Synthetic STM Imaging: a Pipeline for Deep-Learning Analysis

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Outline

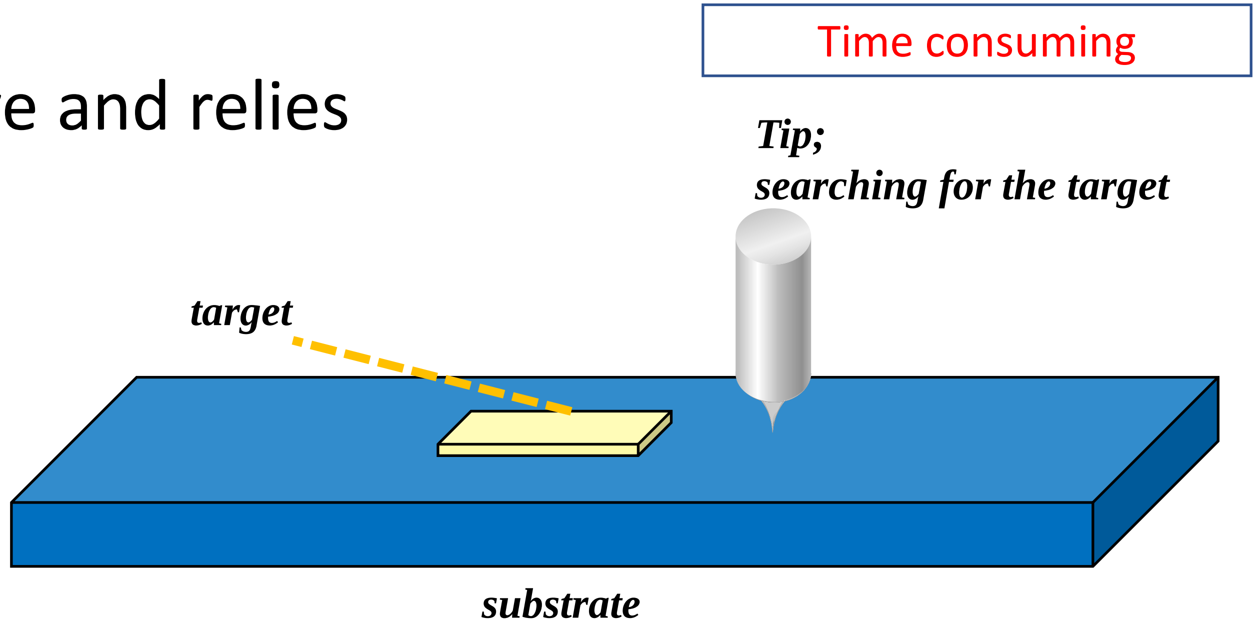
- Introduction and Motivation
- Simulation of STM Imaging
  - Tunneling Current
  - Tight-Binding Method
  - Feedback Control
  - Distortions
- Application and Results
  - Defect localization
- Conclusion
  - Summary of contributions
  - Limitations and future work

Introduction and Motivation

Introduction

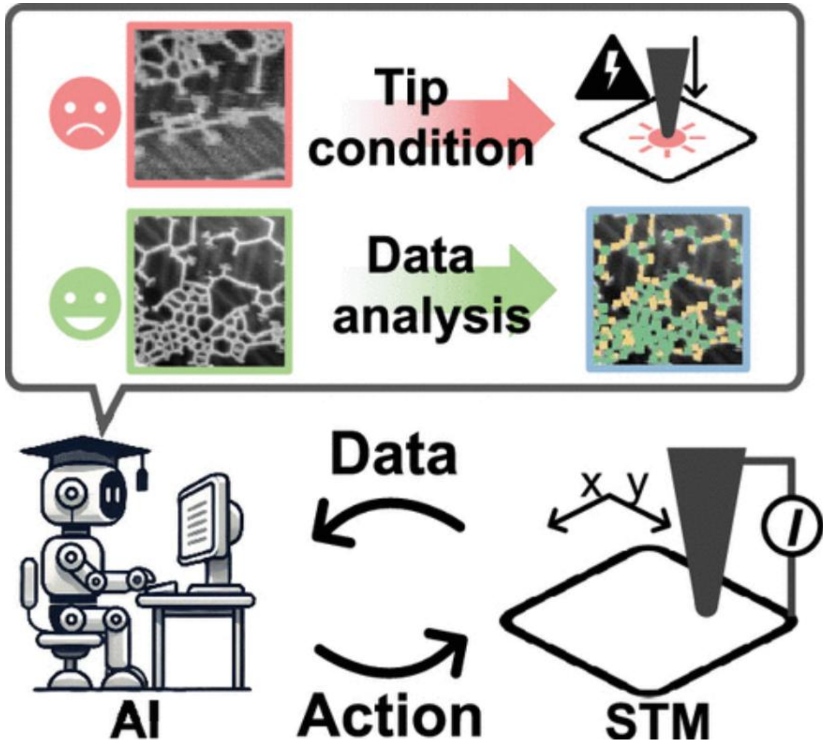
- Scanning Tunneling Microscopes (STM) enables subatomic resolution of materials' electronic structures.

- Measuring is very labor-intensive and relies on guesswork.



Introduction

- Many recent works try to address this via machine learning:
  - Denoising [1]
  - Localizing atomic-scale defects [2]
  - Autonomous tip conditioning [3]
  - Measurement workflow automation [4]
- **Large labeled datasets required.**

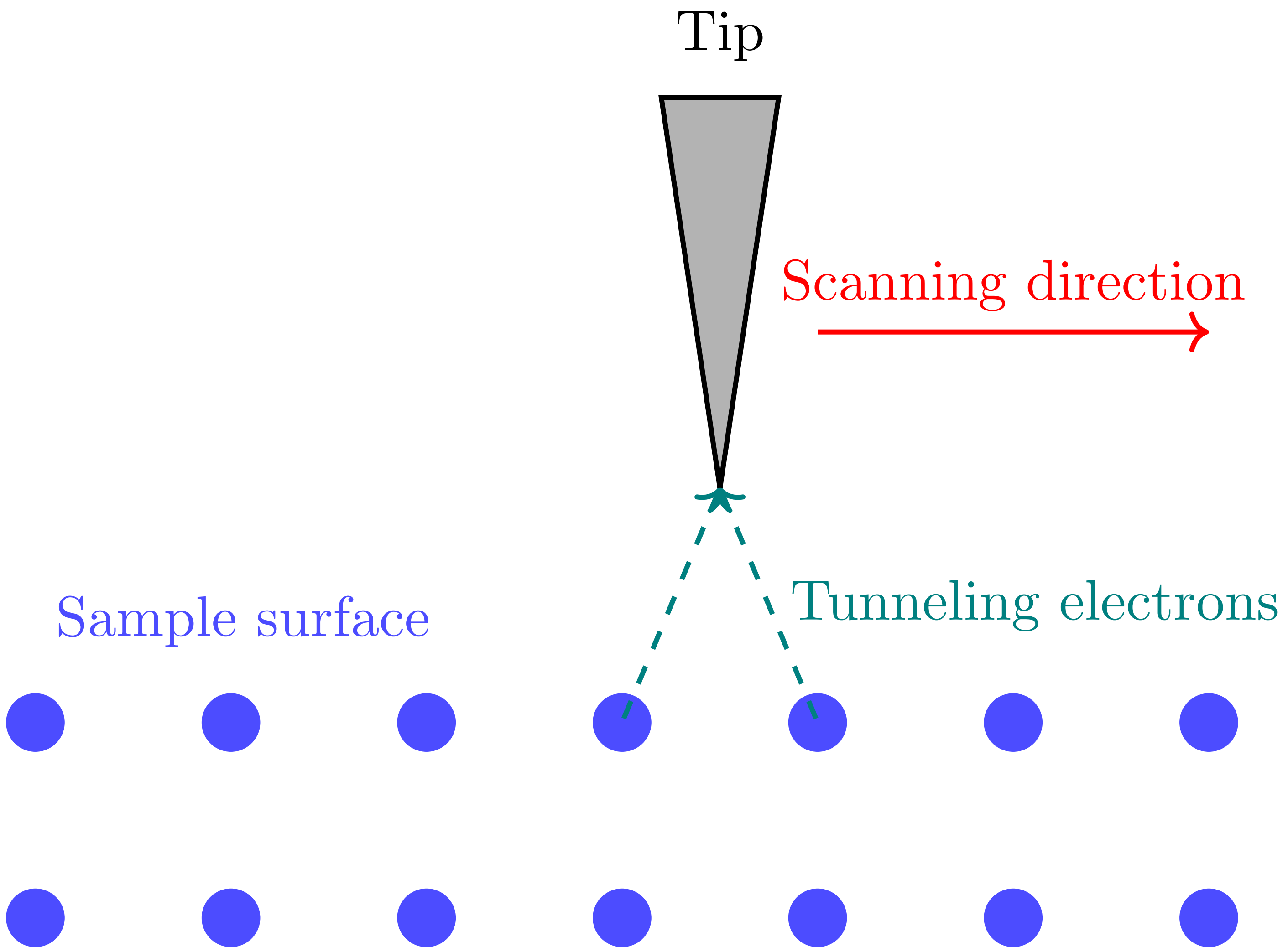


Taken from [4]

