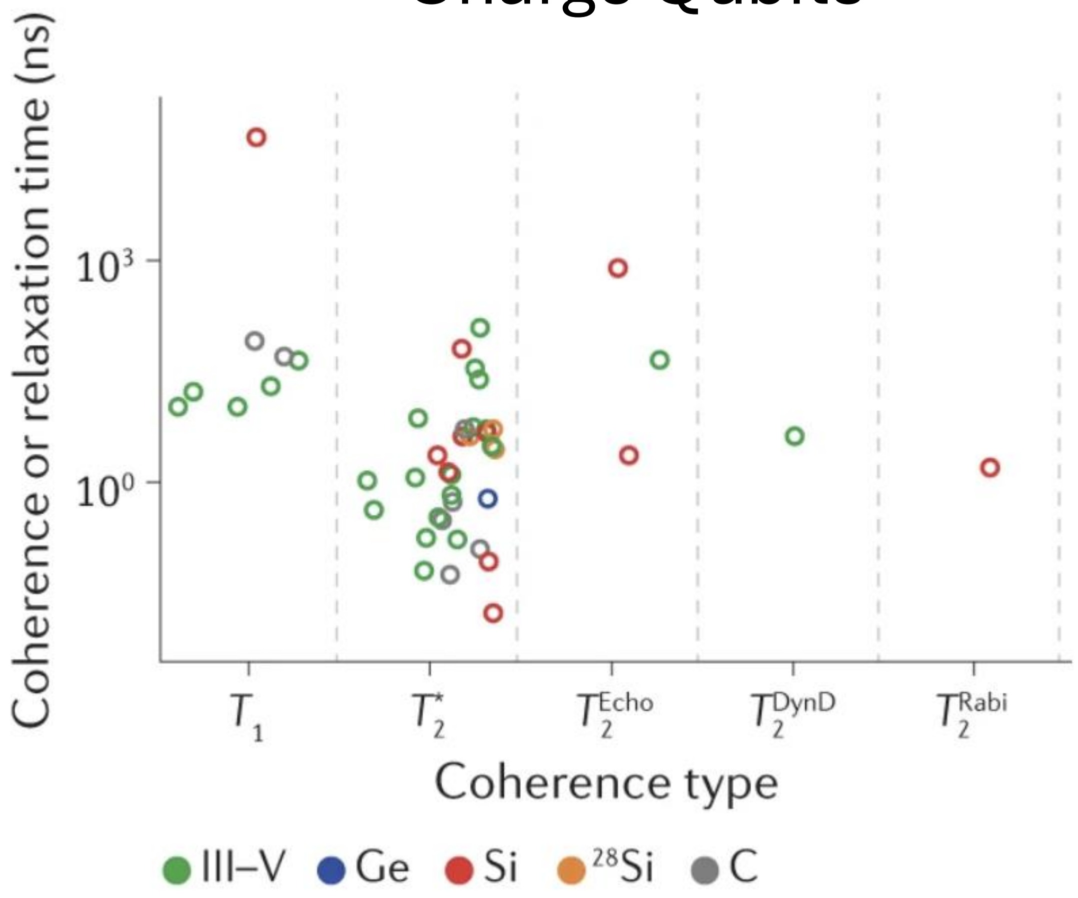
Qubit states = spin orientation



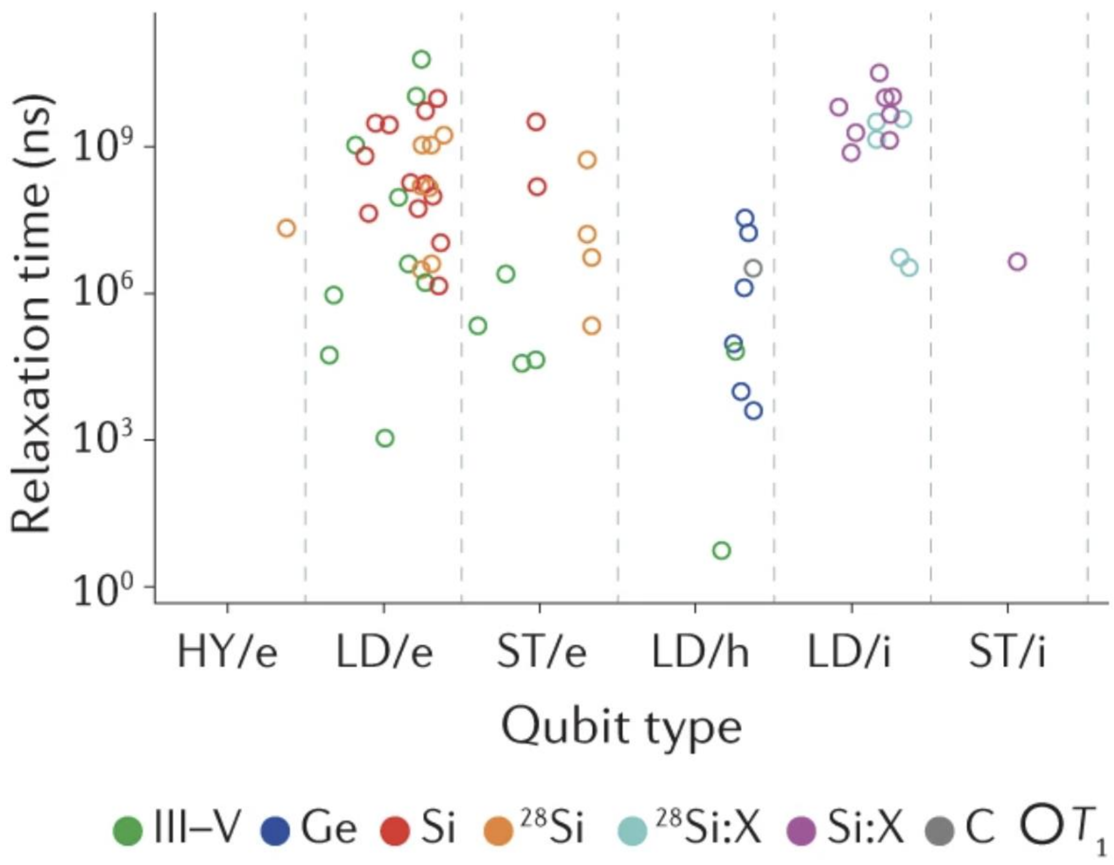
Sensitive to magnetic fields

Quantum Microelectronics – Magnetic Spins

Charge Qubits



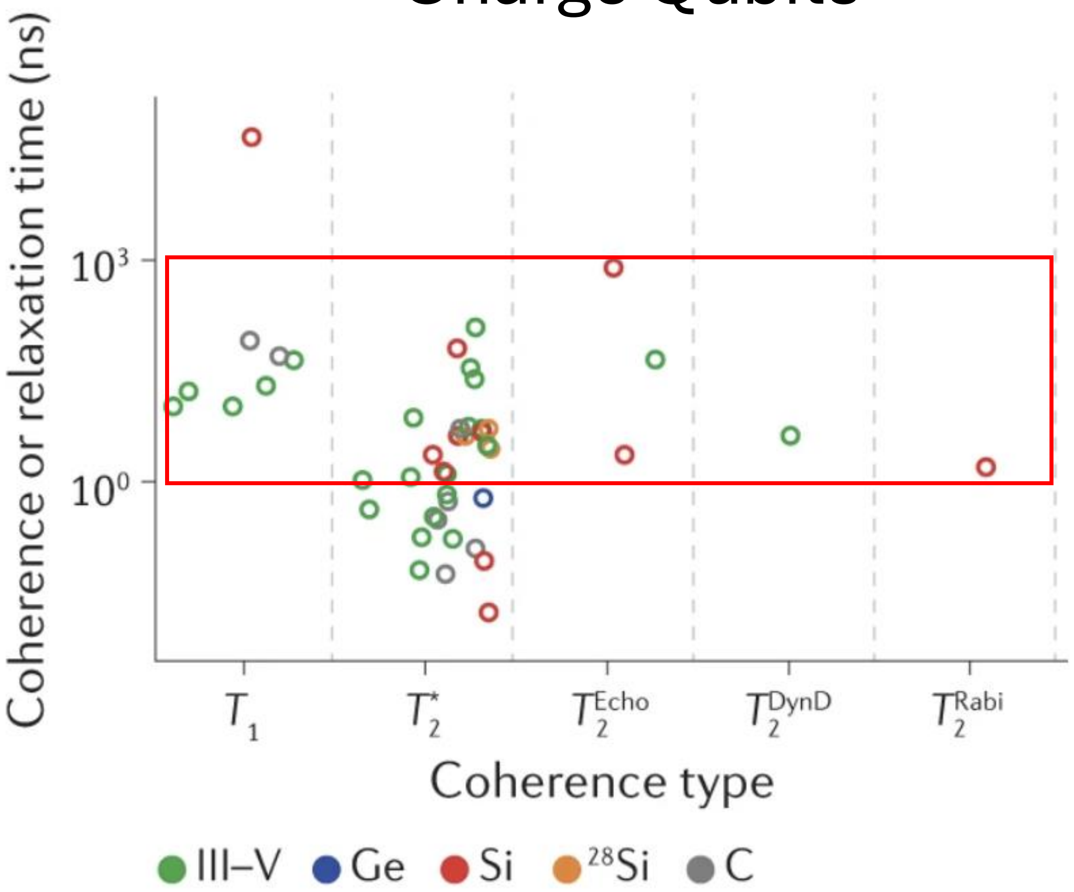
Spin Qubits



https://doi.org/10.1038/s42254-022-00484-w

Quantum Microelectronics – Magnetic Spins

Charge Qubits



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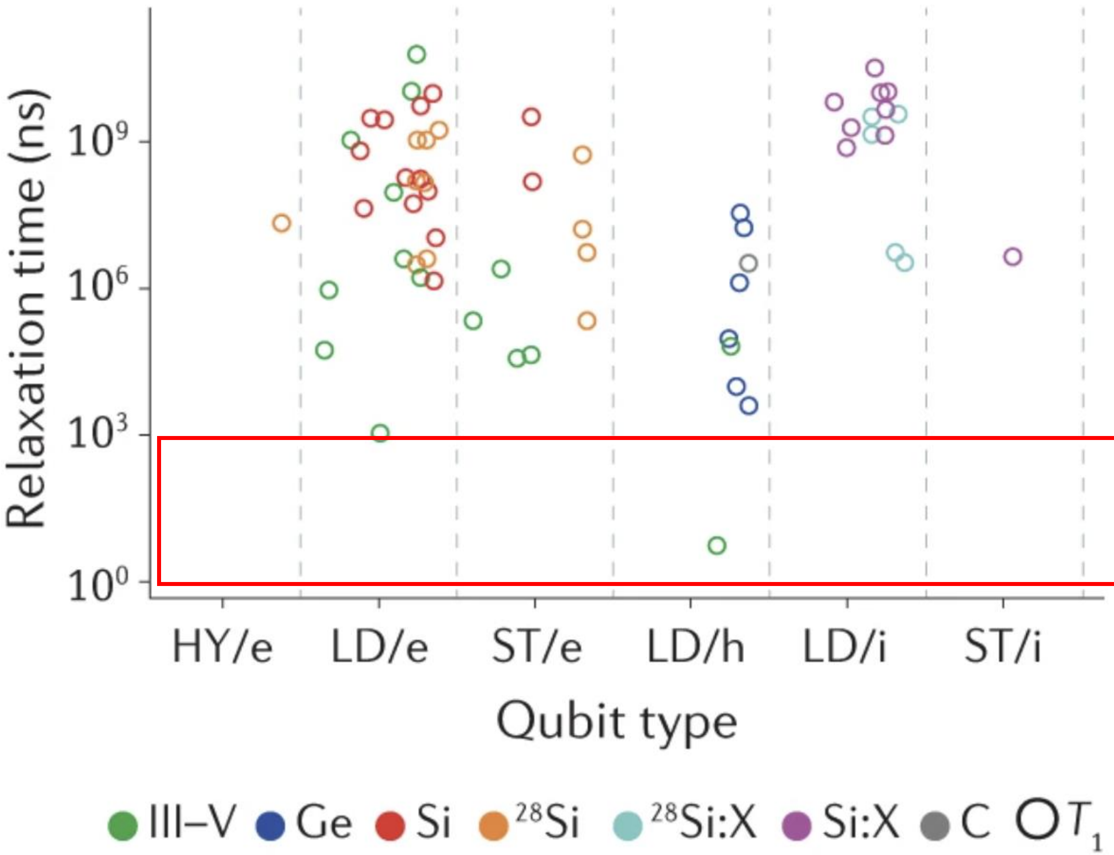
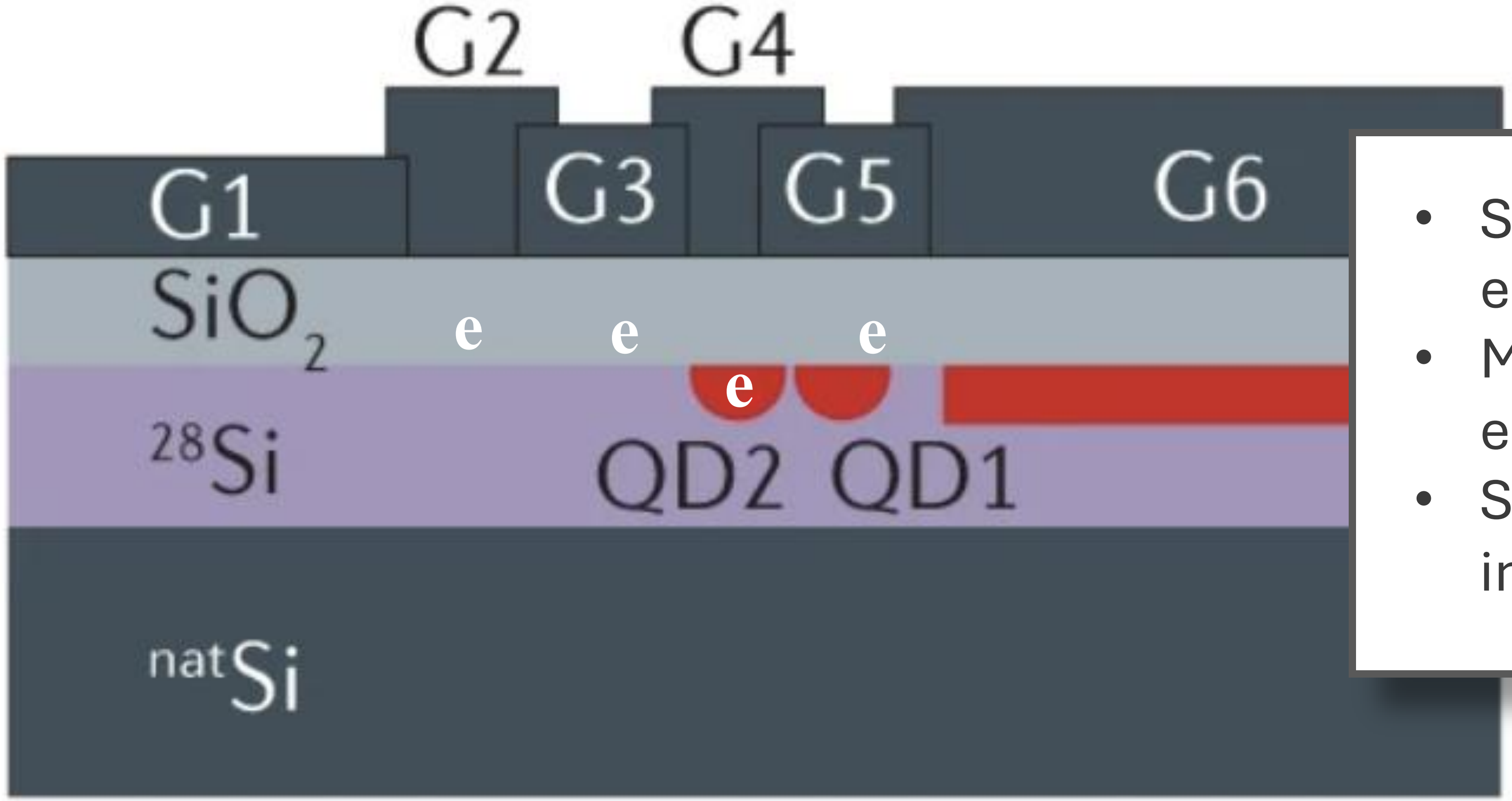


https://doi.org/10.1038/s42254-022-00484-w

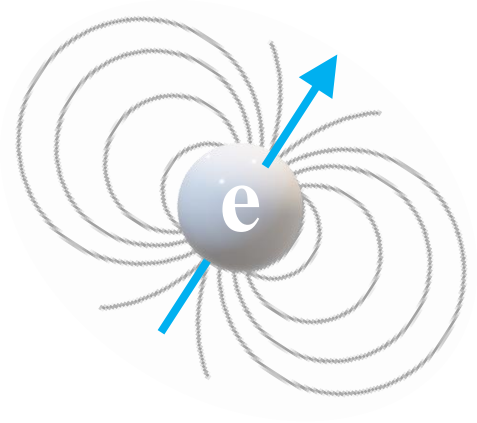
Quantum vs Classic Microelectronics Processing



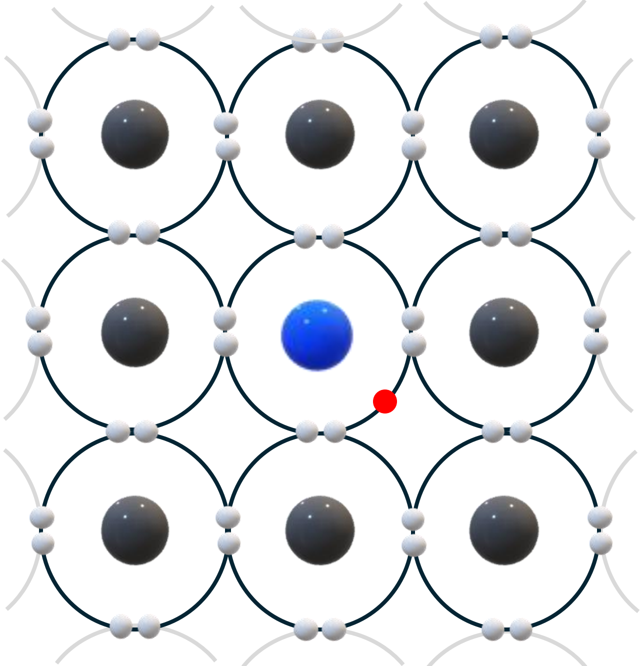
- Similar nanoscale electronic structures
- More sensitive to electronic defects
- Sensitive to magnetic impurities / defects

Not all electronic charges create magnetic fields

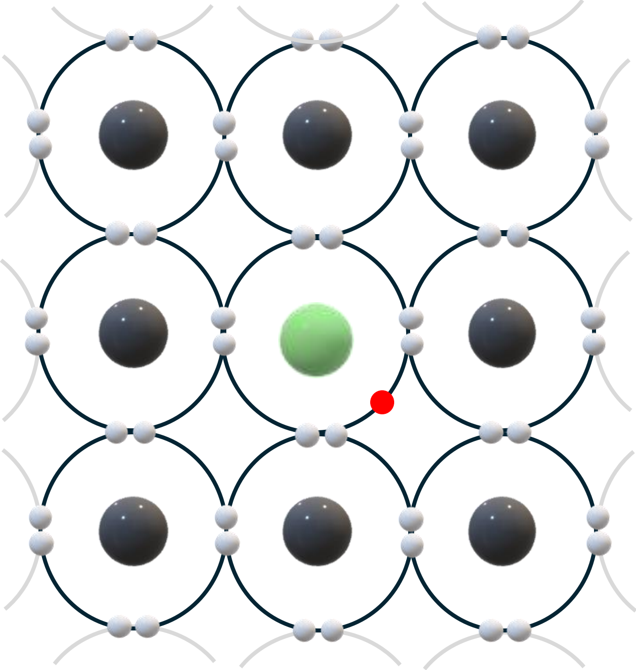
Unpaired electrons
= nanomagnets



‘Free electron’ =
spin ½ nanomagnet

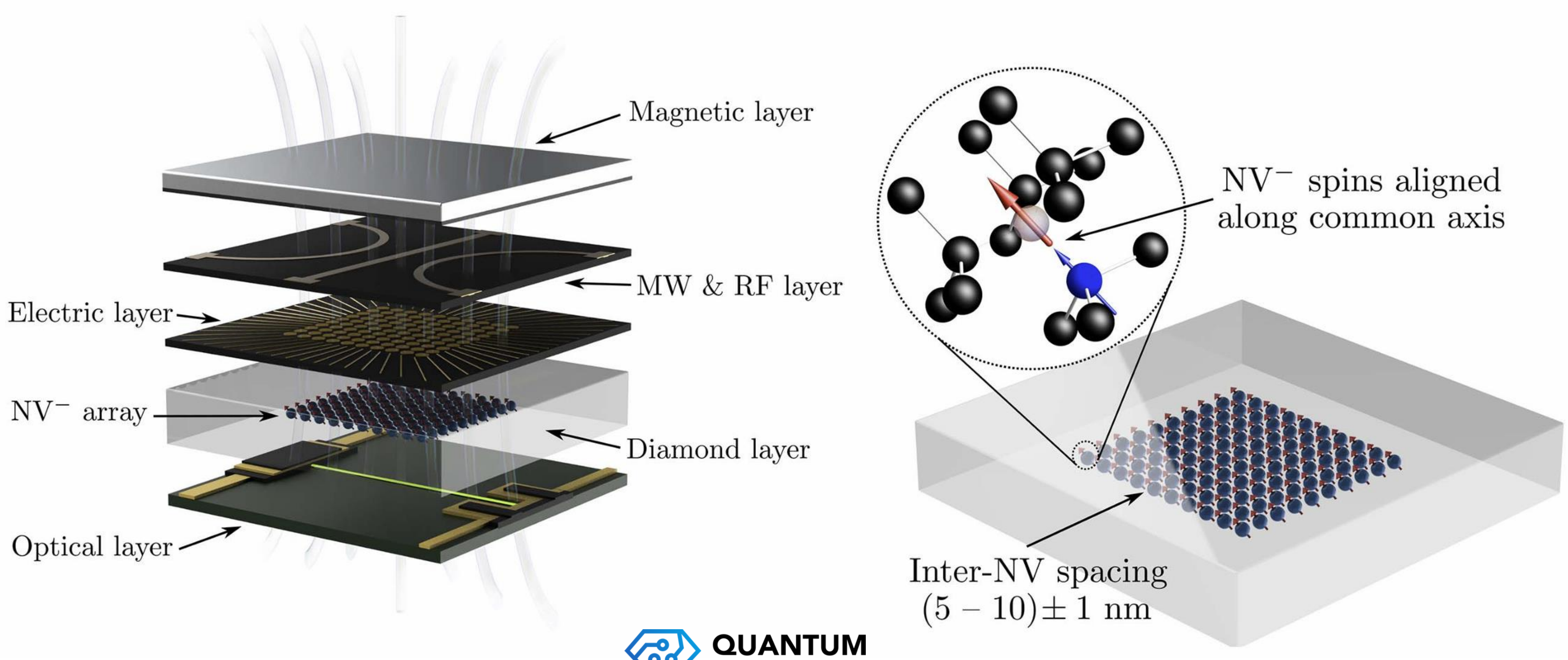


P^0 in **Si** = spin but no charge
P^+ in **Si** = charge but no spin



C– in **Si** = spin and charge

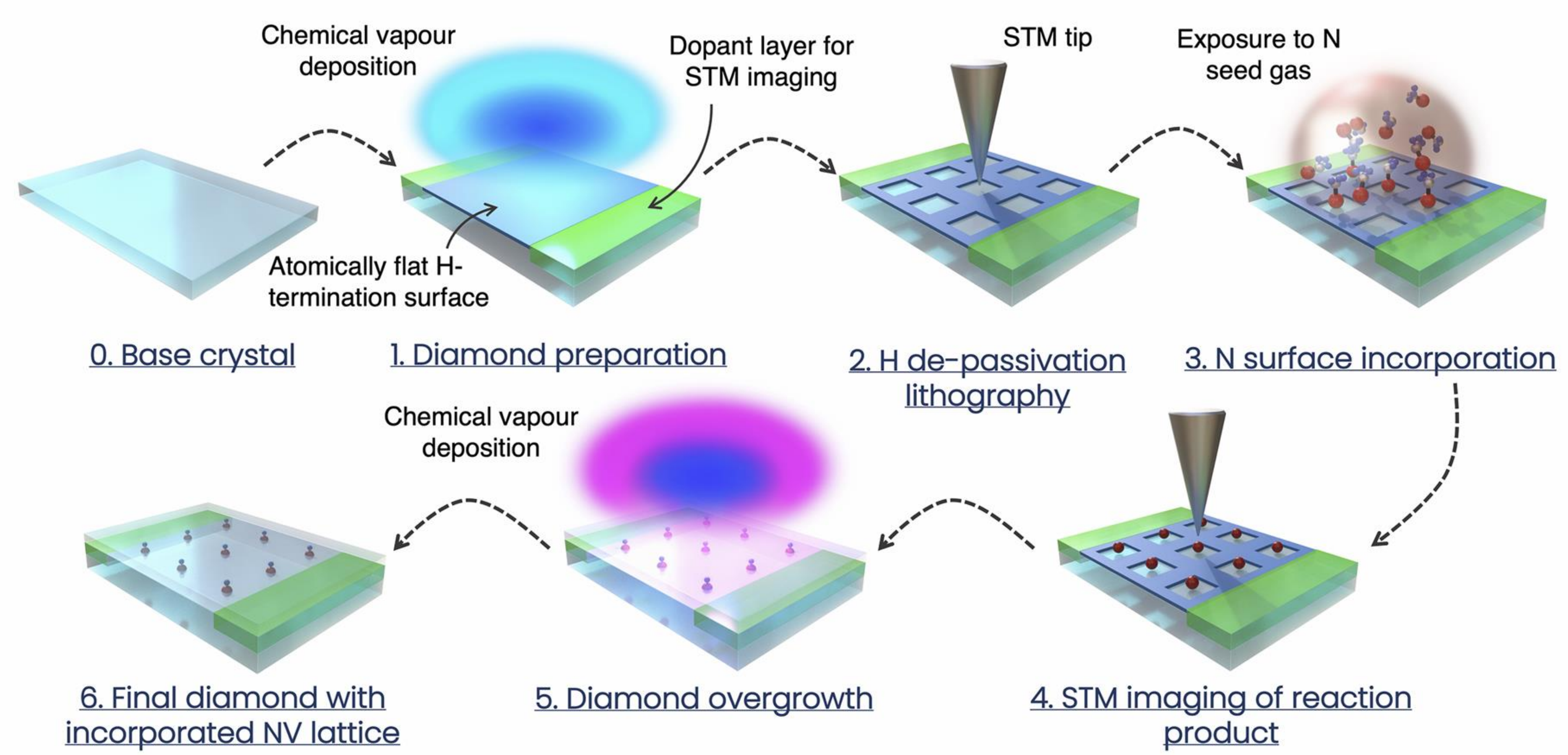
QIS: Using Quantum States



QUANTUM BRILLIANCE

https://doi.org/10.1088/2633-4356/ade359

Plasma processing for QIS: Fabrication



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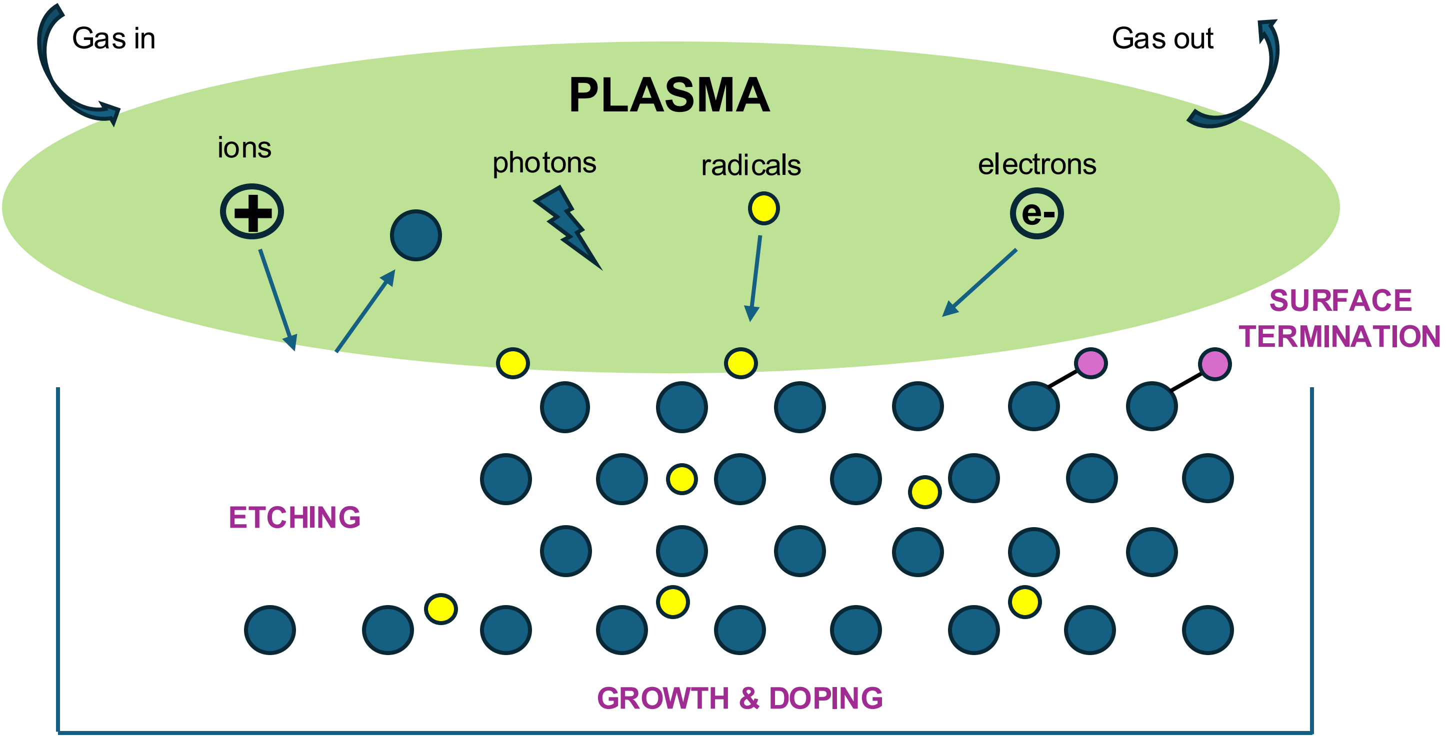
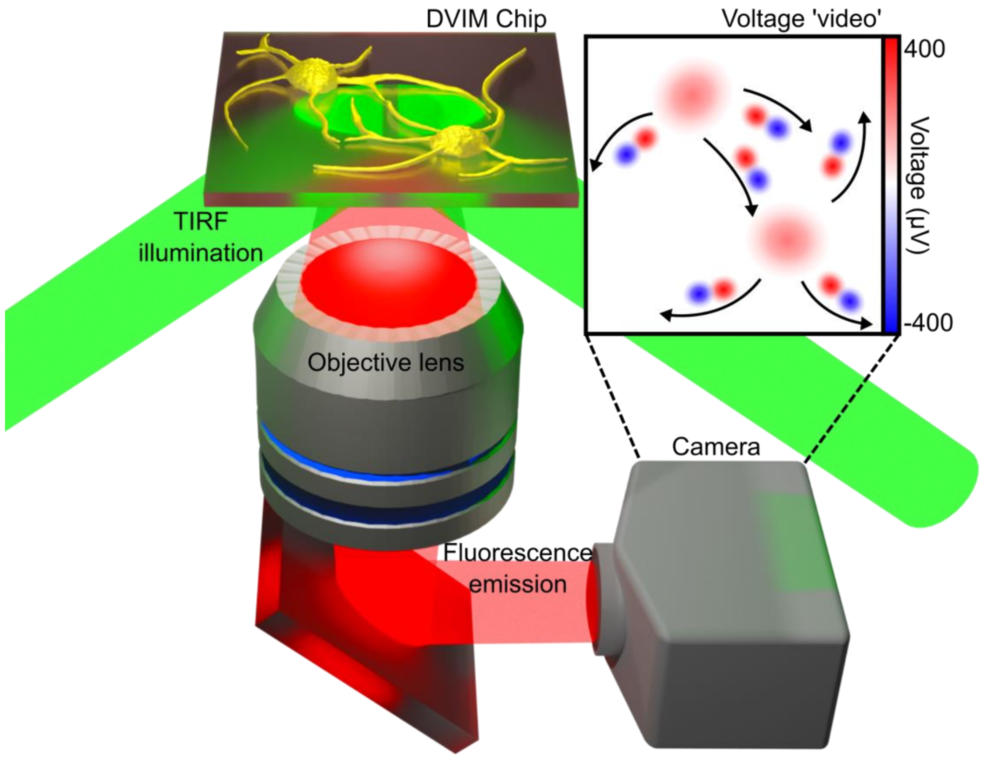
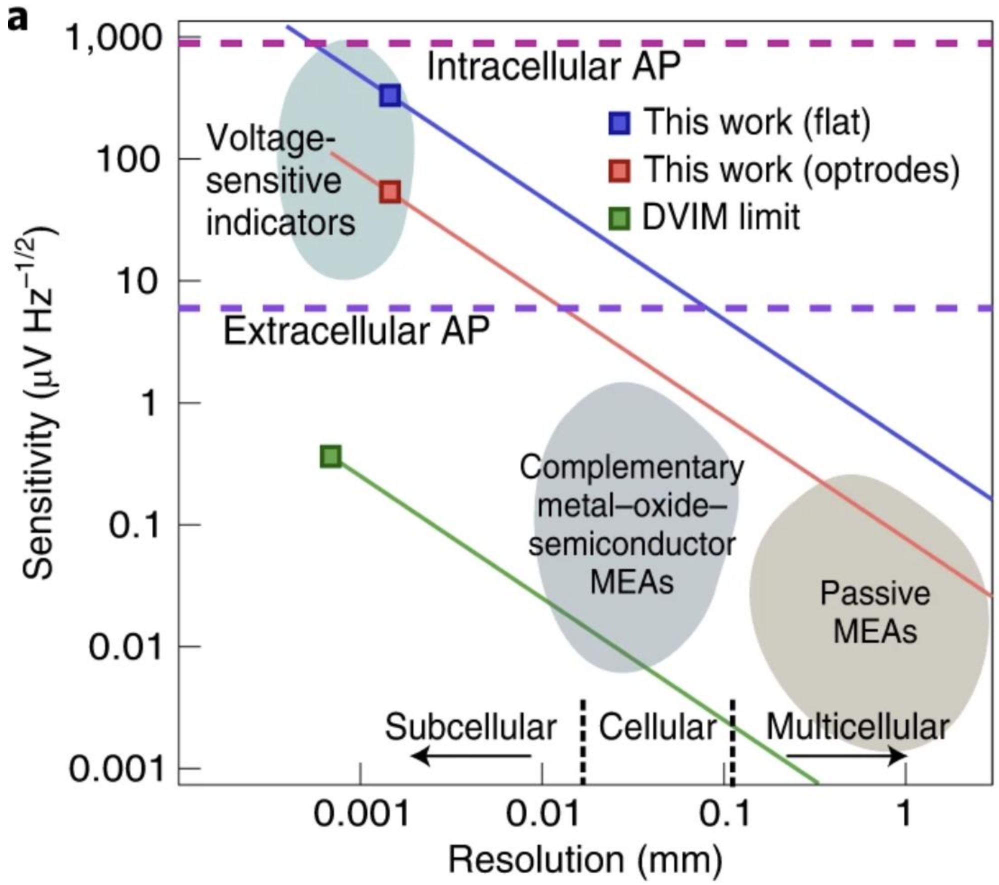


Plasma Processing for:

