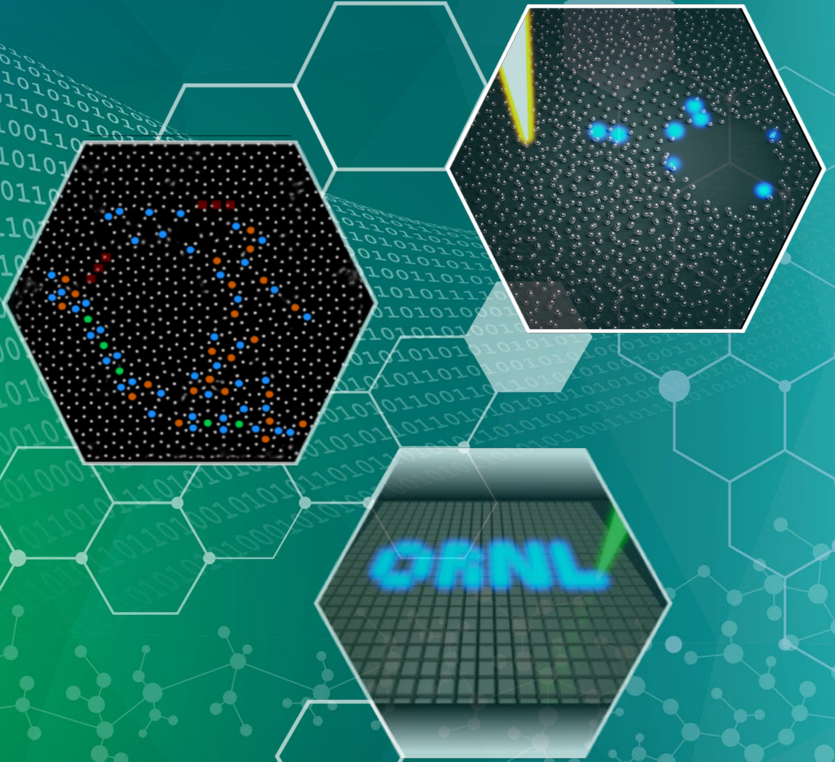
Lecture 04

From post-acquisition to real-time analysis: ensemble networks and automated experiment



Kevin Roccapriore[1,2] and Sergei V. Kalinin[3,4]

[1]Center for Nanophase Materials Sciences, Oak Ridge National Laboratory
[2]AtomQ
[3]University of Tennessee Knoxville
[4]Pacific Northwest National Laboratory

Motivation

Robust post-experiment **analytics**

Hyperspectral measurements of **sensitive materials**

Measurements of **specific atoms**

Selective **atom removal**

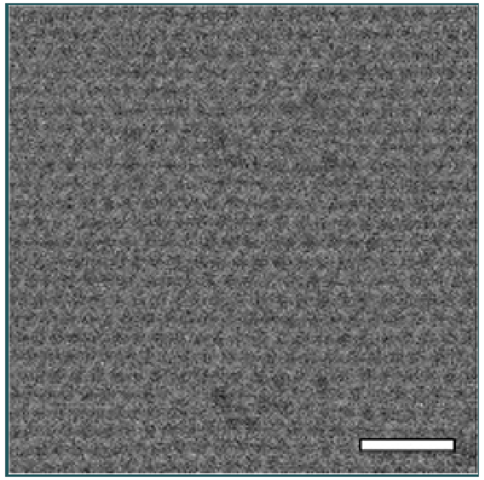
Extract **beam-matter interactions**

Creation of various **defects**

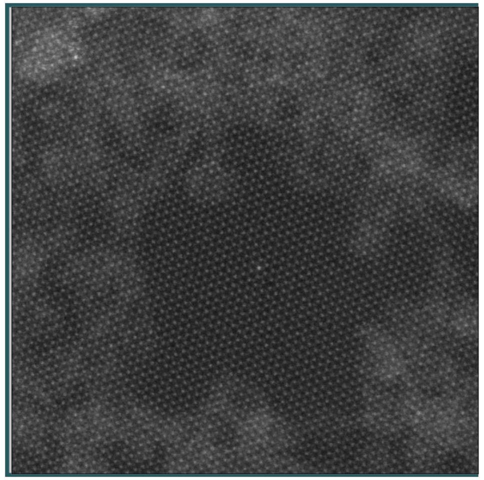
Measurements of created **defects**

Variation of STEM images

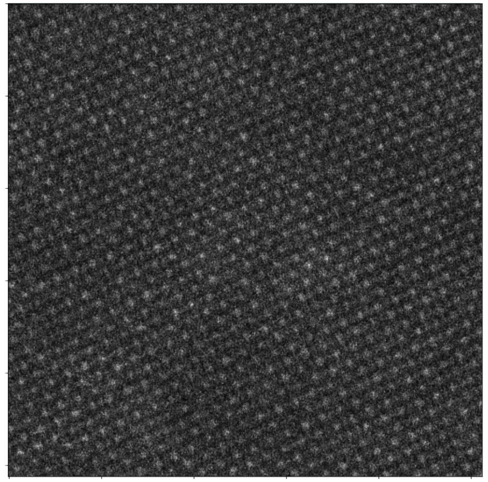
Graphene (MAADF)



MoS_2 (HAADF)

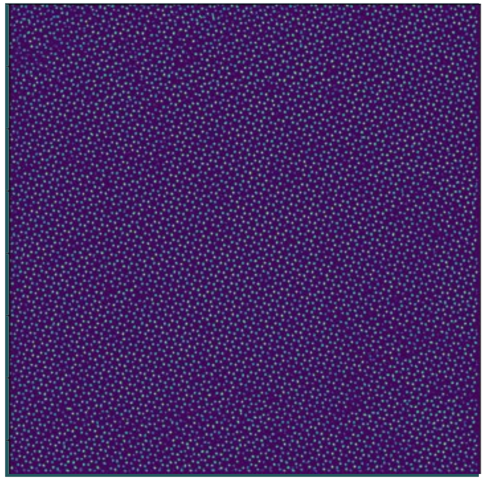


CrSBr (multilayered) (HAADF)

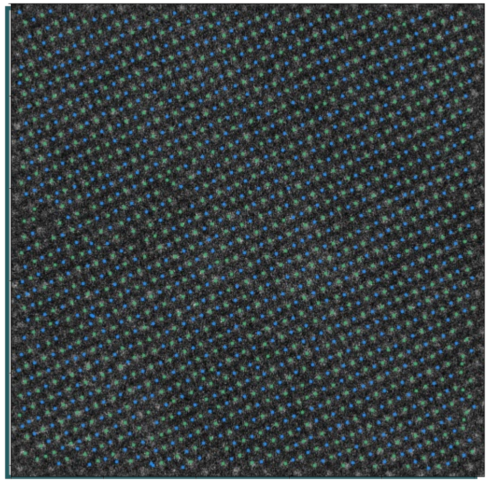


Model prediction

Model prediction



Model prediction



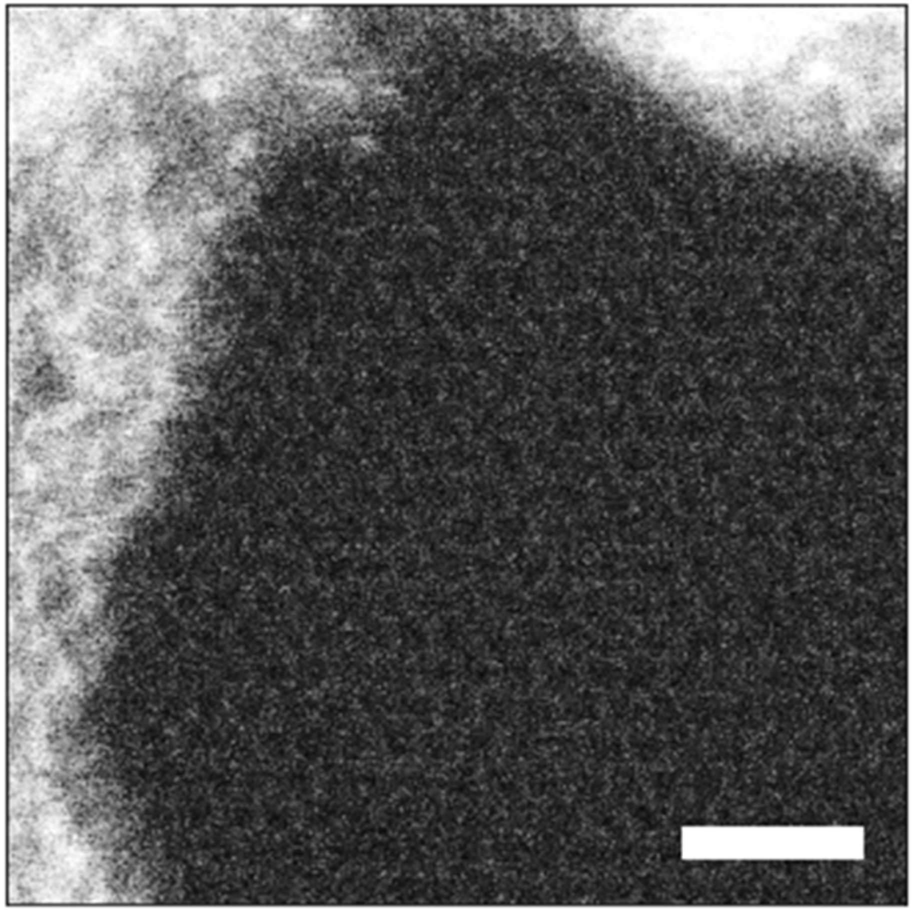
Handling different...

- Materials
- Microscope settings
- Environmental behaviors (uncontrollable):
  - Contamination
  - Electrical noises
  - Other items that cause changes to the image signal
- Material evolution of material under stimuli (e.g., beam itself!)