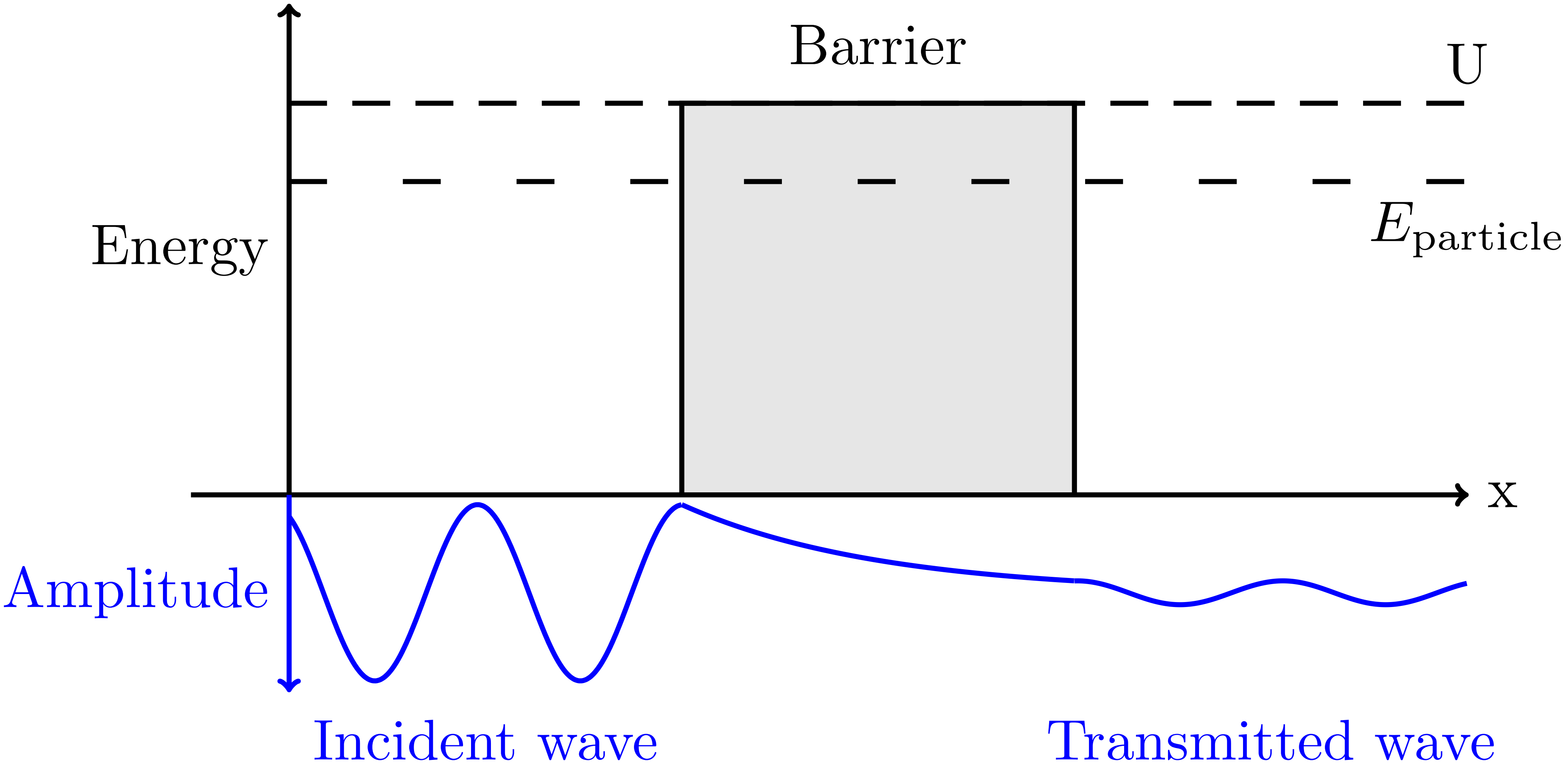
[1] Joucken, F., Davenport, J. L., Ge, Z., Quezada-Lopez, E. A., Taniguchi, T., Watanabe, K., Velasco, J., Lagoute, J., & Kaindl, R. A. (2022) *Physical Review Materials*
[2] D. Smalley, S. D. Lough, L. Holtzman, K. Xu, M. Holbrook, M. R. Rosenberger, J. C. Hone, K. Barmak, and M. Ishigami. (2024) *MRS Advances*
[3] M. Rashidi and R. A. Wolkow. (2018) *ACS Nano*
[4] Zhu, Z., Yuan, S., Yang, Q., Jiang, H., Zheng, F., Lu, J., & Sun, Q. (2024) *Journal of the American Chemical Society*

Simulation of STM Imaging

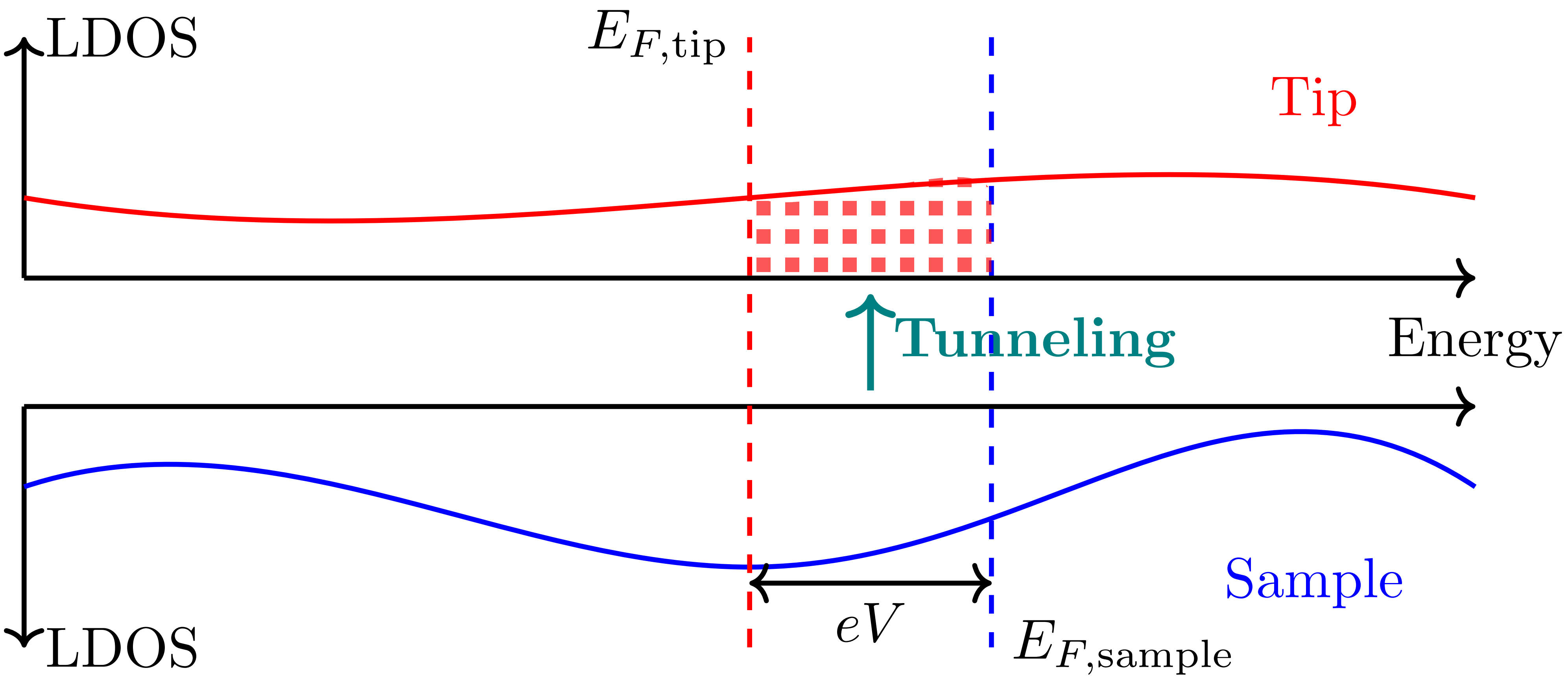
Scanning Tunneling Microscope (STM)