A. Etching structures
B. Smoothing surfaces
C. Growing material
D. Surface termination

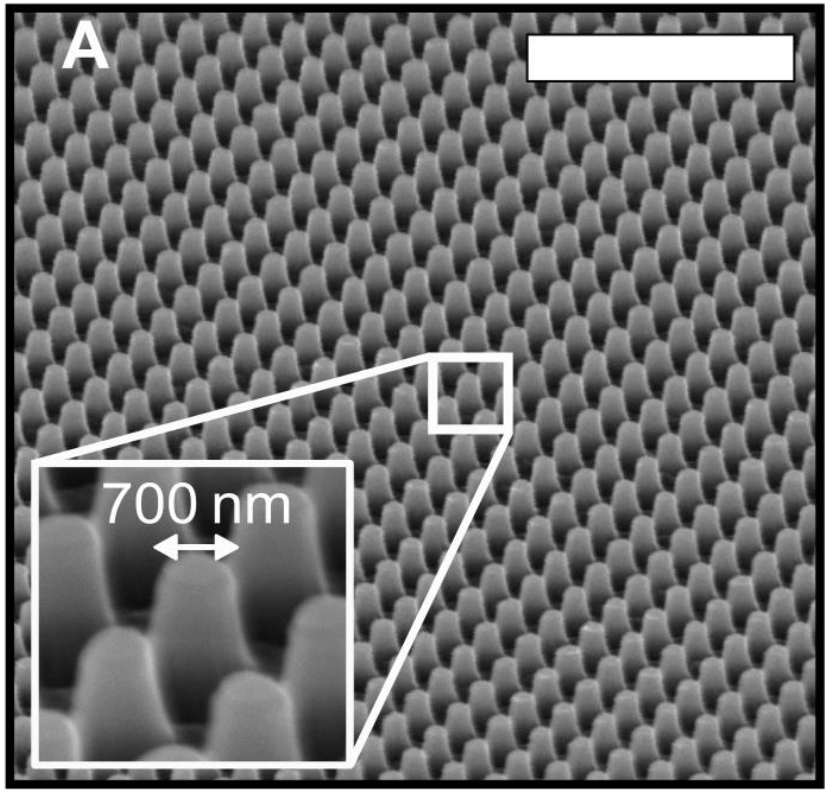
Plasma processing for QIS: Photonics

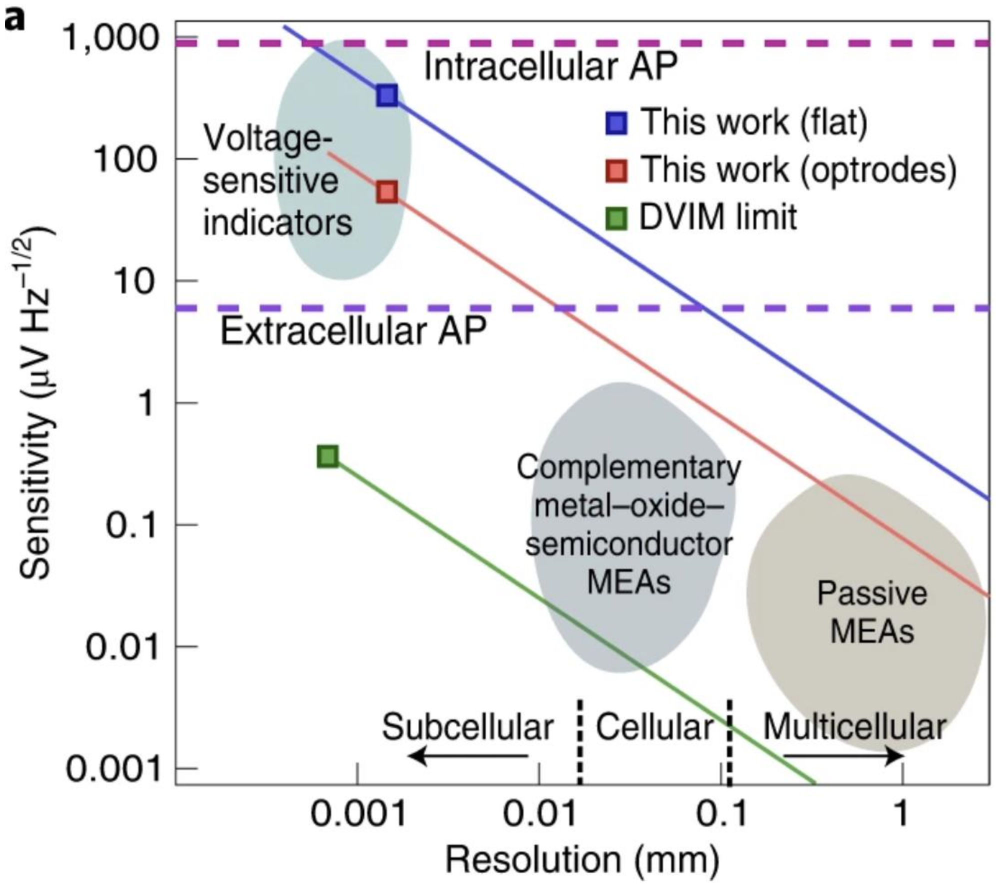




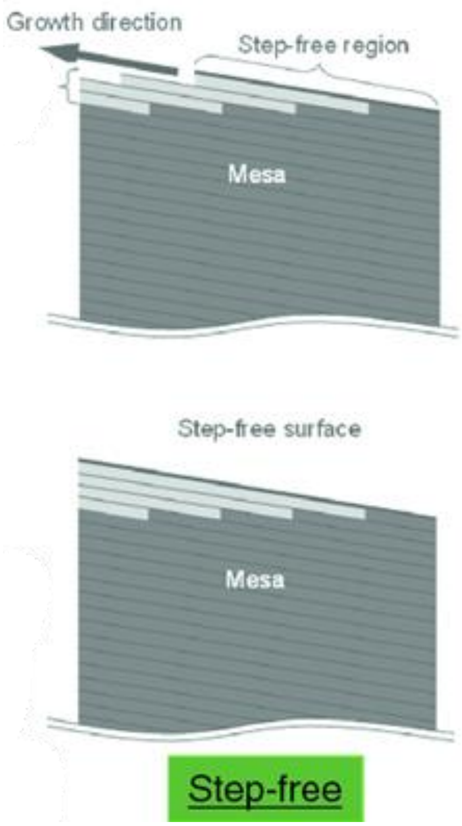
https://doi.org/10.1038/s41566-022-01064-1

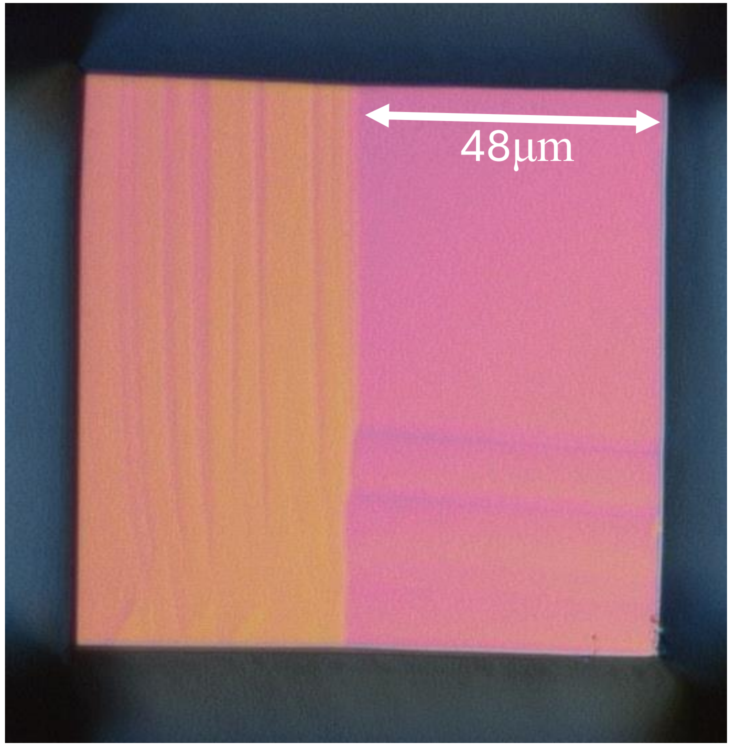
Plasma processing for QIS: Photonics





Plasma processing for QIS: Surface Smoothing



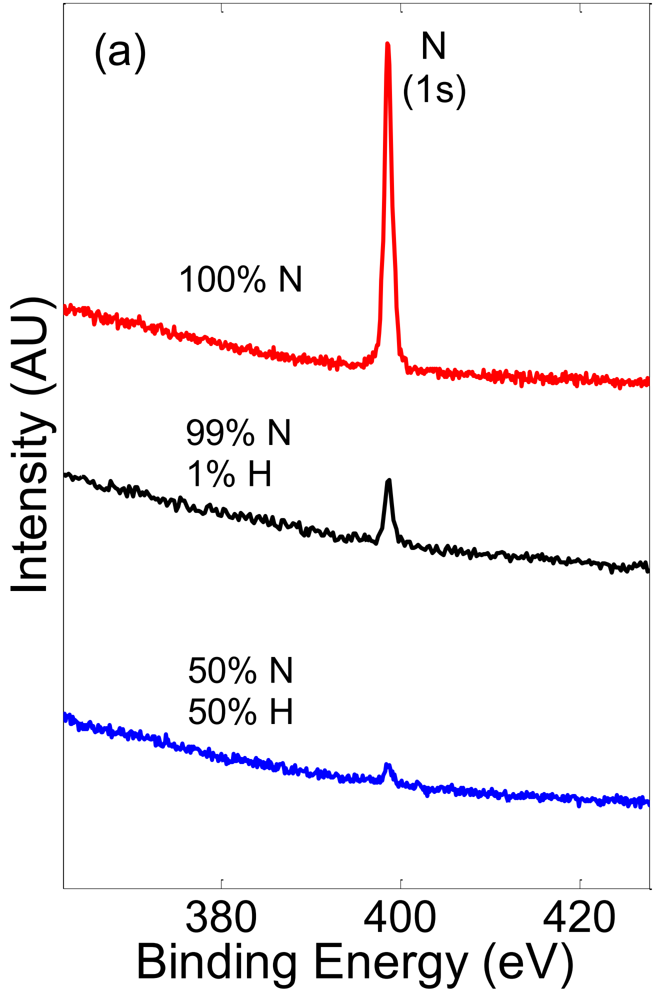


Credit: James Belcourt (RMIT University)

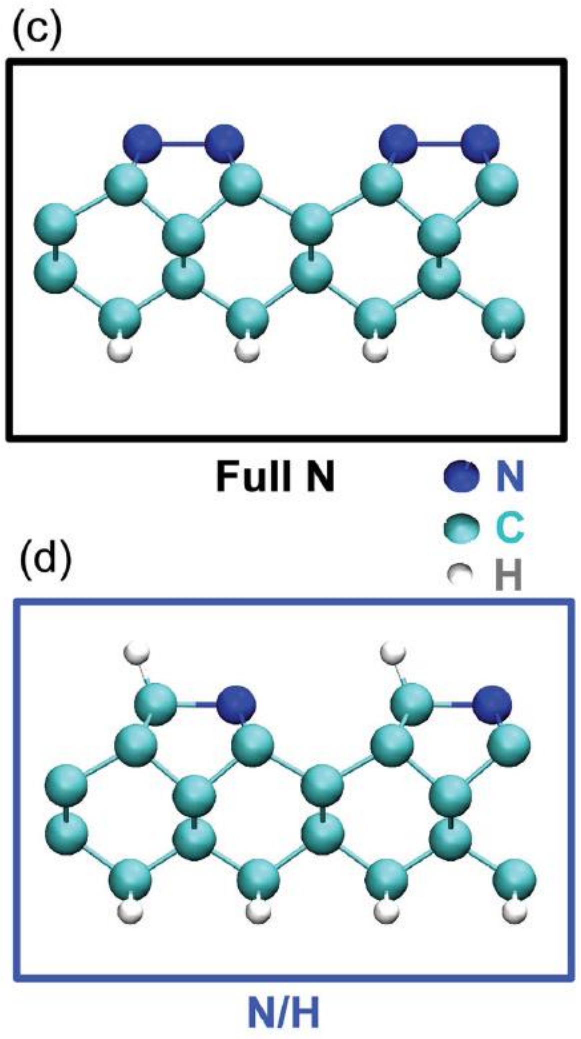
Technique proven on {111} diamond

Tokuda, N., et al., *Topics in applied physics* (2019) pp. 1–29

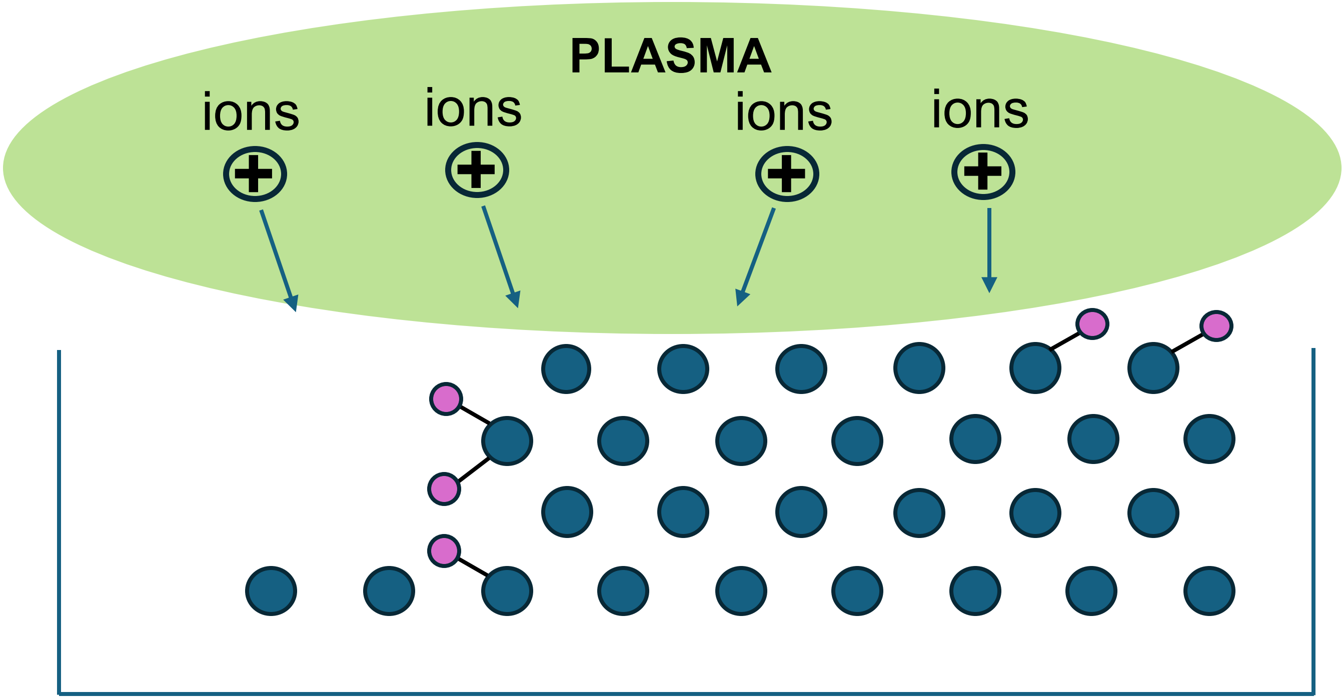
Plasma processing for QIS: Surface Termination



Mixed N and N/H termination 'observed'

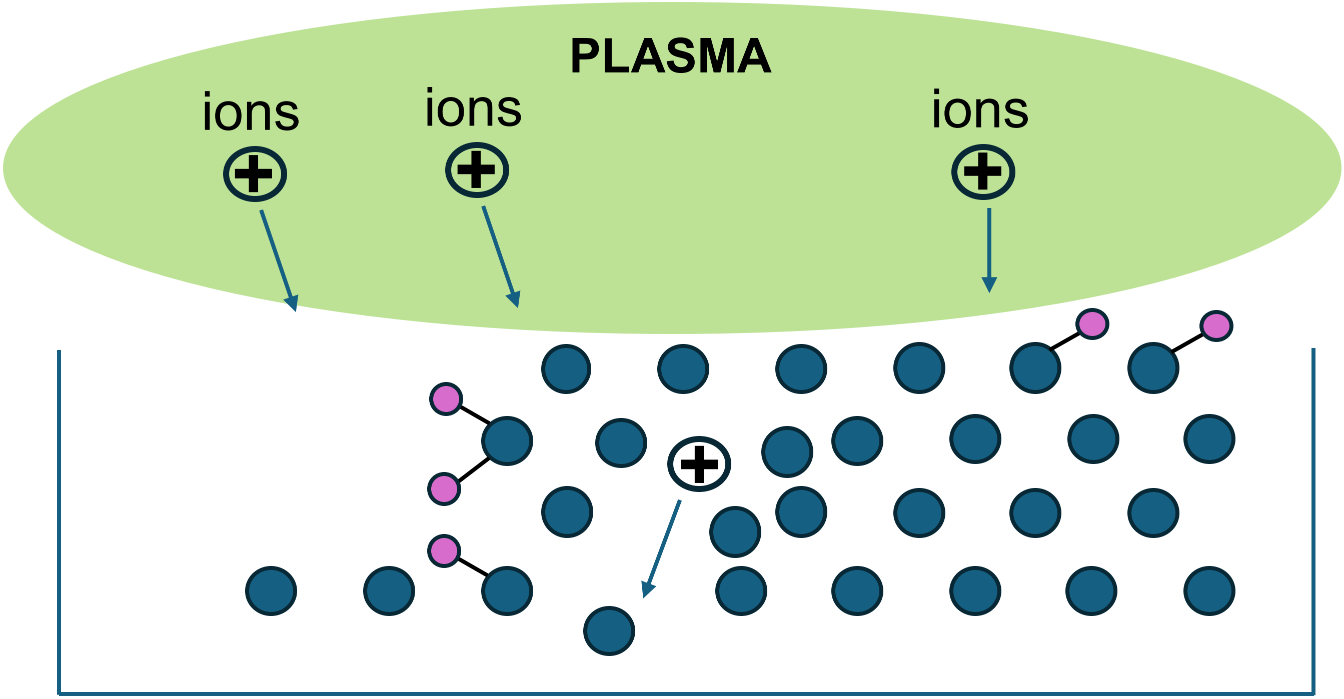


QIS: Surface and Damage Sensitivity



#1 Surface Roughness and Chemistry

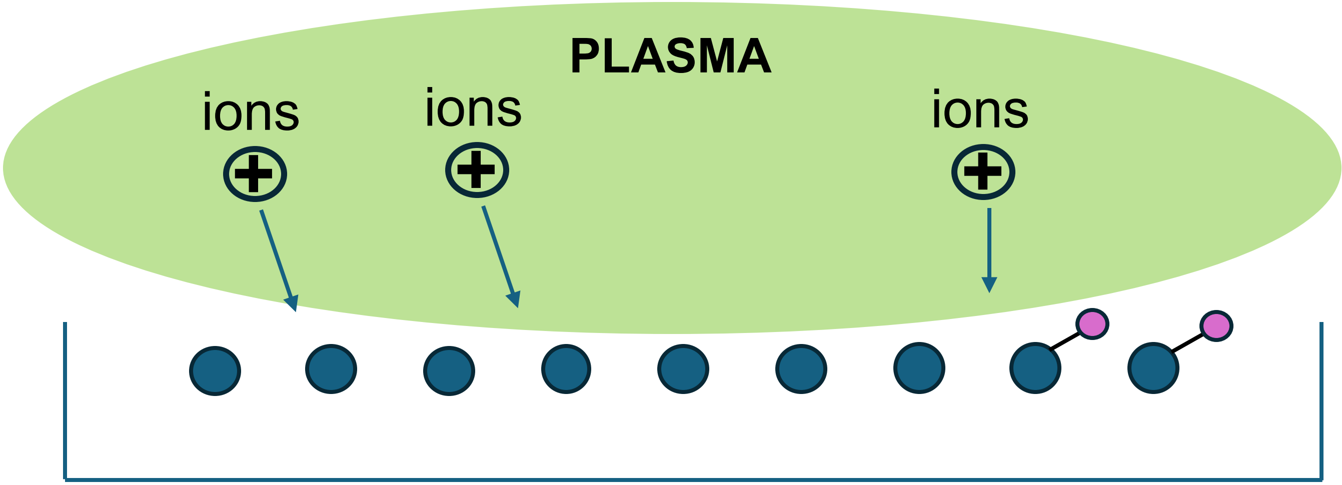
QIS: Surface and Damage Sensitivity



#1 Surface Roughness and Chemistry

#2 Ion Damage May be Unrecoverable

QIS: Surface and Damage Sensitivity

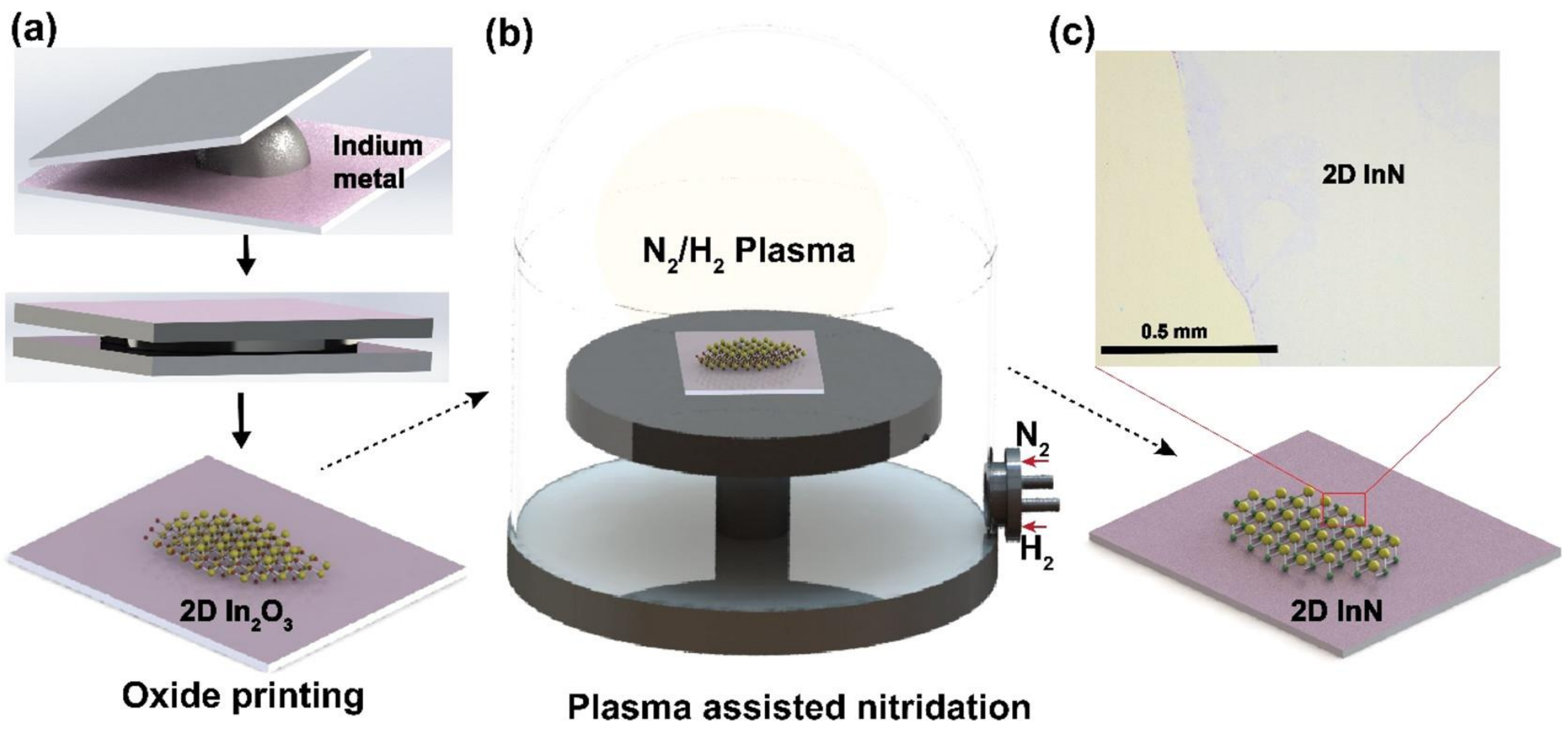


#1 Surface Roughness and Chemistry

#2 Ion Damage May be Unrecoverable

#3 Sample May be THIN

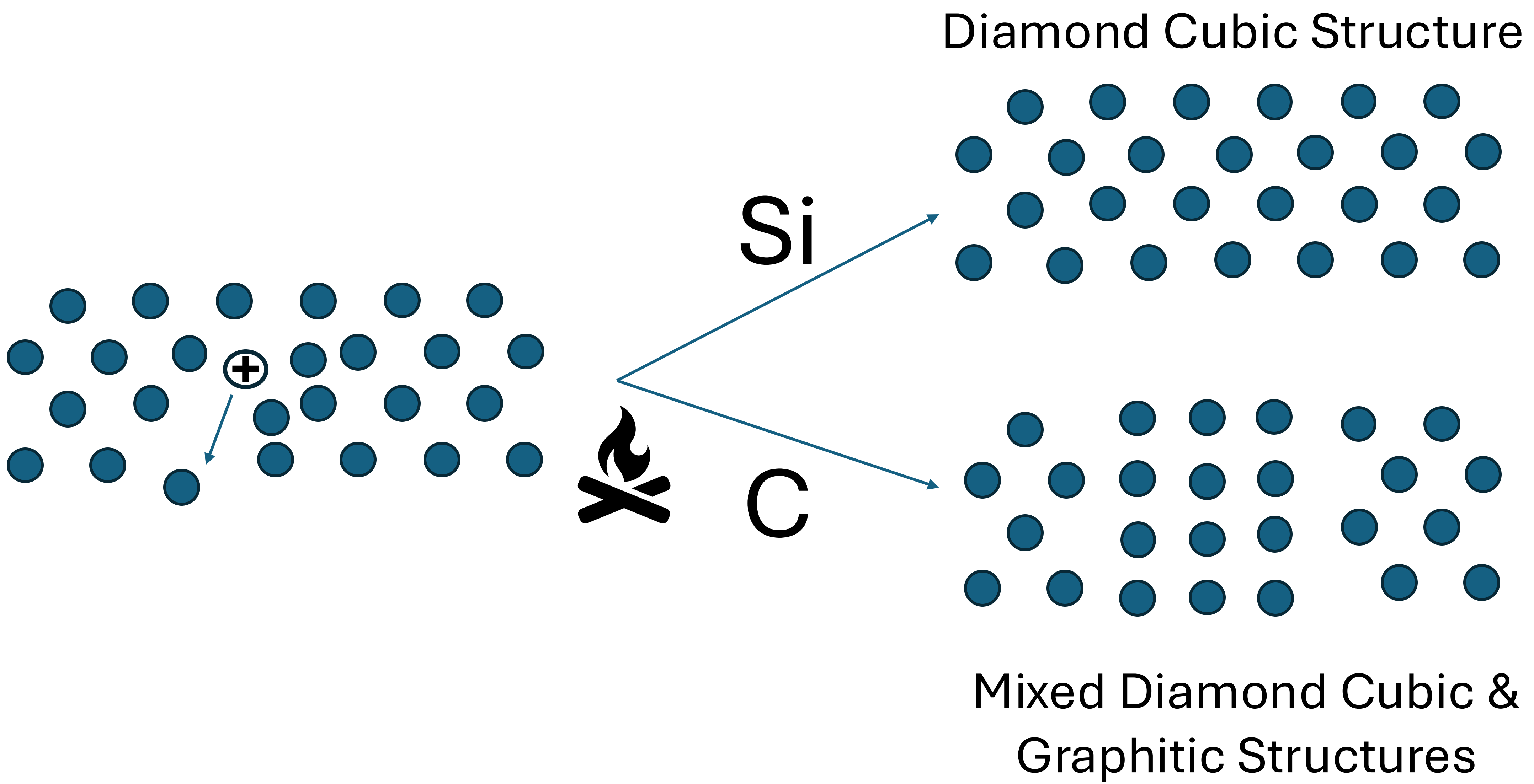
#3 Example: Sample may be THIN



~6 atomic layers thick

https://doi.org/10.1021/acsnano.1c09636

#2 Ion Damage May be Unrecoverable



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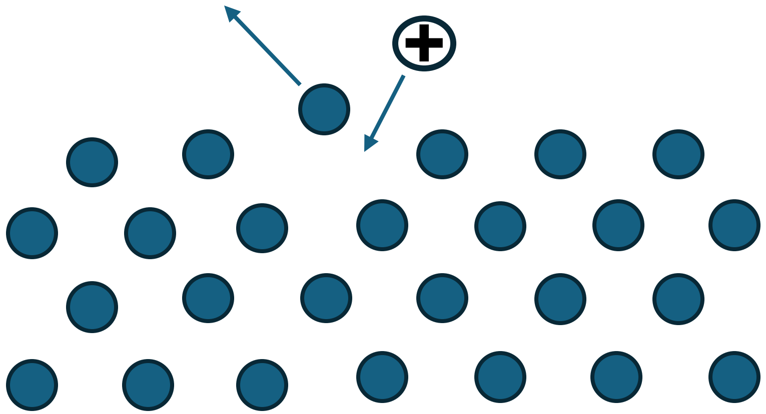
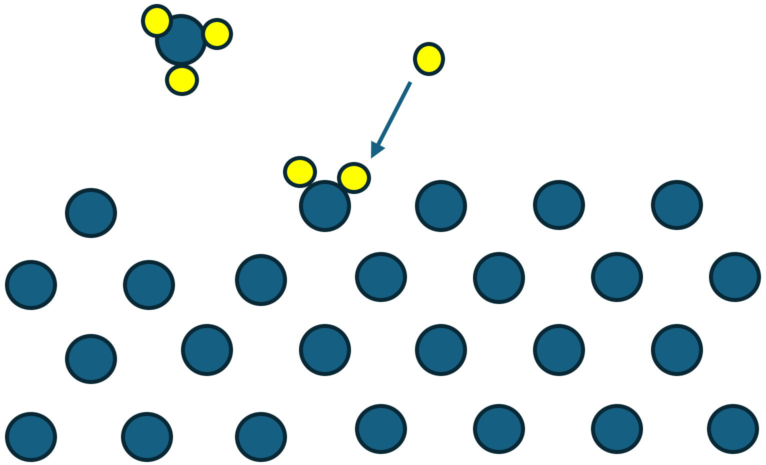




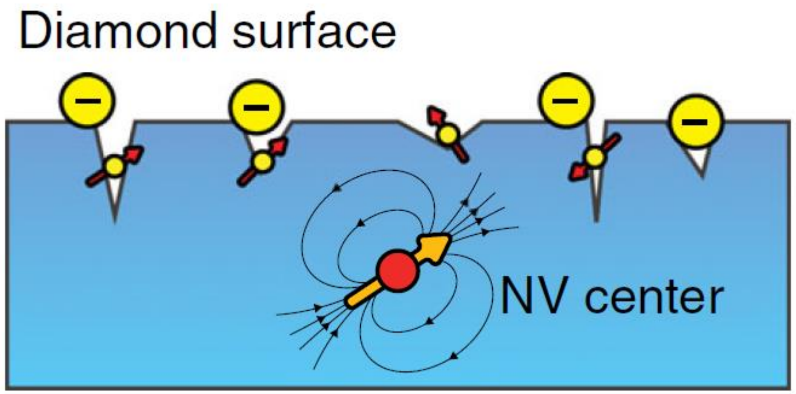
Option 1: Shallow Ions Damage

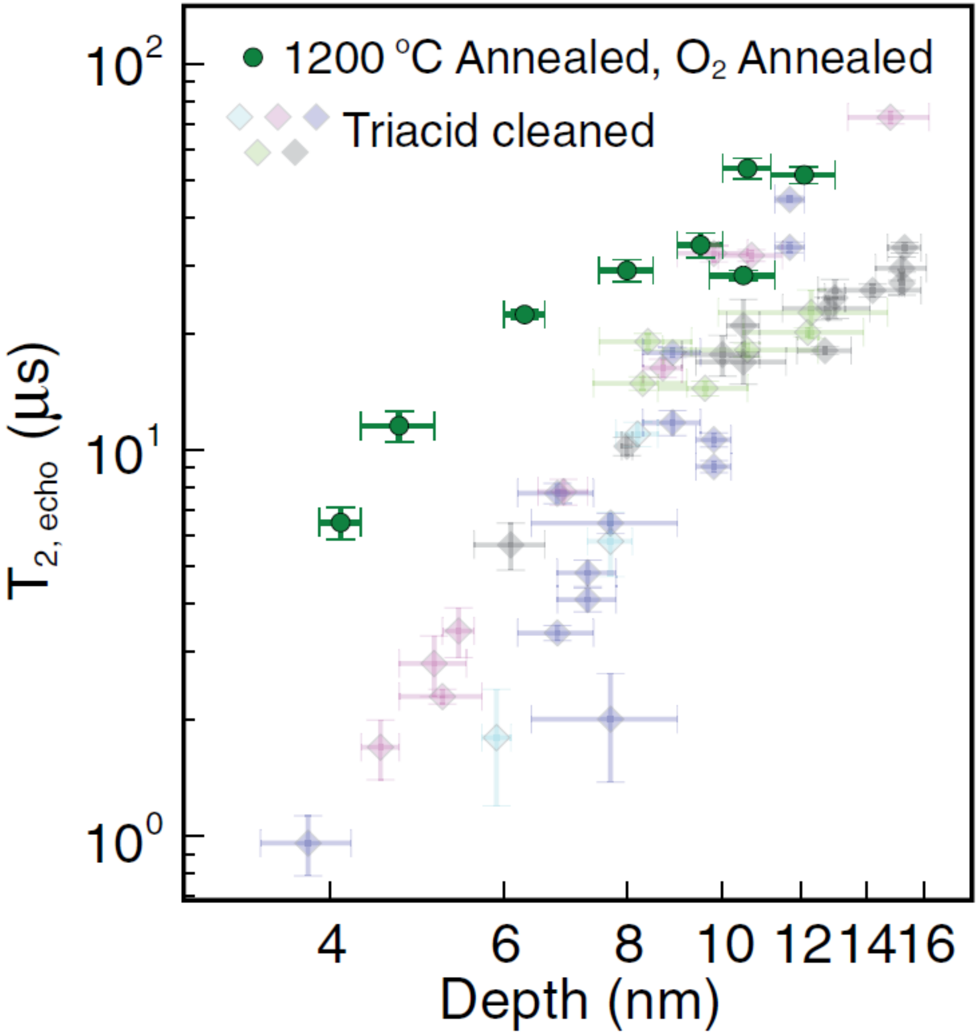
- Low Energy Ions OR Clusters
- Difficult or Slow Etching