Ensemble learning

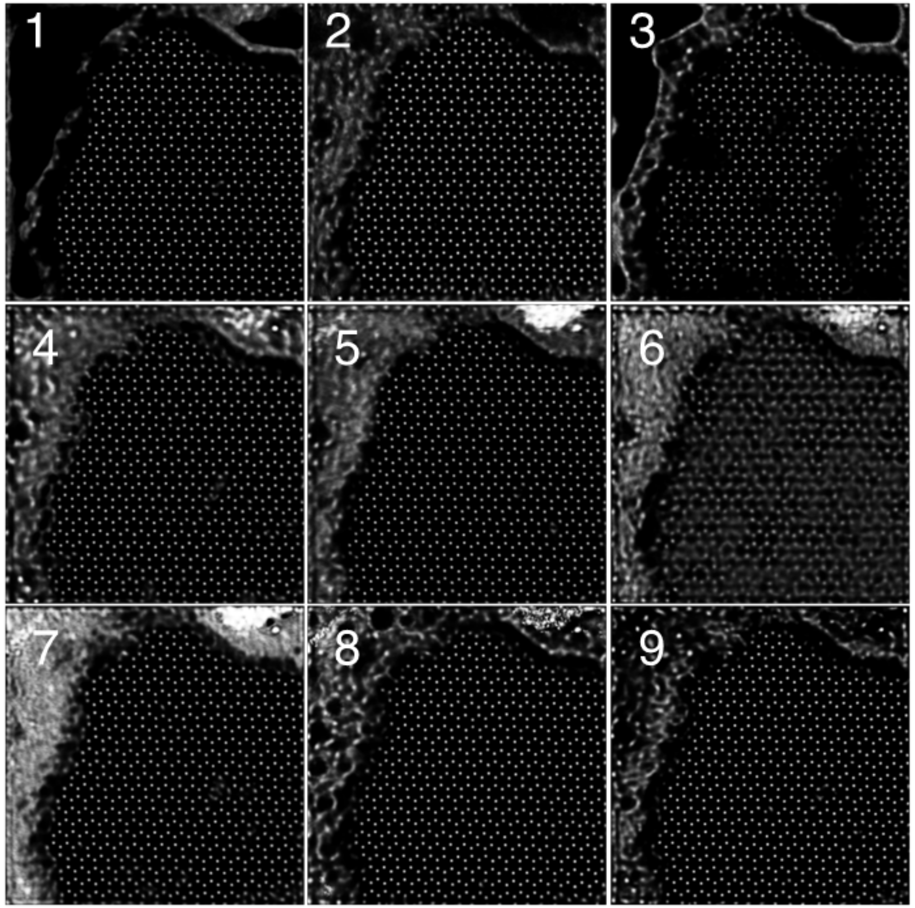
Key for robustness and enables on-the-fly predictions

Live Raw data (ADF)

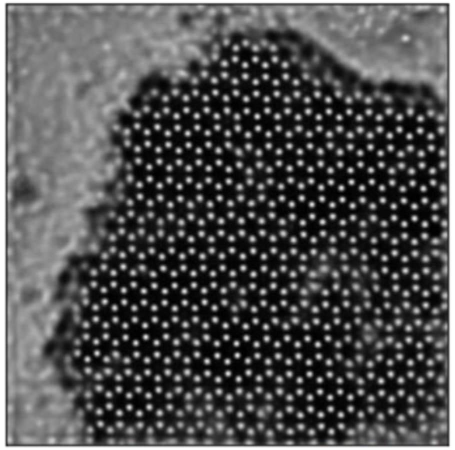


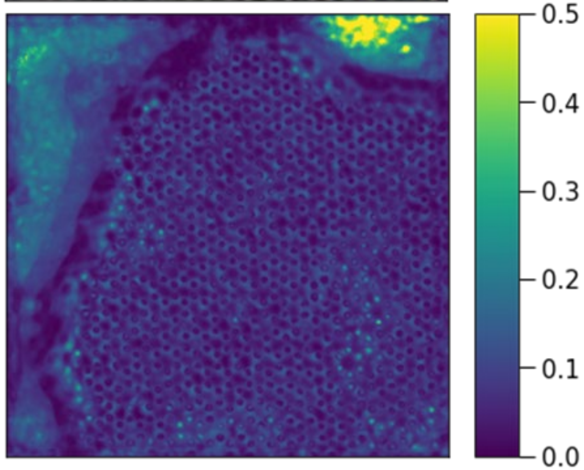
Graphene

Individual model predictions



Ensemble prediction





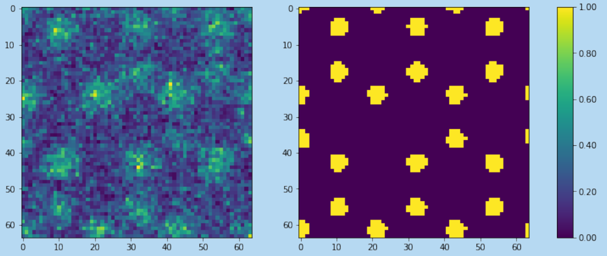
Ensemble uncertainty

(Note: may have single or multiple classes)

Atomic classification

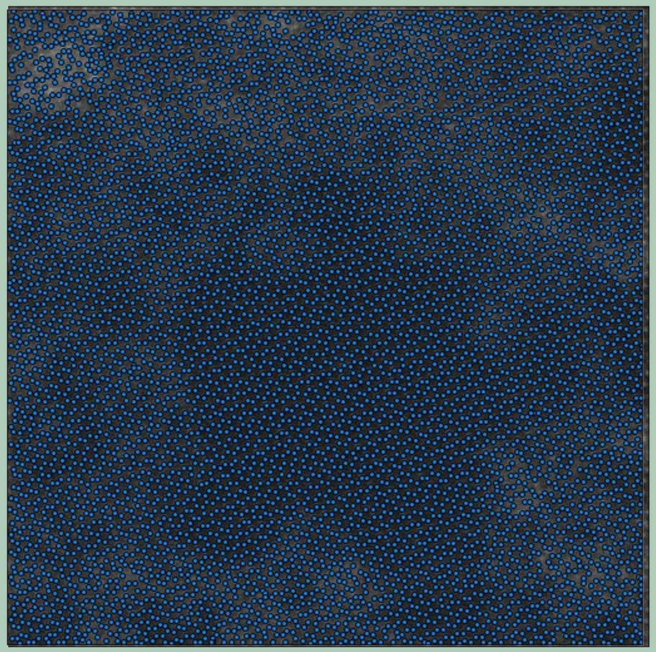
1. <u>Train with **simulated** data</u>

Simulated Labeled



e.g., MoS_2, all are just "atoms" (no classes)

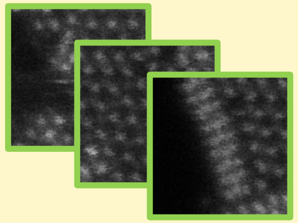
2. <u>**Experimental** image prediction with initial model</u>

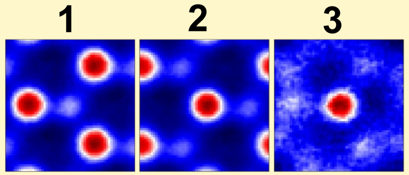


3. <u>Augmentation</u>

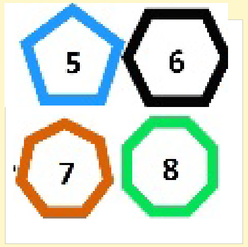
retrain using experimental image

Image patch local analysis

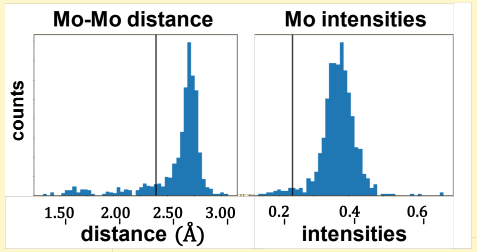




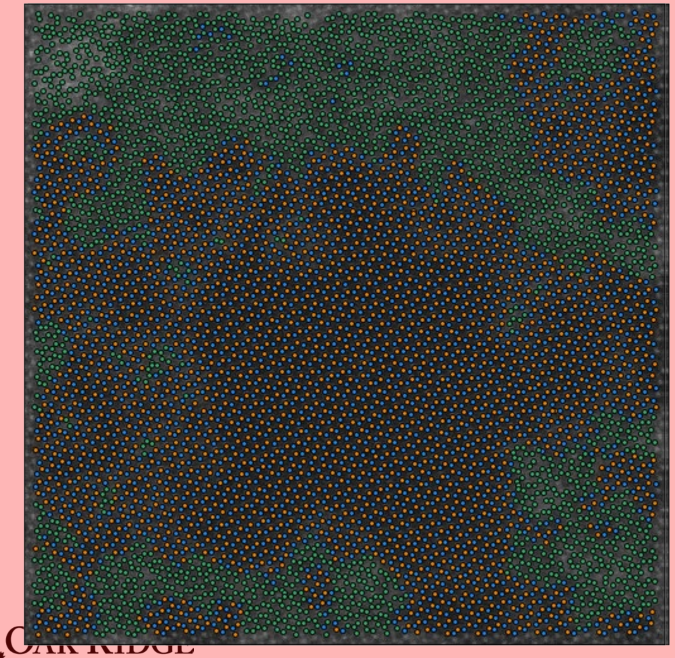
Graph analysis



Intensity & distance comparison



4. <u>Redeploy new model</u>



"I might have more atom types appear during my experiment"