Tunneling Current – Barrier Dependency



Tunneling Current – Local Density of States (LDOS)



Tunneling Current

- Final formula:

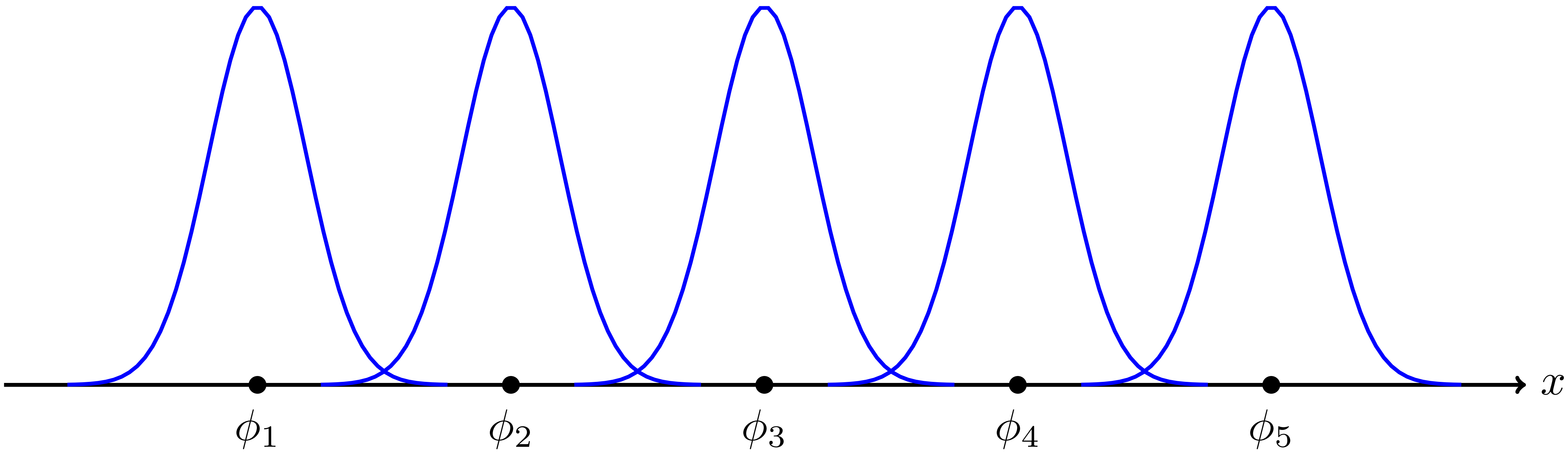
$$I(\rho_T, \rho_S, z) \propto \quad e^{-2\kappa z} \int_0^{eV} \rho_T(E_F - eV + \epsilon)\rho_S(E_F + \epsilon)d\epsilon$$

- Discretized expression, summed over all atoms:

$$I_{ij}(V) \sim e^{-2\kappa D_{ij}} \sum_{\epsilon=0}^{eV} \rho_i(E_F - eV + \epsilon)\rho_j(E_F + \epsilon)$$

$$I(V) = \sum_{i \in \text{sample}} \sum_{j \in \text{tip}} I_{ij}(V)$$

LDOS Calculation – Tight Binding Method

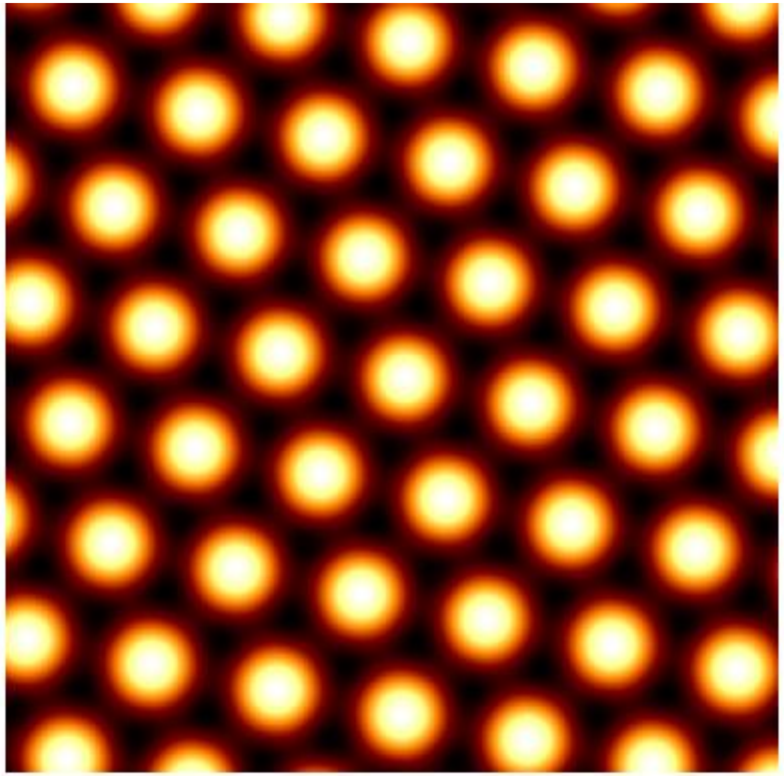


- The system is projected onto a localized orbital basis: $H_{ij} = \langle \phi_i | \hat{H} | \phi_j \rangle$
- LDOS is then calculated: $\rho_i(E) = \sum_n |\langle \phi_i | \psi_n \rangle|^2 \, \delta(E - E_n)$

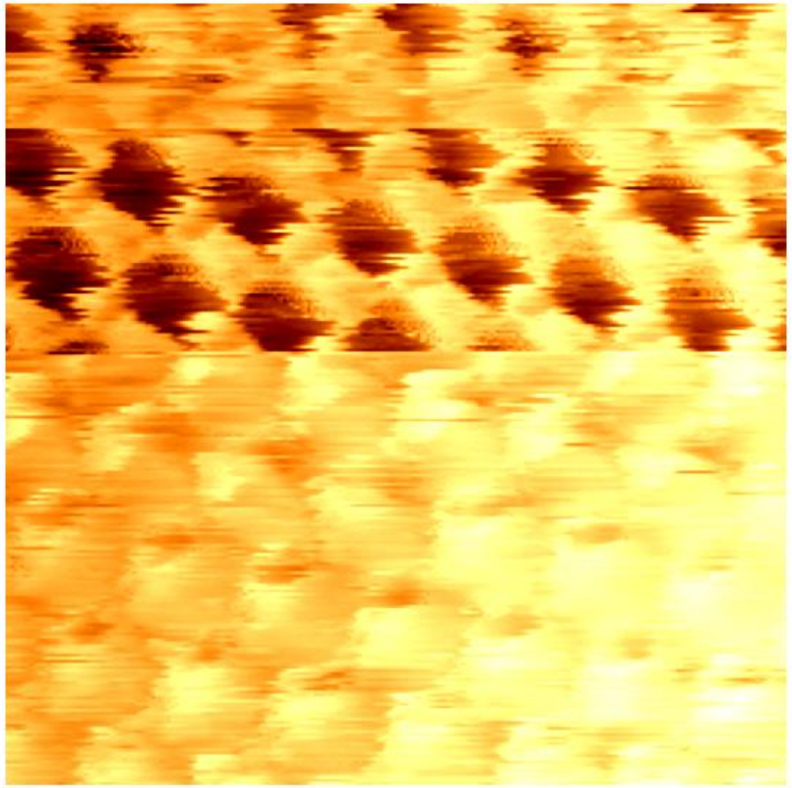
Idealized Constant Height Mode

- Tunneling current can now be calculated for each position in a grid.