Option 2: Chemical Etching

- Bespoke Chemistry
- Anisotropy Likely

#1 Surface Roughness and Chemistry





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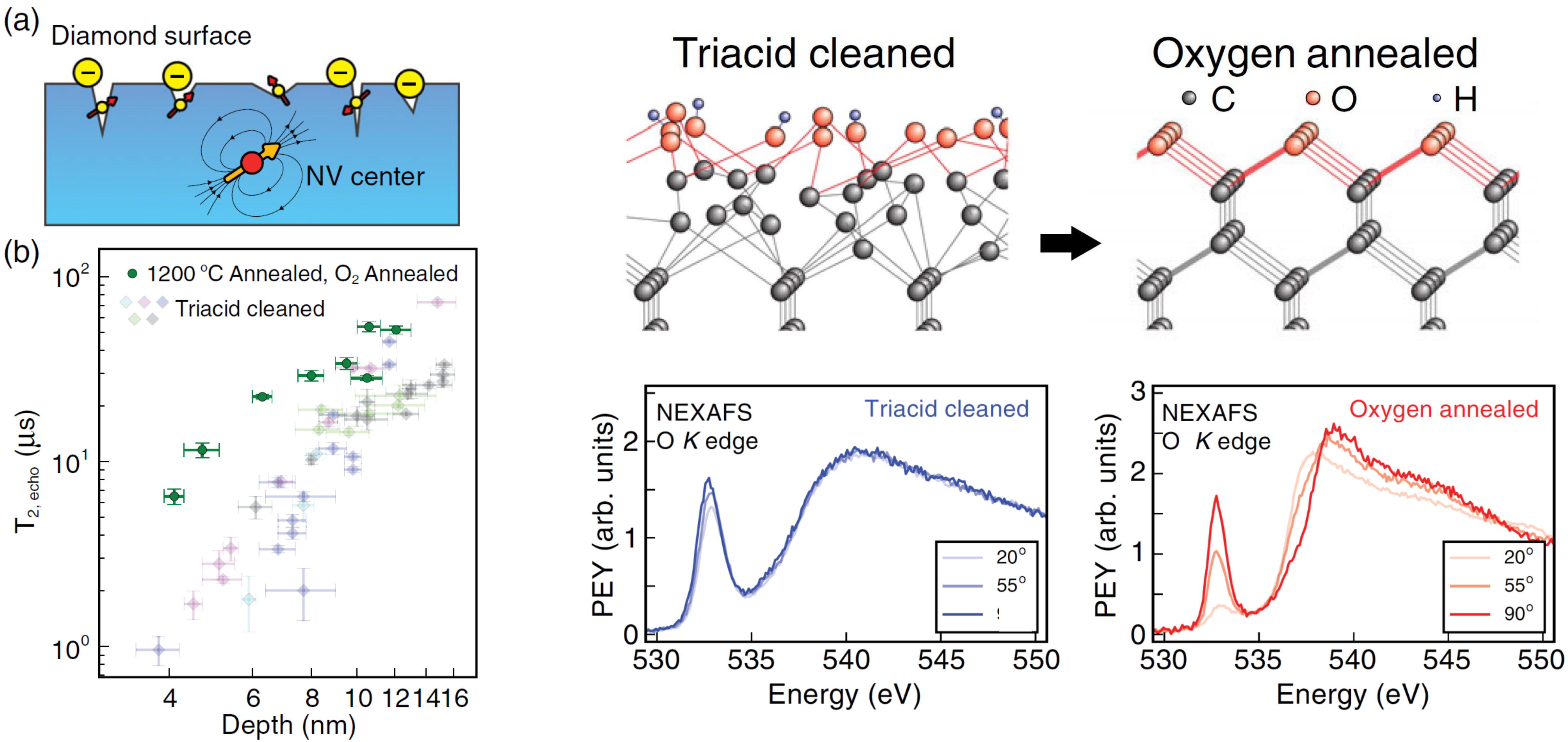

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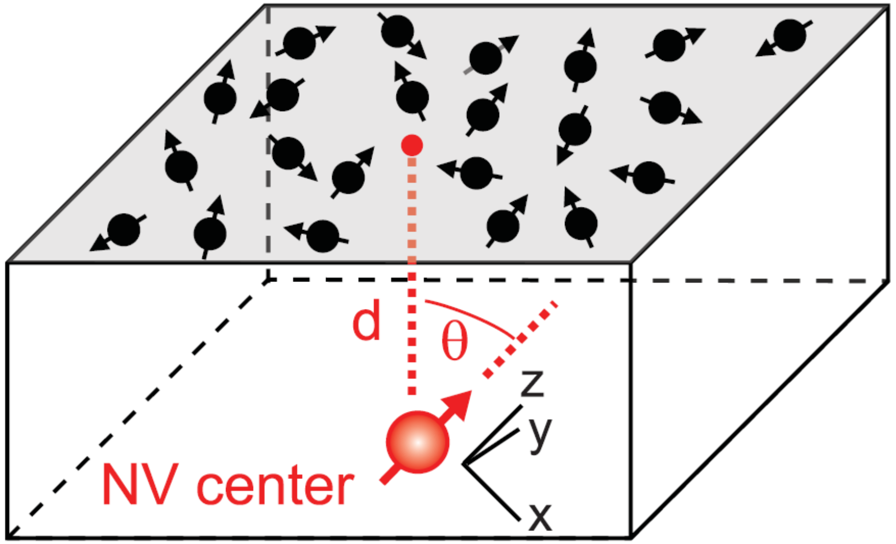


T. Rosskopf, *et al.*, PRL **112**, 147602 (2014)

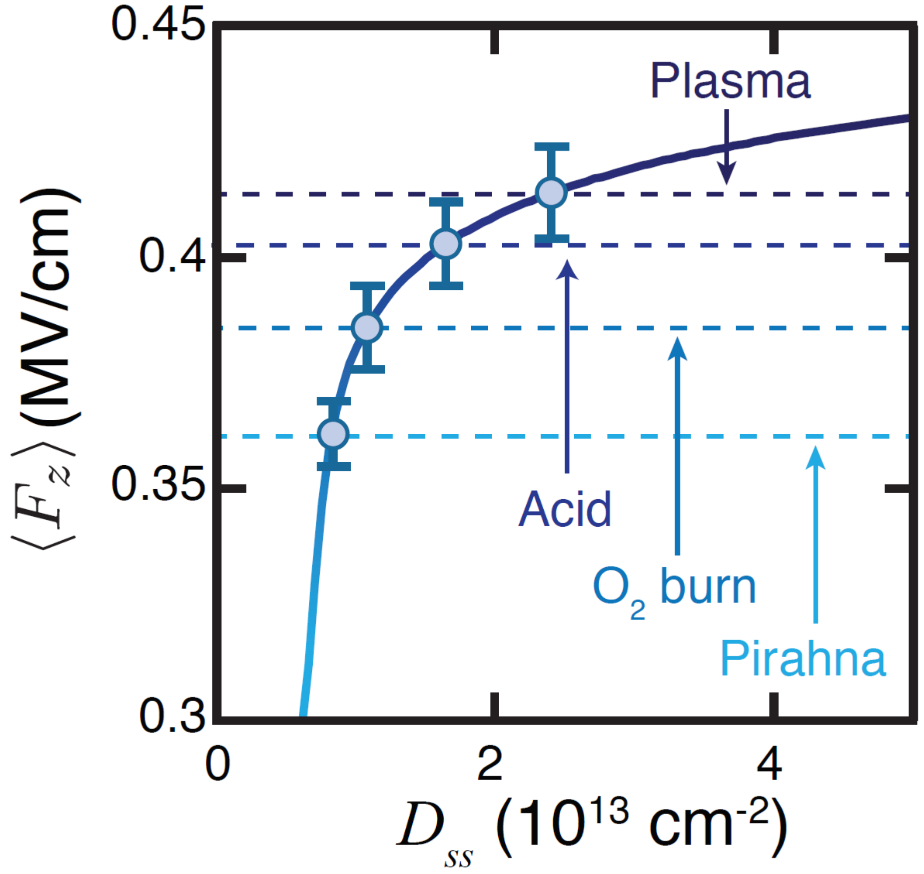
0.01-0.1 μ_B/nm^2

B. A. Myers, *et al.*, PRL **113**, 027602 (2014)

0.04 spin/nm²

Y. Romach, *et al.*, PRL **114**, 017601 (2015)

2-3 nm electron spacing



D_{ss} = surface trap density model

D.A. Broadway, et al., Nat. Elec. **1** (2018) 502-507

Diamond 0.01 / nm² = 10^{12} cm^{-2}

c.f. Silicon/SiO_2 ~ 10^9 cm^{-2}

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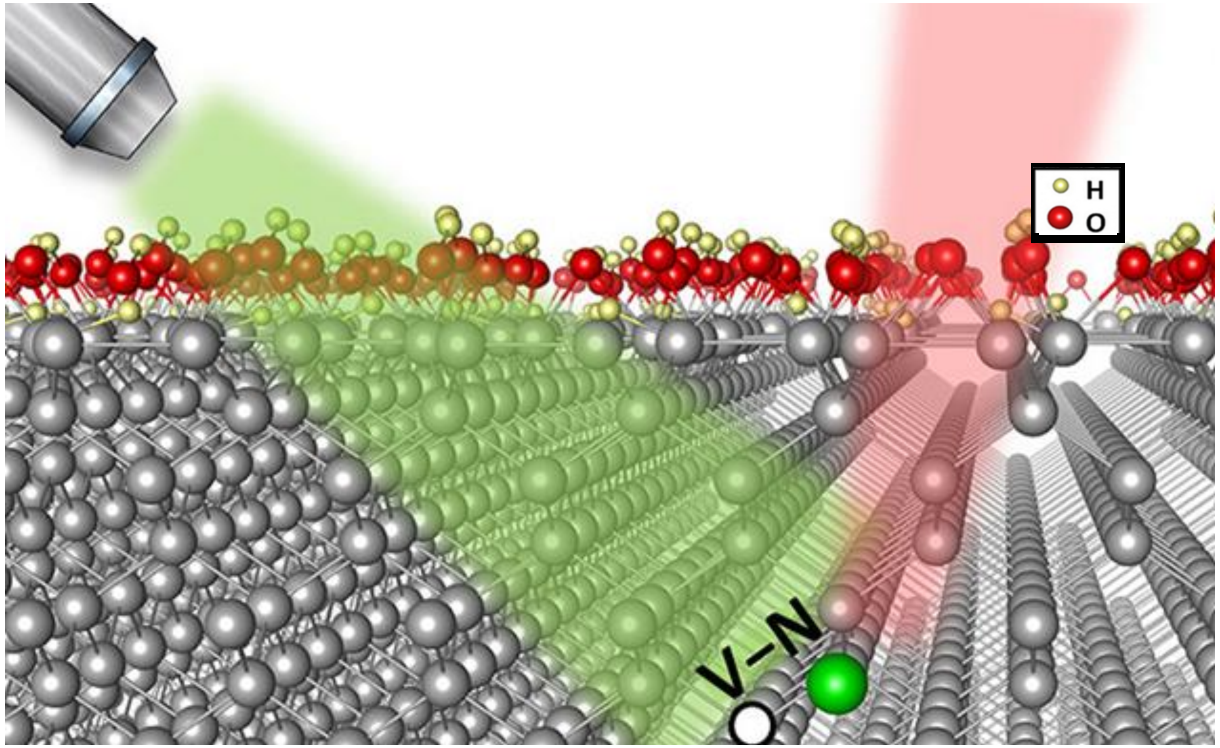

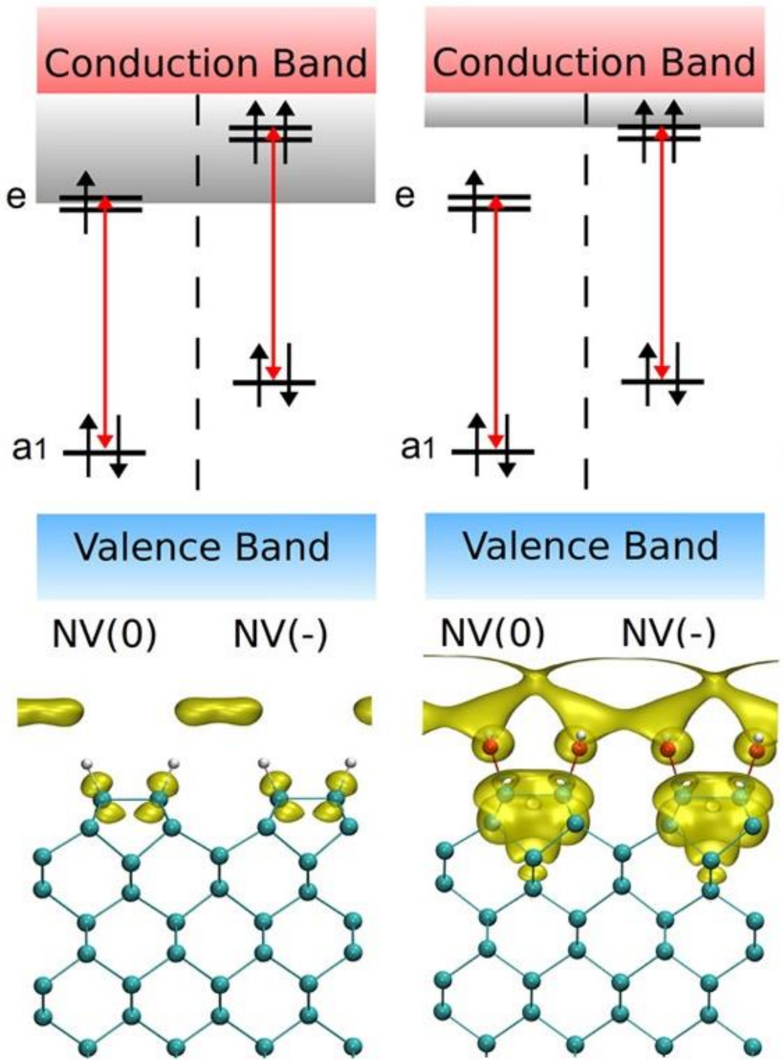

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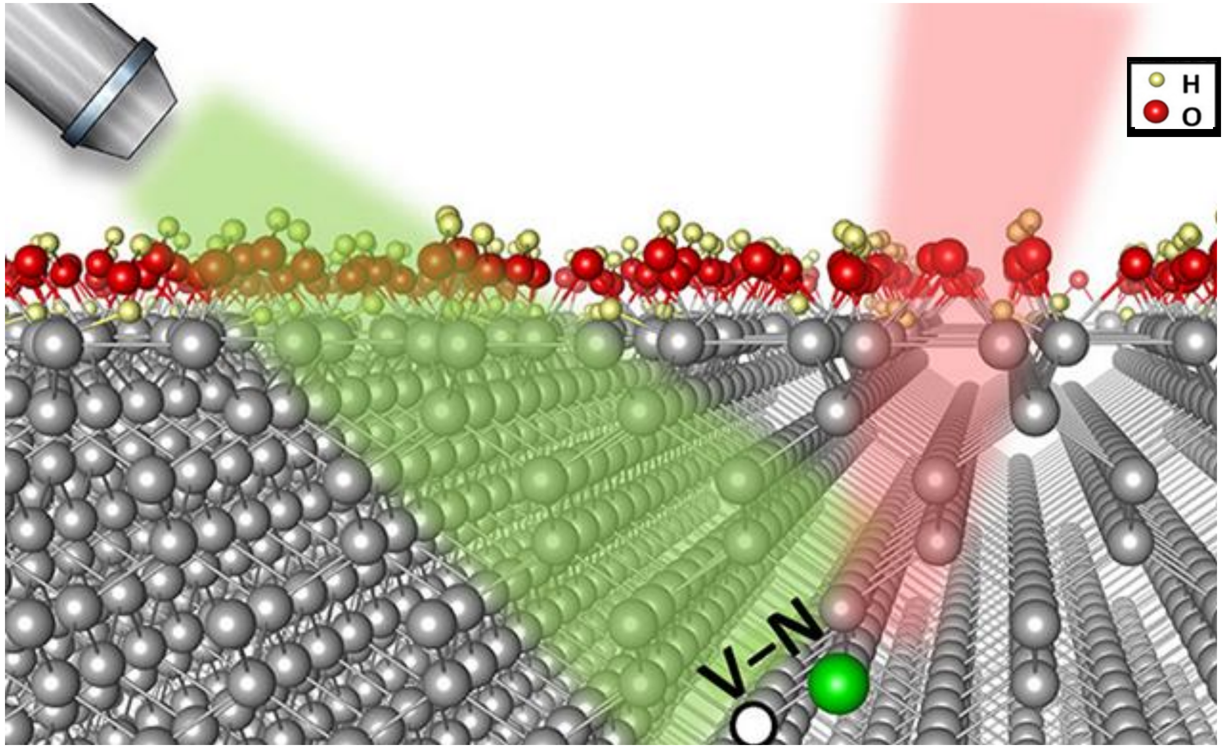

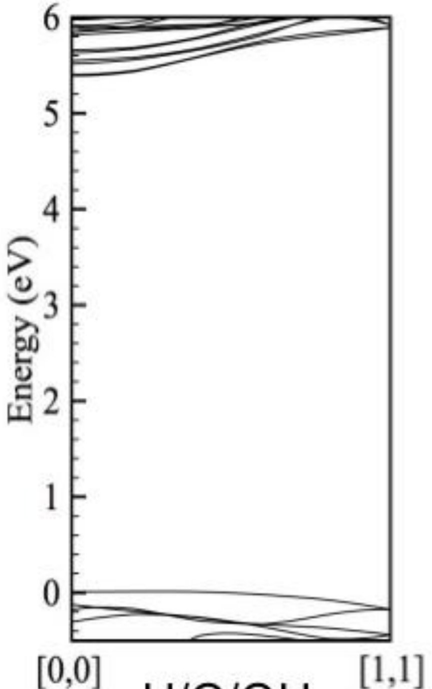

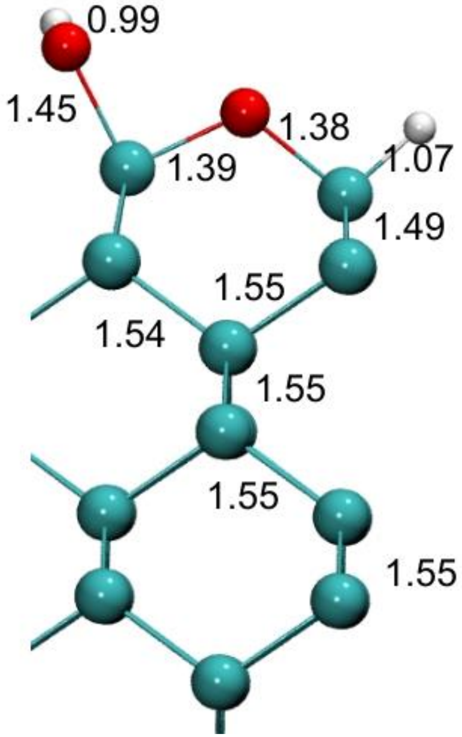

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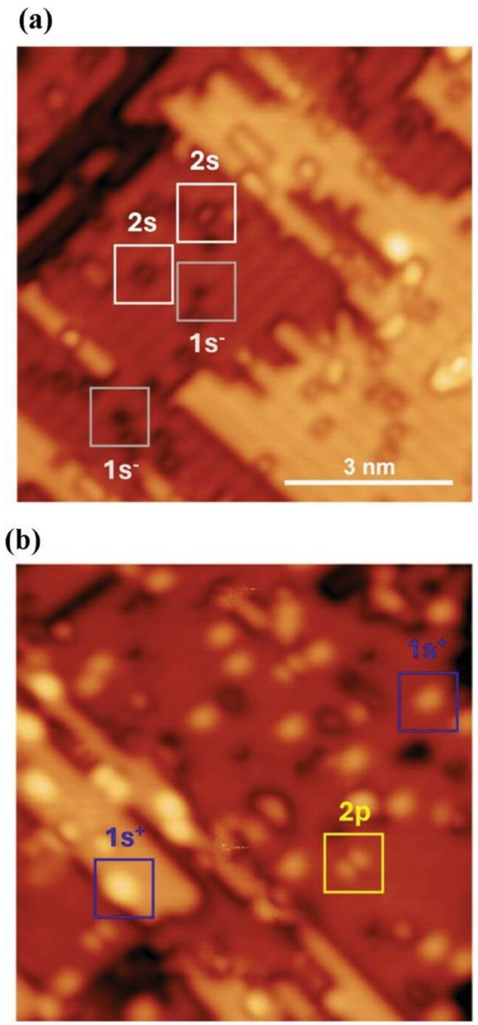
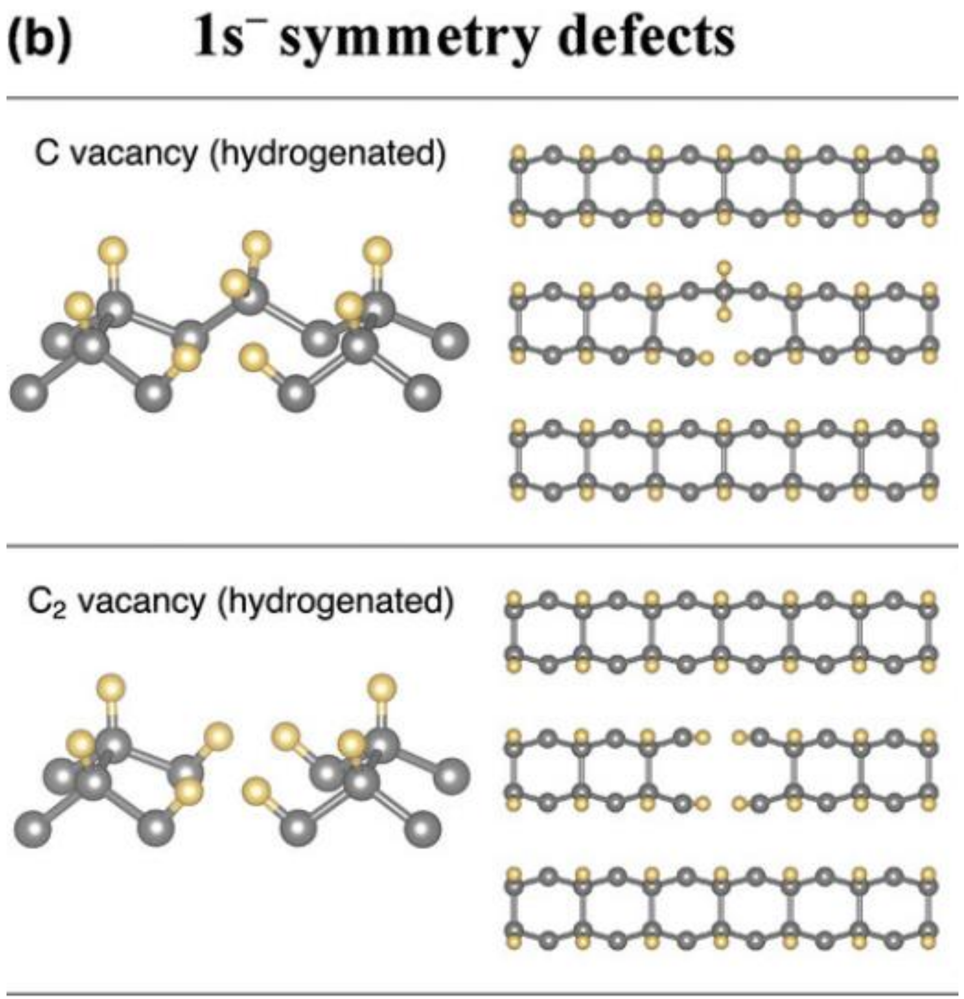




doi.org/10.1002/admi.202400695

~3nm terraces

~5% sites are step-edges

Plasma Processing for QIS

- QIS devices extremely sensitive to defects, including magnetic
- Kinetic vs Chemical etching
- Etch, growth & surface termination chemistries
- Charge, spin & optical-transition defects

#1 Surface Roughness Affects Chemistry

#2 Ion Damage May be Unrecoverable

#3 Sample May be THIN

UNUSED SLIDES

Intro to QIS

- Dirty problems with QIS – importance of the surface
  - Superconductors
  - Trapped ion systems
  - Diamond surfaces
  - Silicon surfaces?
- Solid state quantum vs ME
  - Similar materials
  - Similar electrons
  - Similar device construction (layers, E-field control, RF control)
  - Different information encoding → More sensitivity to device perfection
- Examples
  - CMOS transistor (lots of electrons) – insensitive to magnetic field
  - Superconducting qubit (one photon limit) – sensitive to tiny number of electric or magnetic dipoles
  - Spin qubit (two electrons) – sensitive to one nearby electron or a few nearby nuclear spins

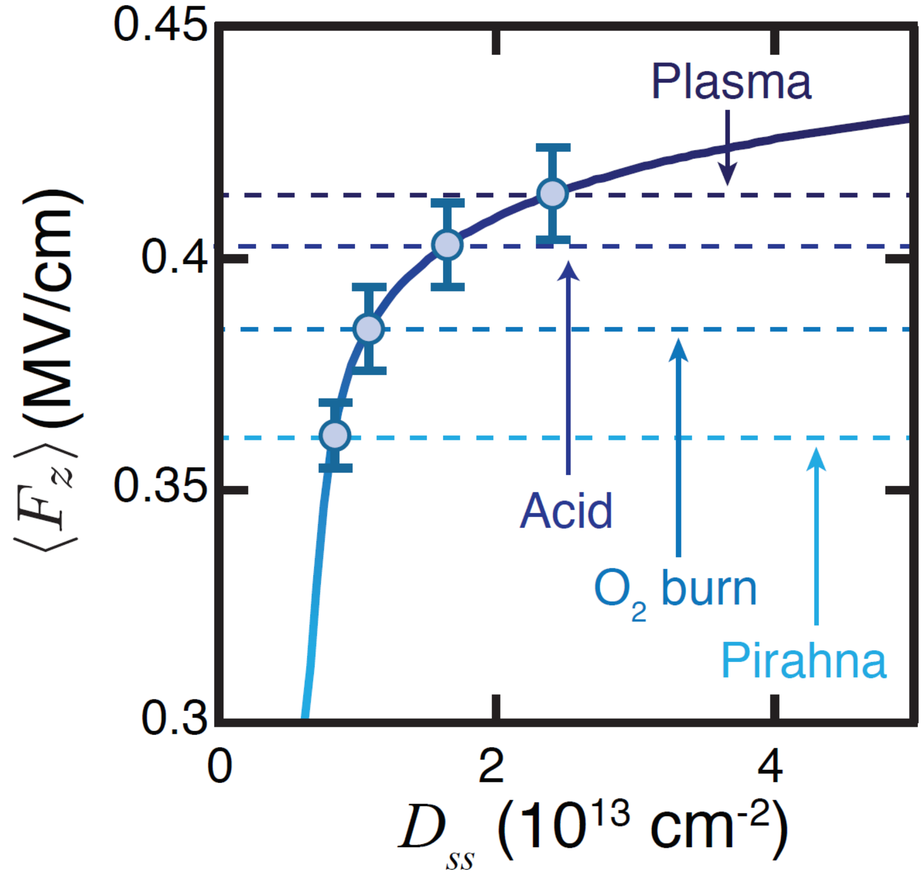
Diamond QIS/surfaces intro

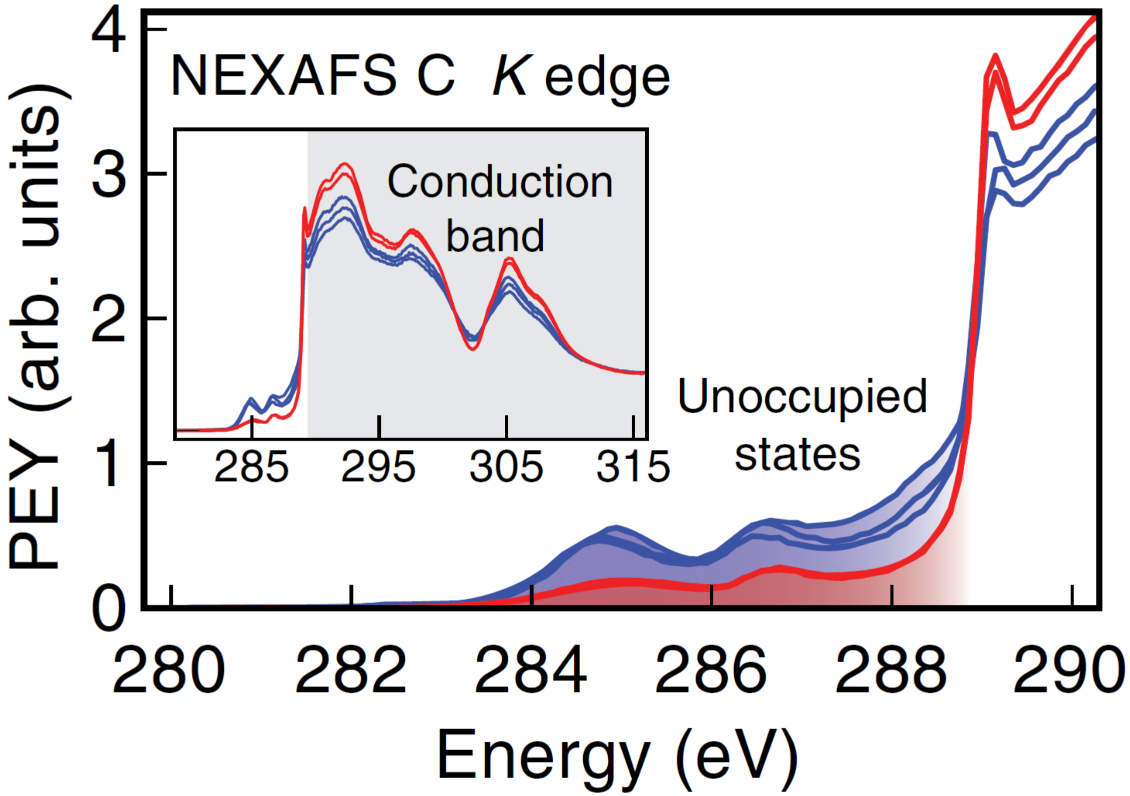
- QIS Example: Diamond quantum systems
  - NV centre – Hamiltonian sensitivity to everything
  - Host material properties
  - Coherence vs field (T2*)
  - Introduce coherence concept (phase accumulation at least – precession argument is sufficient)
  - --- skip T2 but maybe have a slide?---
  - Difference between spin and charge effects
    - (both matter, but spin often accompanies charge)
  - ppm concentrations cause problems
- Diamond QIS applications
  - DVIM
  - Bulk sensing
  - Single-molecule MRI
- Role of surfaces
  - Surfaces affect electronic states
    - Gali work etc.
  - Current estimate of surface spin/electron density
  - Surfaces vs PLE (spectral diffusion)
  - Surfaces vs coherence
- What do real surfaces look like?
  - STM images
  - Range of chemistries we've achieved – mainly non-plasma

Diamond QIS plasma processing

- Beginnings of nanofabrication structures
  - Optical structures
  - PDMR structures
- Concept of chemical vs kinetic etching
  - Diamond is metastable… but needs ~30eV+ to damage lattice
  - Unlike silicon, can't use energetic ions and then 'fix' the lattice afterwards
  - Similarities with 2D materials
  - This removes an easy path to non-chemical isotropic etching
- Putting it together
  - Exact chemical state needs to be controlled
  - Crystal defects and impurities at <ppm levels
  - Damage often unrecoverable
  - Atomic precision can be critical (may require CMP and crystal orientation control)
  - Interaction between processed surface and ambient (or post-processing) also critical

Trap state measurements: Quantum sensor results





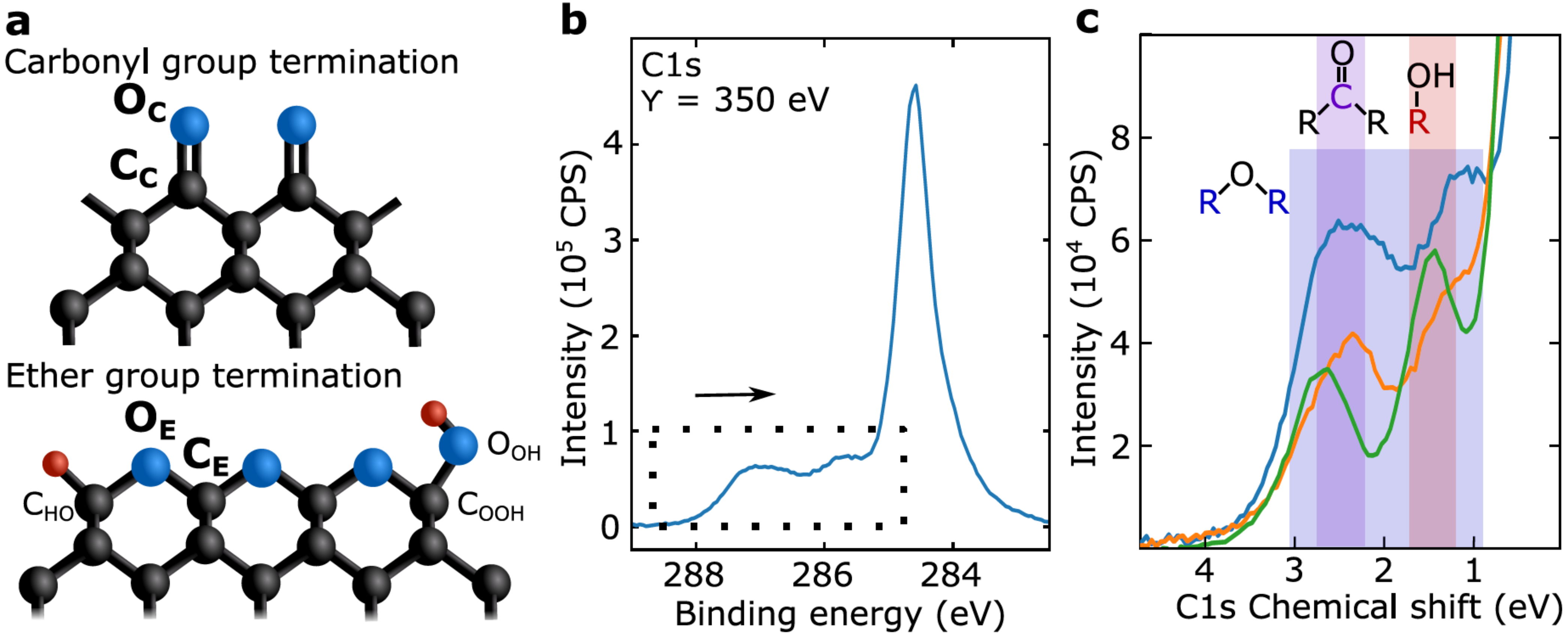
$\langle F_z \rangle$ measured by NV ensemble → D_{ss} density modelled

D.A. Broadway, et al., Nat. Elec. **1** (2018) 502-507

Sangtawesin, et al. PHYS. REV. X **9**, 031052 (2019)

→ Trap states estimated ~ 10^{14} /cm^2 → Filled states ~ 10^{13} /cm^2

Oxygen quantification with XPS?