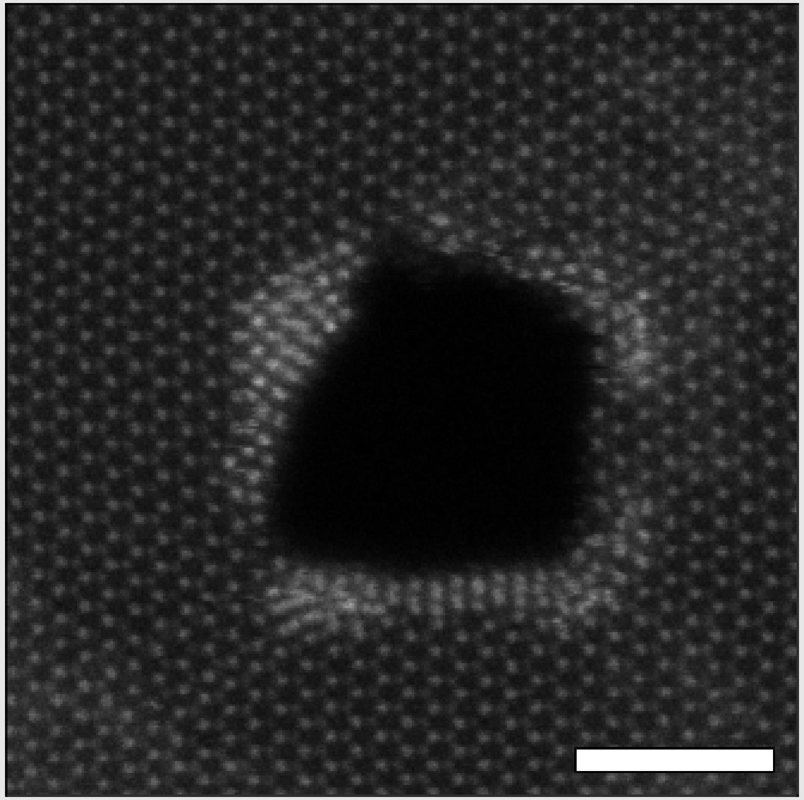
or

"Microscope conditions might change day to day for reason XYZ…"

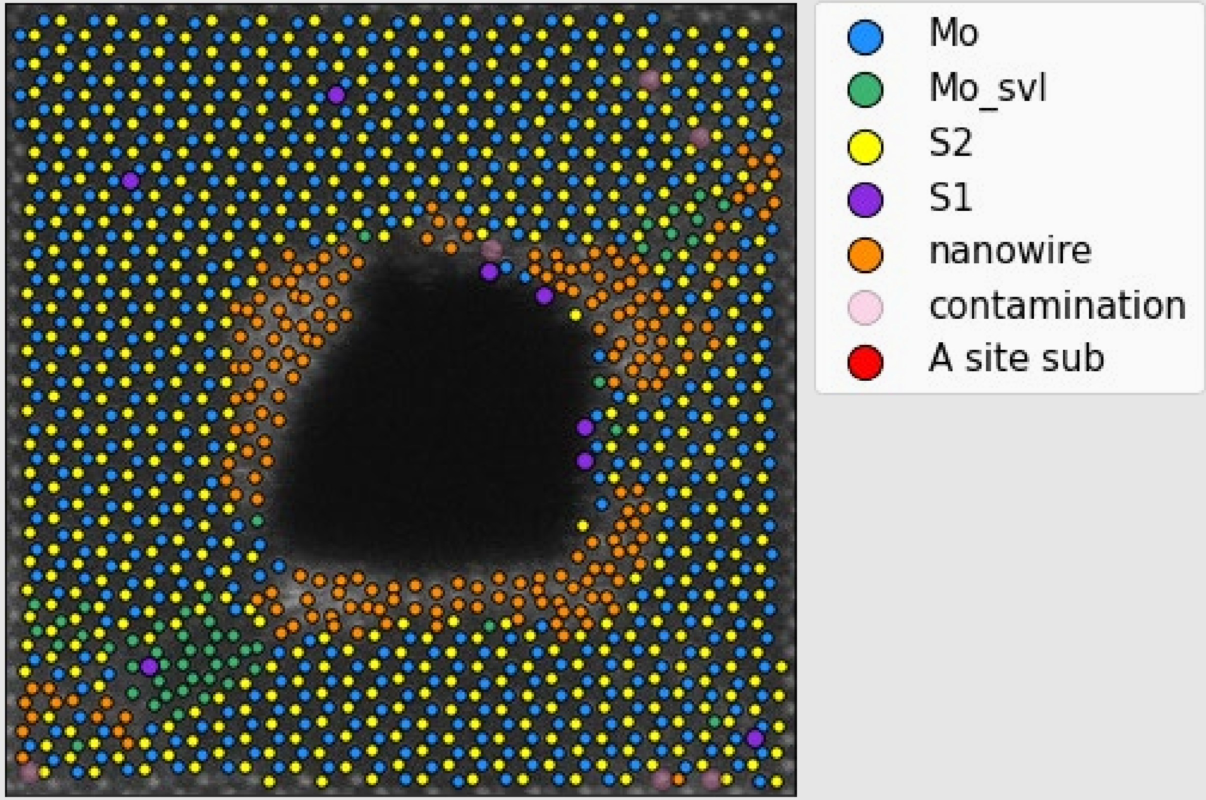
Atomic classification

- Many atom and defect types can evolve
- Defect configuration space is large!
- We do not need to train new models for each new type (impractical)

New HAADF-STEM image



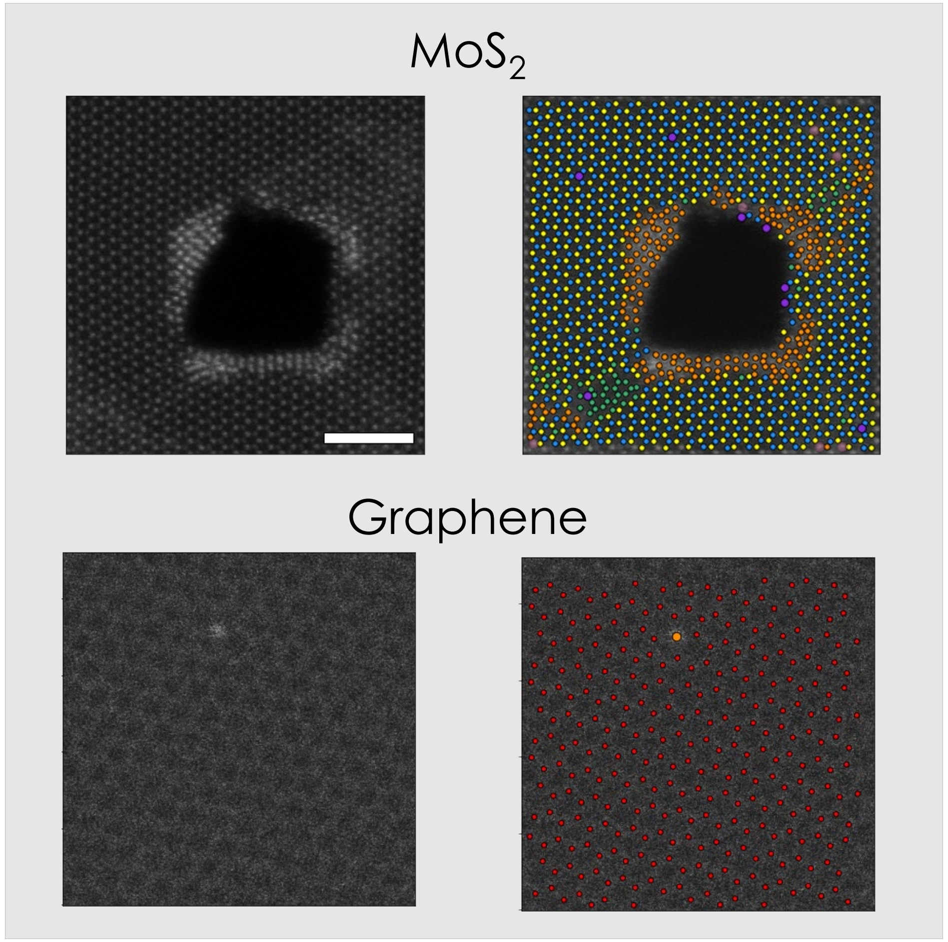
With augmented ensemble network



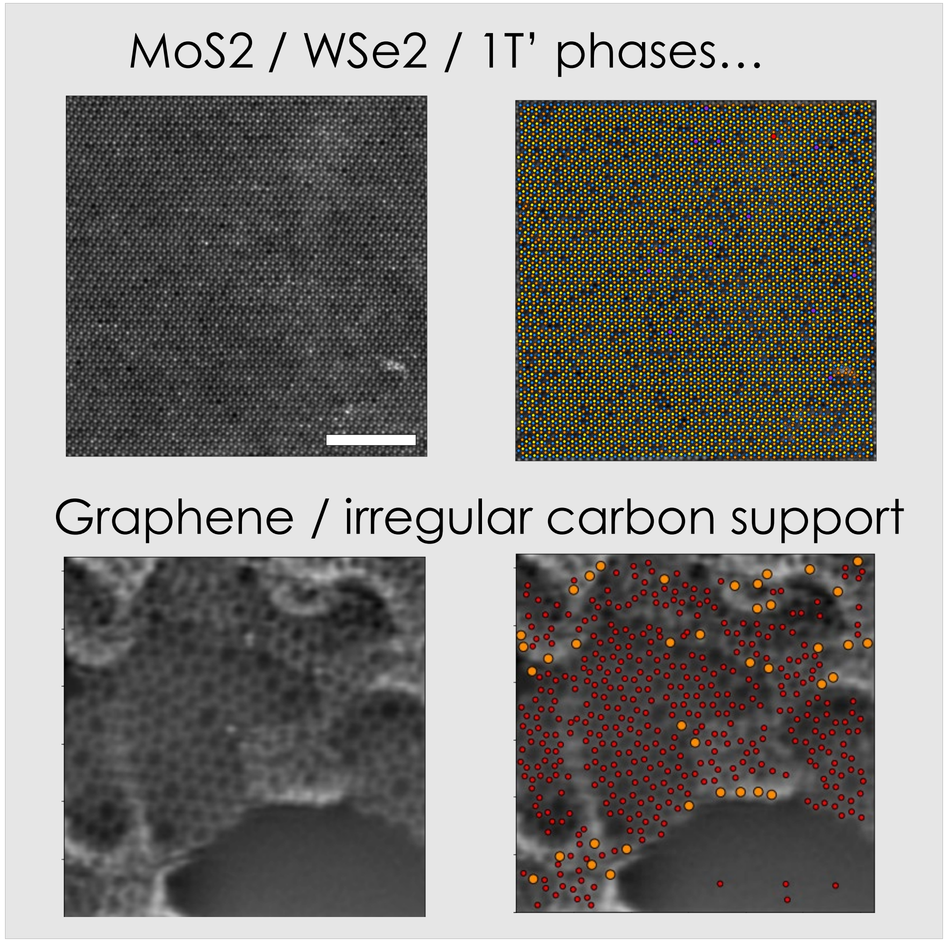
Model transfer

A quick note…

- Similar symmetry? **Maybe you do not need to train a new model from scratch**
  - at least, maybe not the image simulation part!
- Imaging modalities and conditions: HAADF, MAADF, ABF, a-ADF, CoM, etc.