Simulated Image

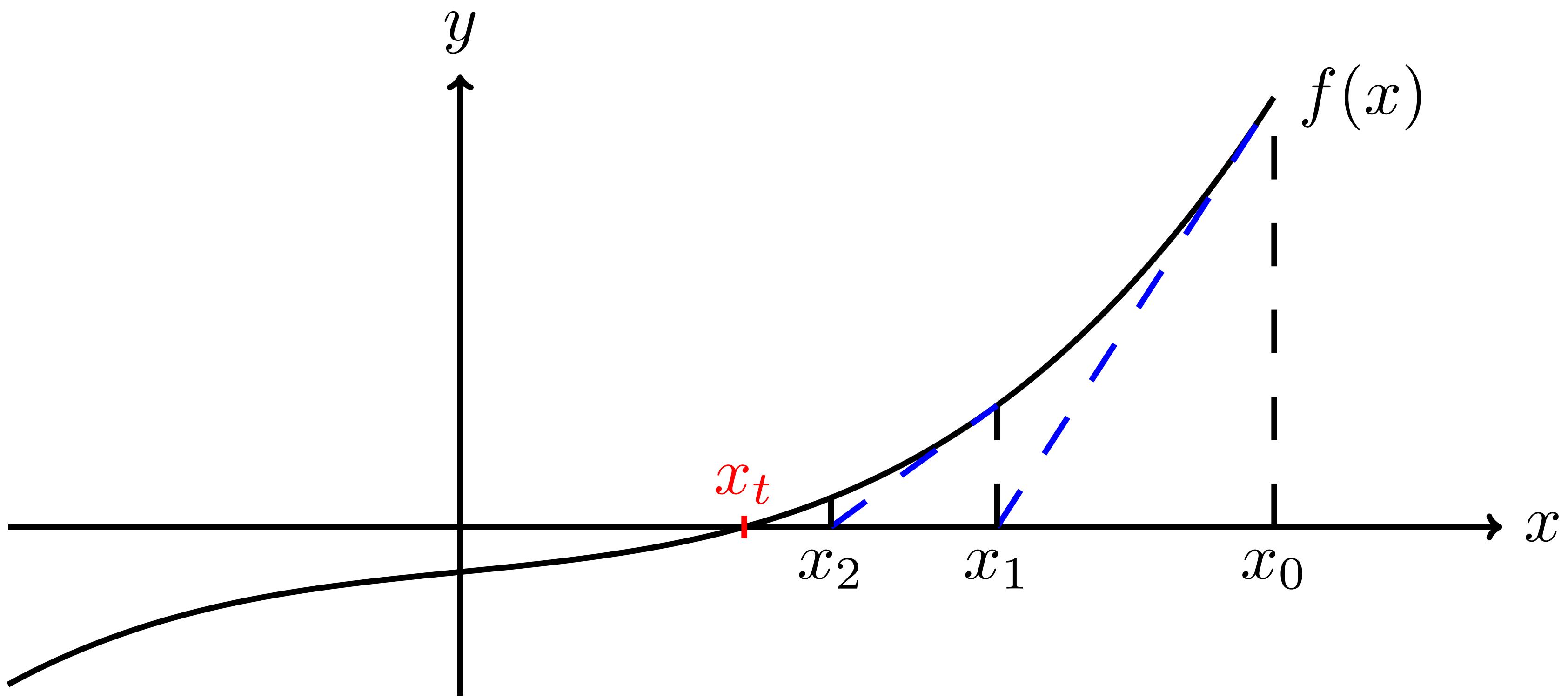


Real Measurement



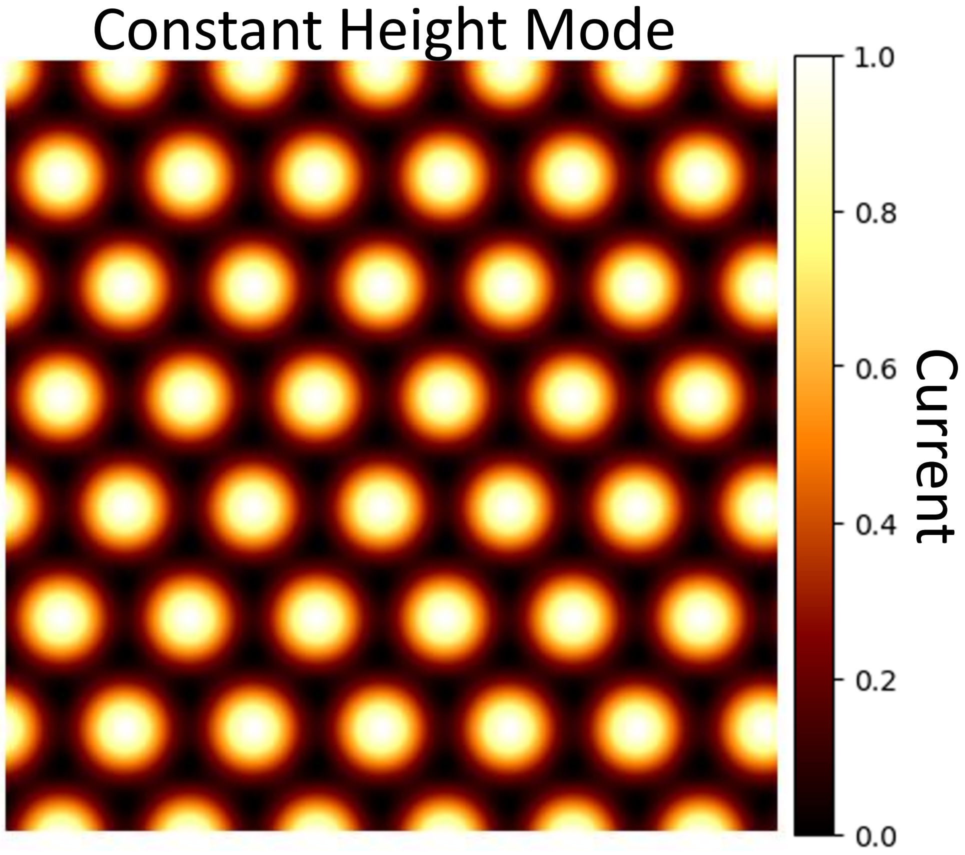
Idealized Constant Current Mode

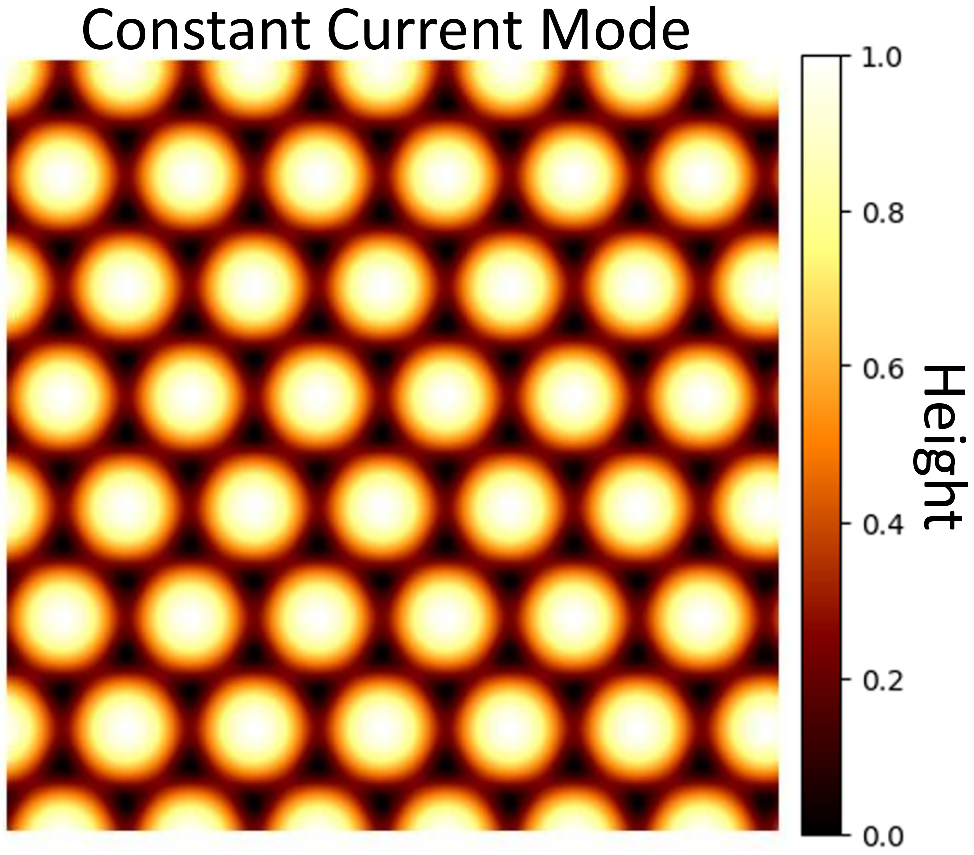
- Iterative Newton's Method can be used to calculate constant current mode