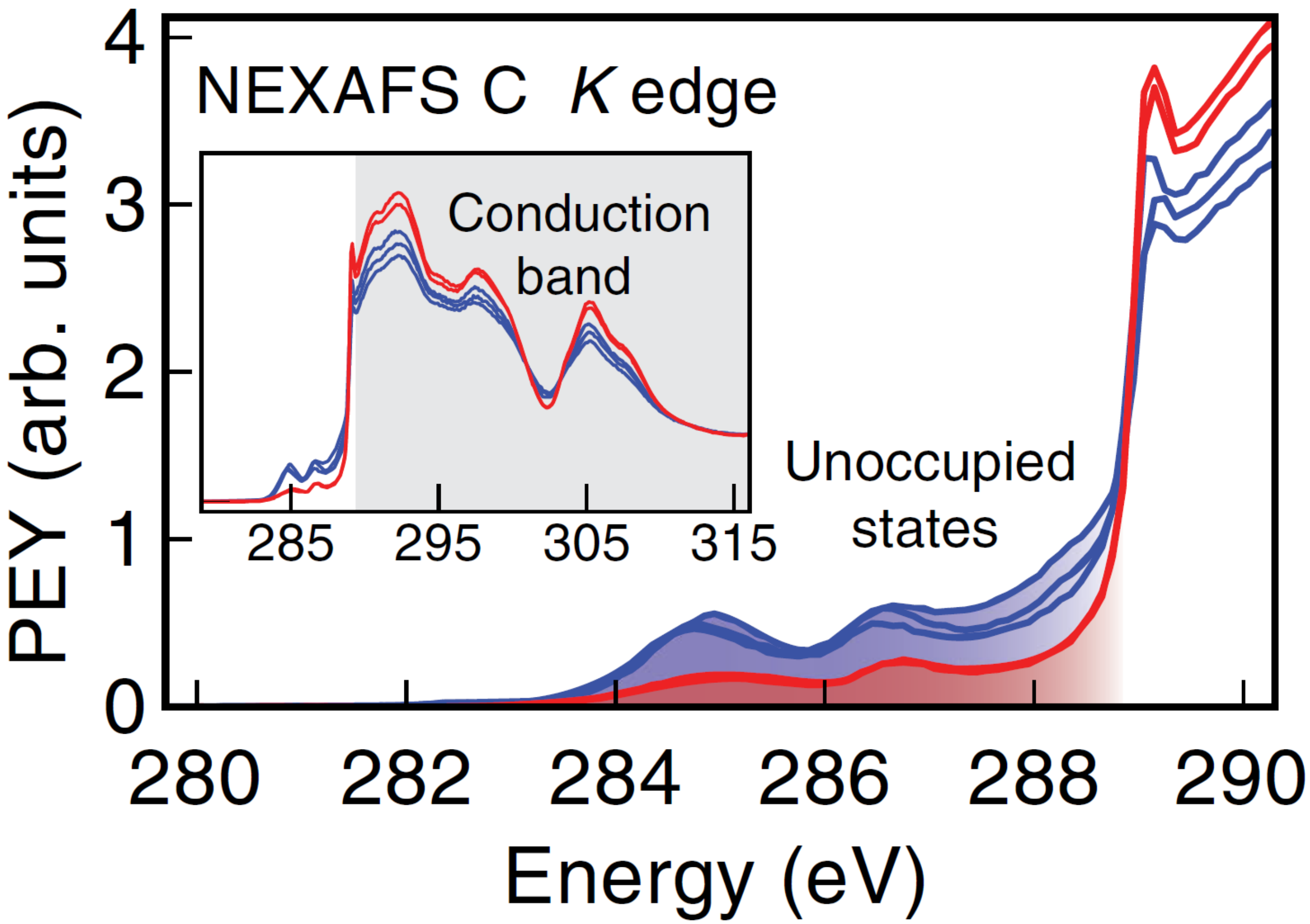


→ C1s XPS is difficult to ascribe to specific chemical states
→ C1s and O1s spectra are often inconsistent

Confirming oxygen chemistry is difficult

- **O-C-O may look identical to C=O**
- **C-H is not directly observed**

Chemical origins of surface noise



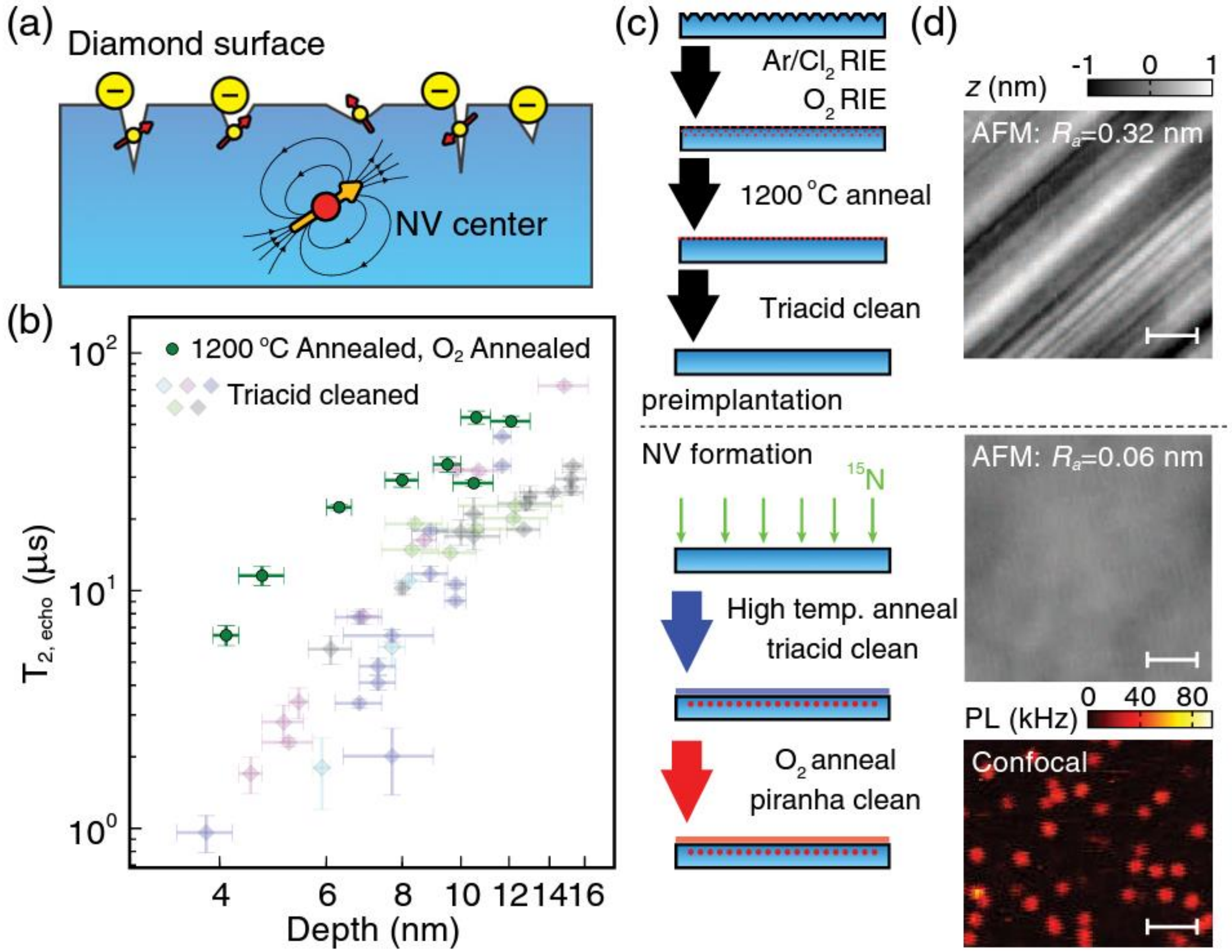
Oxygen annealed

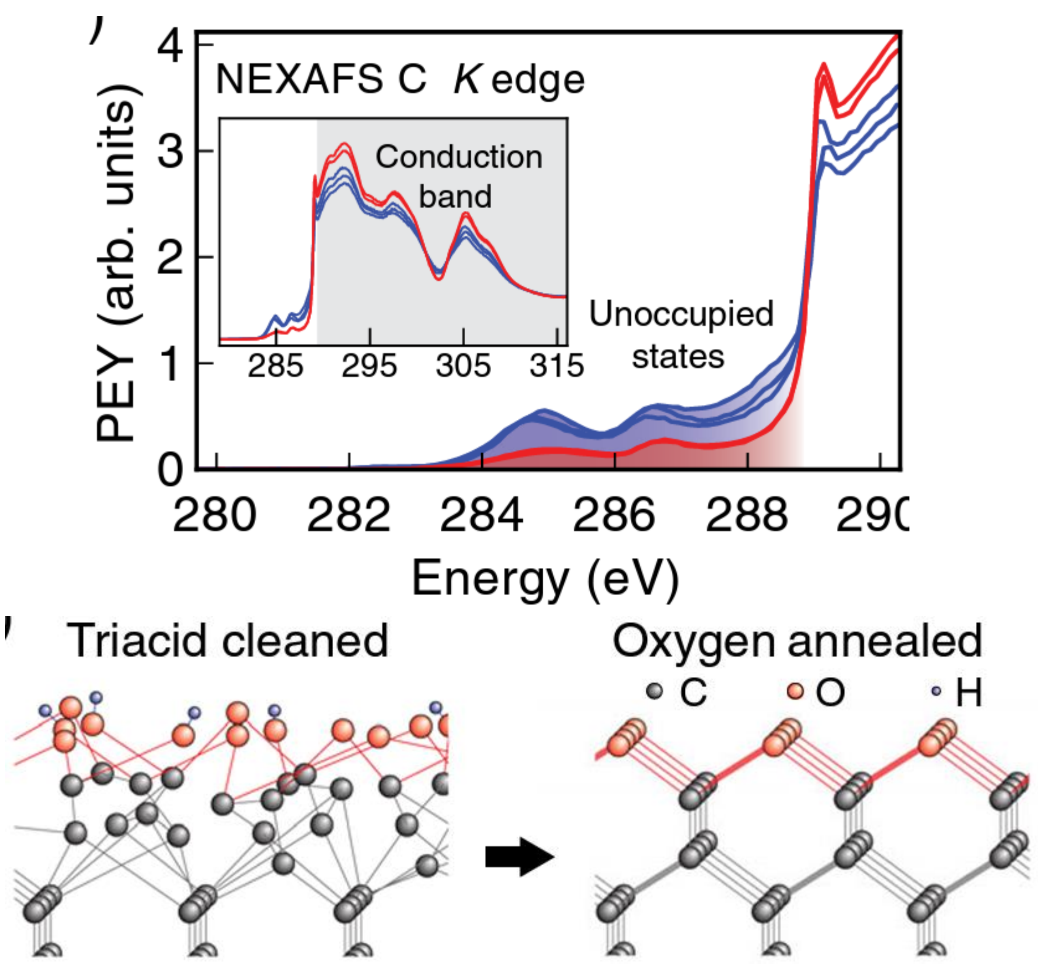
Triacid cleaned

Better oxygen process
→ Decrease in unoccupied states
→ Increase in coherence

→ C=O related trap states quantified (4 - 15% ML ≈ 10^{14} /cm^2)
→ C=C related sites 2 – 15% ML

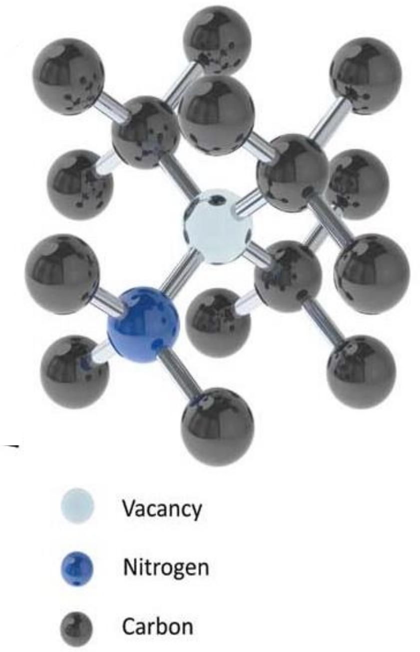
Engineered surfaces to reduce surface traps

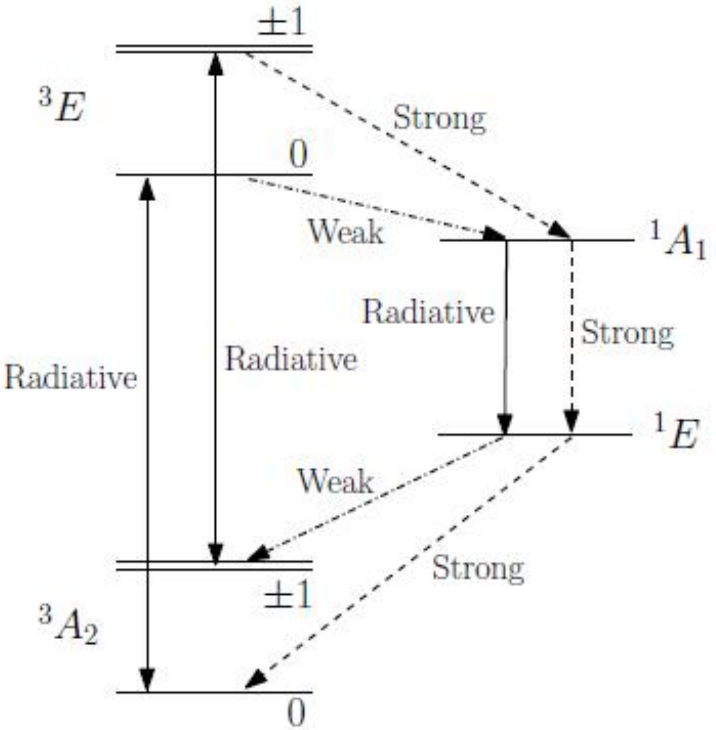




Sangtawesin, et al PRX 2019

Solid state





lightboxjewelry.com

Atomic-like emission and absorption spectra

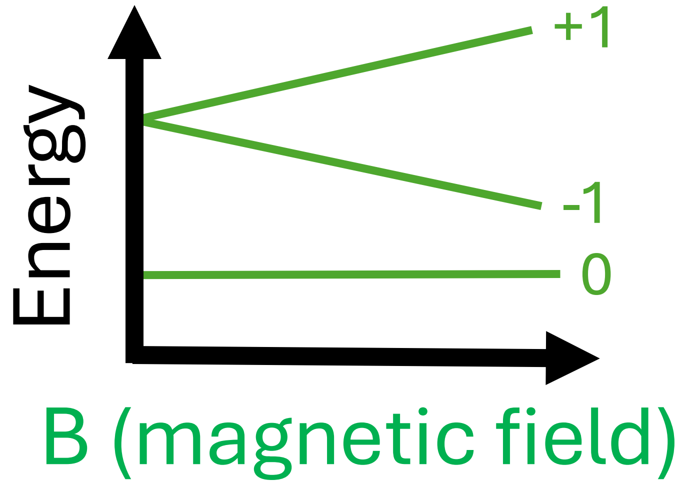
→ Colour in gems

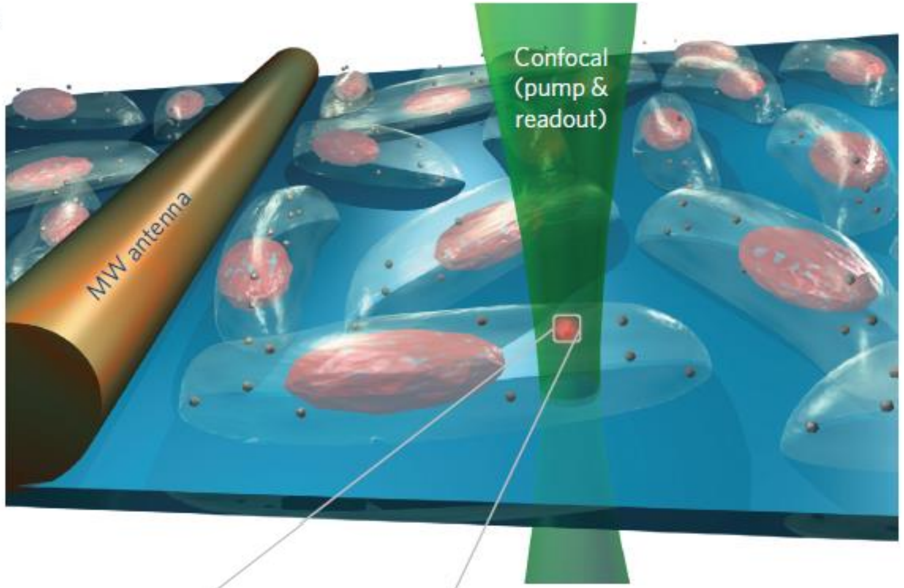
→ Single defects can generate single photons

→ Quantum-secure communications/communication

→ True single-photon interference

Quantum Sensing at the atomic scale



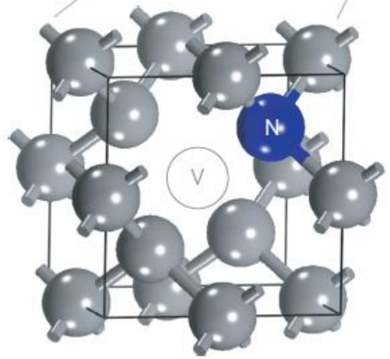


$$H = D(T)S_Z^2 \quad + \gamma_e \vec{B} \cdot \vec{S} \quad + d_\parallel [E_Z + \mathcal{E}_Z(\overleftrightarrow{\sigma})] S_Z^2$$
$$+ d_\perp [E_X + \mathcal{E}_X(\overleftrightarrow{\sigma})](S_X S_Y + S_Y S_X)$$
$$- d_\perp [E_Y + \mathcal{E}_Y(\overleftrightarrow{\sigma})](S_X^2 - S_Y^2)$$

Zero-field splitting (T)

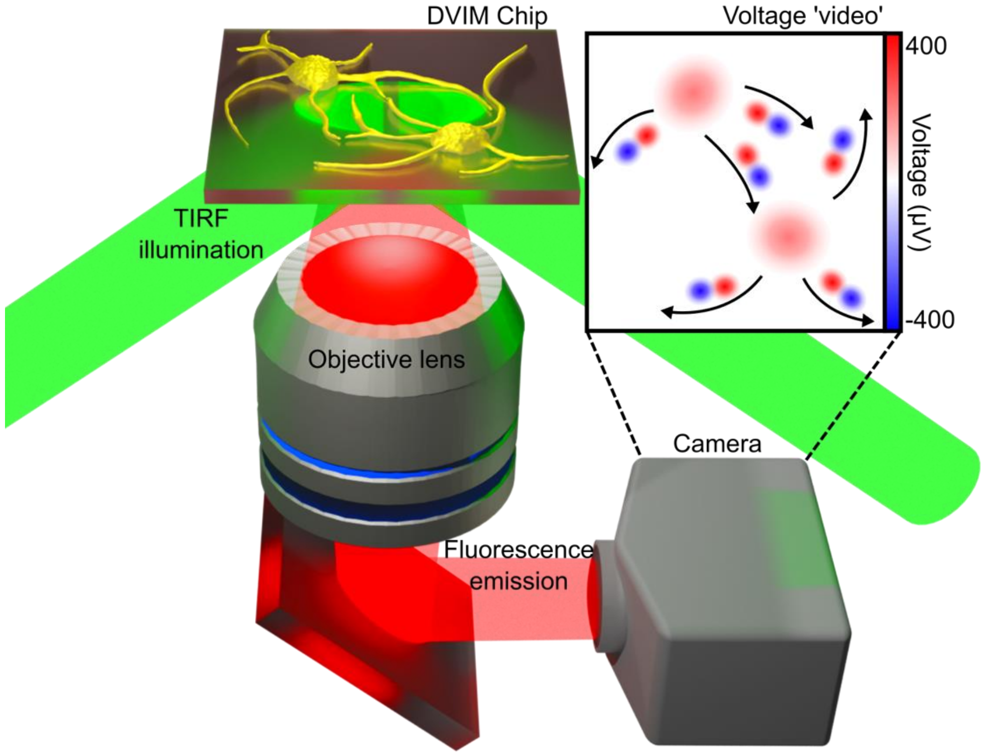
Zeeman effect $(\vec{B})$

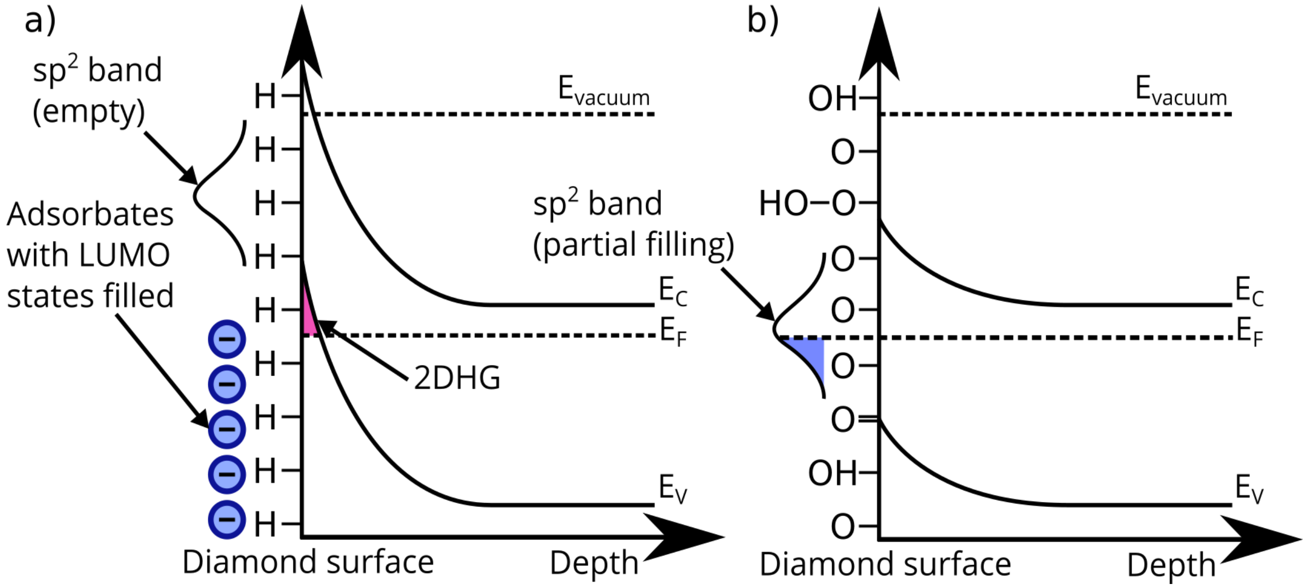
Stark effect $(\vec{E}, \overleftrightarrow{\sigma})$



L.P. McGuinness et al, Nat. Nano (2011)

Impact of defects: Electrostatic pinning



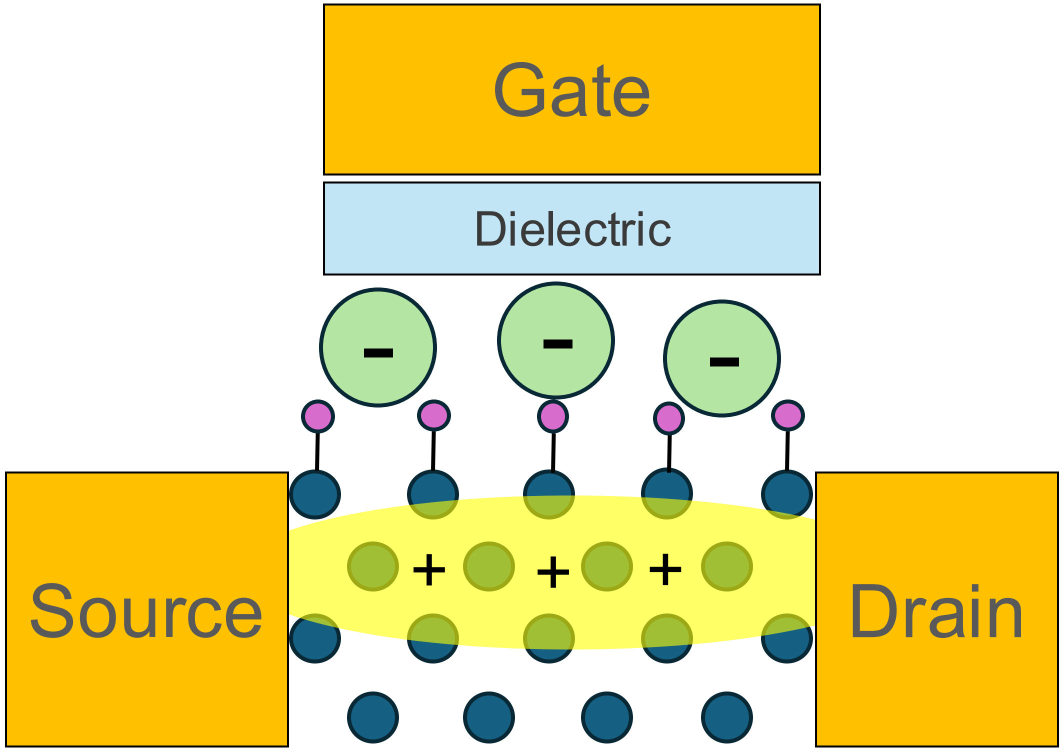


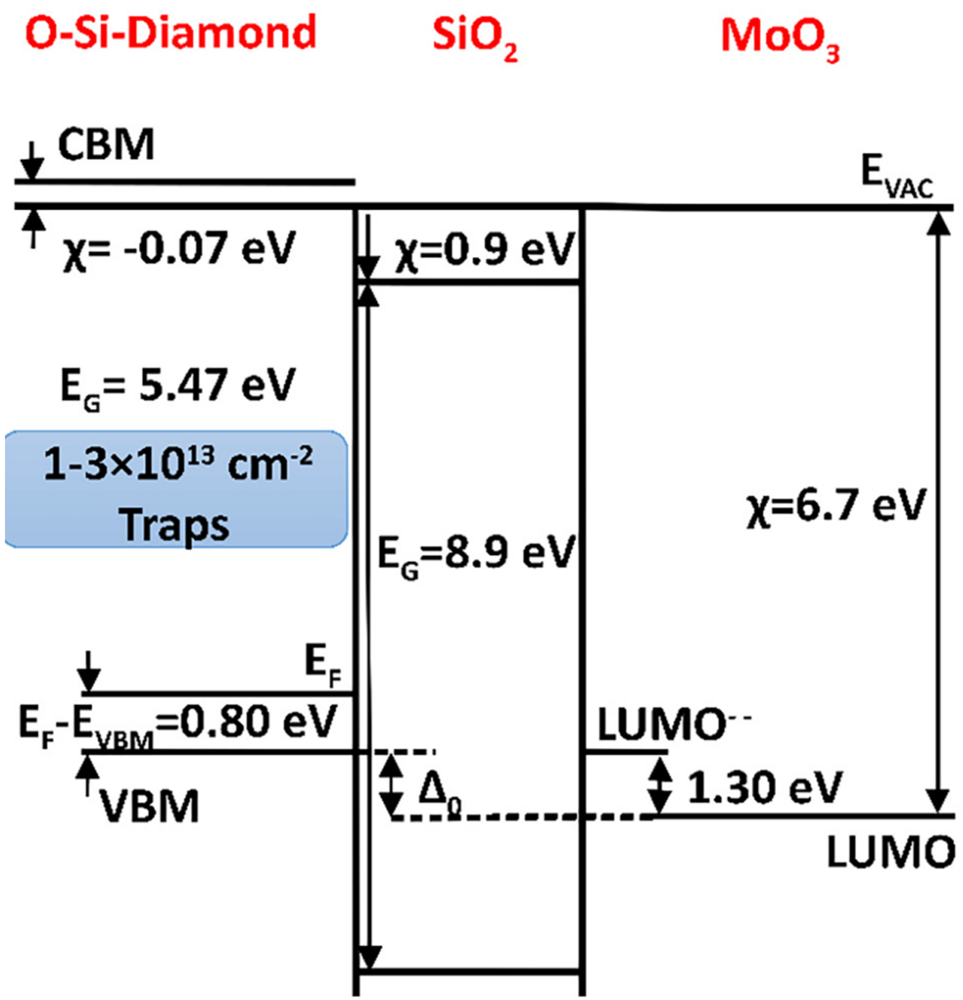
Surface traps can 'pin' the Fermi energy

Diamond Voltage Imaging Microscope D. J. McCloskey, et al., Nature Photonics 16 (10) 730-736, 2022

Electrophysiological recording → high resolution, long-term stability and wide field-of-view

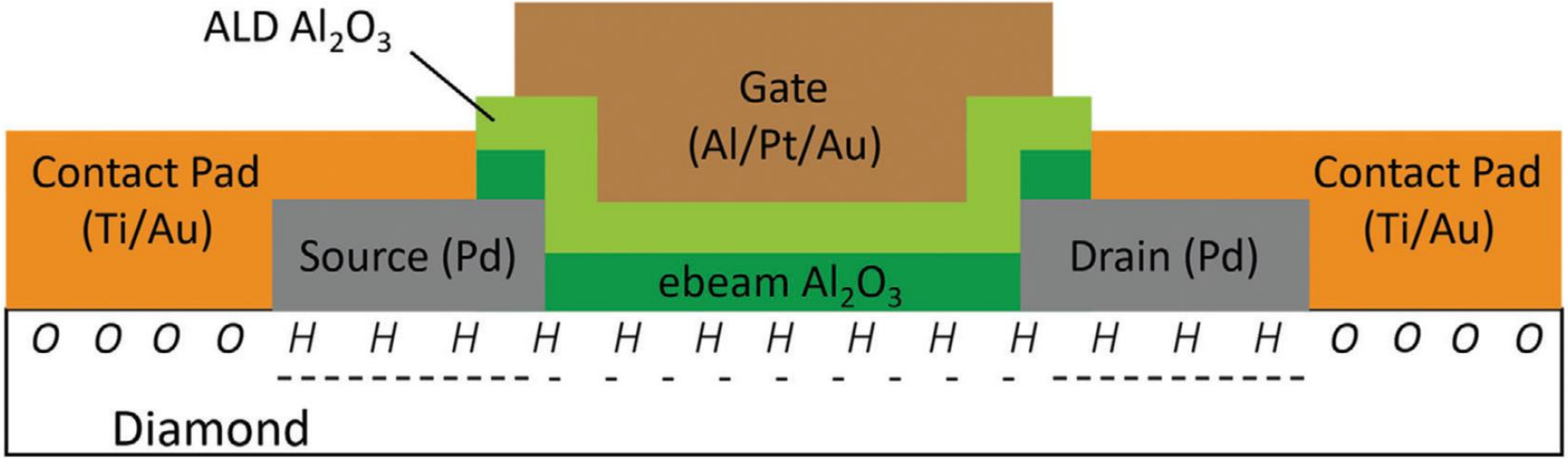
Diamond surface transfer doped FETs





Osslinker et al., Diam. & Relat. Mater.133 (2023) 109712

Accumulation mode FET trials



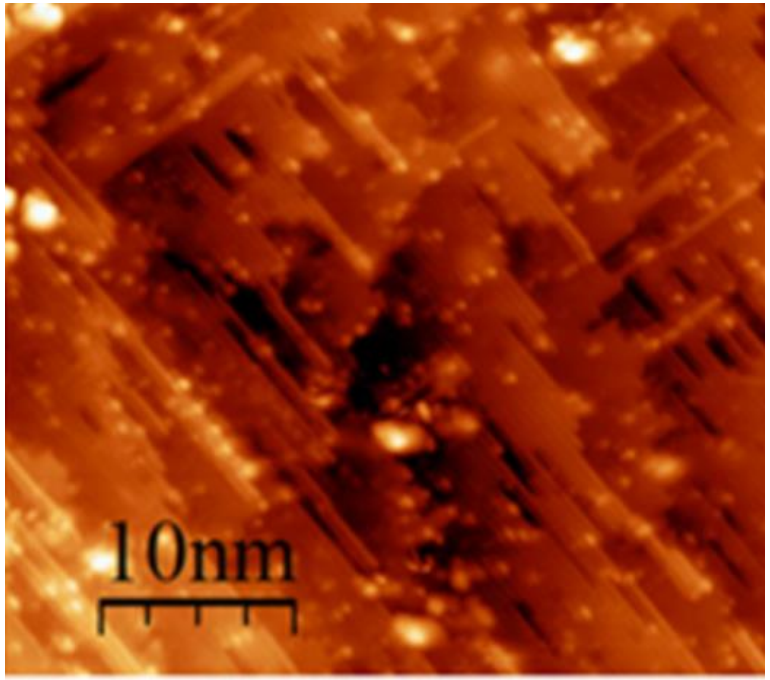
D. Moran group Glasgow U.

C. Qu et al. Adv. Electron. Mater. **2024**, 2400770

→ **Need to suppress transfer doping AND surface defects**

Origins of surface traps?