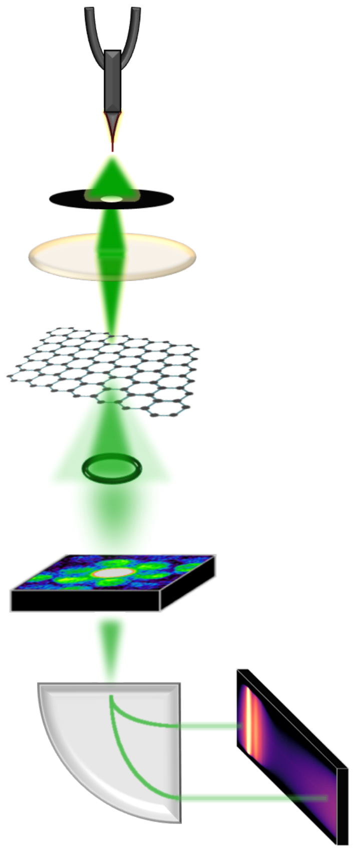
Versatile / multiple uses



Automated Experiments

What is smart control and what does it do for us

in the scanning transmission electron microscope



1. Deciphering image data

2. Precise beam placement

must occur during experiments
(fast, reliable)

SOFTWARE SOLUTIONS

Image recognition,
dose control,
structure-property extraction...

HARDWARE SOLUTIONS

Scan control,
beam blanking…

Enable new class of experiments

- Drive and study reactions, atom by atom
- Measurements of dynamic processes in sensitive materials
- Single atom-beam time dynamics
- “Self-driving” experiments?
- …?

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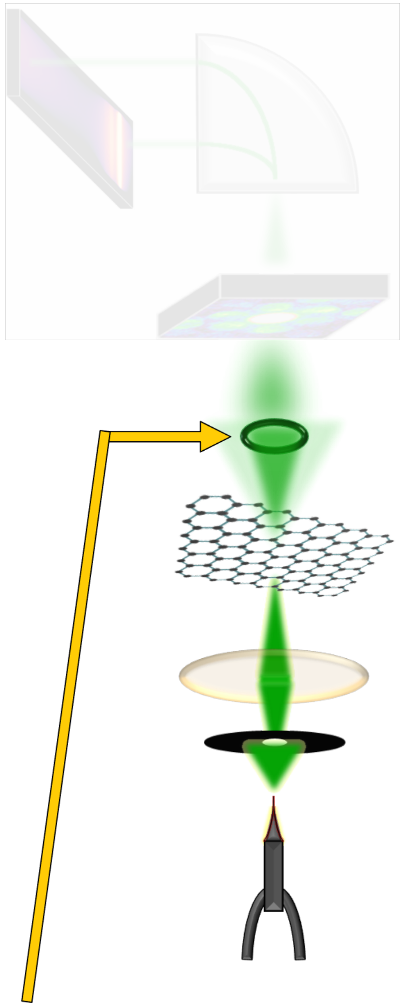
Open slide master to edit

Atomic coordinates

- *Robust* technique to locate (and classify) atomic species from a noisy detector image
- Single layer graphene, 60kV
- Key: ensemble learning with deep convolutional neural networks

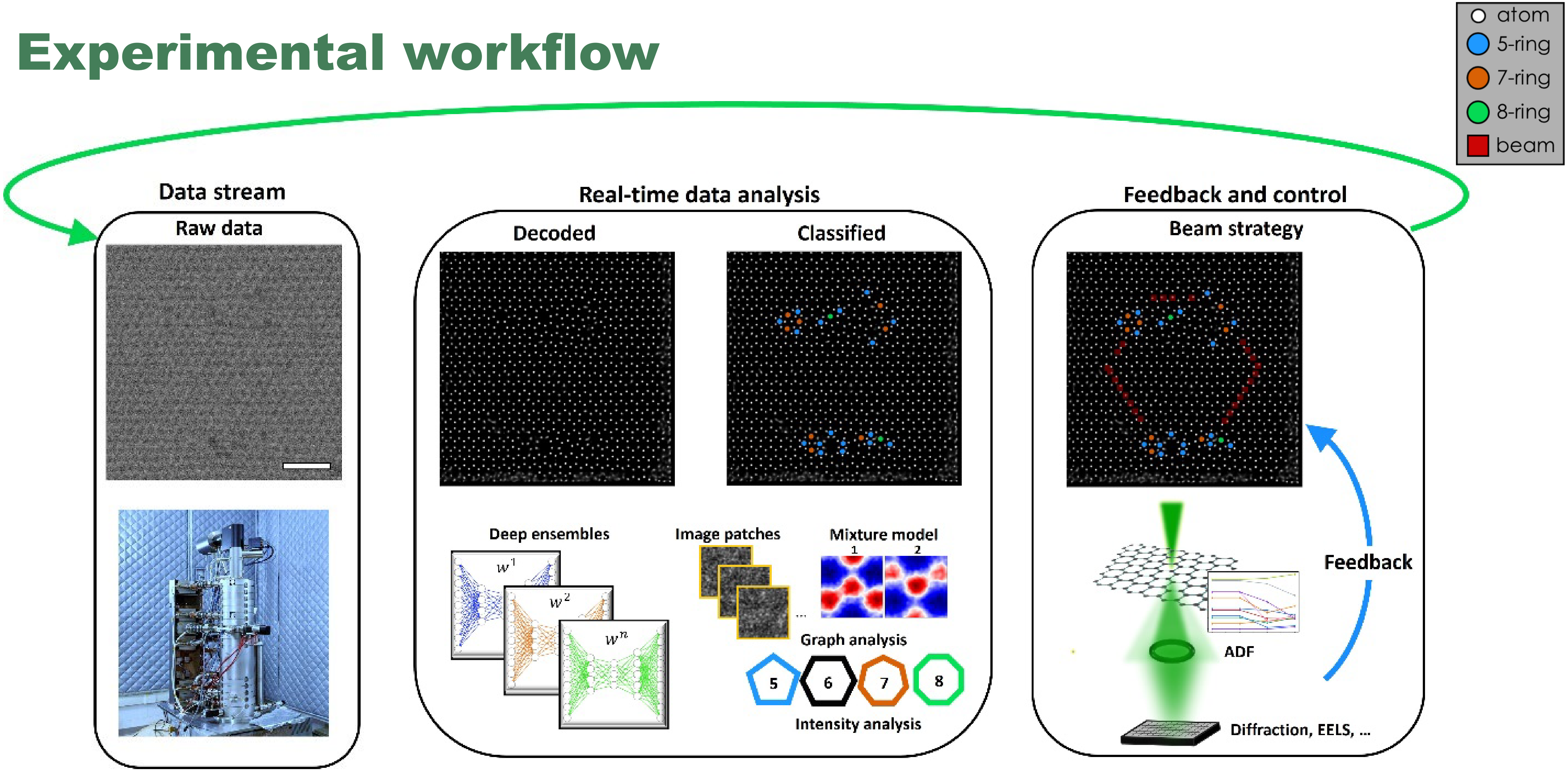
Predicted image (screen record video) Raw image



Usually: STEM images from annular dark field (ADF)

Roccapriore K.M, et. al., ACS Nano 2022, 16, 10, 17116–17127

Experimental workflow



Ensemble learning intro: **Ghosh A. et. al., npj Comput. Mater. 2021, 7 (1), 1– 8**

Use in real-time on microscope: **Roccapriore K.M, et. al., ACS Nano 2022, 16, 10, 17116–17127**