Idealized Constant Current Mode

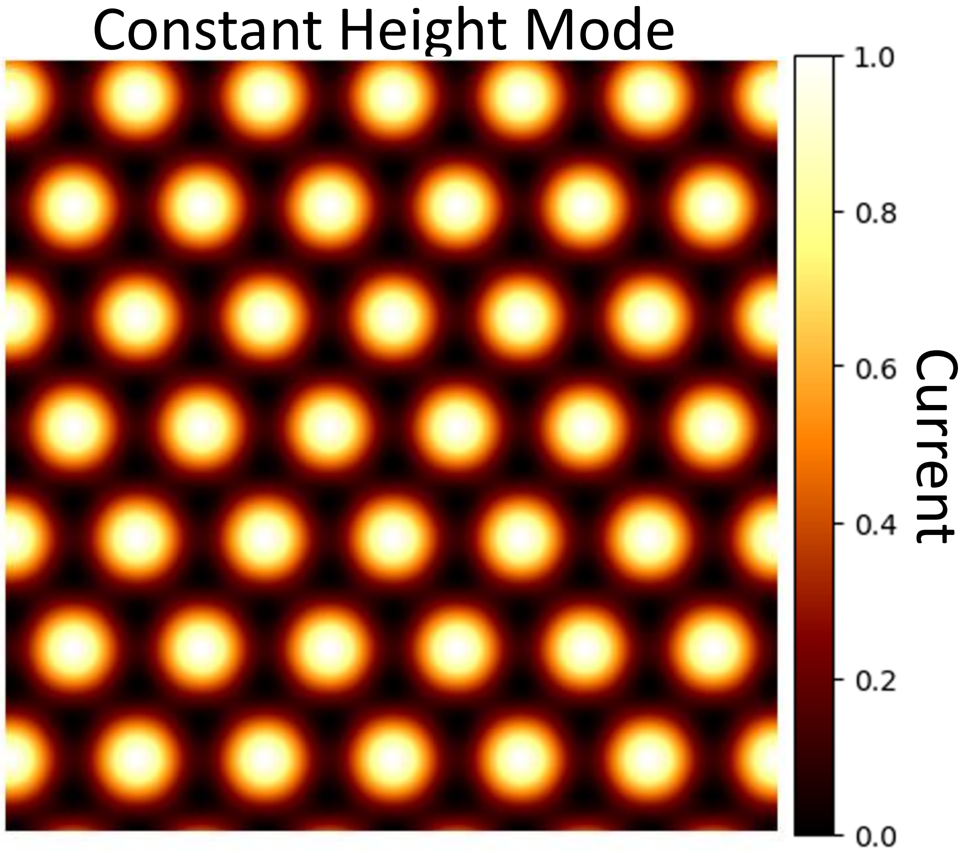
- Small initial height (1 Ångström):

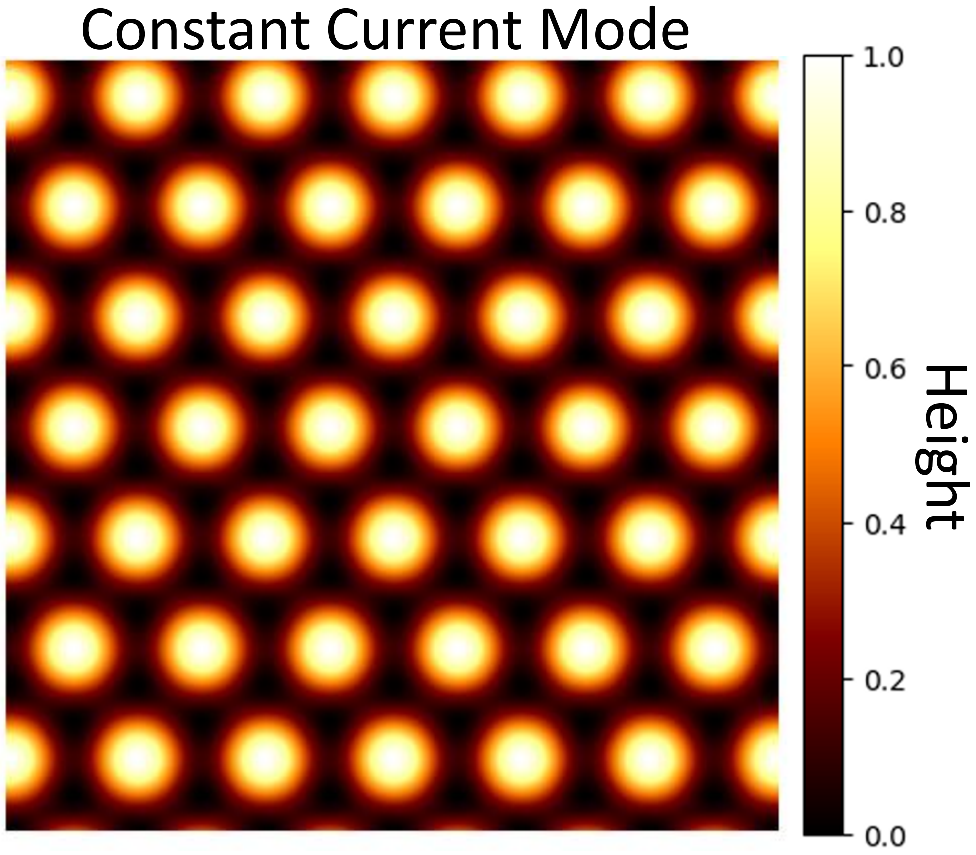




Idealized Constant Current Mode

- Large initial height (5 Ångström):