{100} surface



~3nm terraces
~5% sites are step-edges

Schenk et al., LaTrobe University

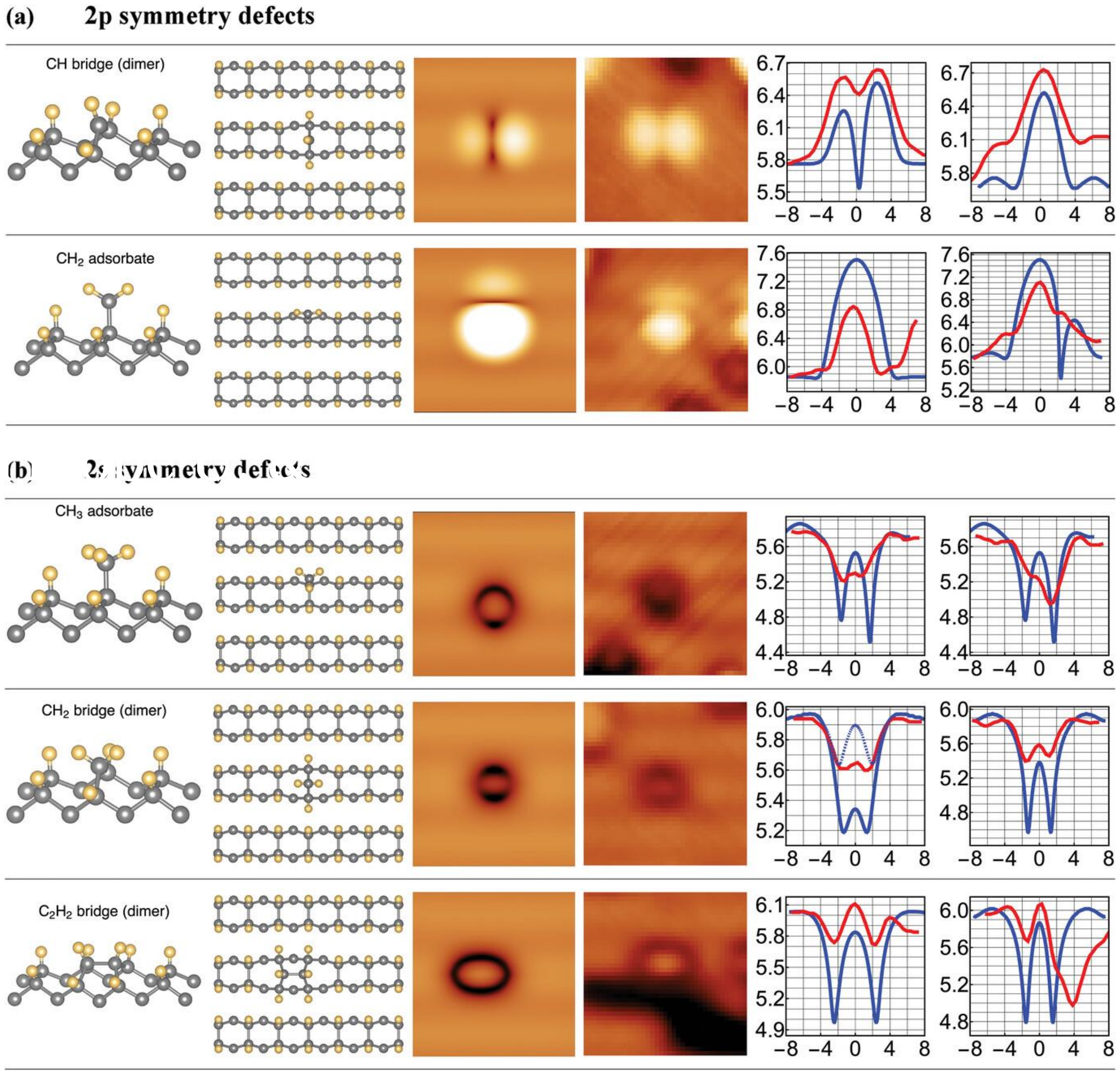
Surface vacancies can reconstruct into C=C bonds

Confirmed with calibrated X-ray absorption > **10^{13} /cm²**

A. Stacey, et al., Adv. Mater. Inter. **6** (2019) 10801449

→ Step edge defects & reconstructed surface vacancies

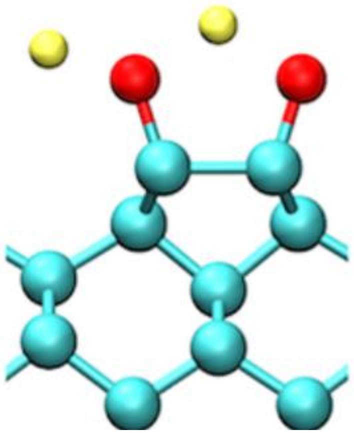
Surface defect imaging – ‘as-grown’



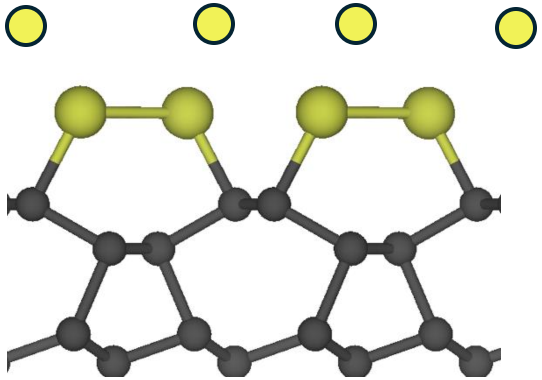
10.1002/admi.202400695

→ Other C=C defects are possible

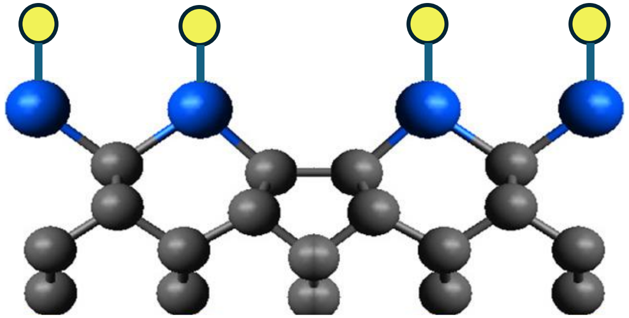
Other (non-H) polar layers are possible



Mg / O
-2.0eV NEA



Si / O
-0.07eV NEA

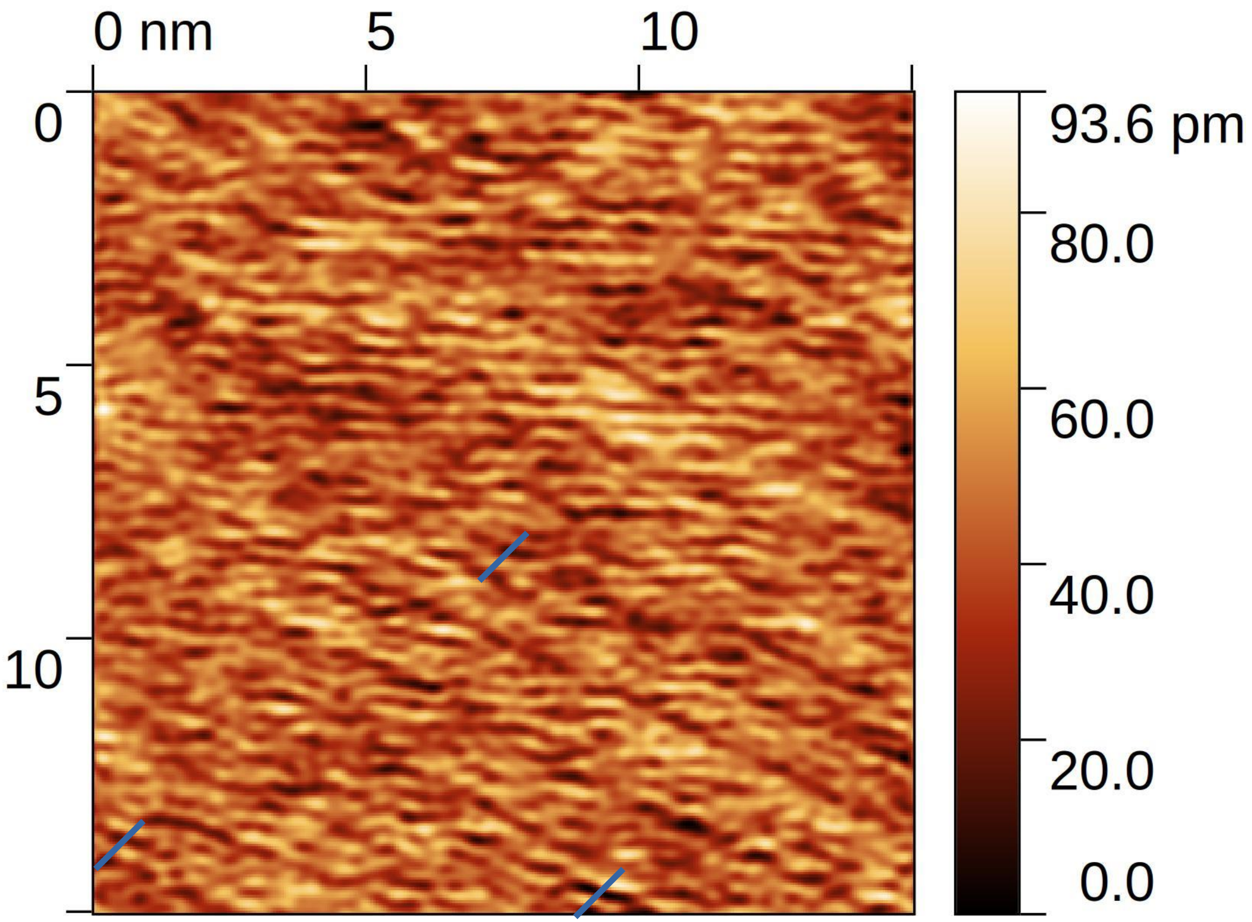


Si / O
-0.25eV NEA

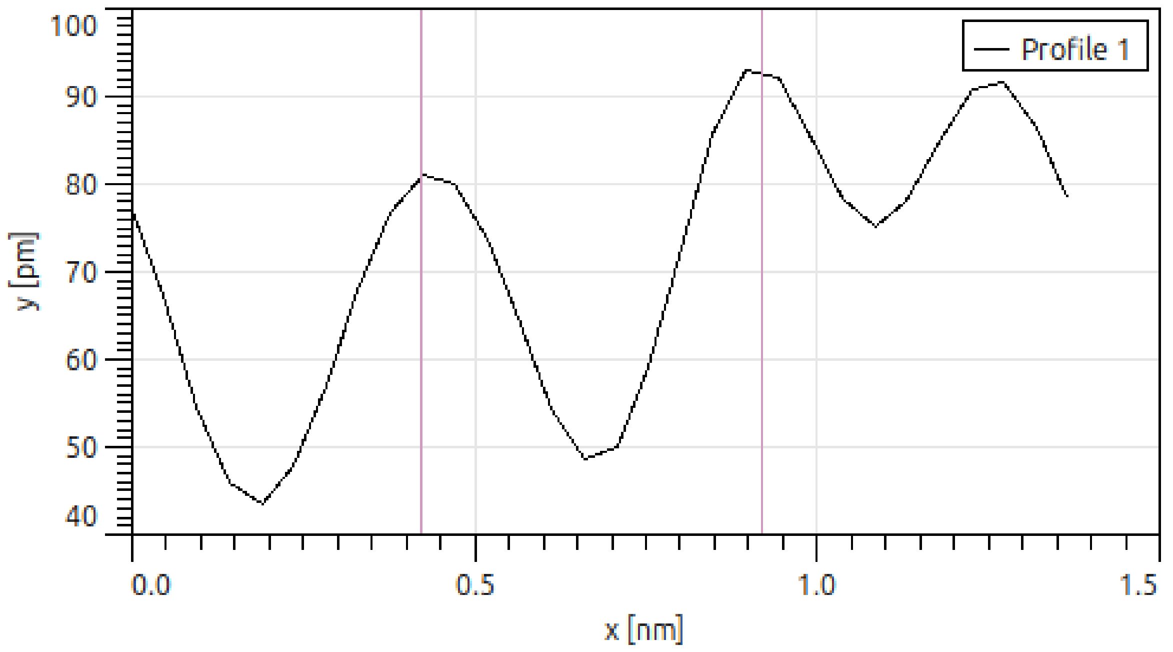
- NEA sufficient to transfer dope in ambient
- No doping of the diamond is observed

- MoO3 hetero-layer acts as electron acceptor for Si-O terminations
- 111 surface showed partial Fermi pinning

→ Better surface state control and understanding required

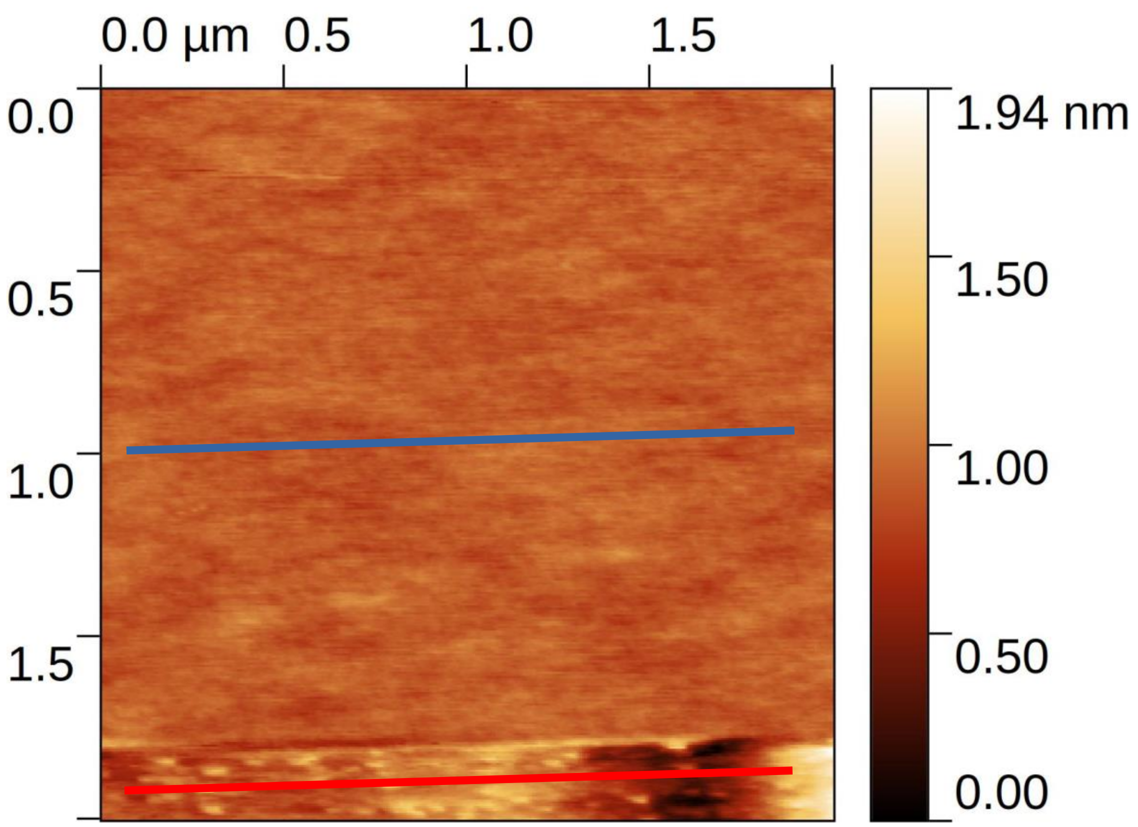


- Spacing of 400-600 pm
- Dimer spacing of 500 pm

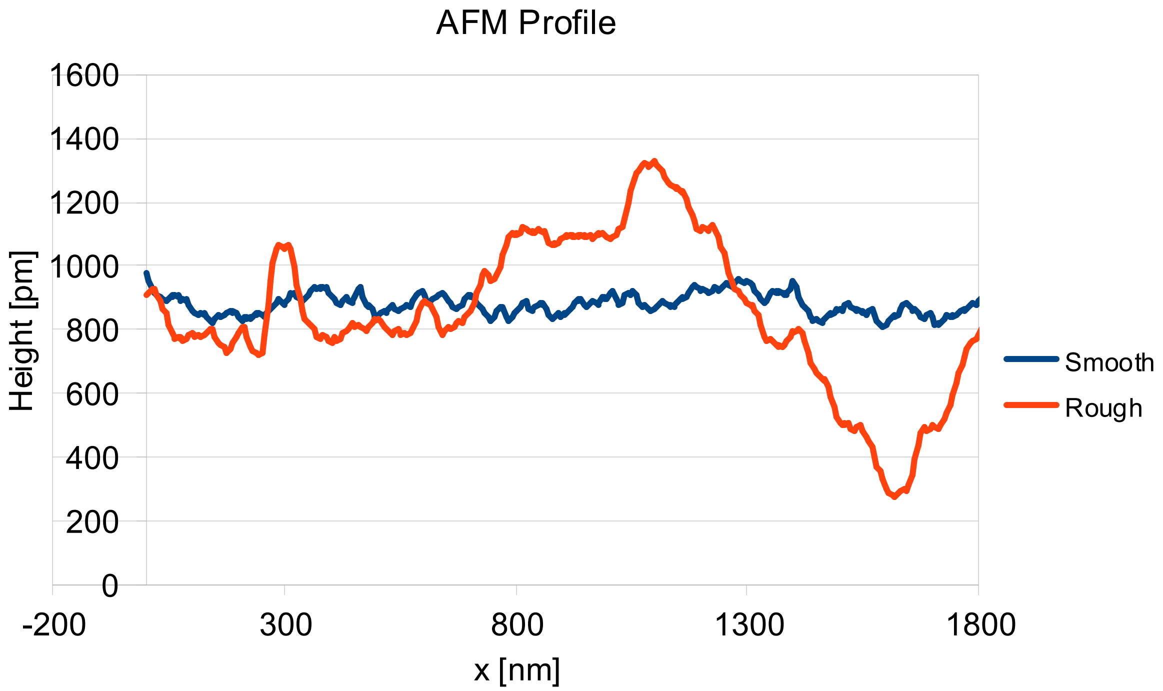


PhD Student: James Belcourt

"smooth" region featureless, rough region clear step-bunch

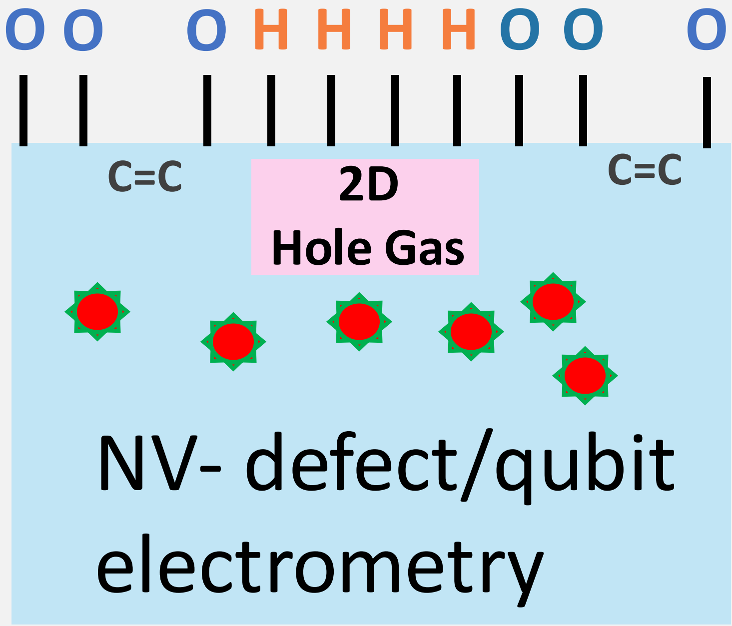


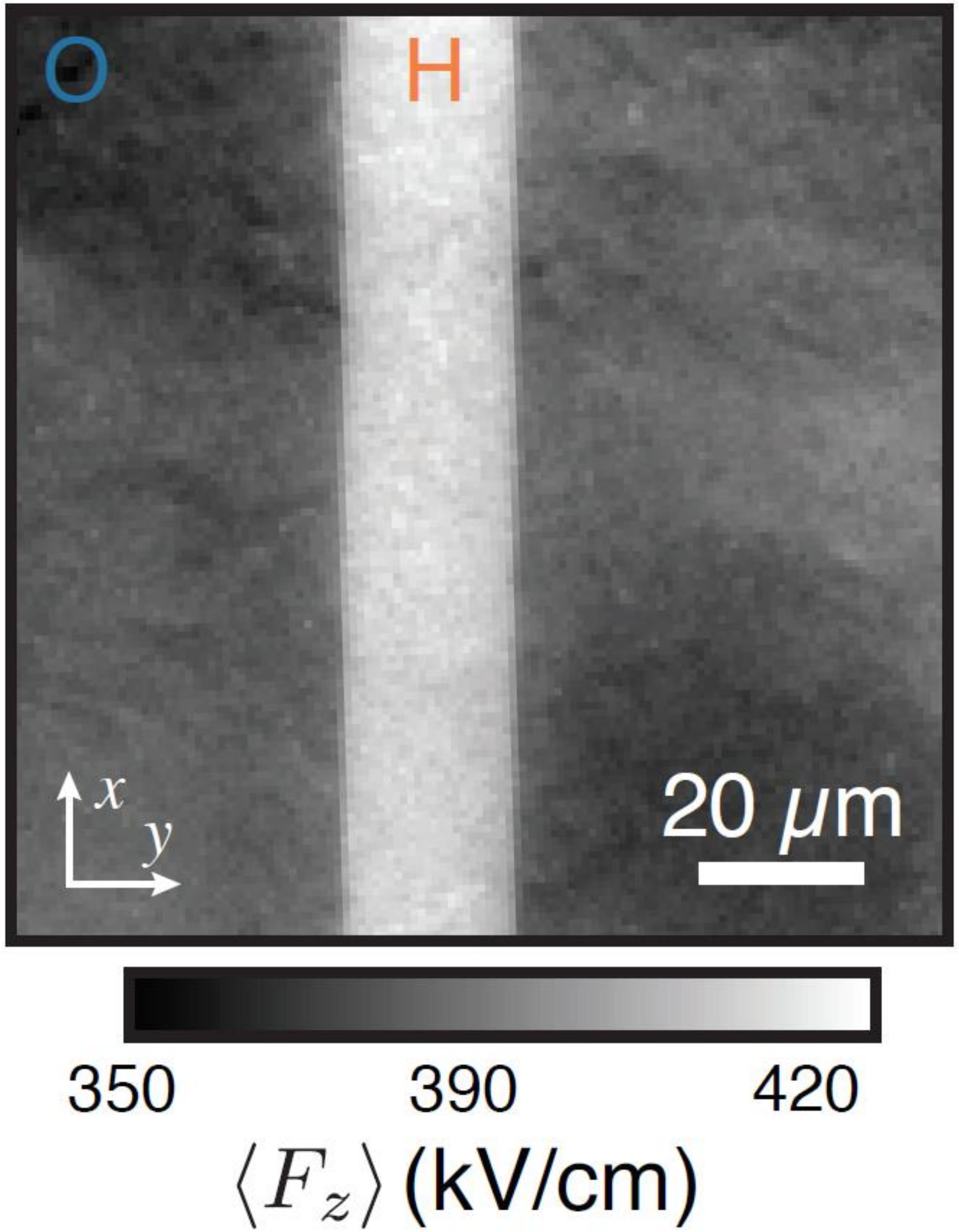
Roughness of 20 pm in Smooth Region



PhD Student: James Belcourt

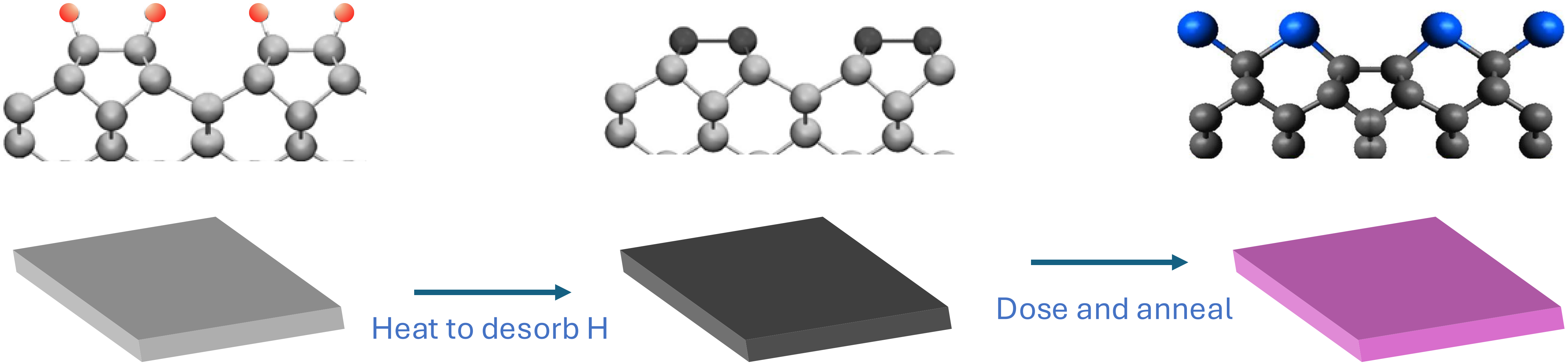
Seeing defects with quantum probes





D.A. Broadway, et al., Nat. Elec. **1** (2018) 502-507

UHV based surface termination approach

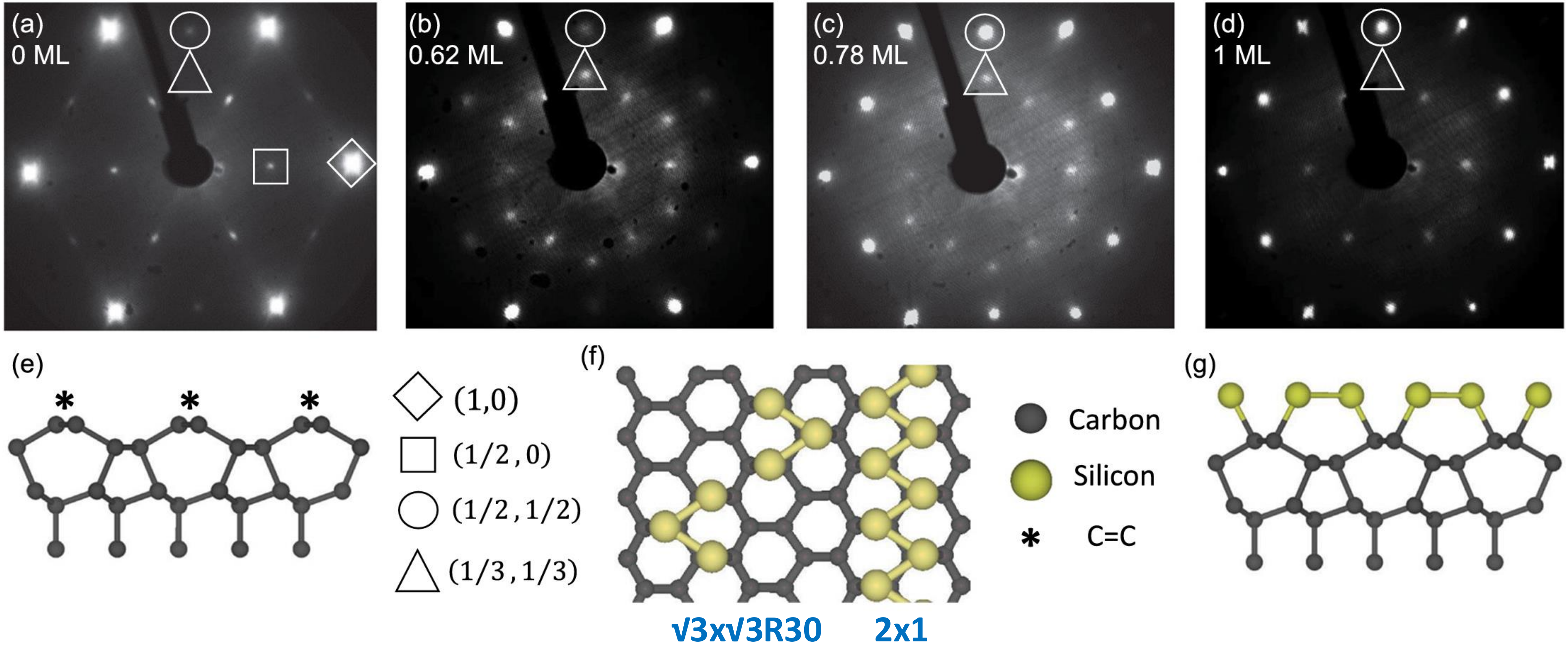


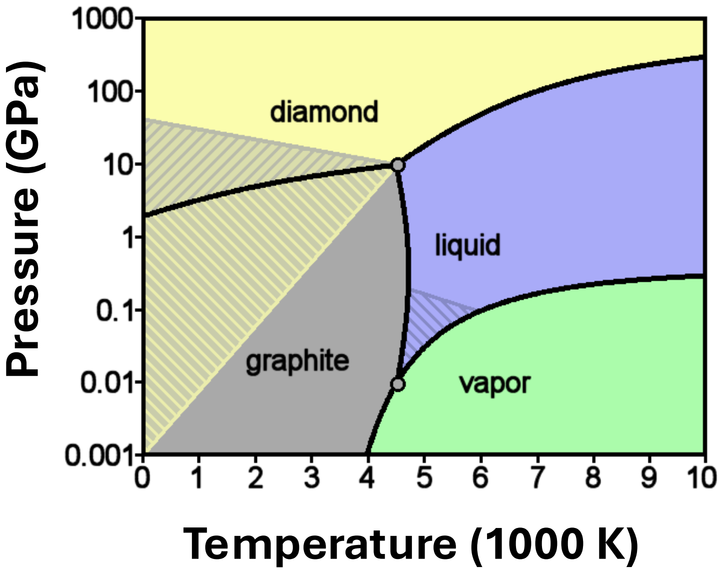
Fresh H-termination

Clean reconstruction

New chemistry & reconstruction

UHV example: Si-terminated {111} diamond





→ **Diamond is only metastable at S.T.P.**

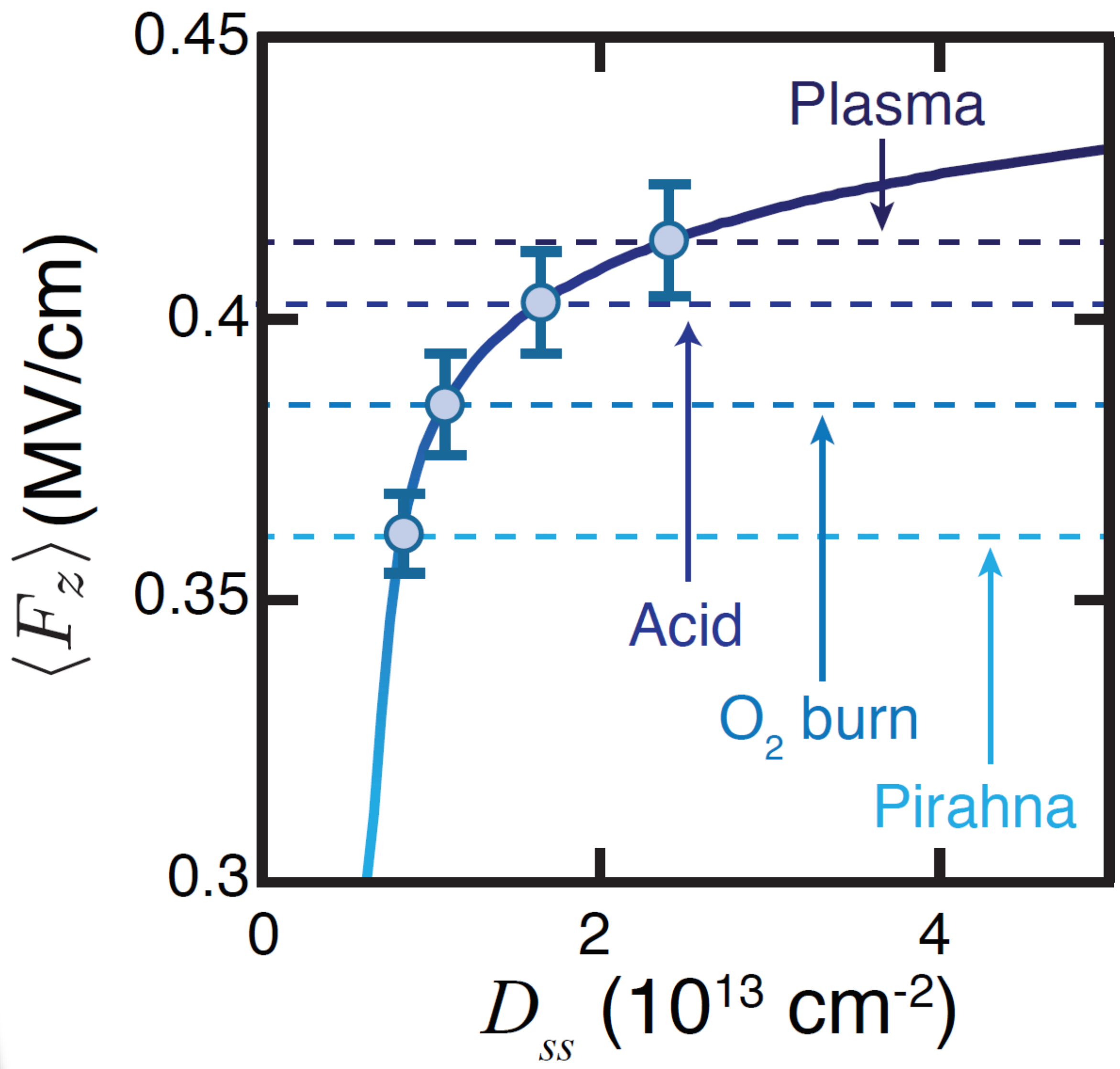
elementsix™ a De Beers Group Company

Surface traps observed with quantum electrometry

Sample Treatment	Acid	+ O_2 Burned	+ Piranha
C=C fraction %	18.6	7.7	1.8

$\langle F_z \rangle$ modelled vs D_{ss}

→ for an ensemble NV implant

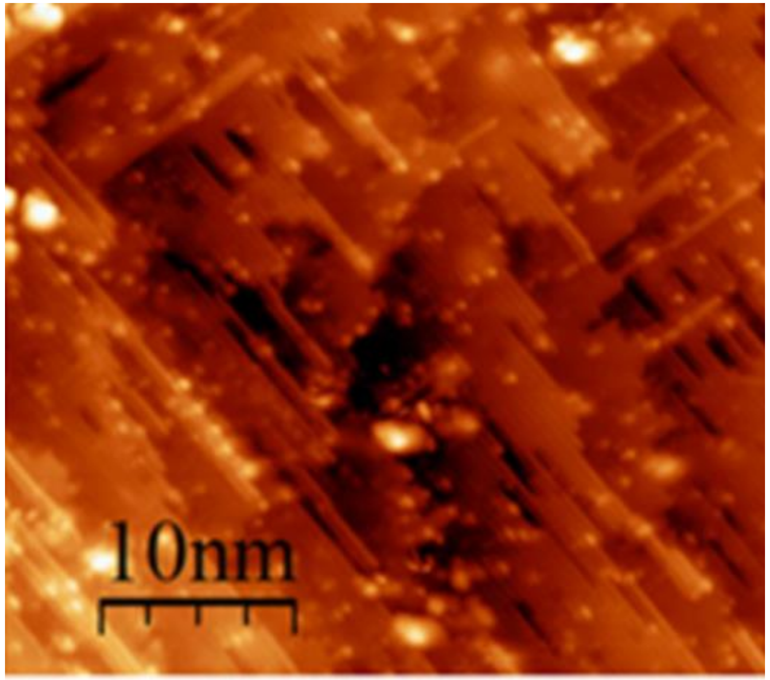


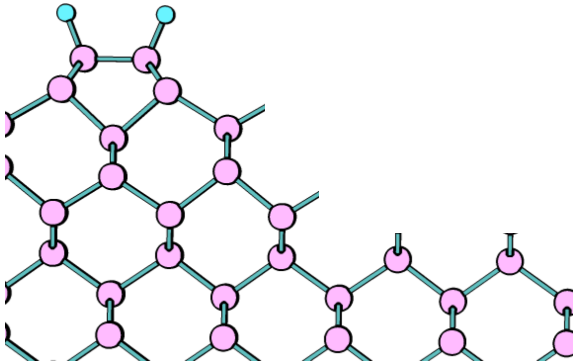
A. Stacey, et al., Adv. Mater. Inter. **6** (2019) 10801449

D.A. Broadway, et al., Nat. Elec. **1** (2018) 502-507

Origins of surface traps?