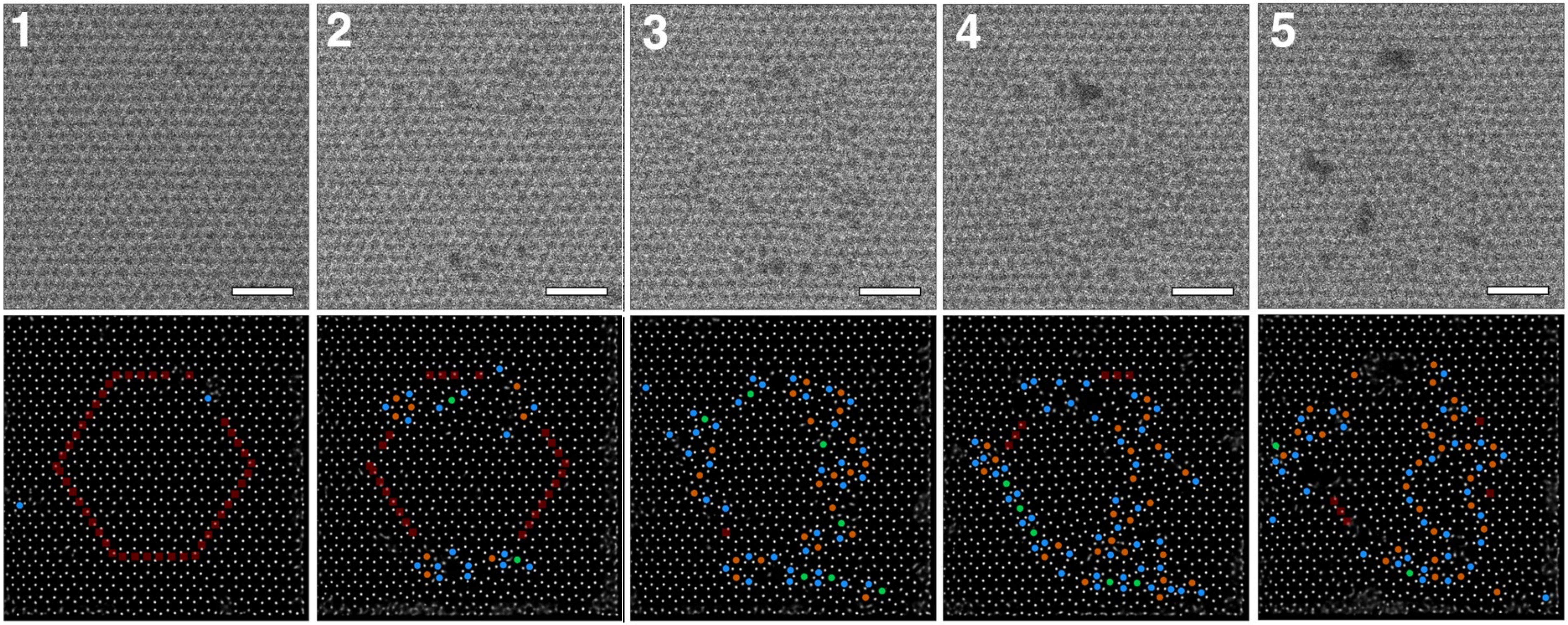
Avoidance patterning

Topological defects

Graphene

Specify desired **end-state**

Image → Action → Image → Action…



https://doi.org/10.1021/acsnano.2c07451

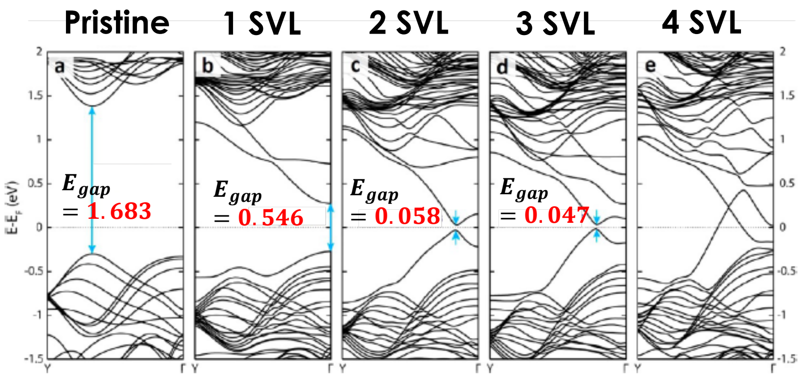
Selective atom removal

MoS_2: single vacancy lines (SVL) → functional properties

- ➡ Atomic band structure engineering?
- ➡ Atomic circuitry?

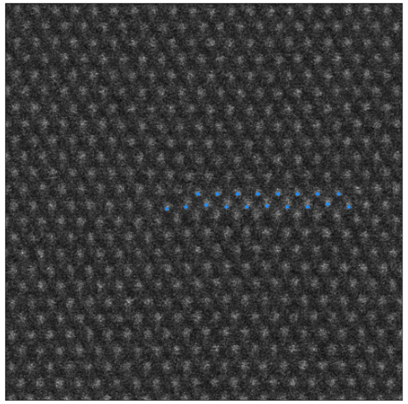
Requirements:

- ✓ **Location of these columns (classified atomic coordinates)**
- ☑ **Feedback & sensitivity: one (not two!) sulfur ejection**

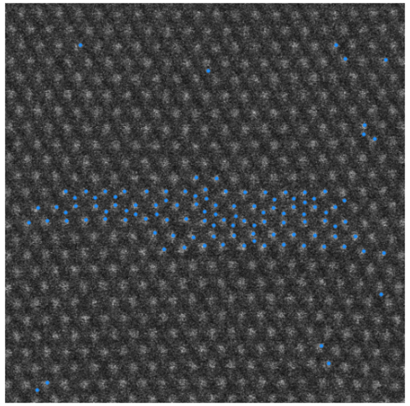


Shanshan Wang, Gun-Do Lee, Sungwoo Lee, Euijoon Yoon, and Jamie H. Warner
ACS Nano 2016 10 (5), 5419-5430
DOI: 10.1021/acsnano.6b01673

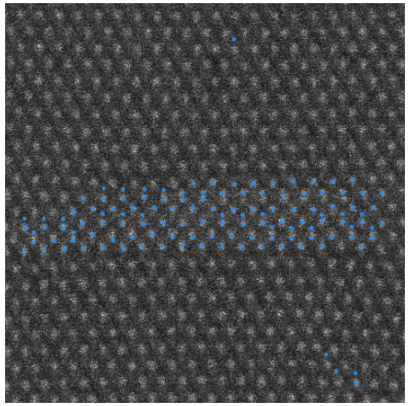
(video)



$E_{gap} = 0.546$



$E_{gap} = 0.04 - 0.05$

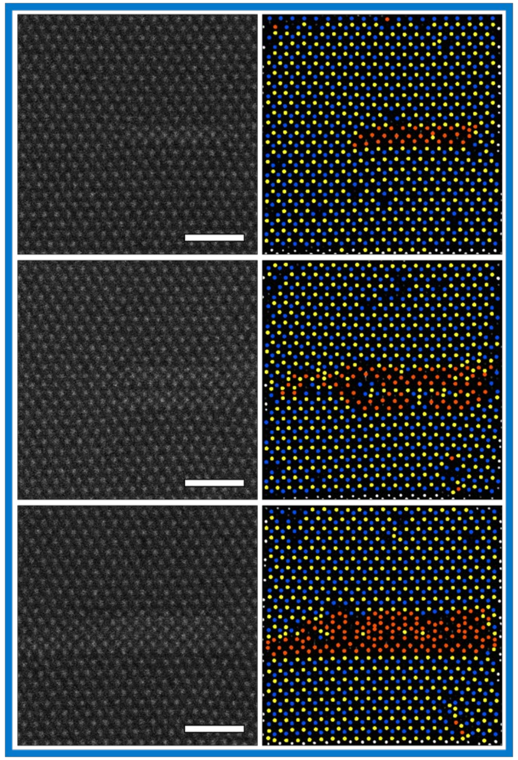


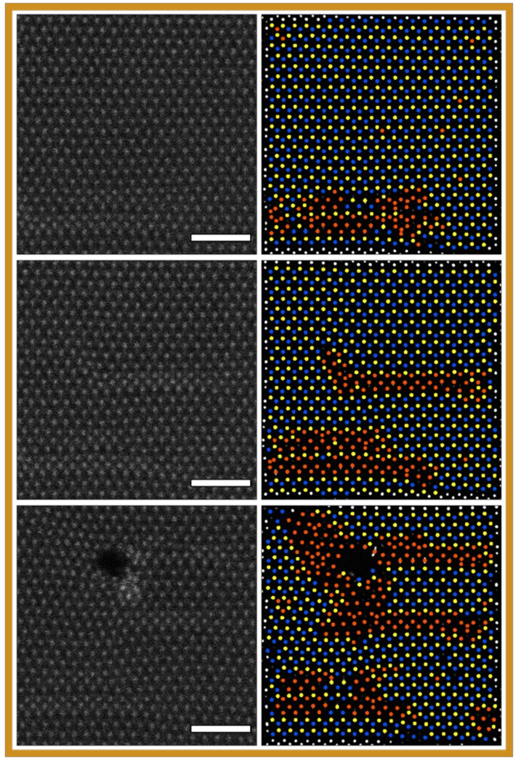
$E_{gap} = METALLIC??$

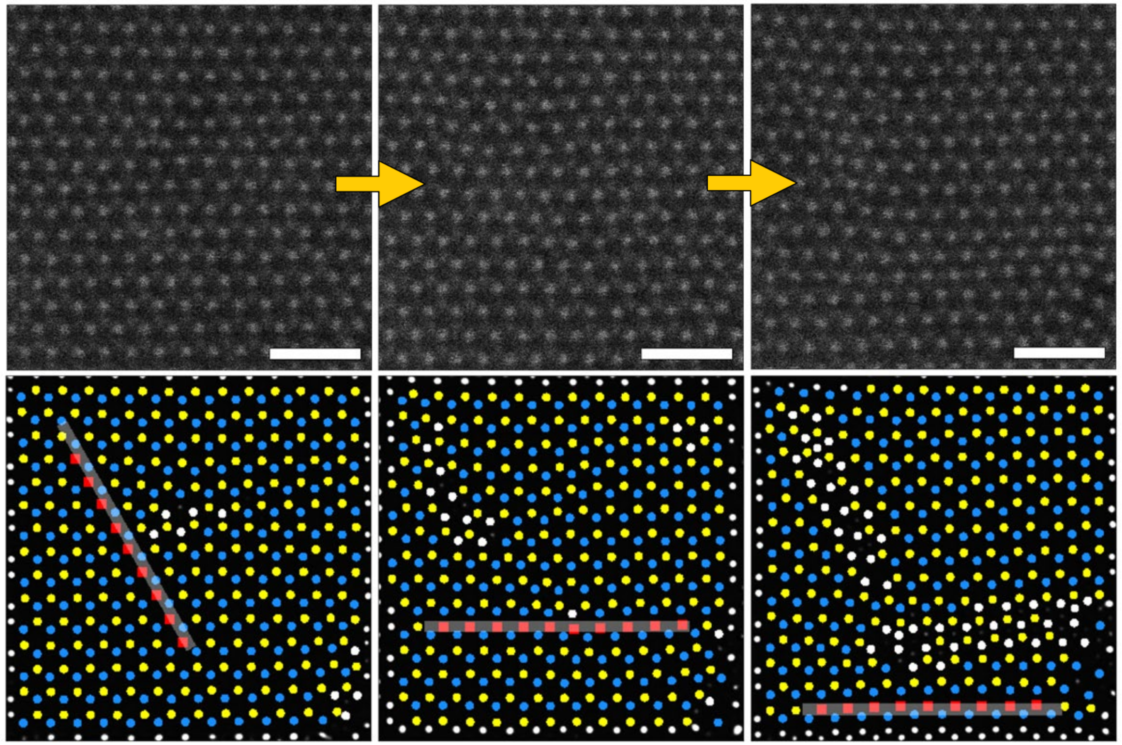
Selective atom removal

Towards complex geometries (SVLs)