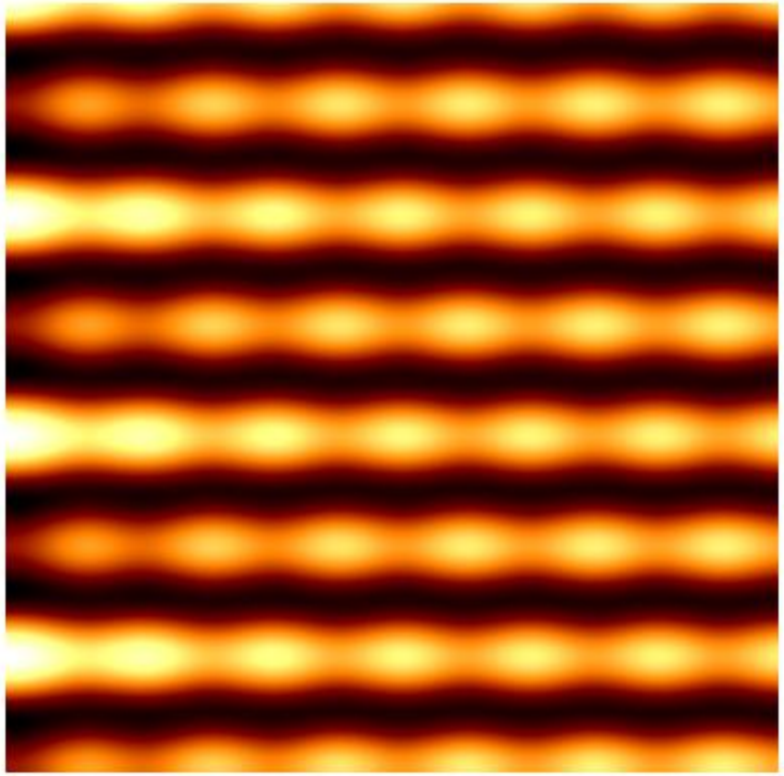




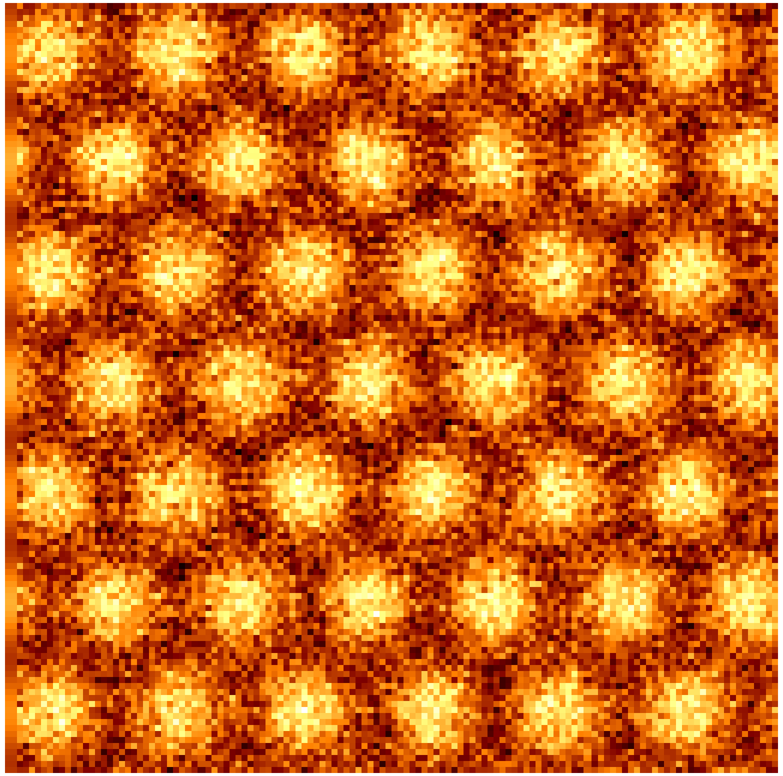
Feedback Control

$$\Delta z(t) = \boxed{K_p \cdot e(t)} + K_i \cdot \int_{t-t^*}^{t} e(\tau)d\tau + K_d \cdot \frac{de(t)}{dt}$$