{100} surface

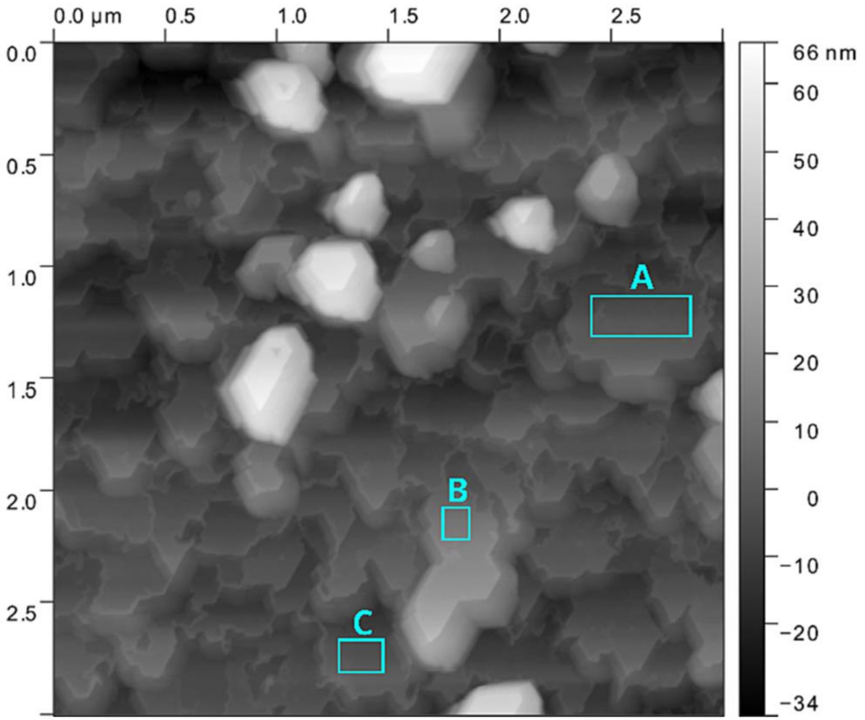




~3nm terraces
~5% sites are step-edges

Schenk et al., LaTrobe University

{111} surface



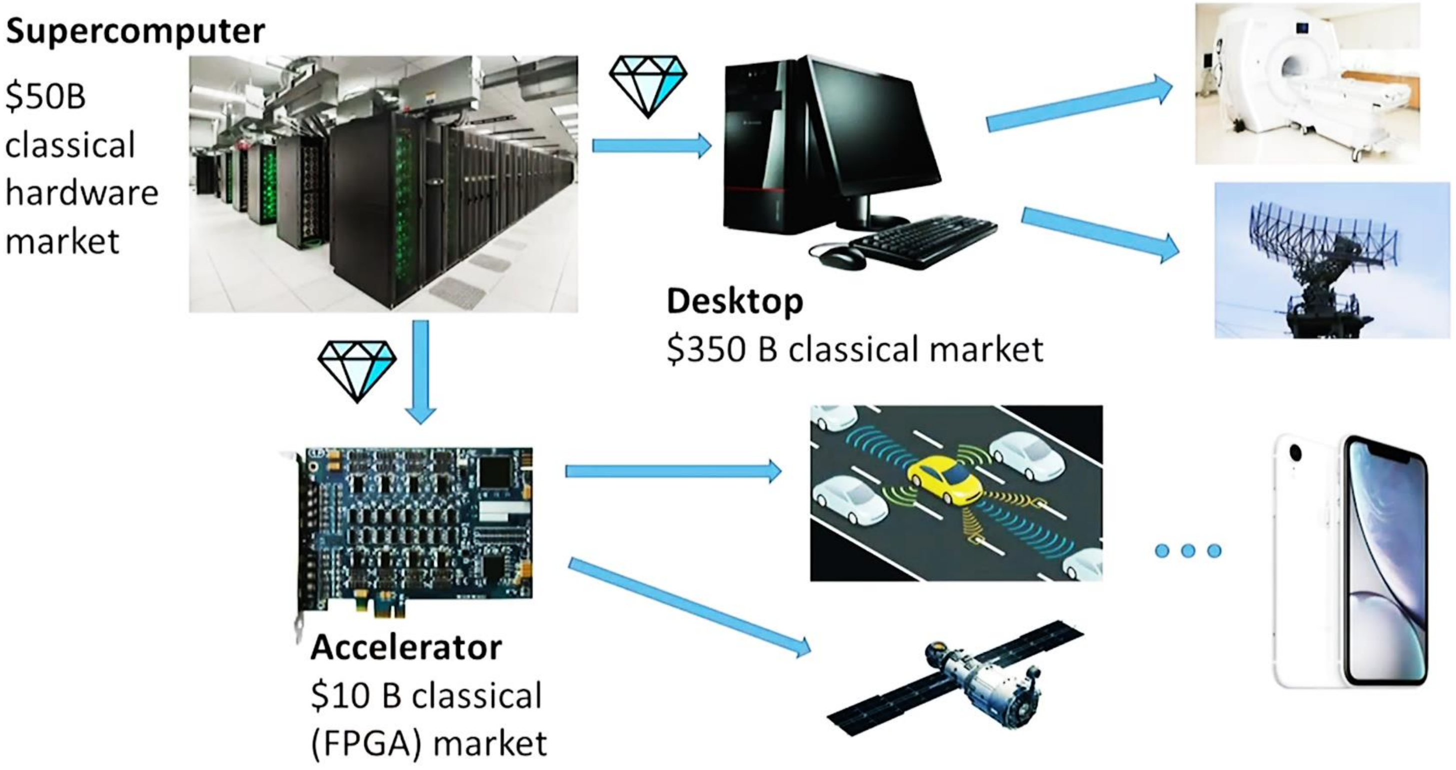
>100nm terraces ?
<0.1% sites are step-edges

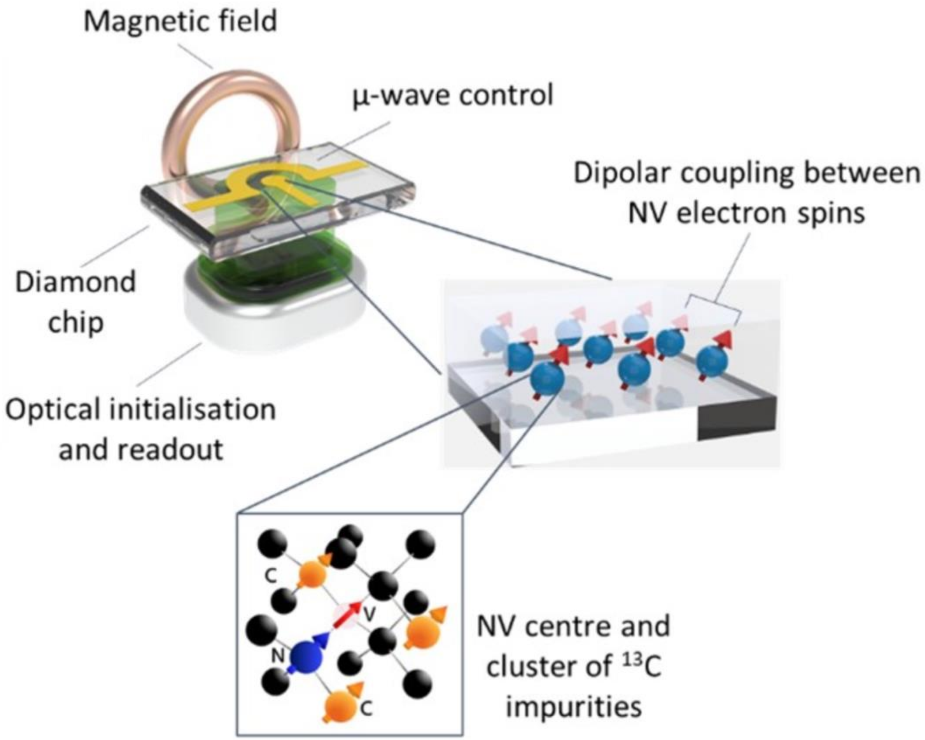
Ossliinker et al., Diam. & Relat. Mater.133 (2023) 109712

→ **What about surface *quality and charge traps?***

QUANTUM BRILLIANCE

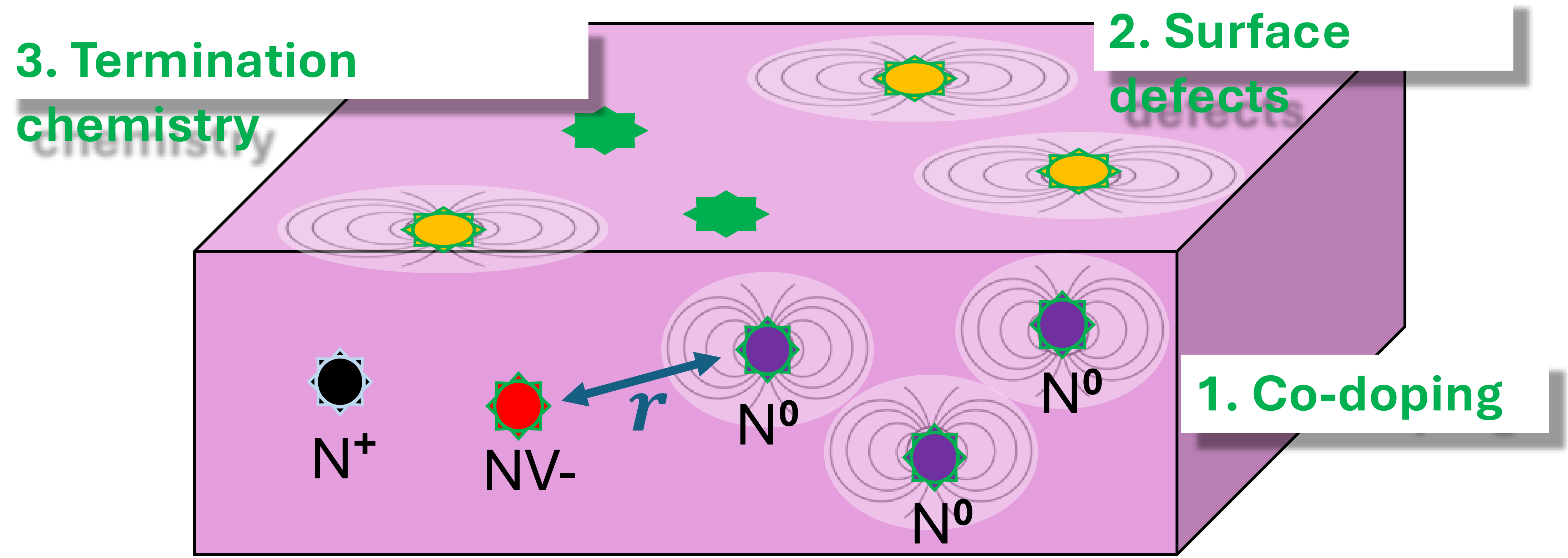
Our vision: quantum computing for all



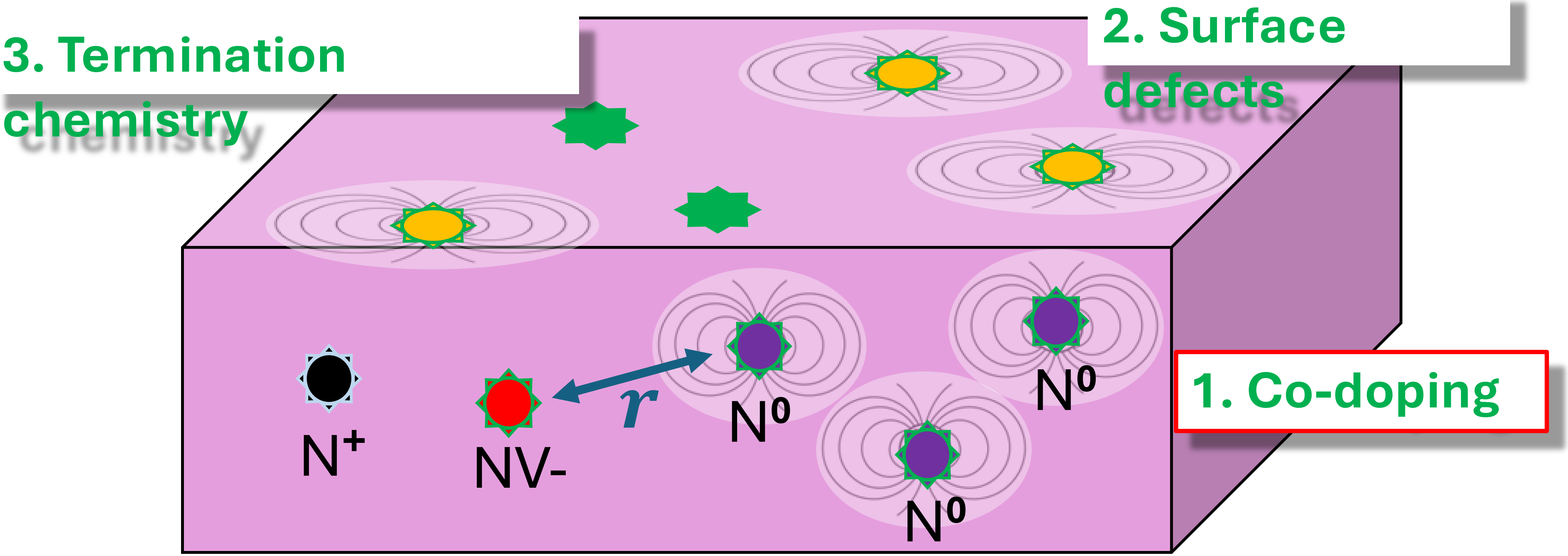


https://quantumbrilliance.com/

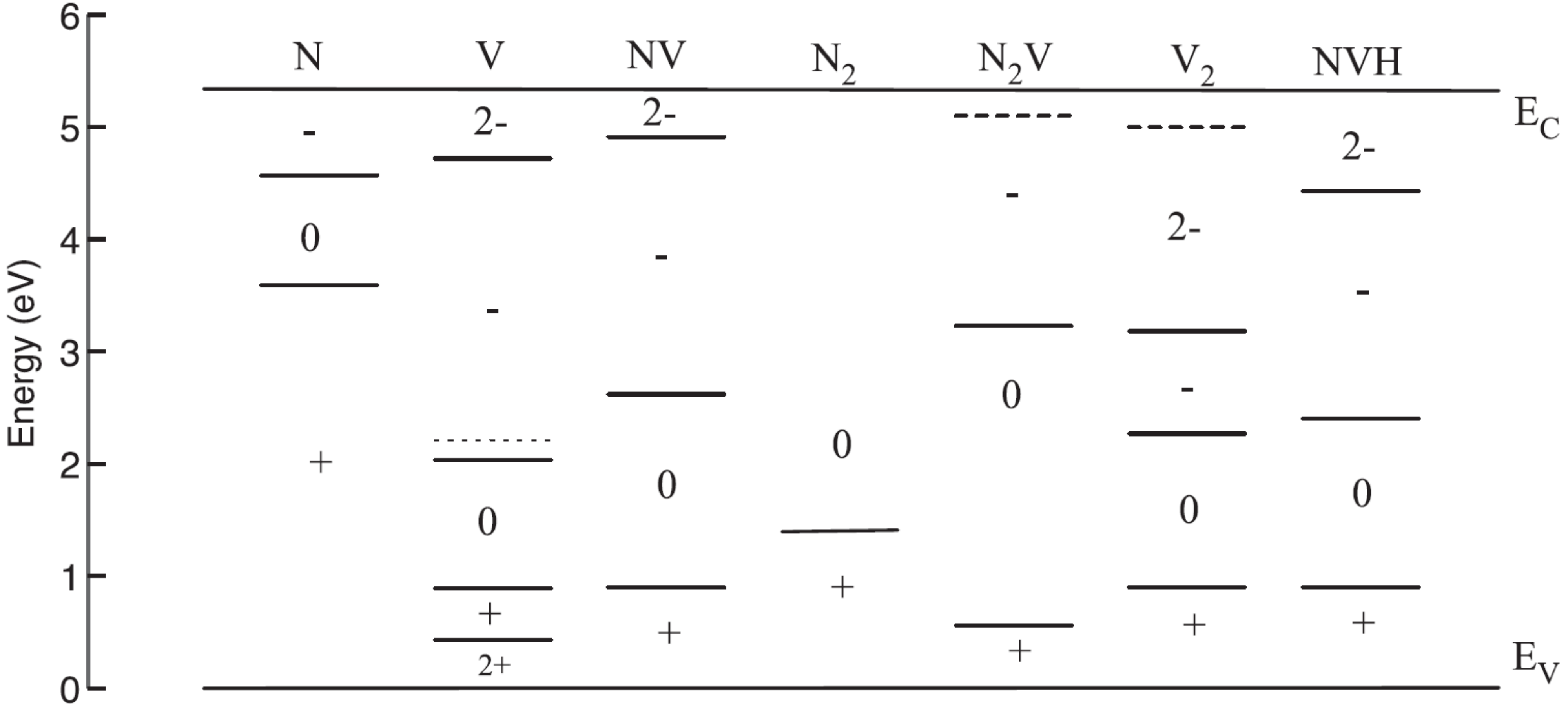
Diamond materials for quantum devices (magnetometry)



1. Bulk co-doping

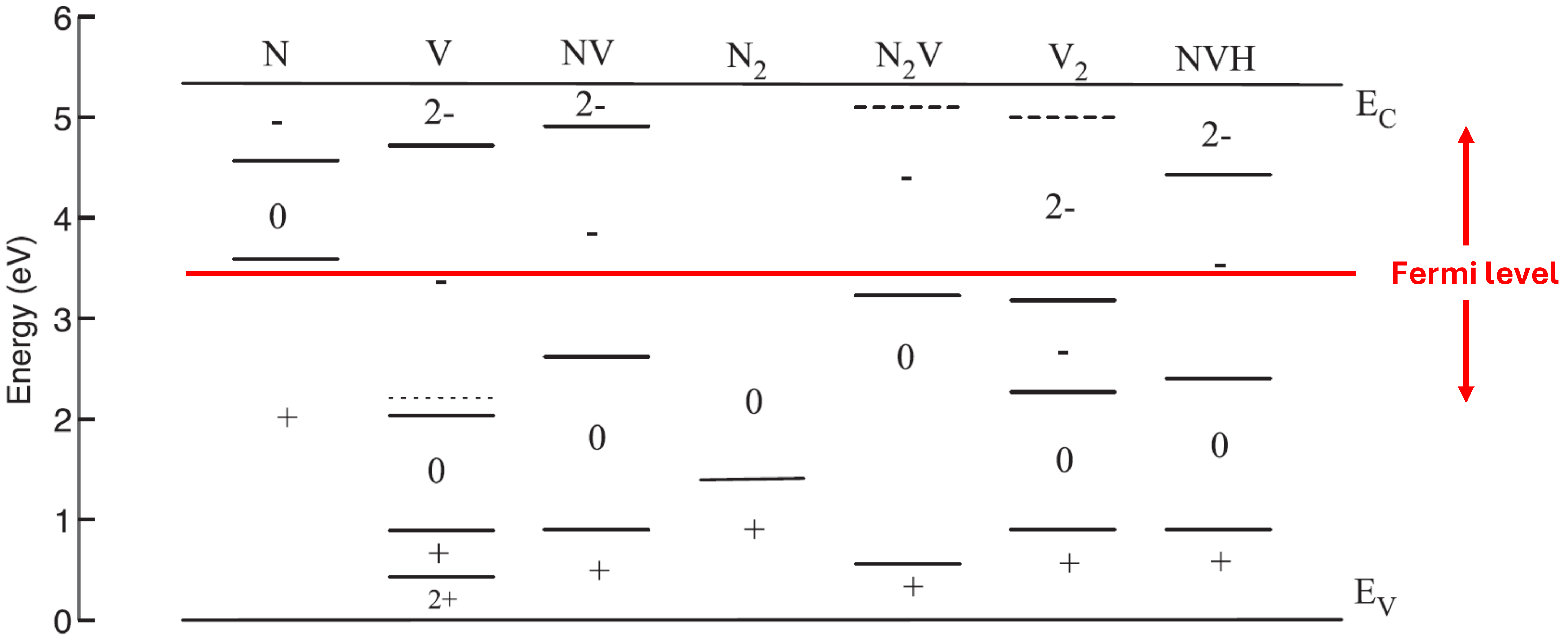


What is 'co-doping'



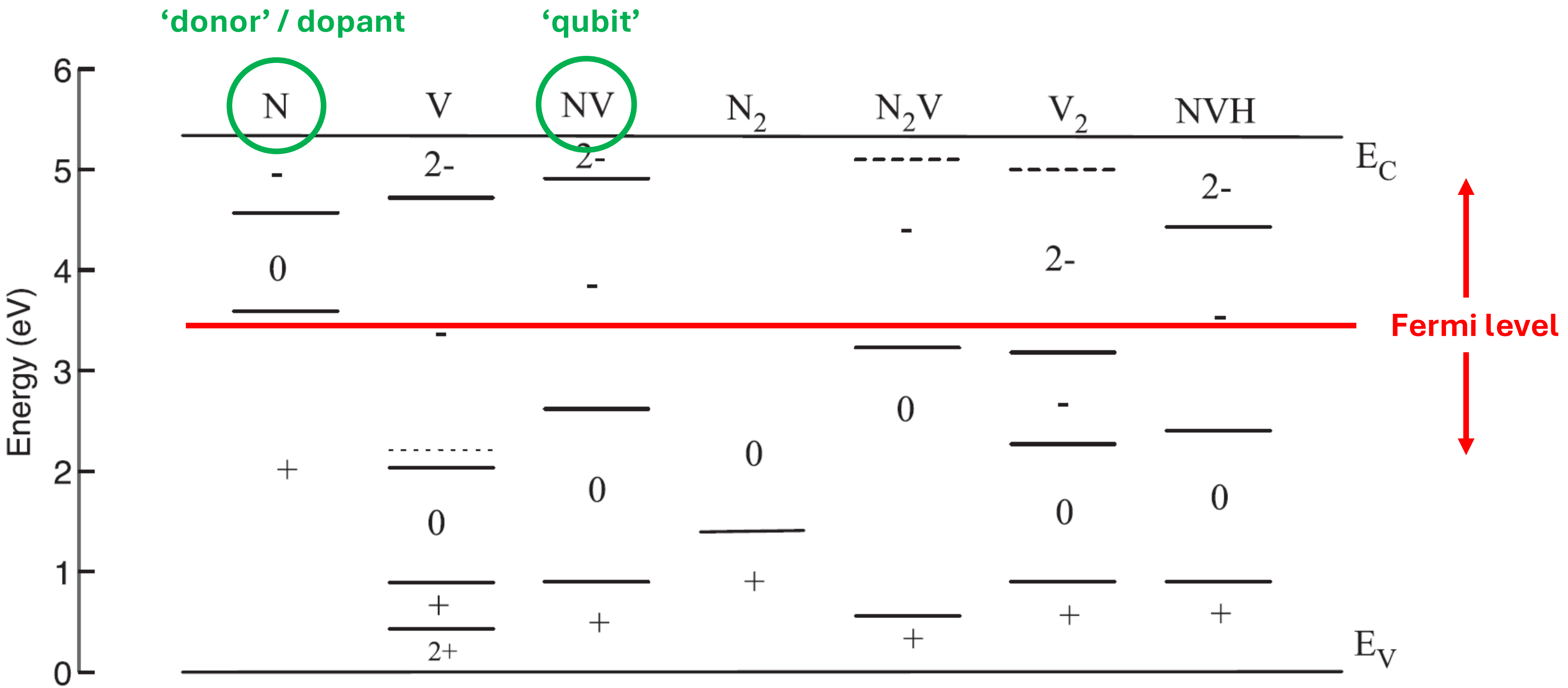
P. Deák, et al., Phys. Rev. B 2014, 89, 075203

What is ‘co-doping’



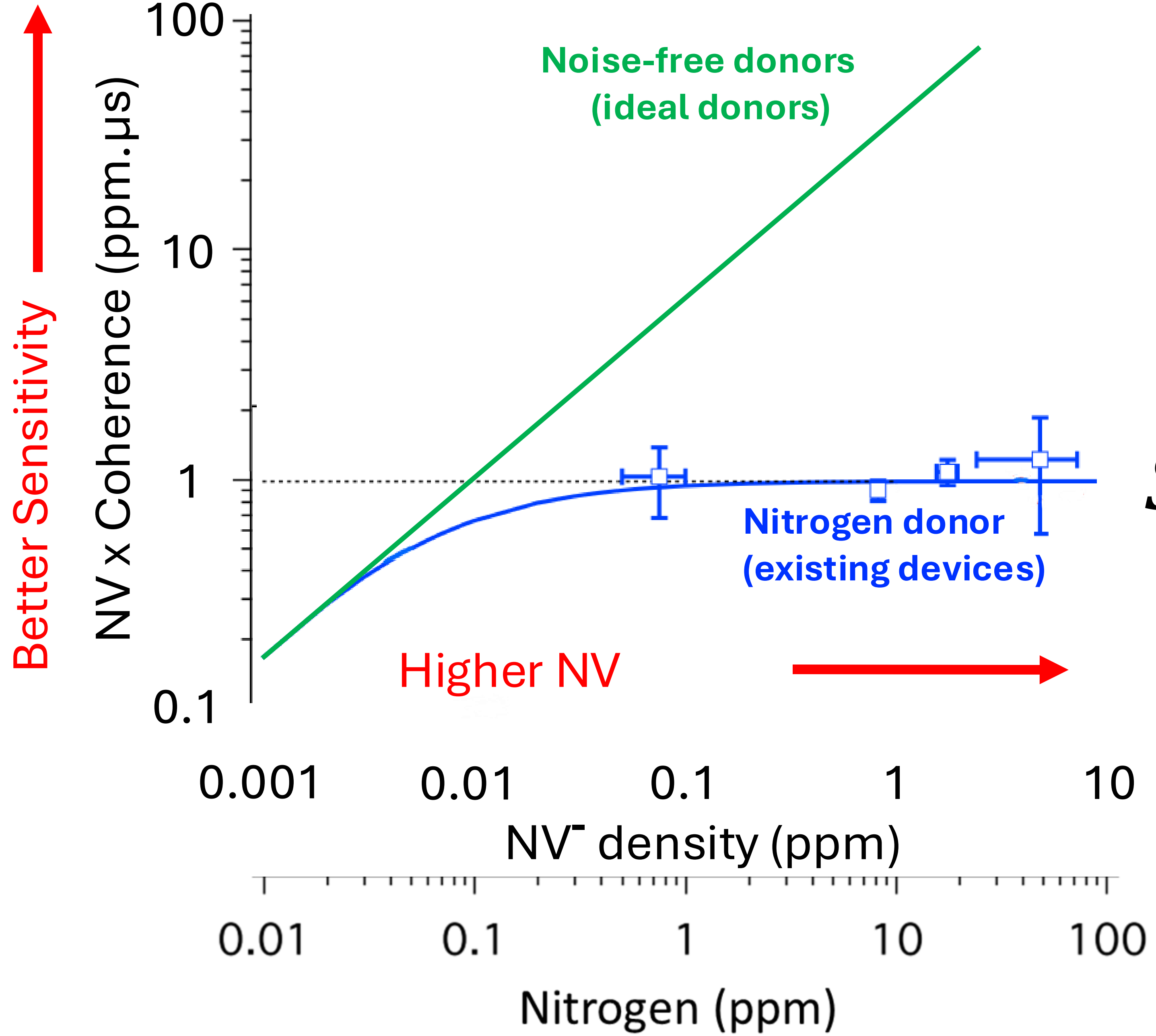
P. Deák, et al., Phys. Rev. B 2014, 89, 075203

What is 'co-doping'



P. Deák, et al., Phys. Rev. B 2014, 89, 075203

Scourge of donor spins: poor scaling



$$S \propto \frac{1}{\sqrt{[NV] \times T_2^*}}$$

$$S \propto 1$$

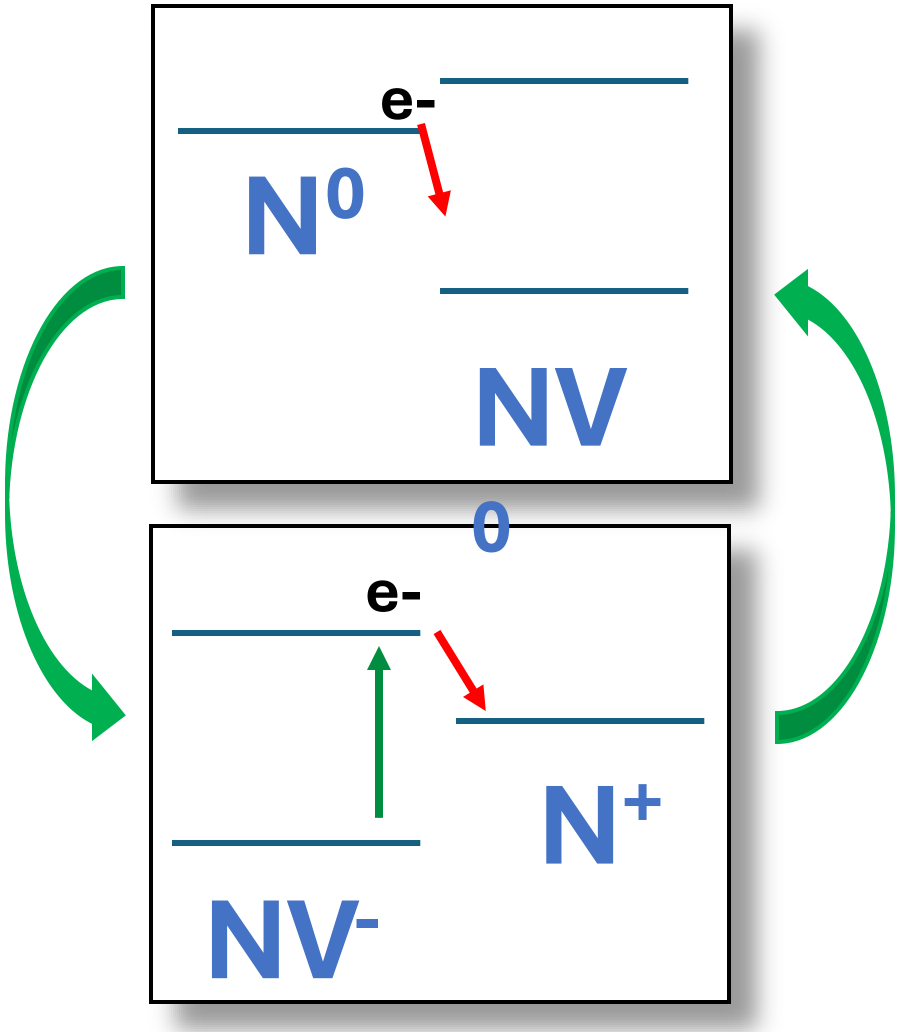
J. Barry, et al., RMP **9** (2020) 015004

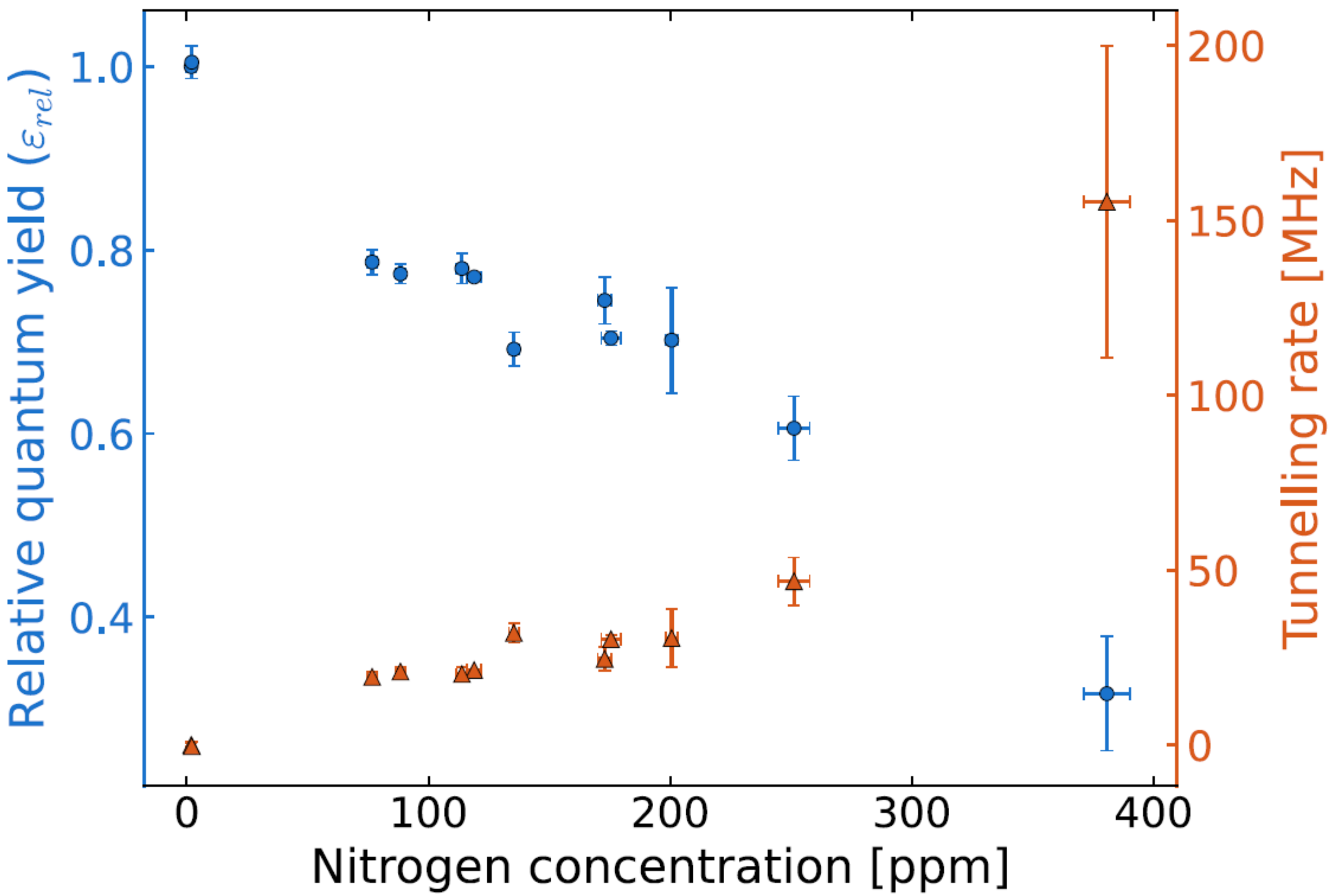
____ post-presentation modification actual reference is:

https://doi.org/10.1088/2633-4356/abd88a

Andrew M Edmonds et al 2021 Mater. Quantum. Technol. 1 025001

Scourge of donor spins: charge instability

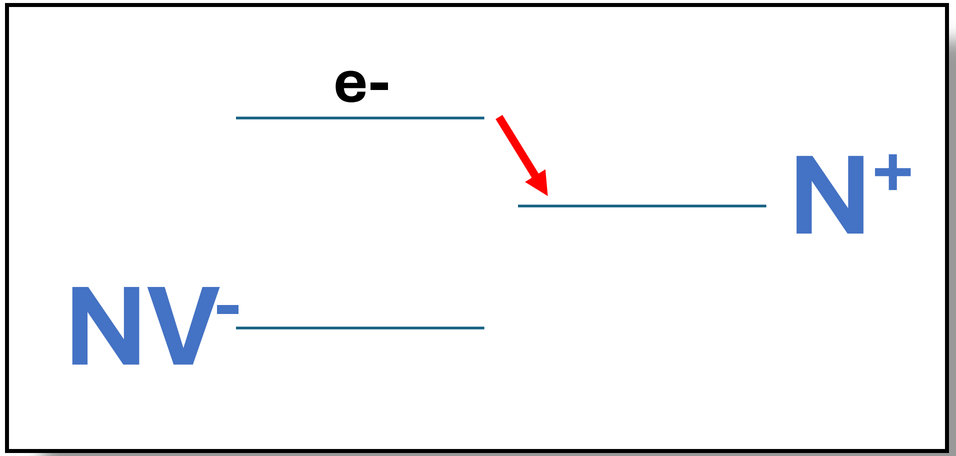


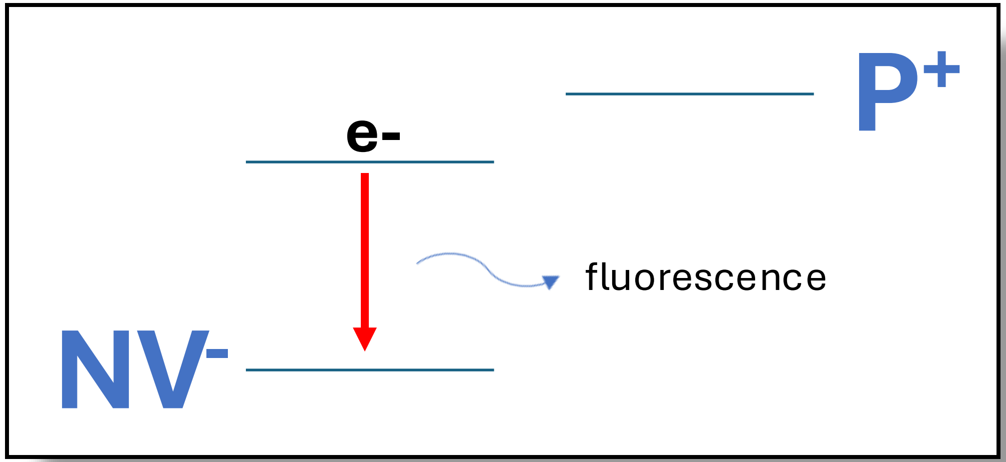


M. Capelli et al., New J. Phys. **24** (2022) 033053

Promise of shallow donors

→ Nitrogen only 'loans' the electron

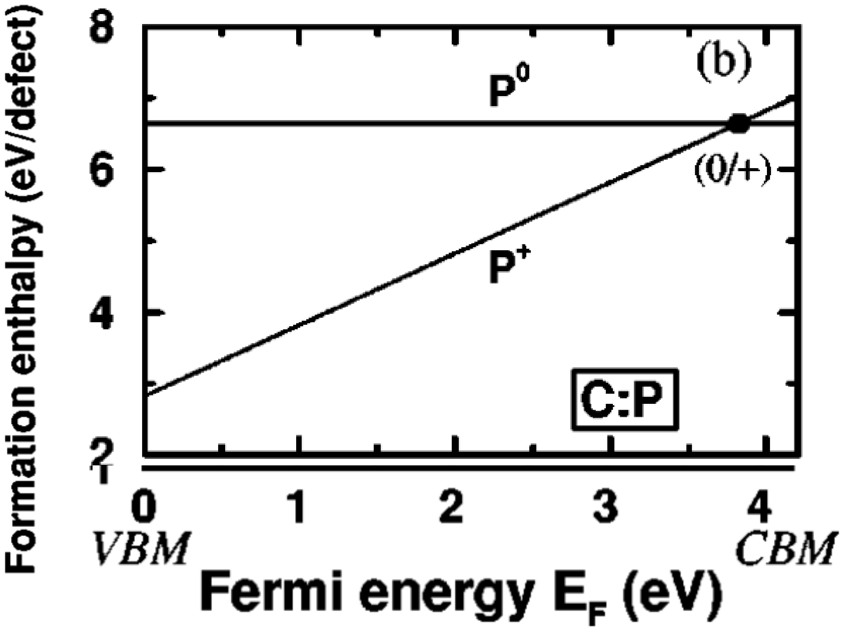




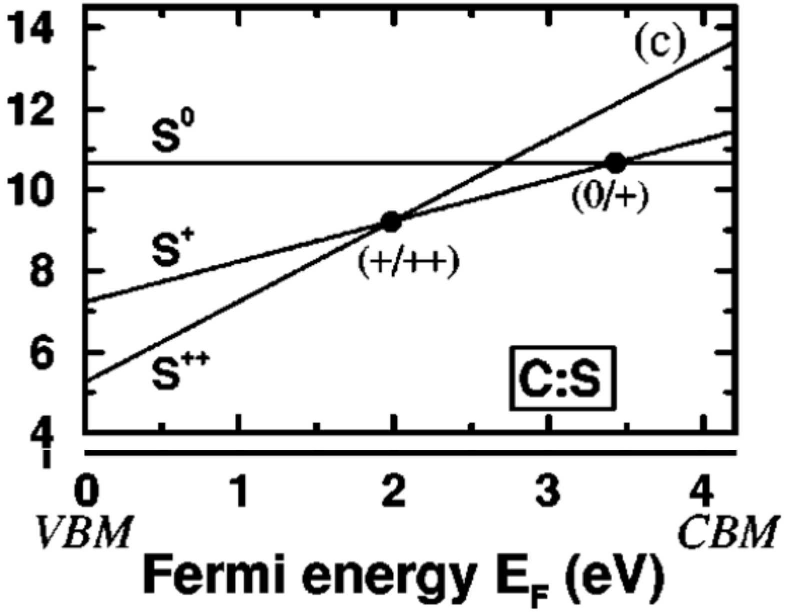
→ Phosphorus really doesn't want the electron back