- Precise sulfur targeting
- Feedback assisted (ADF) to **ensure only single sulfur removal**







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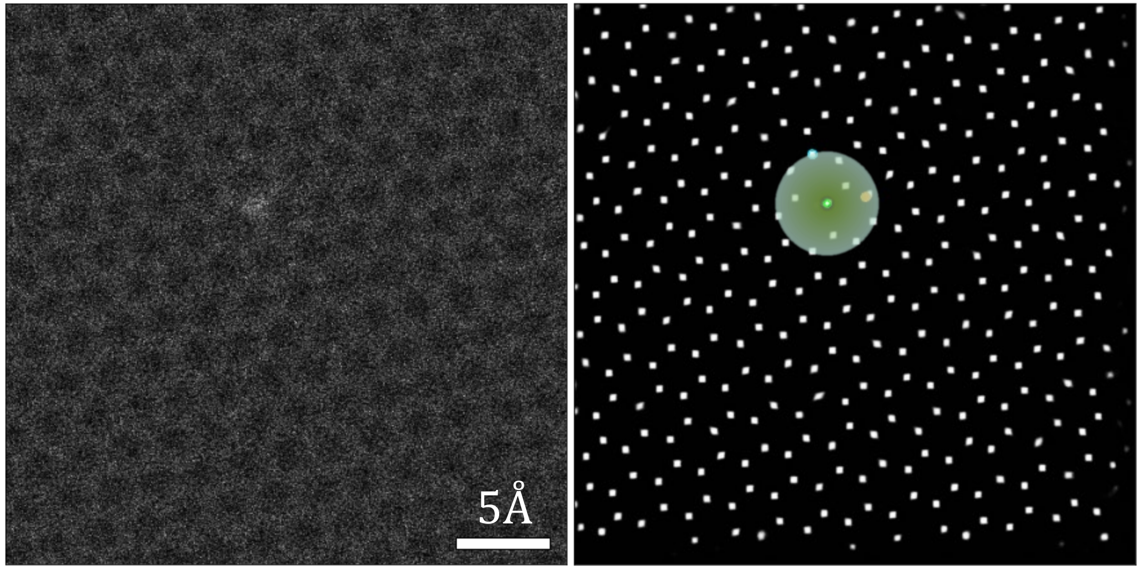
Where should we place beam?

Causal relationships may not be obvious

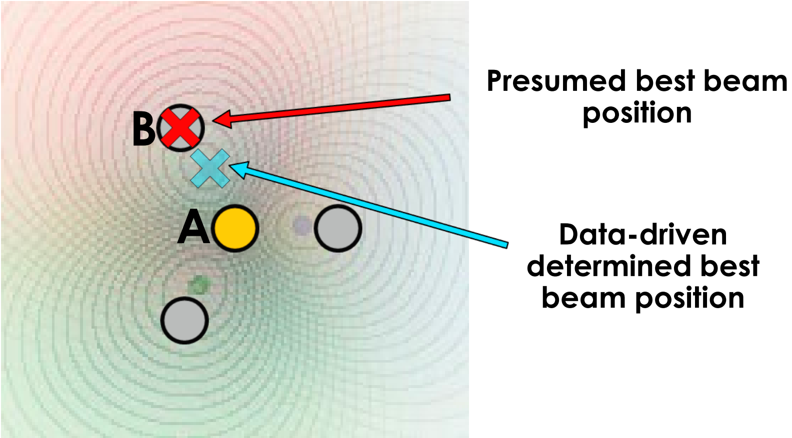
Atomic manipulation rules

Where to place the beam to achieve desired state?

Data driven approach to 'learn' the rules

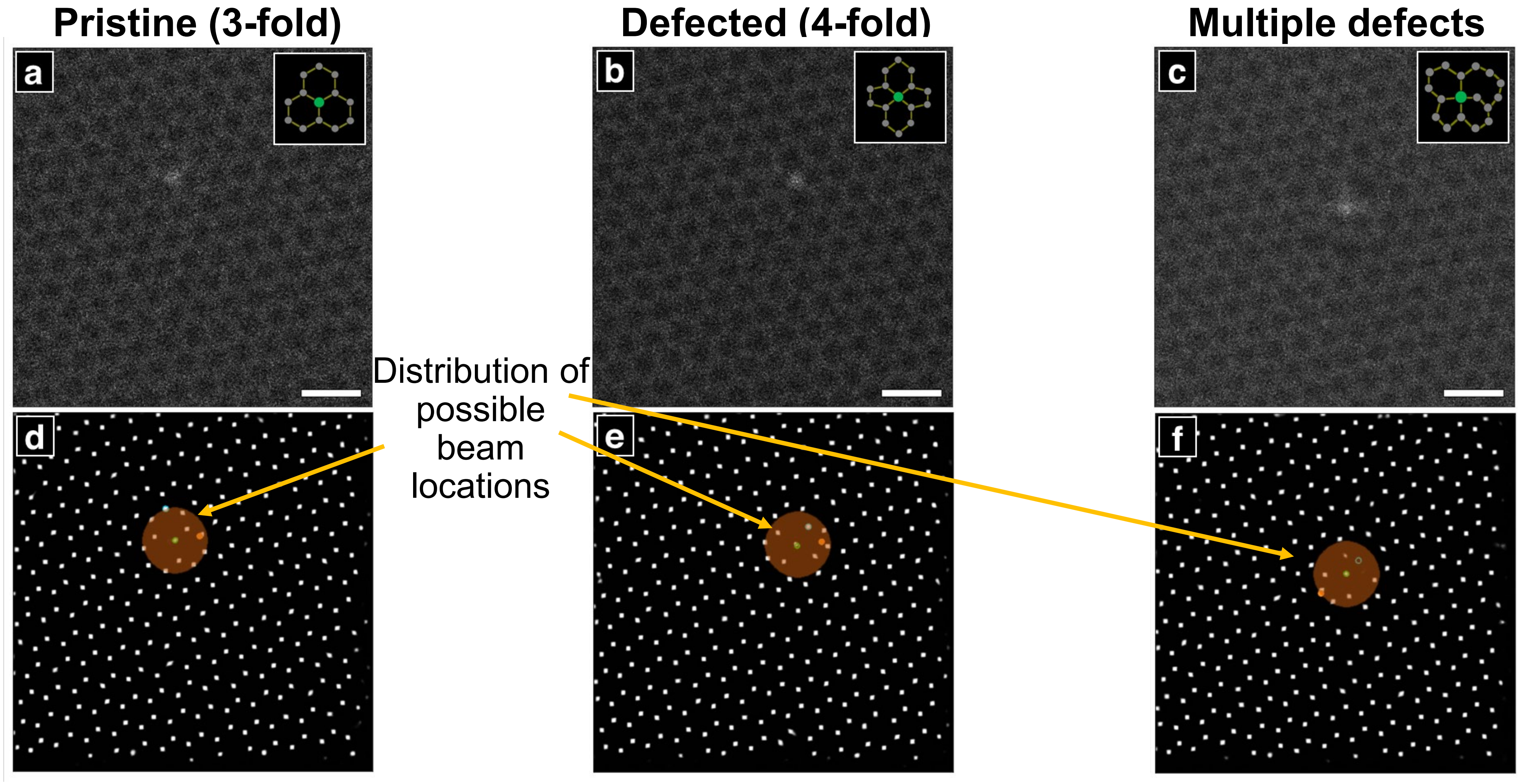


Goal:
Move Si from **A** to **B** most efficiently



- We cannot do this reliably manually, **requires automated experiment!**
- **Robust, fast,** and **LIVE** atomic coordinate extraction is needed (out of distribution drift!)
- Drift, stability, concerns.

Learning atomic manipulation rules: Si in graphene

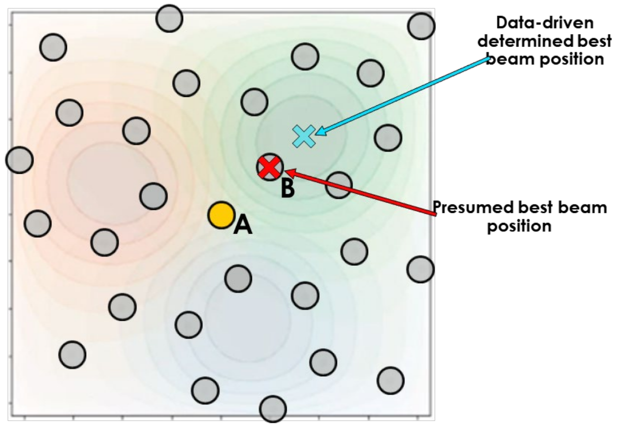


Generally, a real “goal” is not one jump, it might be many jumps

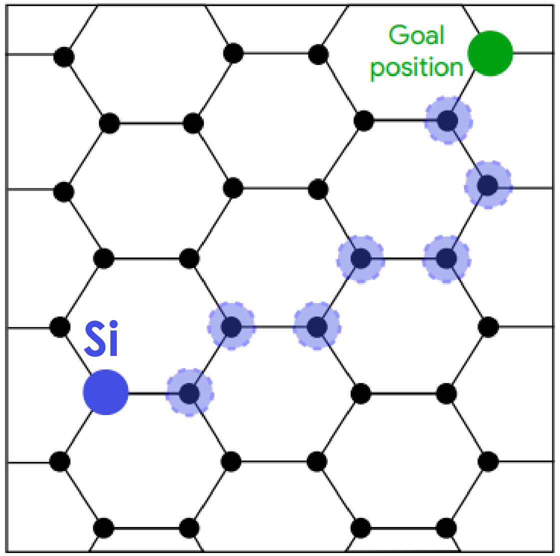
Gather those statistics!