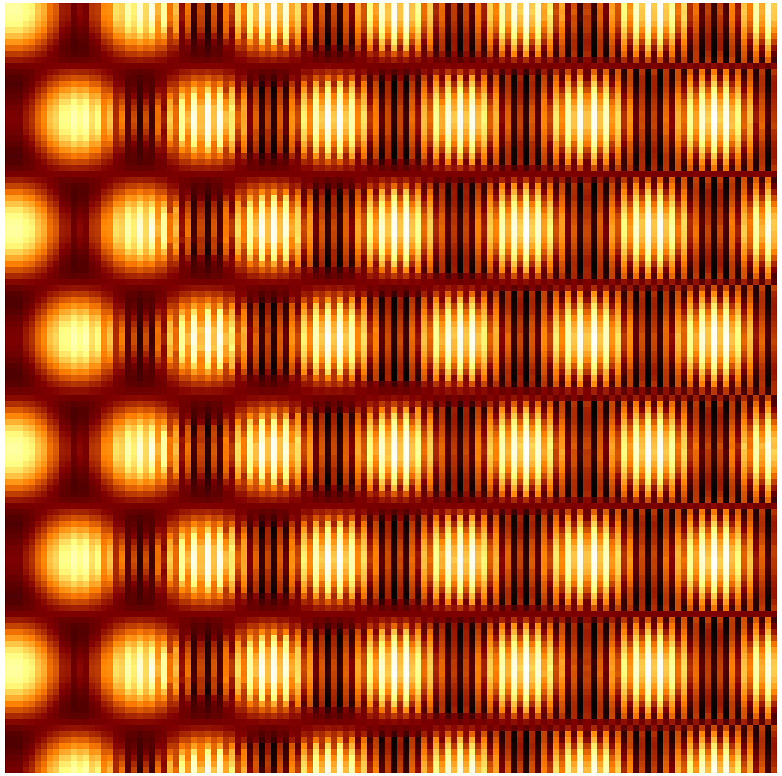
K_p too low



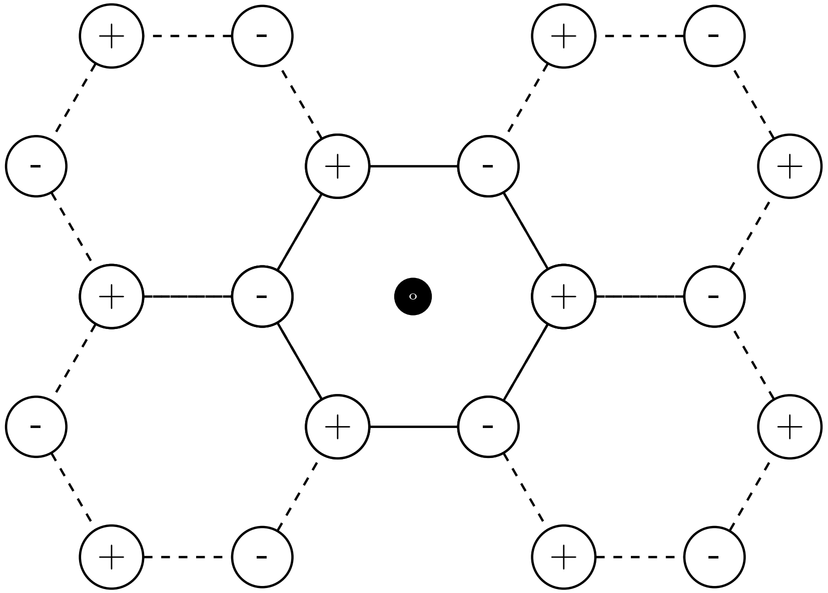
Current too noisy

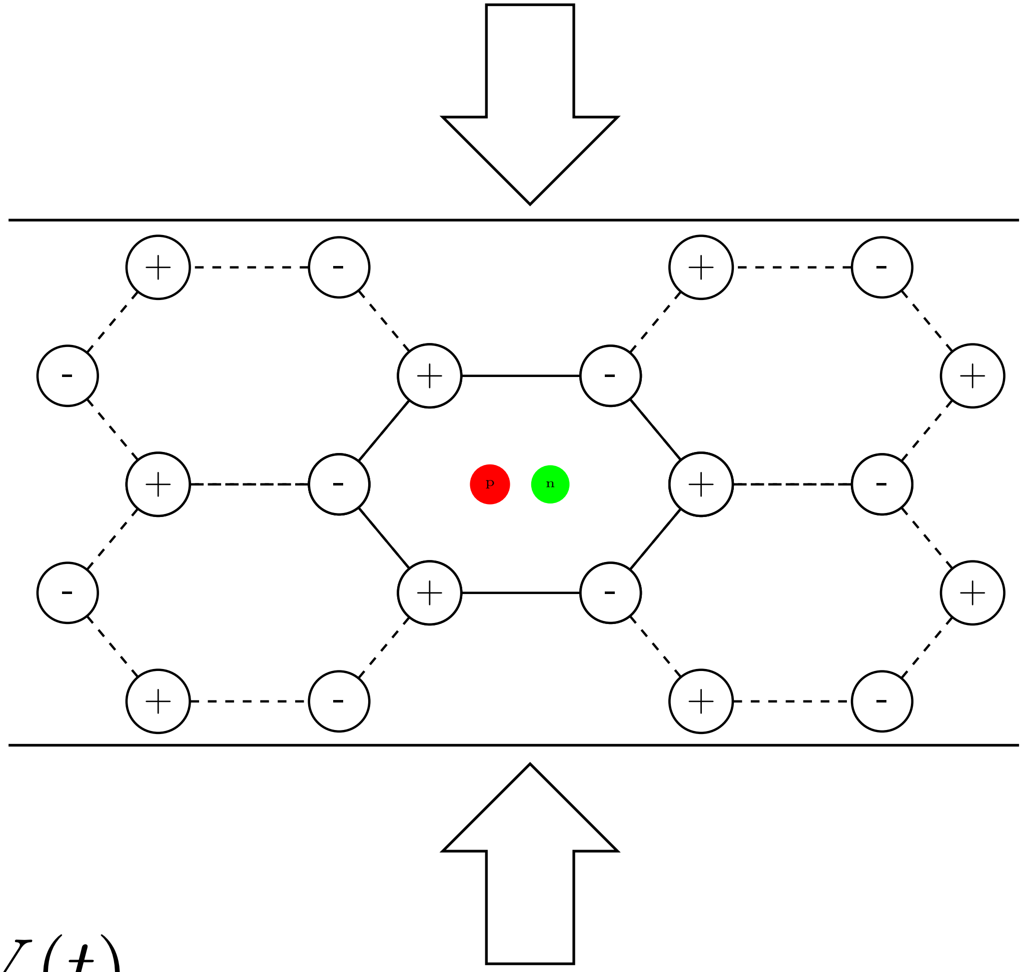


K_p too high



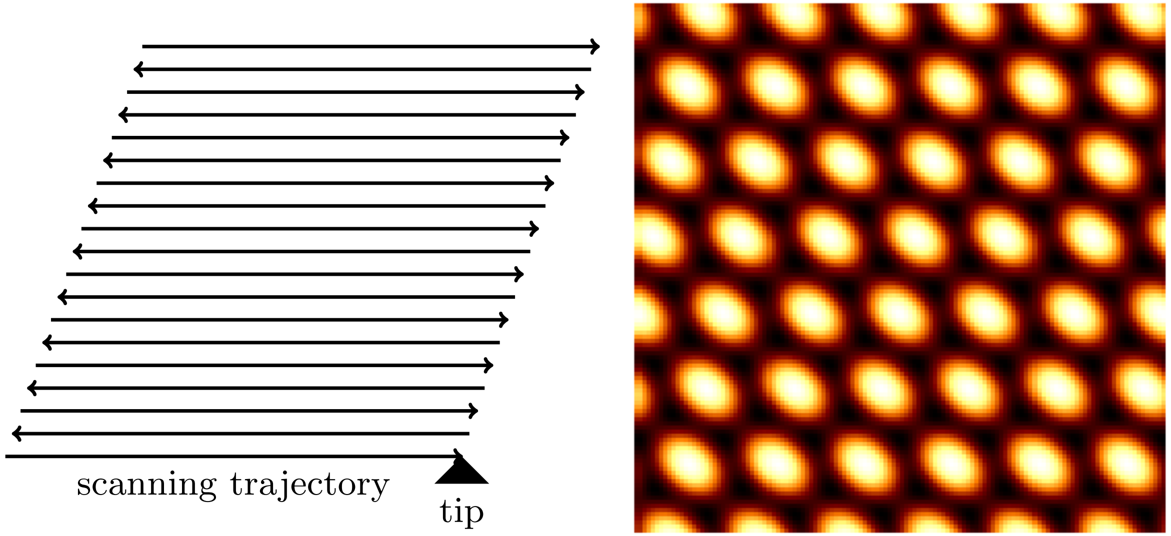
Piezoelectric Actuator



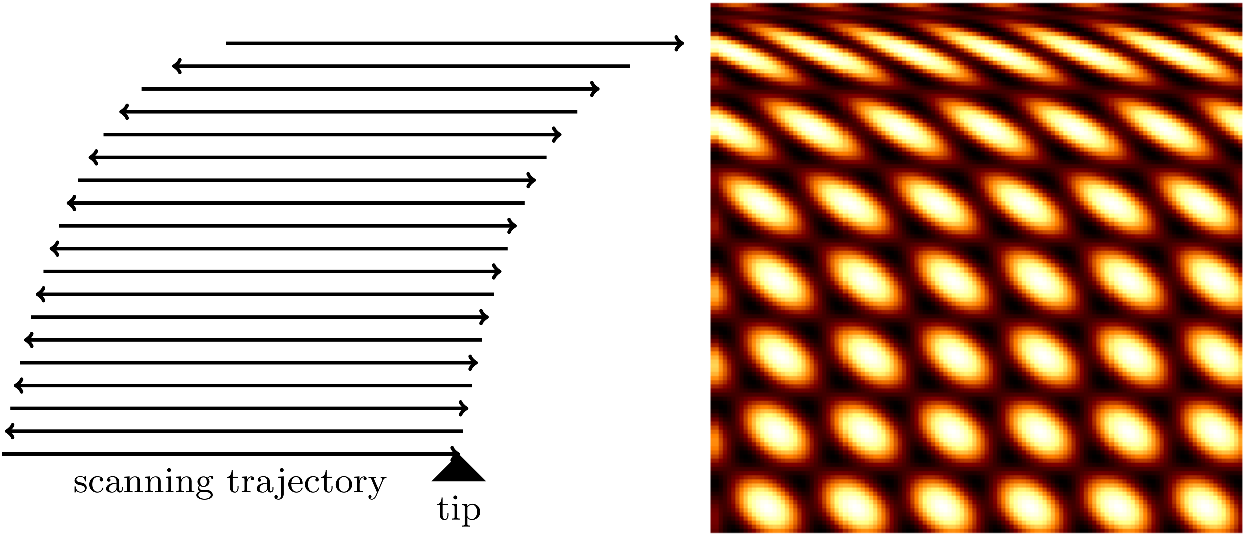


$$y(t) \propto V(t)$$

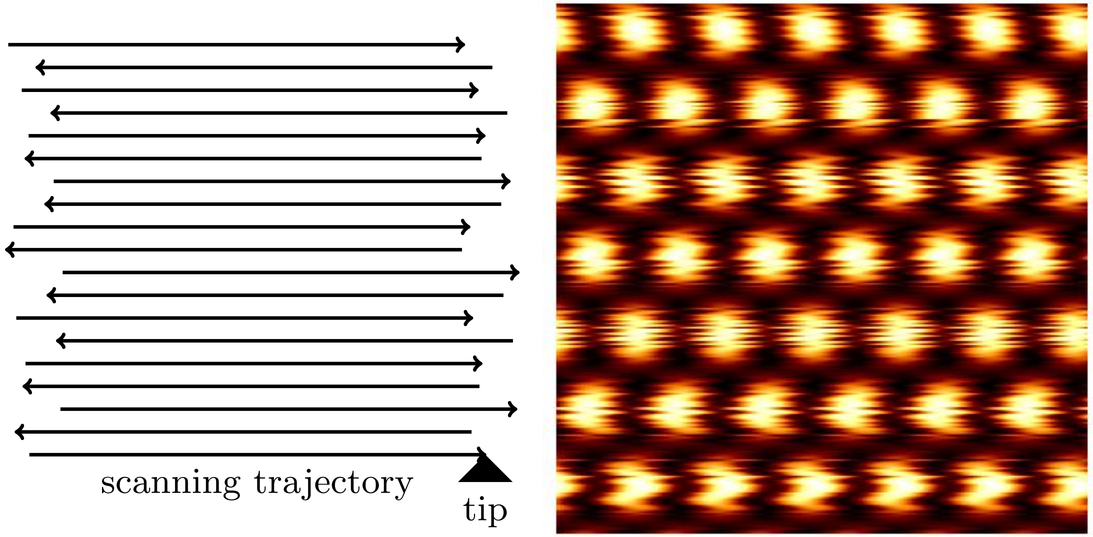
Lateral Distortions



$$y(t) = y_0 + \alpha t$$



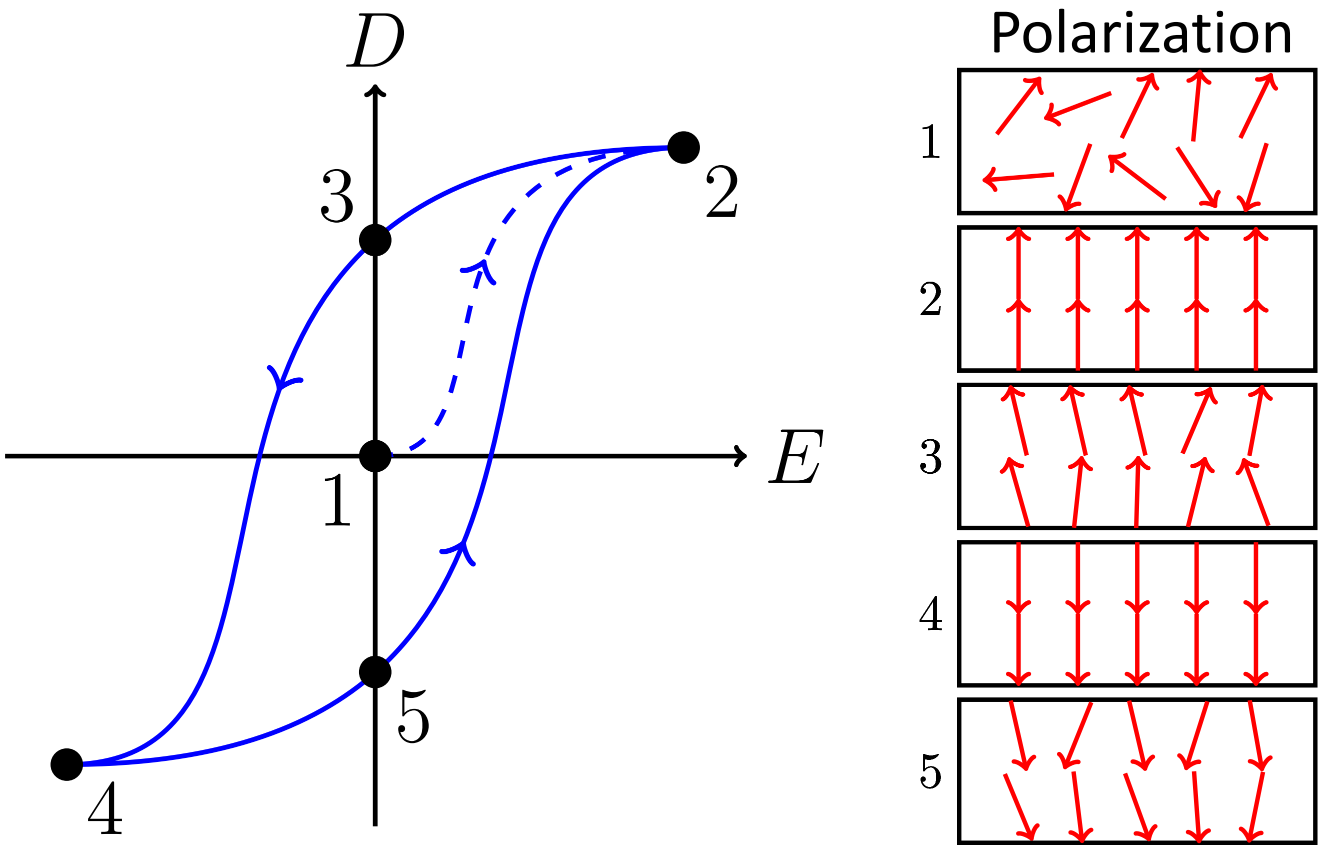