Promise of double donors

P



S



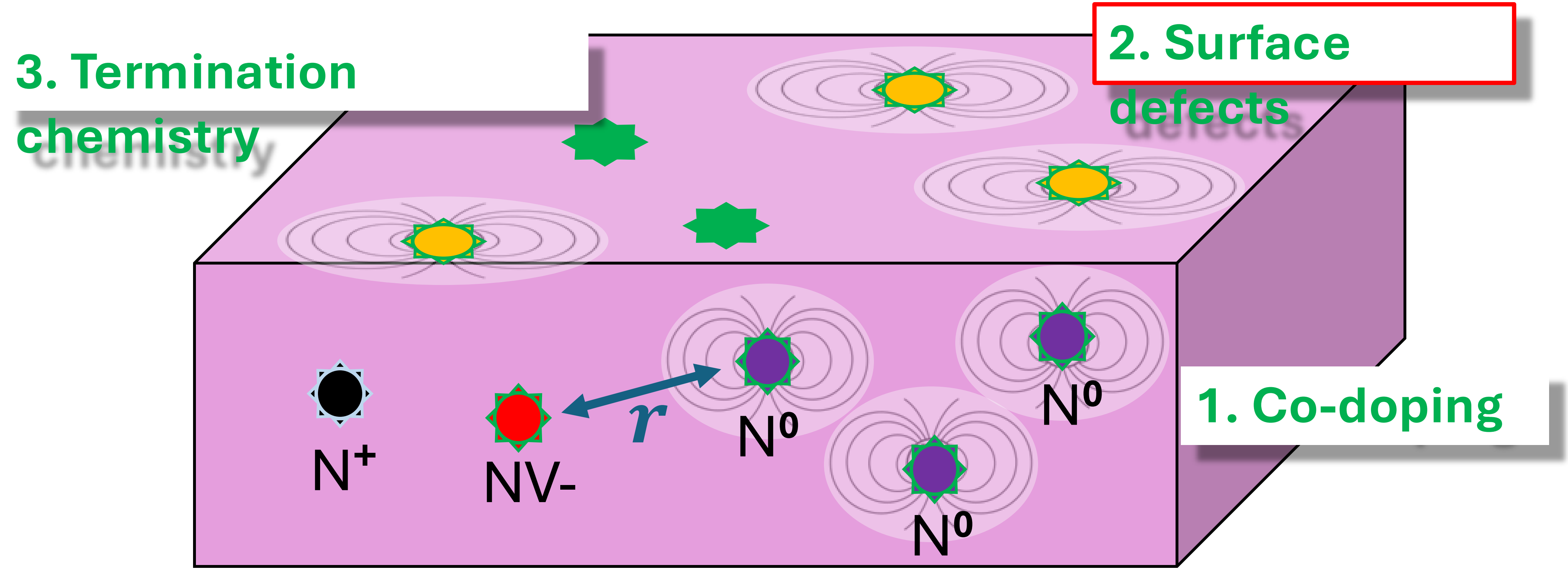
Sulphur: theory says 0.77eV donor level *

Sulphur Questions:

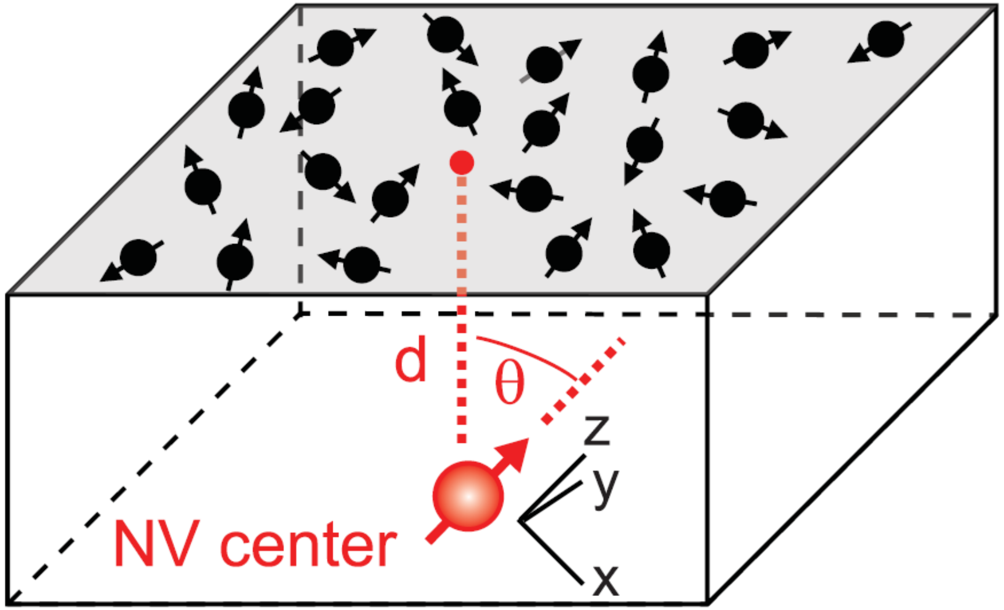
1. **Is (++) transition level higher than NV0?**
2. **Is (0) state spin 0 or spin 1?**
3. **Can we dope with S easily? (it's very big)**
4. **Do SV or SH centres cause trouble?**

* Wang, L. G. & Zunger, A. Phosphorus and sulphur doping of diamond. Phys. Rev. B 66, 161202 (2002)

2. Surface defects



Near surface NVs suffer decoherence



T. Rosskopf, *et al.*, PRL **112**, 147602 (2014)

0.01-0.1 μ_B/nm^2

B. A. Myers, *et al.*, PRL **113**, 027602 (2014)

0.04 spin/nm^2

Y. Romach, *et al.*, PRL **114**, 017601 (2015)

2-3 nm electron spacing

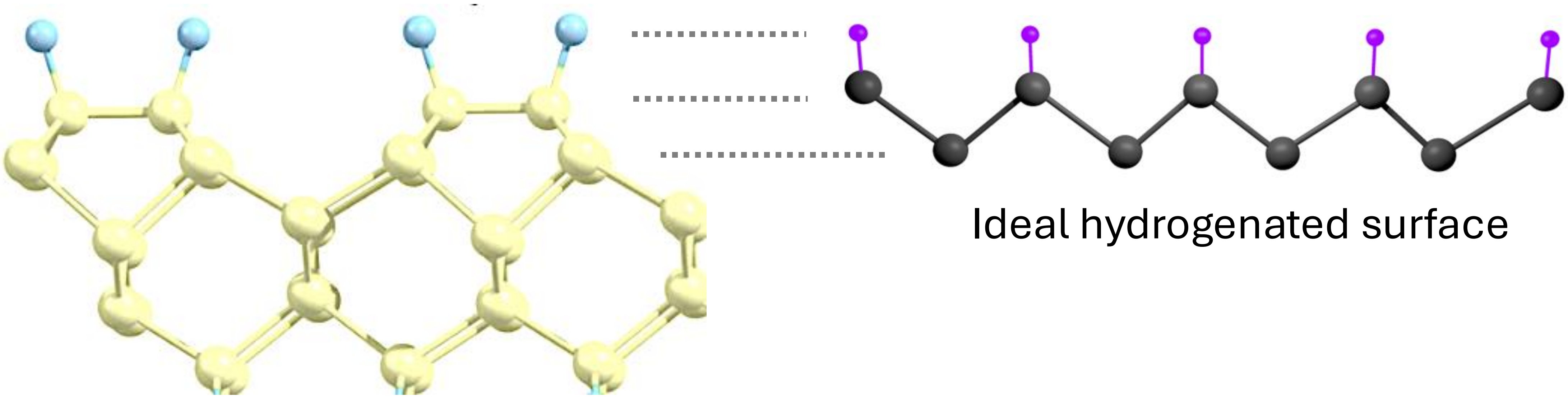
1/d scaling for fast component

M. Kim, *et al.*, PRL **115**, 087602 (2015)

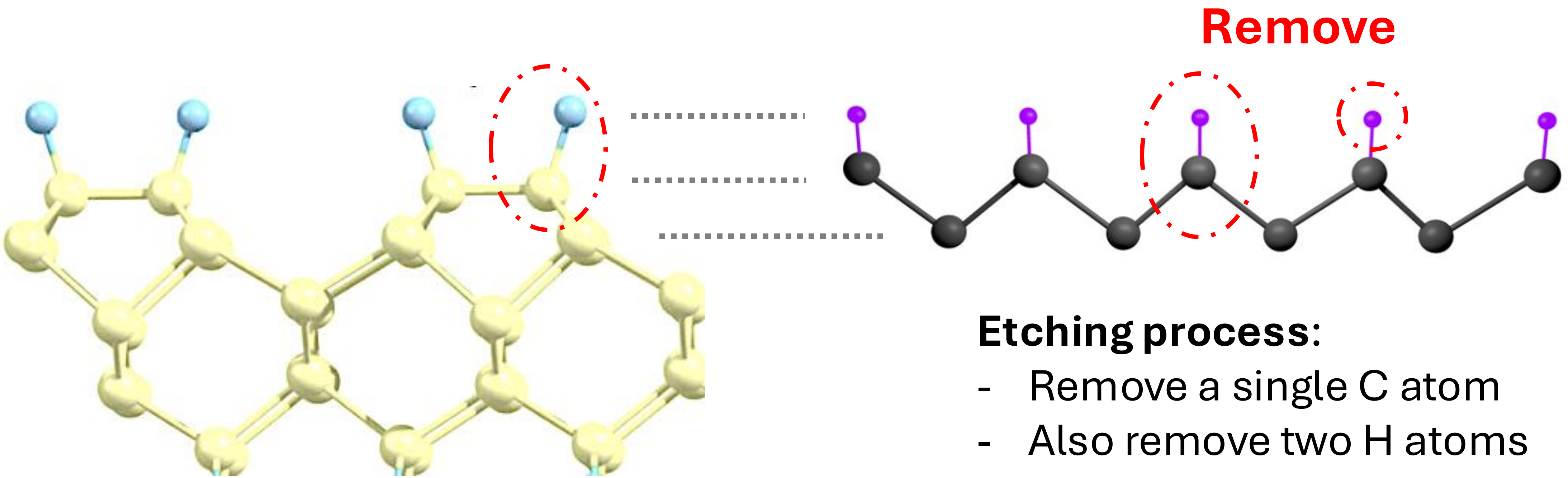
Decoherence limited by electric field noise

Problem = Surface Electrons
'intrinsic' to diamond surface

’sp^2’ surface defects – ubiquitous noise sources



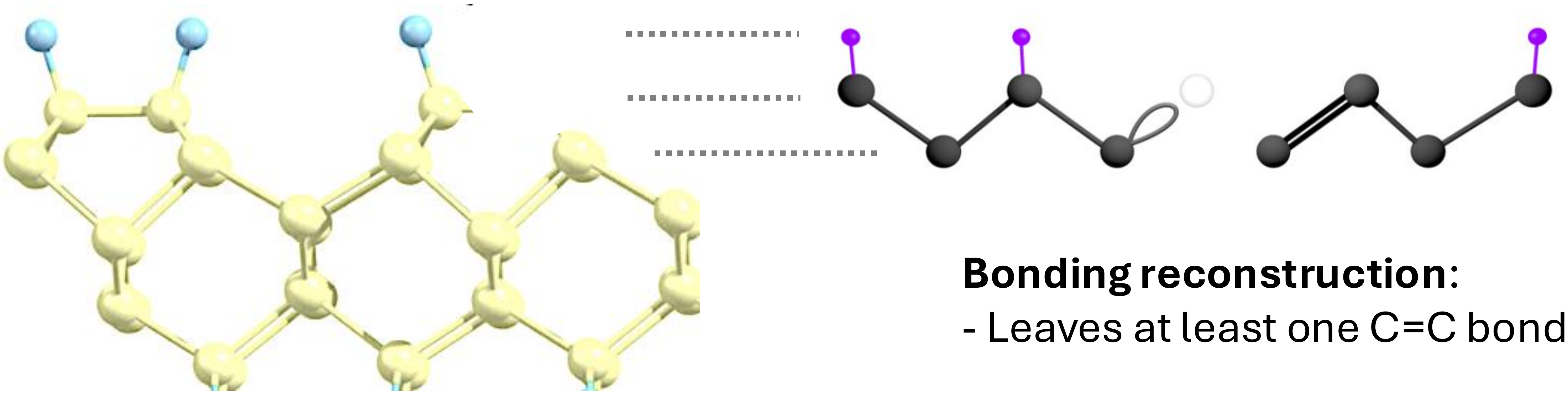
’*sp*^{2}’ surface defects – ubiquitous noise sources



Etching process:

- Remove a single C atom
- Also remove two H atoms

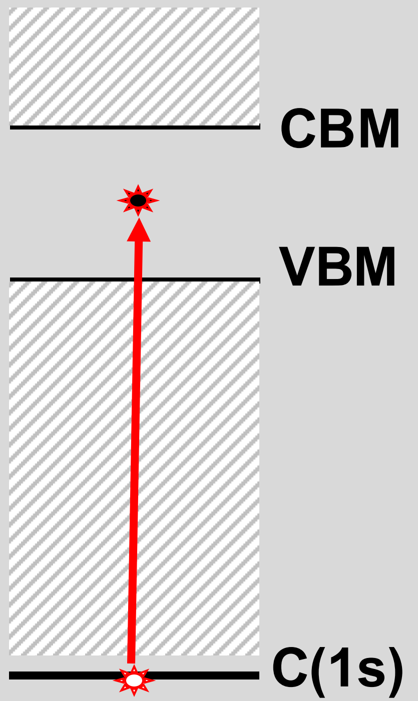
’*sp*^{2}’ surface defects – ubiquitous noise sources



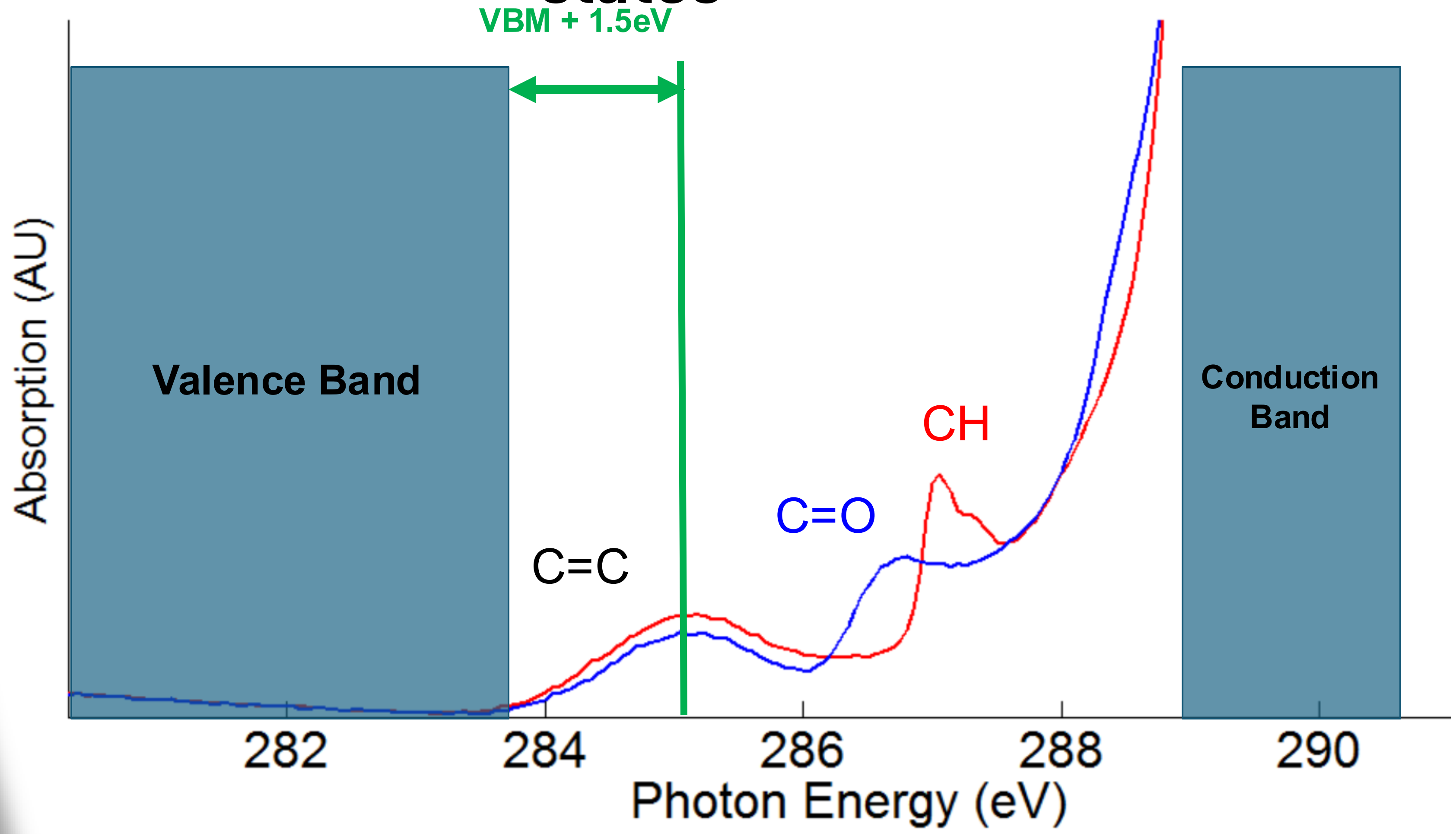
Bonding reconstruction:
- Leaves at least one C=C bond

DFT modelling with Adam Gal

’sp^2’ surface defects – unoccupied electronic states

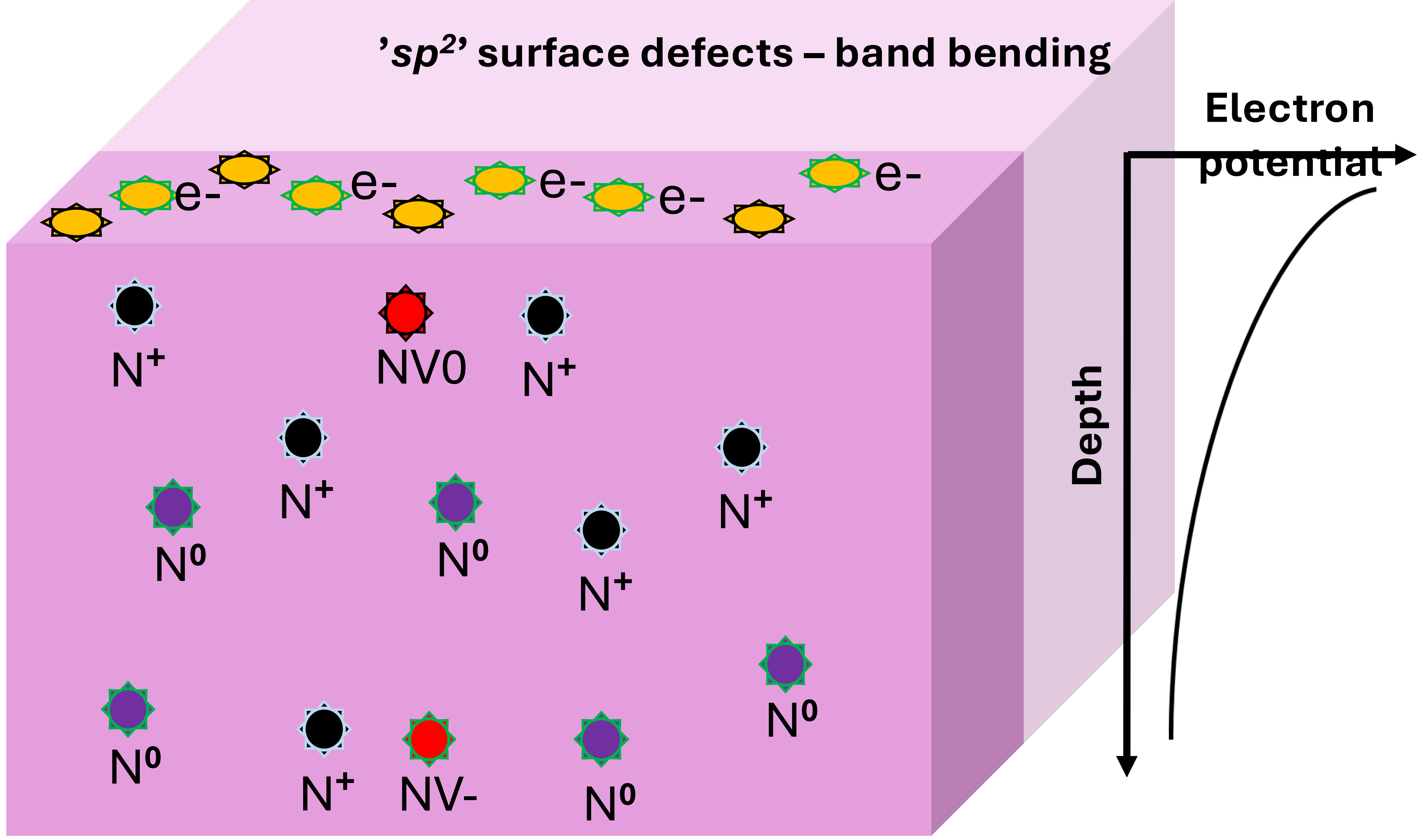


Unoccupied state spectroscopy with NEXAFS

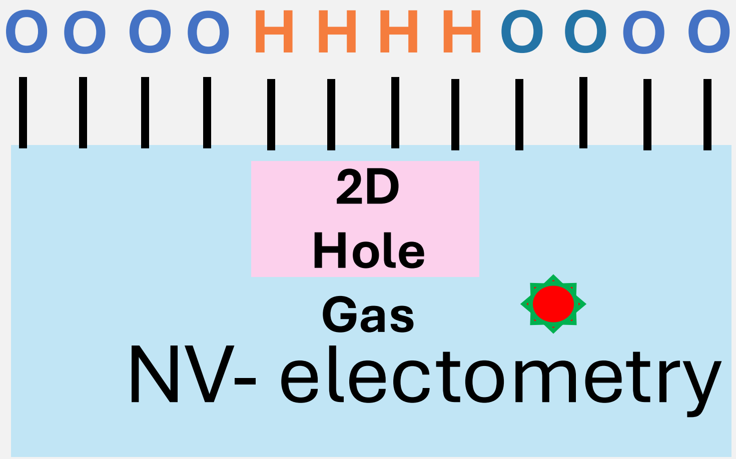


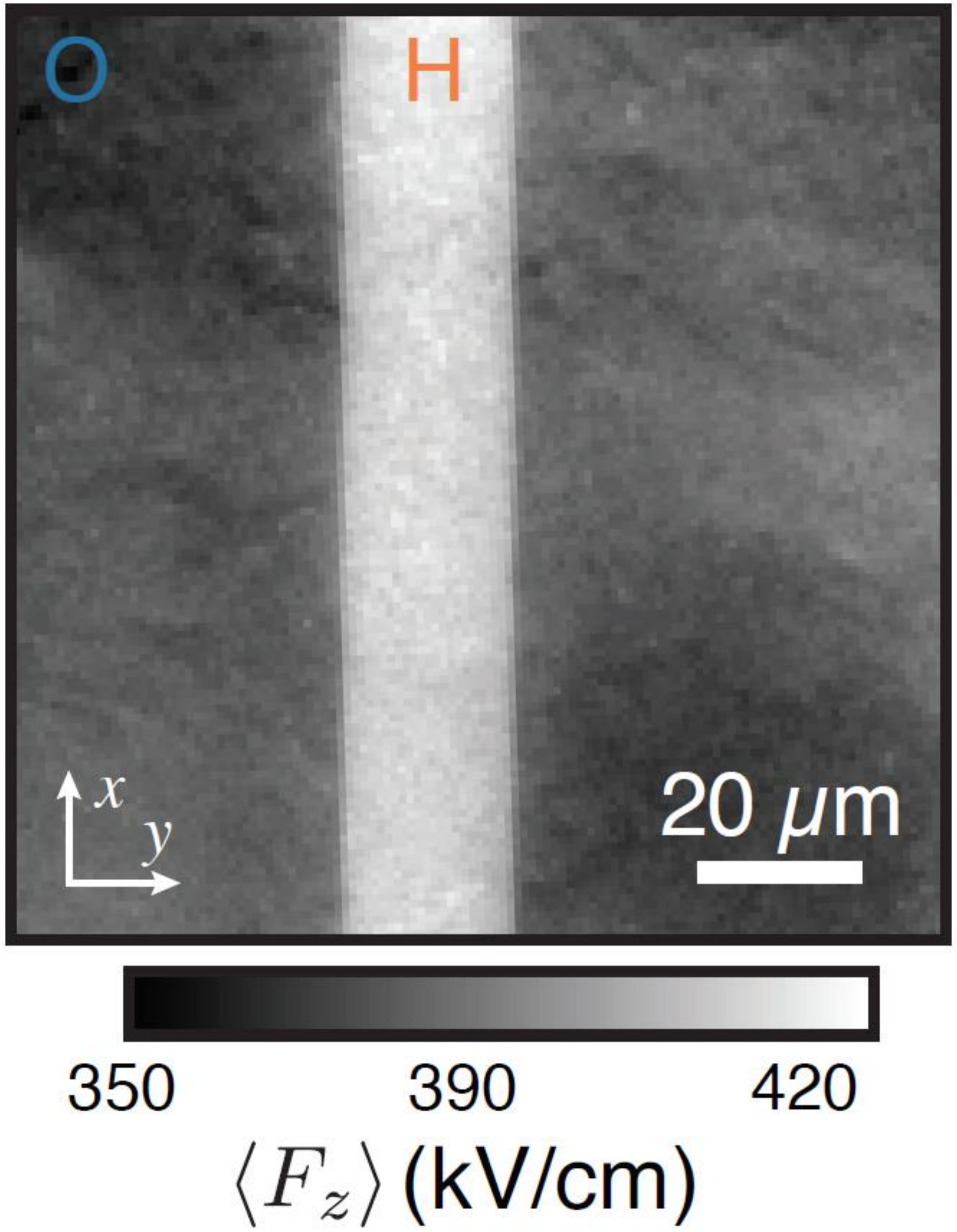
- **C=C bonds regardless of termination chemistry**
- **Defect sits at VBM + 1.5eV**

A. Stacey, et al., Adv. Mater. Inter. **6** (2019) 10801449


'sp²' surface defects – band bending
Electron potential
Depth
e-
e-
e-
e-
e-
N+
NV0
N+
N+
N+
N0
N0
N+
N+
N0
NV-
N0
N0