(screen record video)

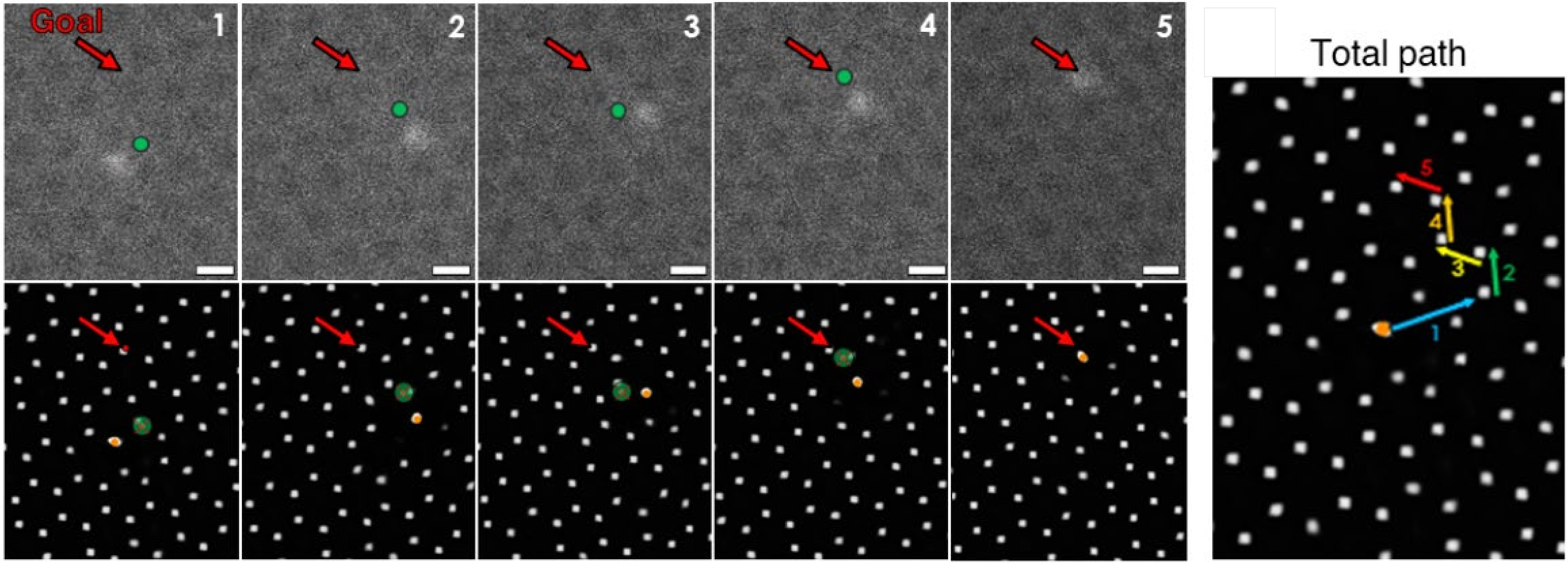
Learned rates for 3-fold Si



Now, let's do the following:



Autonomous manipulation of Si in graphene

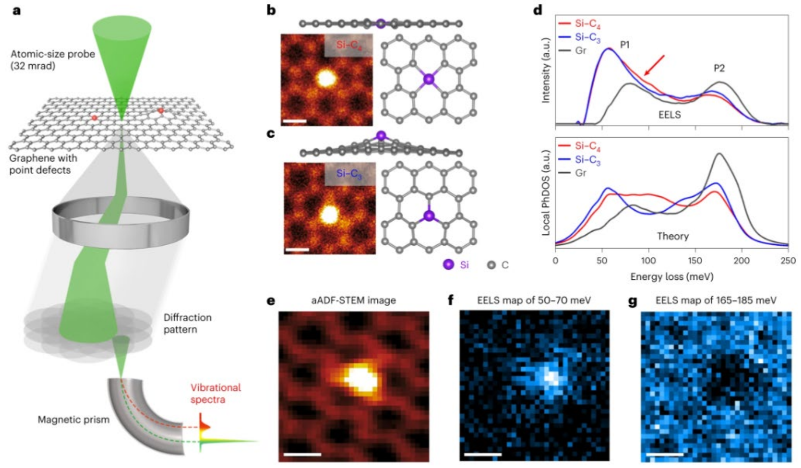


- **Deployment of learned agent allows to "drag" single silicon throughout graphene lattice – *completely automated***

Sensitive & dynamic measurements

Measure native state, not defected state.
Or…
Systematically measure transient states

STEM-EELS: use cases



nature > nature materials > articles > article

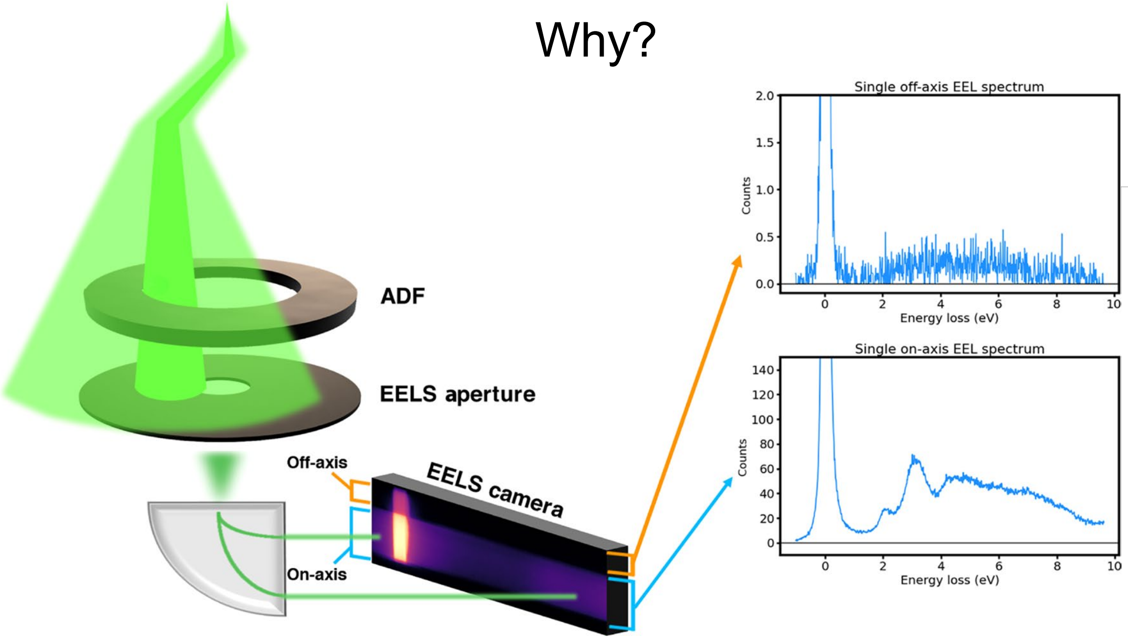
Article | Published: 16 March 2023

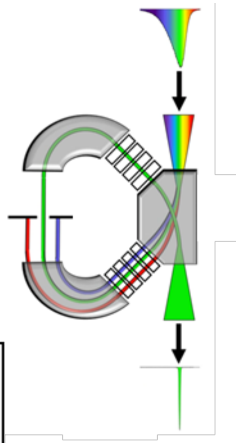
Single-atom vibrational spectroscopy with chemical-bonding sensitivity

Mingquan Xu, De-Liang Bao, Aowen Li, Meng Gao, Dongqian Meng, Ang Li, Shixuan Du, Gang Su, Stephen J. Pennycook, Sokrates T. Pantelides & Wu Zhou

~20-minute exposure!

Why?