Observation of band bending

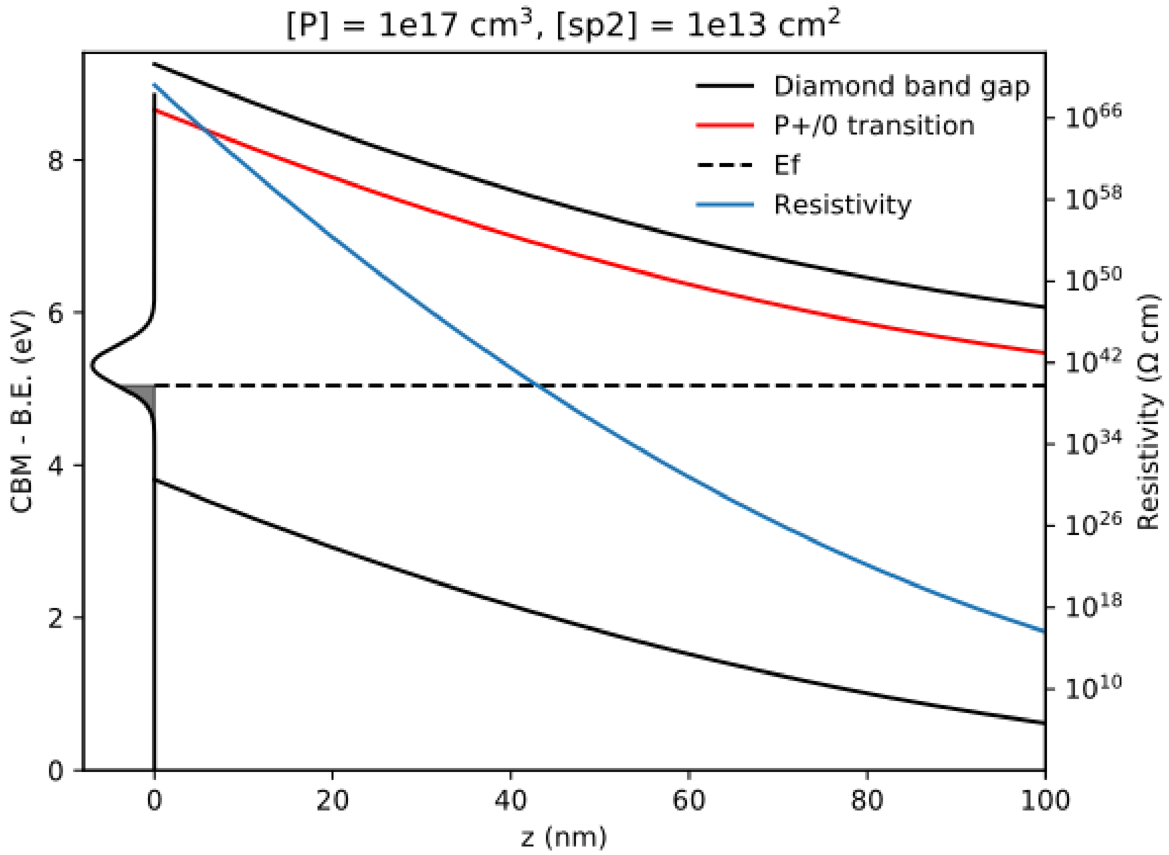




D.A. Broadway, et al., Nat. Elec. **1** (2018) 502-507

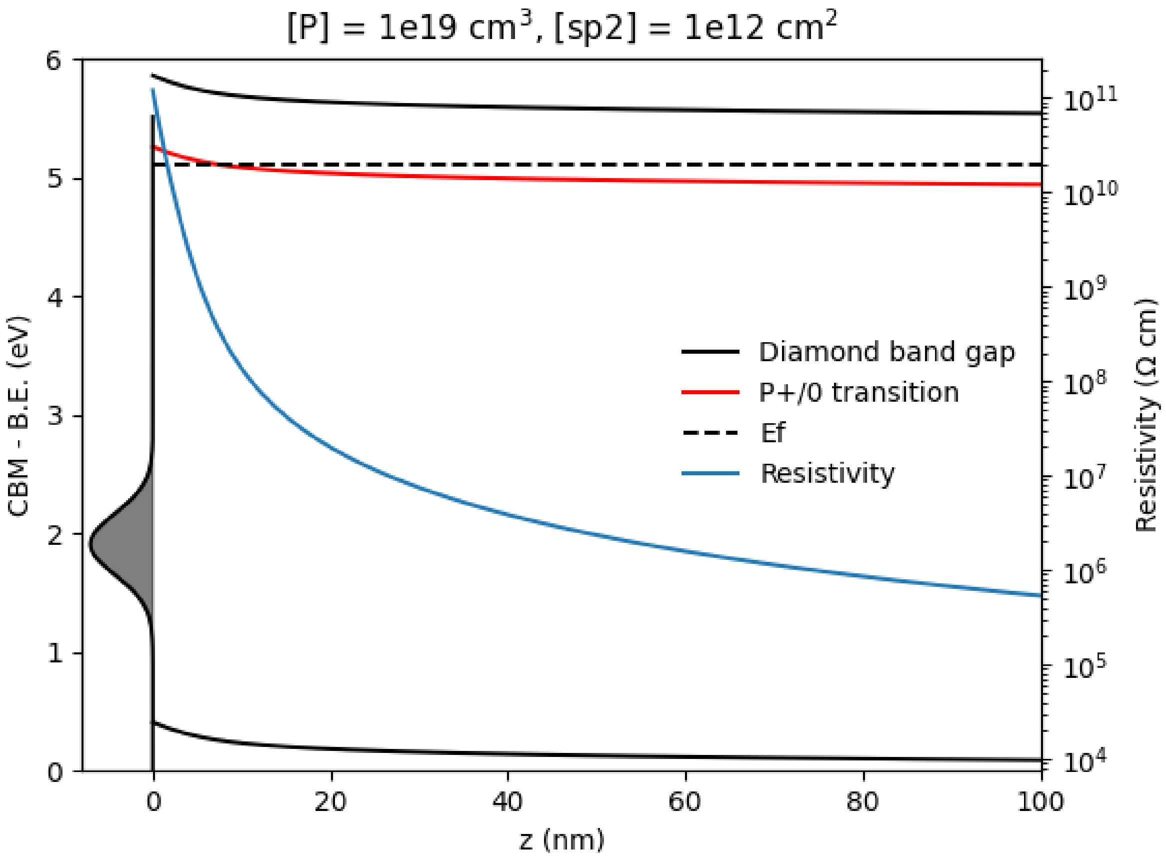
band-bending and co-dopants

Less phosphorus, more *sp²*



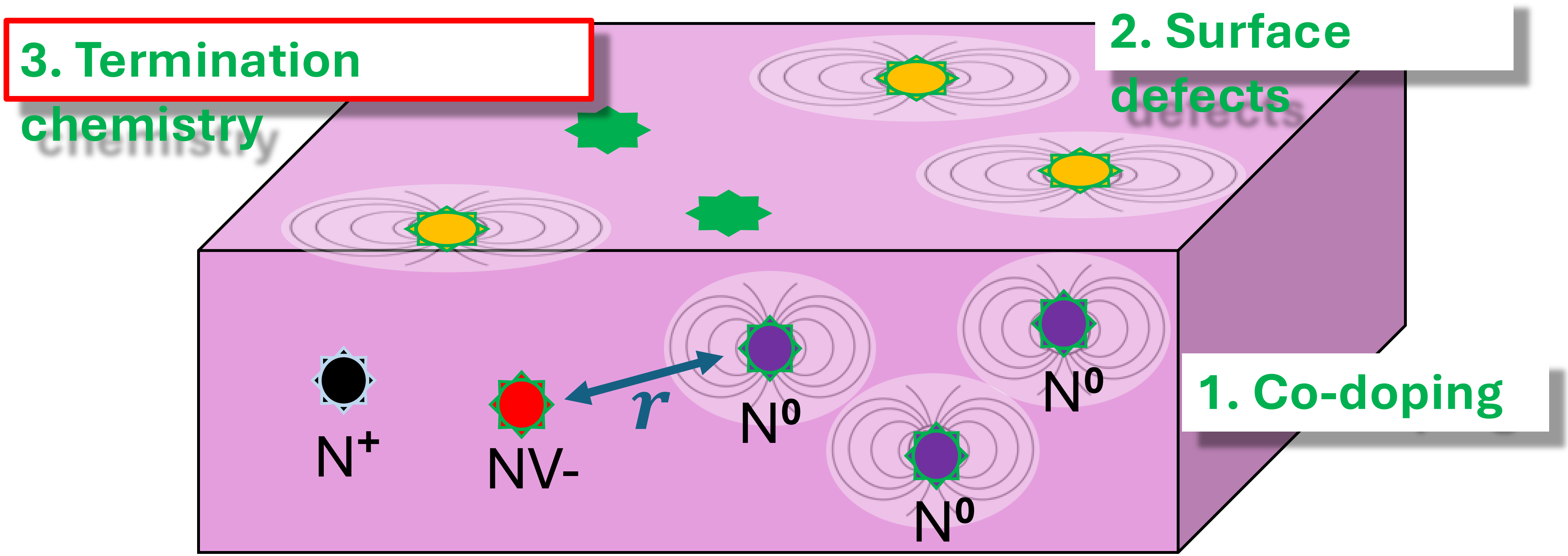
Unfilled surface states
→ Fermi pinning

More phosphorus, less *sp²*



Filled surface states
→ Surface noise sources

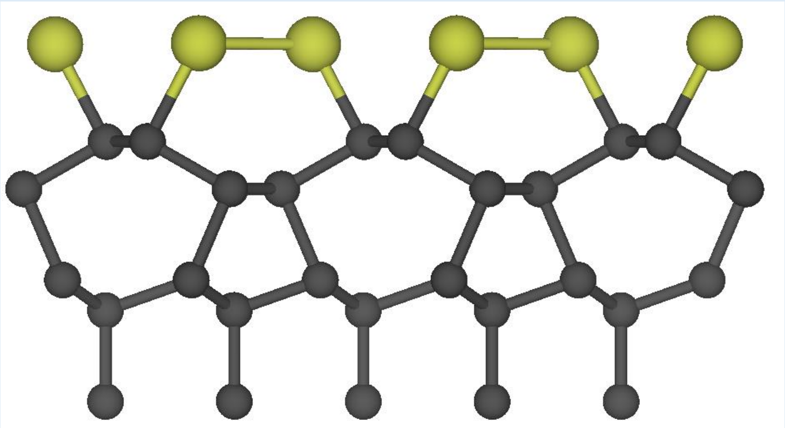
3. Termination chemistry



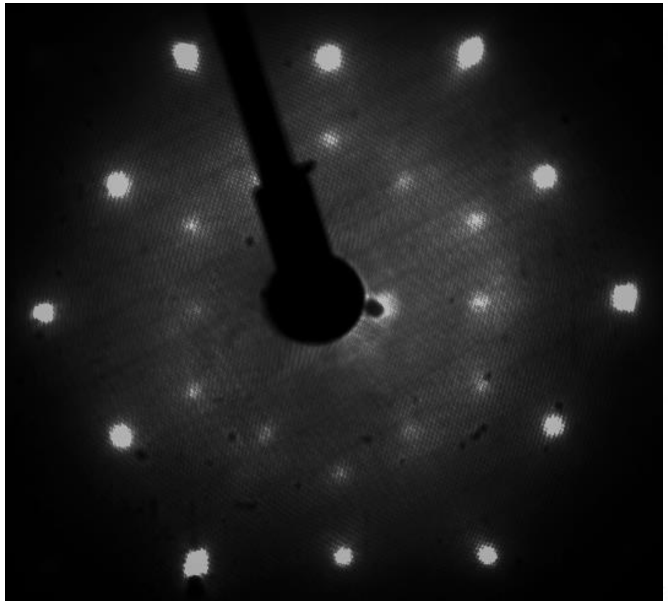
Novel surface toolbox

New Surface terminations

- Silicon
- Germanium
- Fluorine
- Nitrogen
- Lithium/Oxygen
- Magnesium/Oxygen

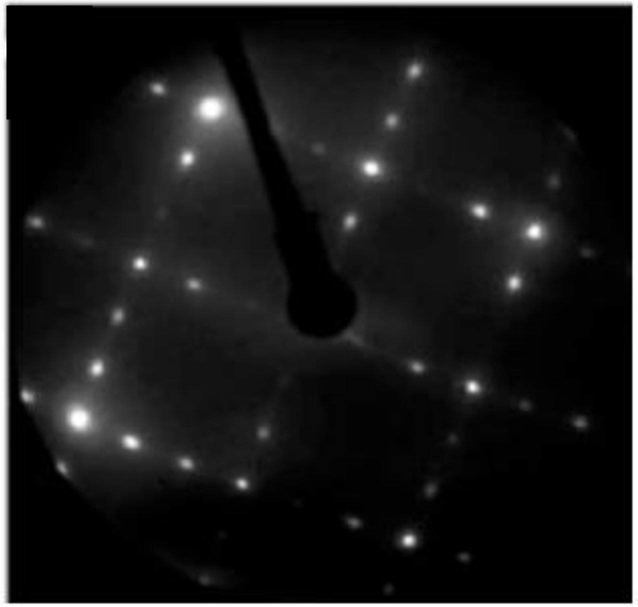


111 diamond



Si 2x1 & √3x√3R30 reconstruction

100 diamond



Si 3x1 reconstruction

Silicon Terminated Diamond

→ Most stable diamond surface measured

Toolbox of Surface Terminations

→ Different surface states

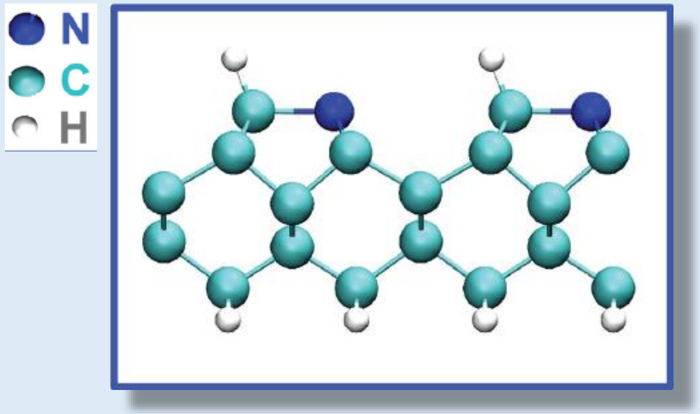
→ Different functionalization pathways

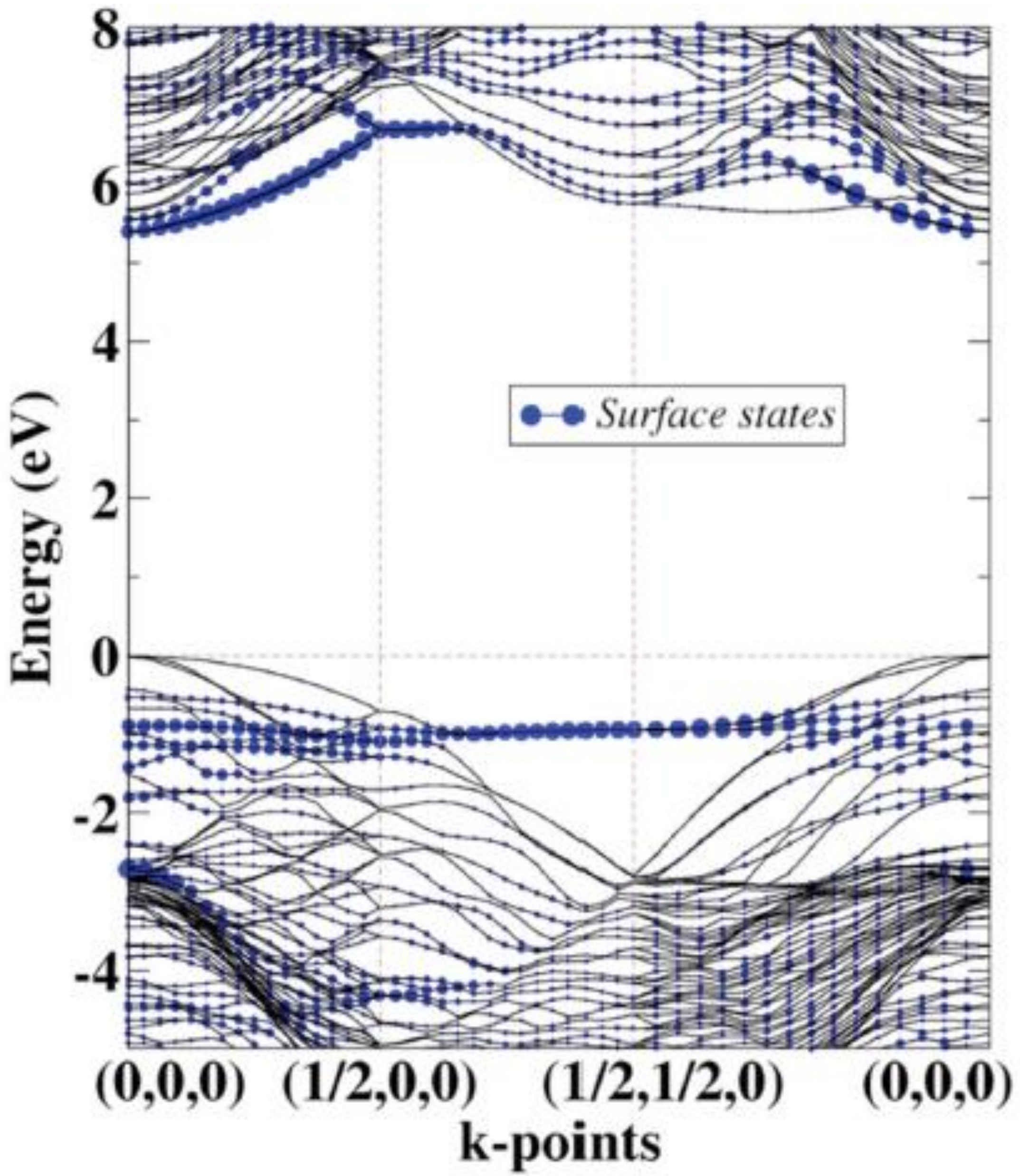
(Schenk et al., Appl. Phys. Lett. 106, 191603 2015)

Novel surface toolbox

New Surface terminations

- Silicon
- Germanium
- Fluorine
- Nitrogen
- Lithium/Oxygen
- Magnesium/Oxygen





A. Stacey, et al., Adv. Mater. Inter. **2** (2015) 1500079

Diamond synthesis for co-doping control

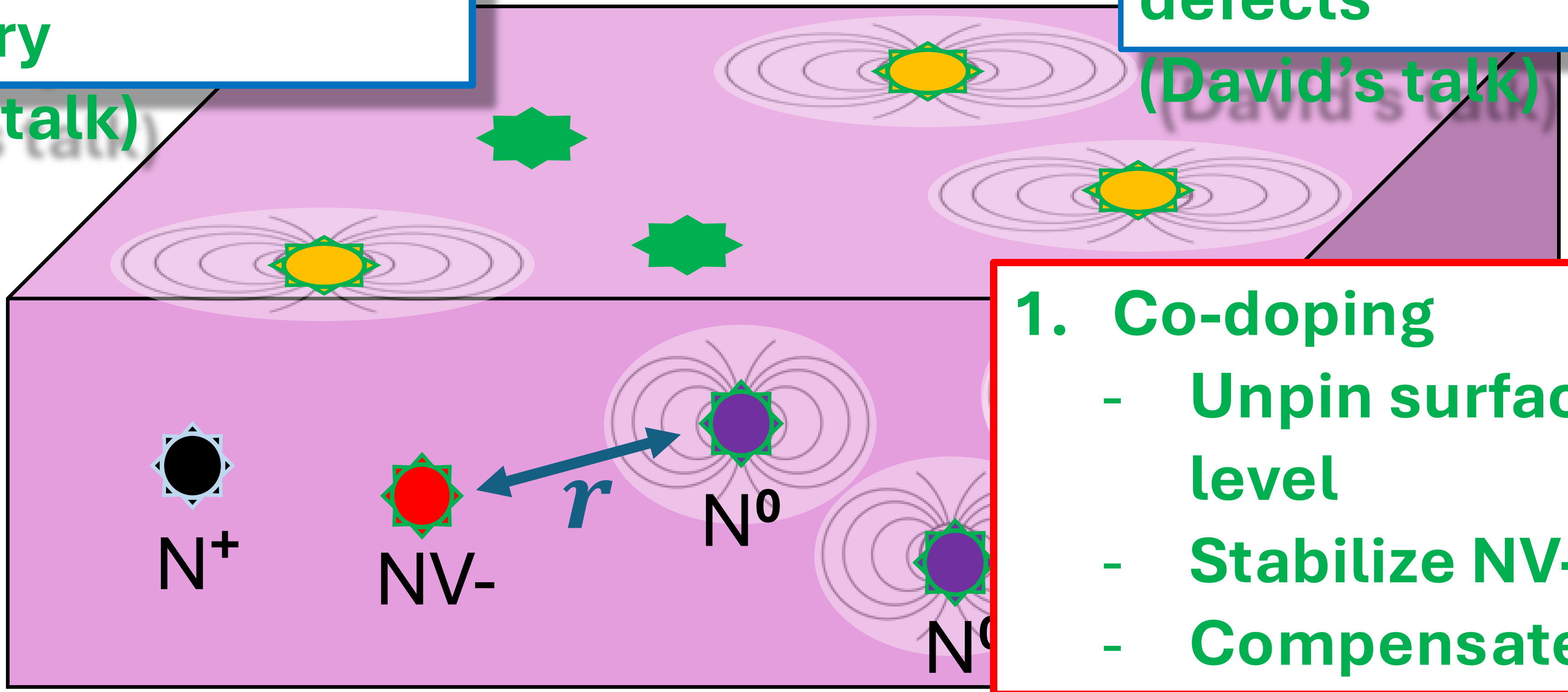
PPPL PRINCETON PLASMA PHYSICS LABORATORY

3. Termination chemistry

(David's talk)

2. Surface defects

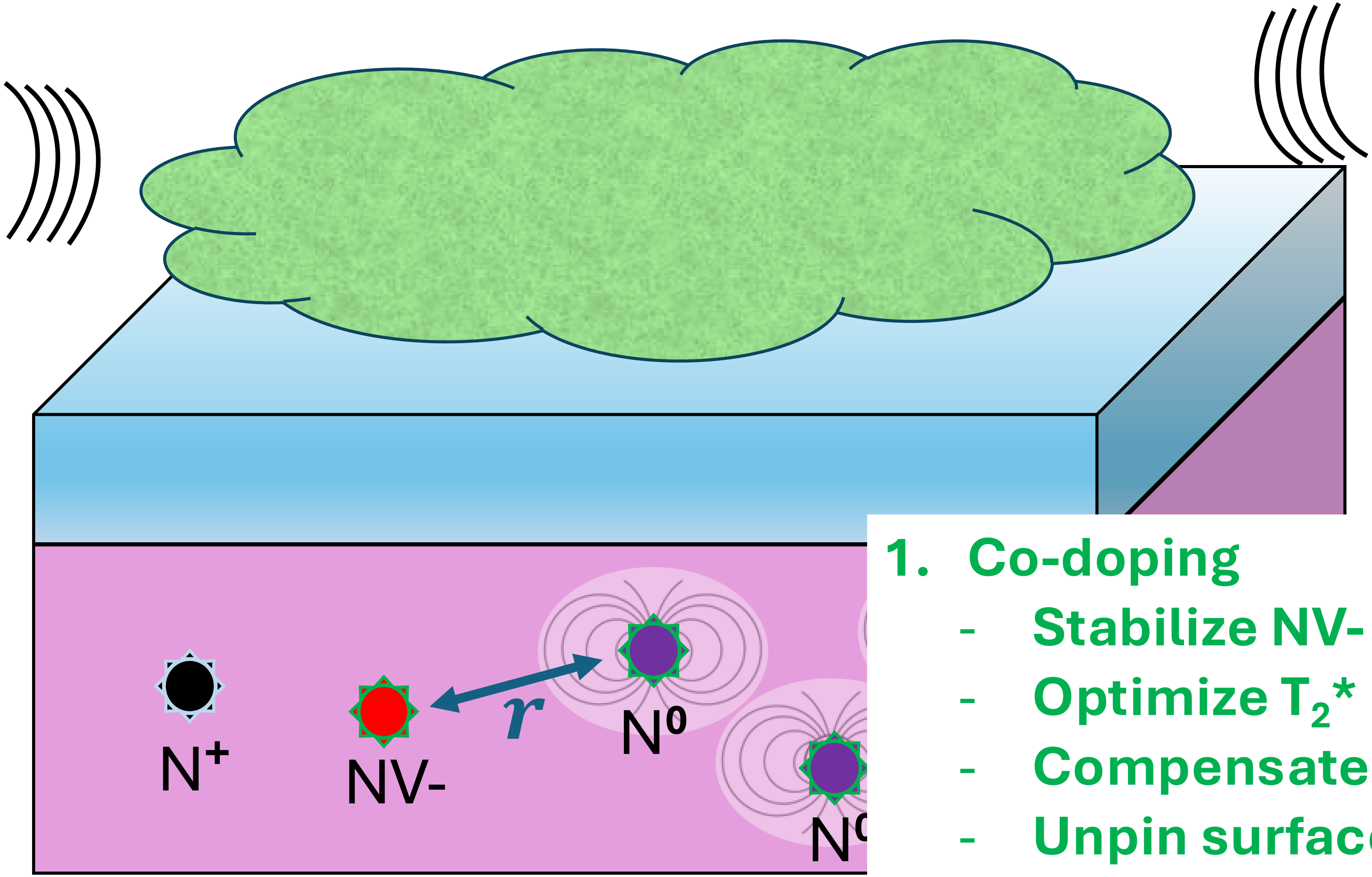
(David's talk)



1. Co-doping

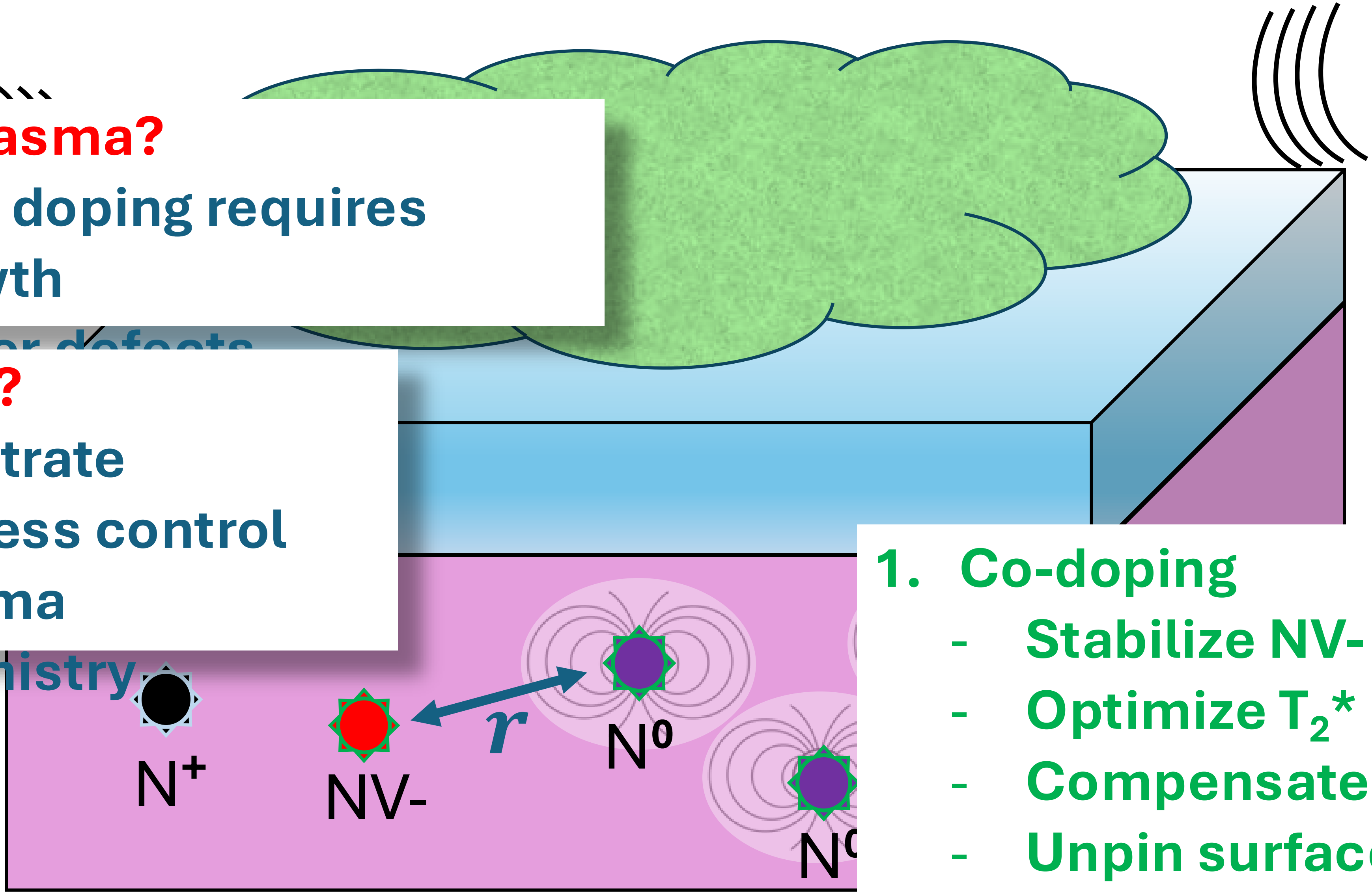
- **Unpin surface Fermi level**
- **Stabilize NV-**
- **Compensate bulk spins**
- **Optimize T_2^***

Plasma synthesis for co-doping control



1. Co-doping
    - Stabilize NV-
    - Optimize T_2*
    - Compensate bulk spins
    - Unpin surface Fermi level

Plasma synthesis for co-doping control



Why Plasma?

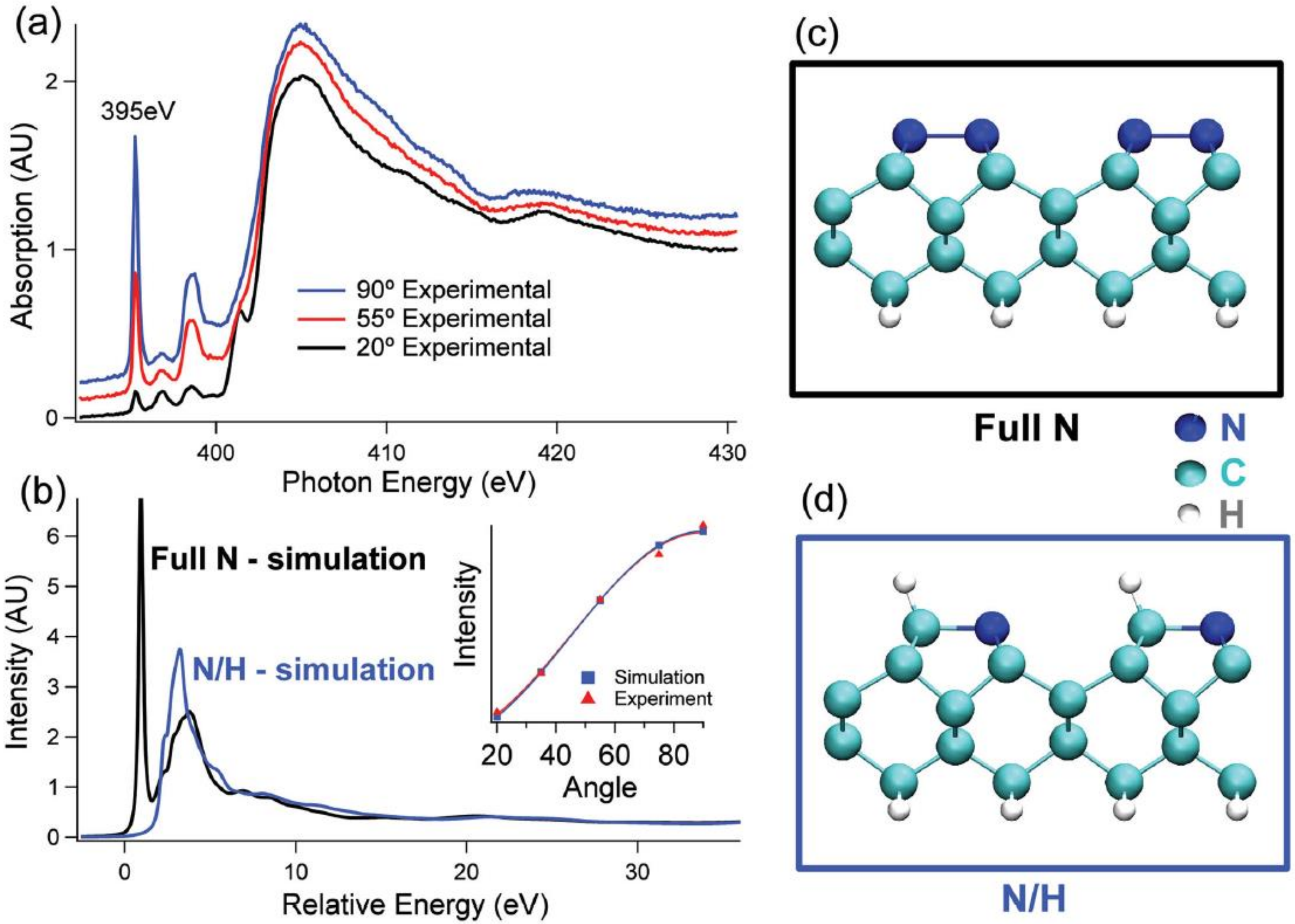
- Bulk doping requires growth
- Fewer defects

Control?

- Substrate
- Process control
- Plasma chemistry

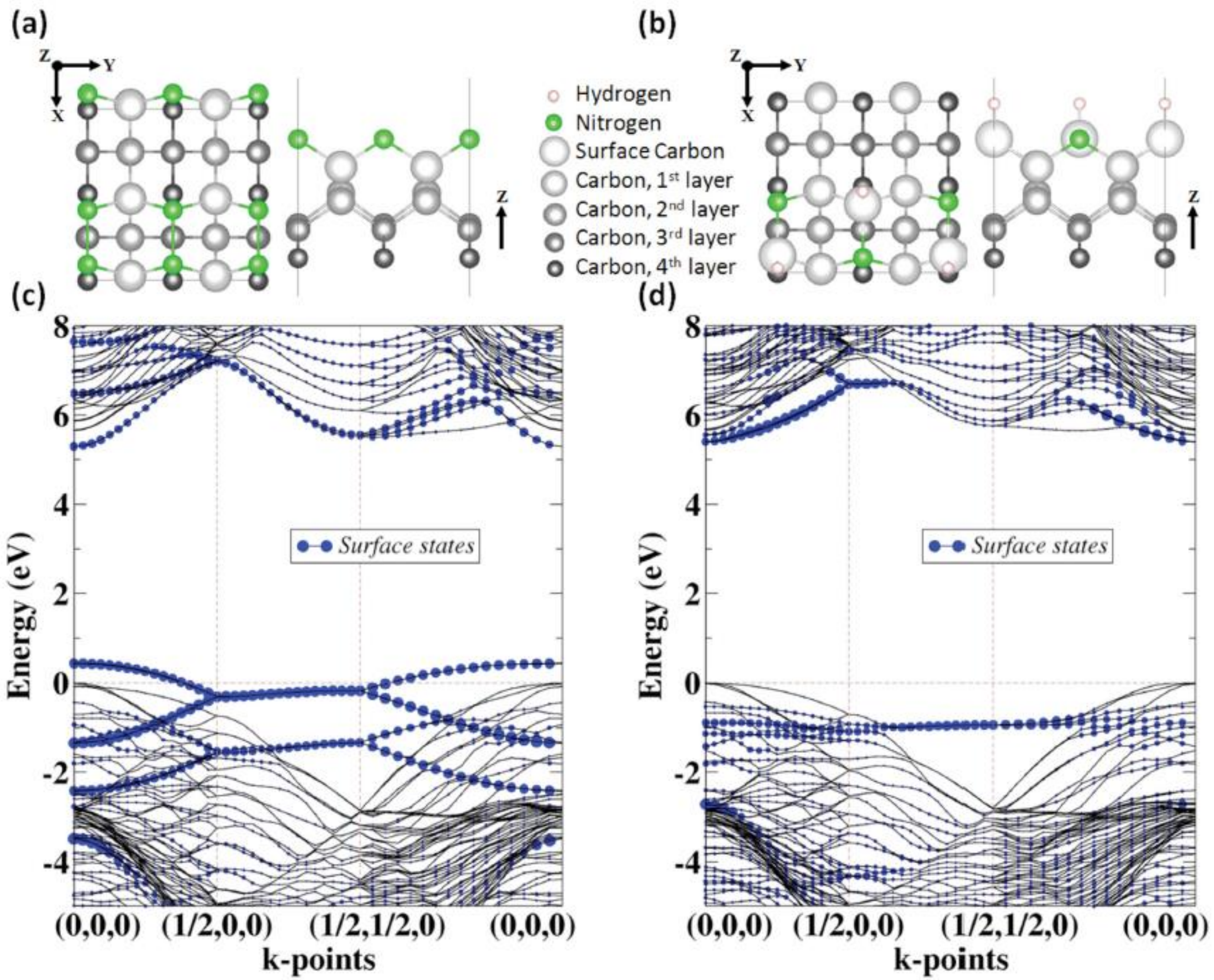
1. Co-doping
   - Stabilize NV-
   - Optimize T_2^*
   - Compensate bulk spins
   - Unpin surface Fermi level

Nitrogen Structure Evidence



A. Stacey, et al., Adv. Mater. Inter. **2** (2015) 1500079

Nitrogen termination



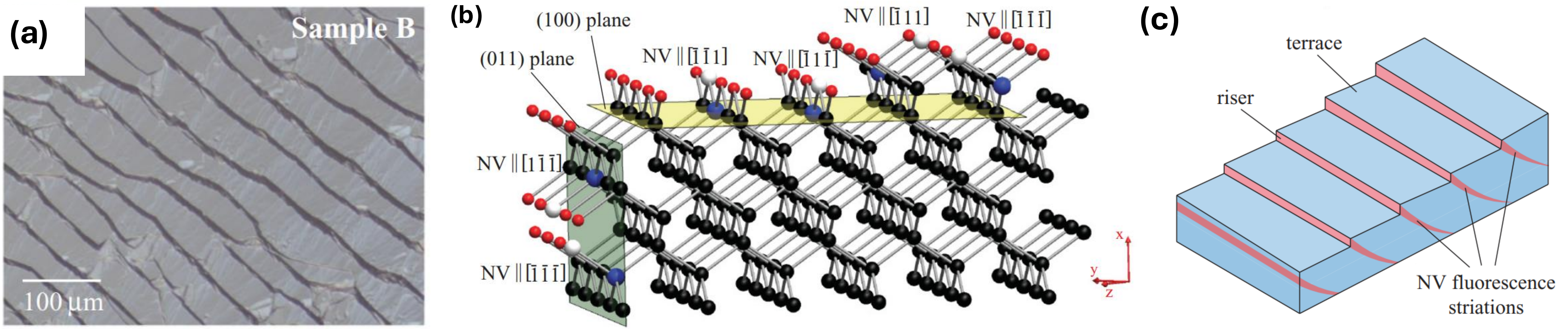
Nitrogen Terminated Diamond

Clear band-gap (almost)

- No additional unoccupied ‘trap states’
- Natural valency = spin-0

A. Stacey, et al., Adv. Mater. Inter. **2** (2015) 1500079

Substrate control: surface miscut

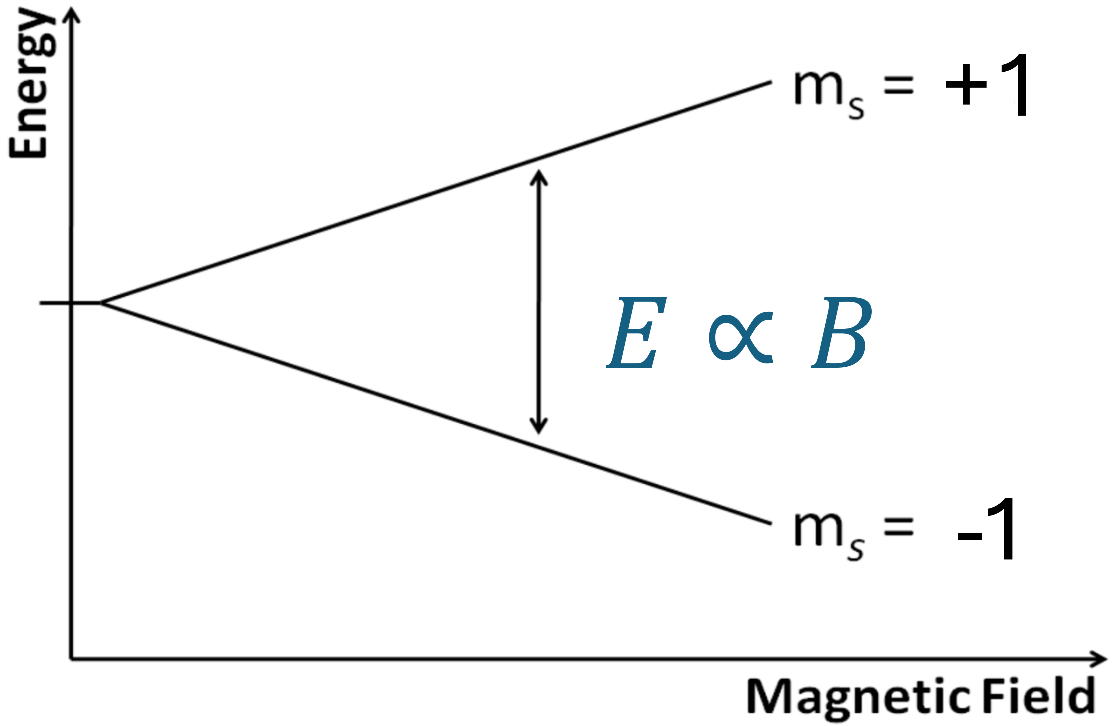


→ **Wrong miscut angle can induce inhomogeneous doping**

L.M. Pham, et al., Phys. Rev. B **86** (2012) 121202(R

Magnetometry with NV centres

Magnetometry = Zeeman effect



$$S \propto \frac{1}{\sqrt{[NV] \times T_2^*}}$$

Shot noise limited readout (real or apparent NV density)

Magnetic noise (real or 'virtual')

→ **Extrinsic noise sources and charge traps limit magnetometry**

$$[N] \propto [NV]$$

N

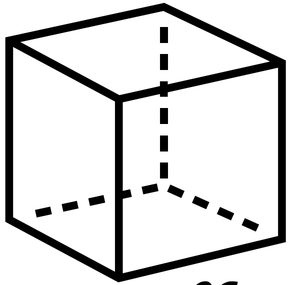
+

NV^-

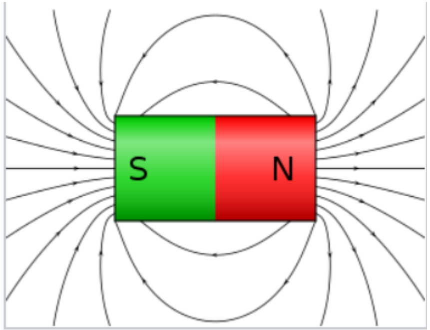
N^0

N^0

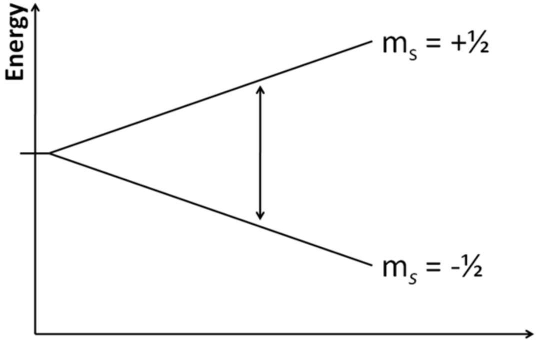
$$\bar{r} \propto \frac{1}{\sqrt[3]{[N]}}$$



$$\bar{B} \propto \frac{1}{\bar{r}^3}$$



$$T_2 \propto \frac{1}{\bar{B}}$$



Scourge of donor spins

$$T_2 \propto \frac{1}{[NV]}$$

$$S \propto \frac{1}{\sqrt{[NV] \times T_2^*}}$$

$$S \propto 1$$