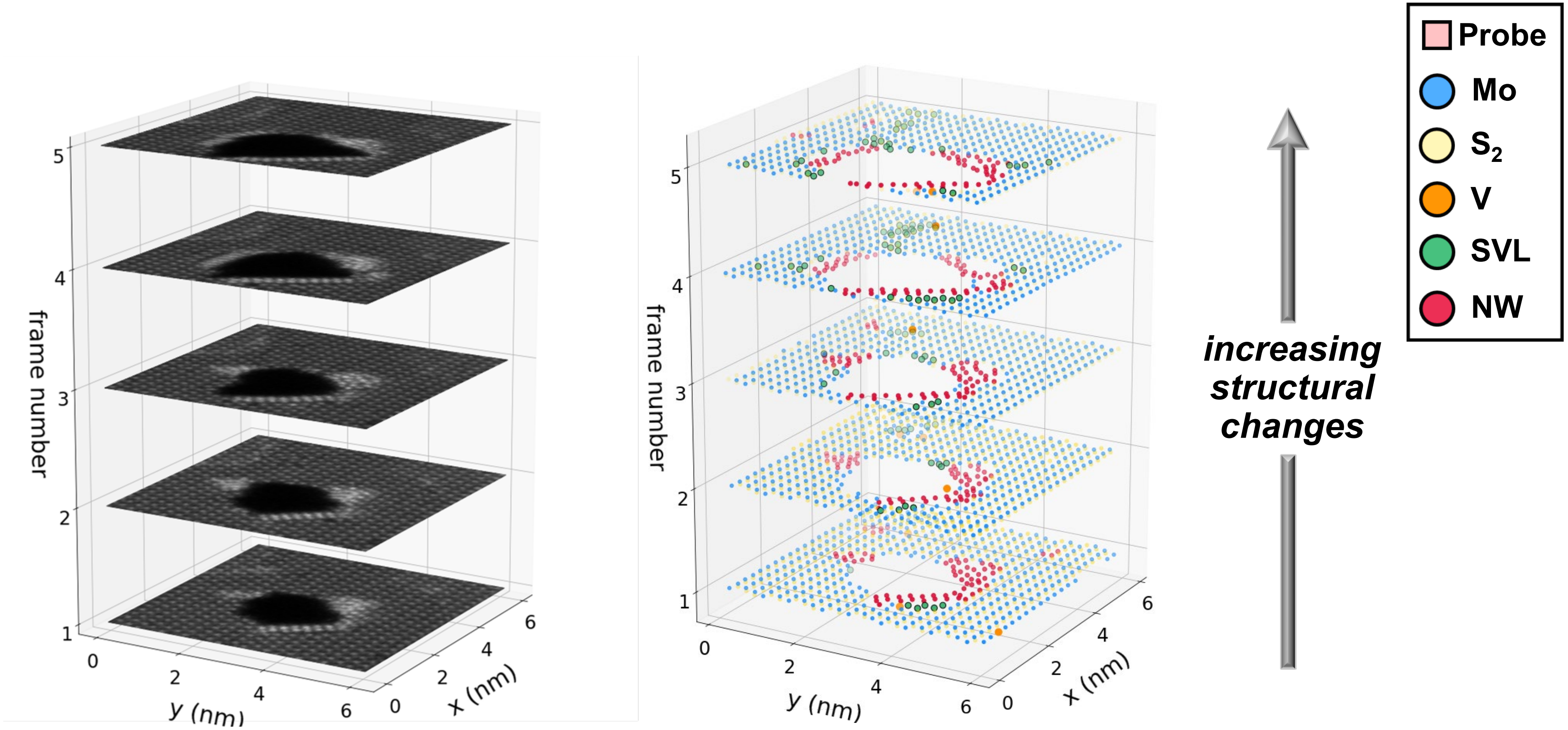




- Monochromation // off-axis → more phenomena to explore but…
- Only viable for **extremely** stable specimens and systems! (these are usually the well-understood ones too!)
- …Unless we probe intelligently

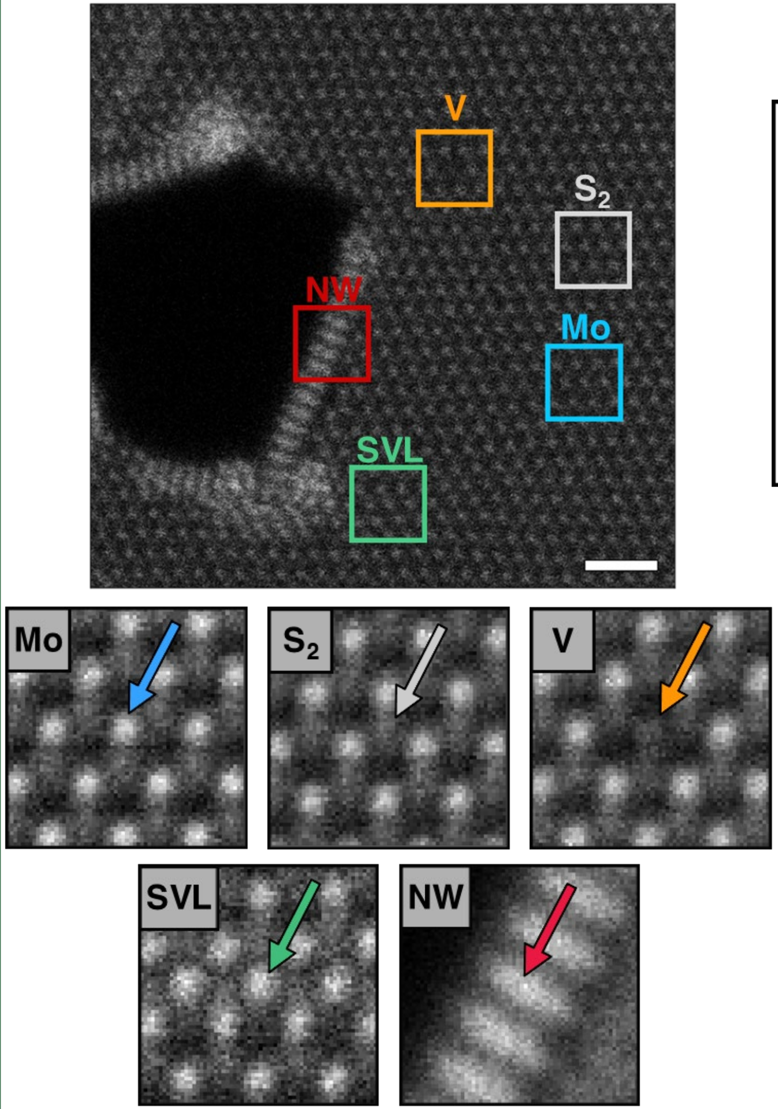
Sensitive & dynamic measurements

Energy loss spectroscopy in single-layer, V-substituted MoS_2

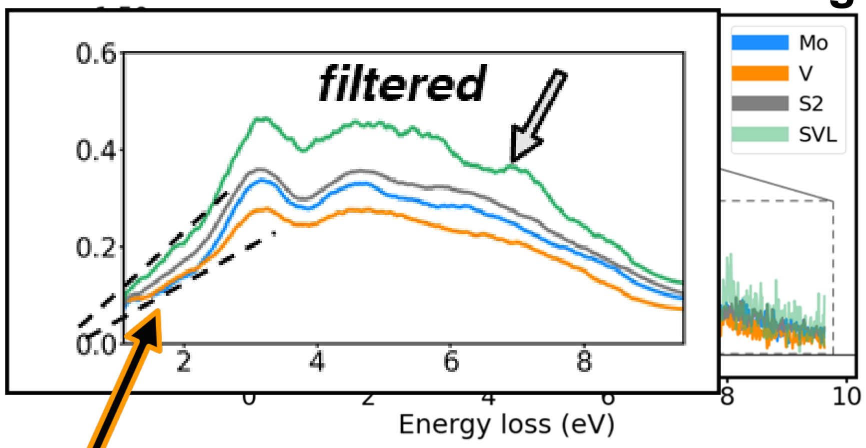


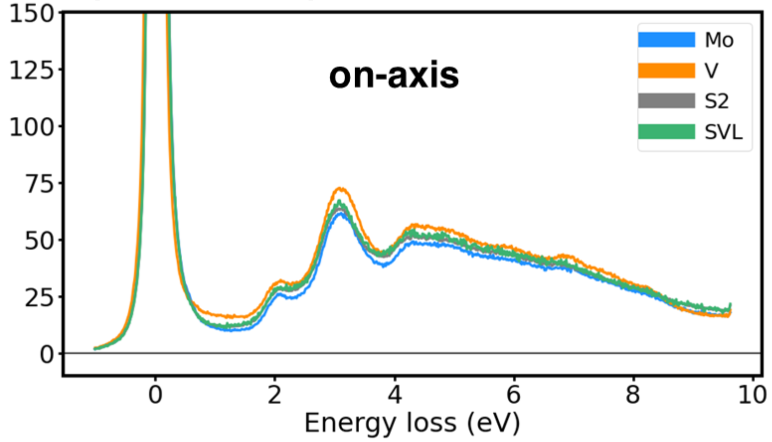
Sensitive & dynamic measurements

Energy loss spectroscopy in single-layer, V-substituted MoS_2



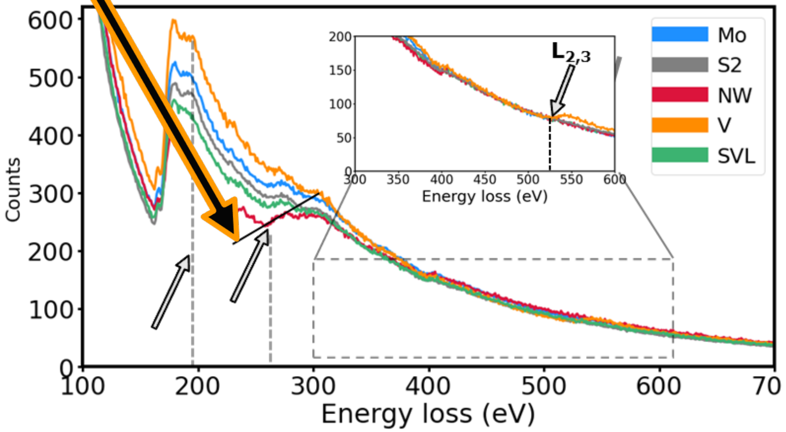
Low loss regime (1 – 10 eV)

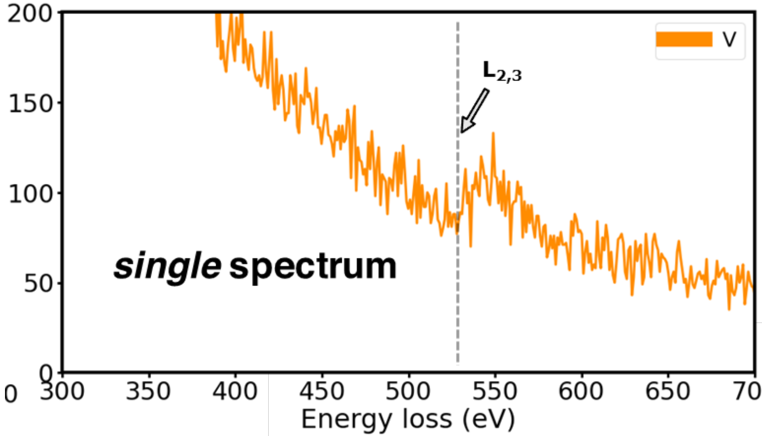




The slopes!

Core loss regime (> 50 eV)





Hands-on notebook!