$$y(t) = y_0 + \gamma \log(\frac{t}{t_0})$$



$$y(t) = y_0 + n(t), \qquad n \sim \mathcal{N}$$

Hysteresis

- Domain reorientation:



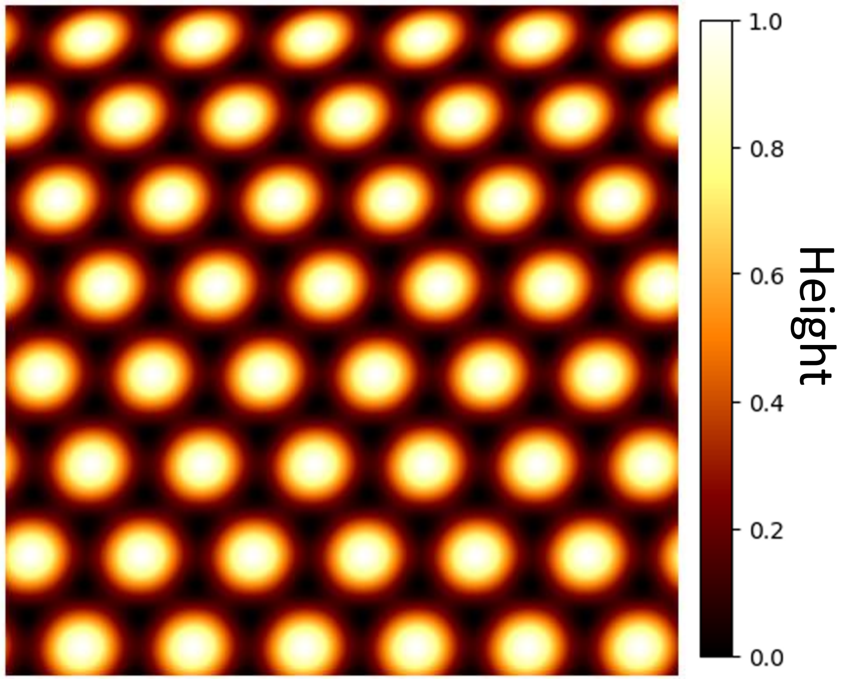
- Bouc-Wen model:

$$y(t) = \alpha V(t) + \beta H(t)$$

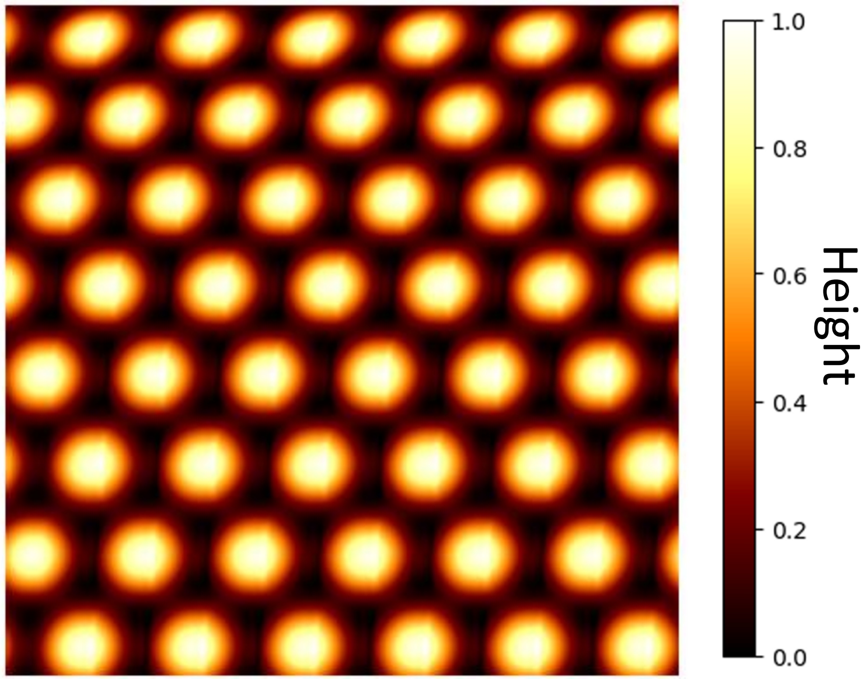
$$\dot{H}(t) = A\dot{V}(t) - B|\dot{V}(t)|H(t) - C\dot{V}(t)|\dot{H}(t)|$$

Hysteresis – Vertical Distortion

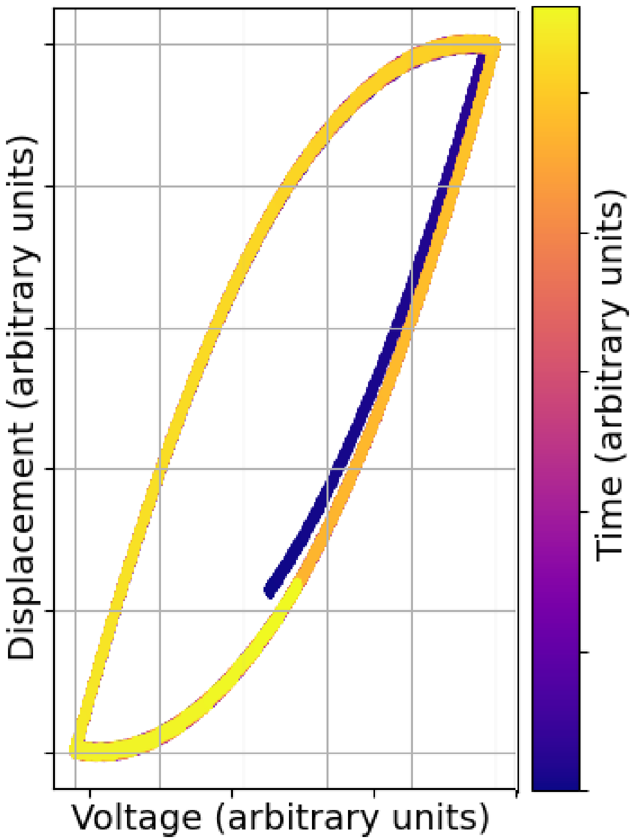
True Topography



Distorted Topography

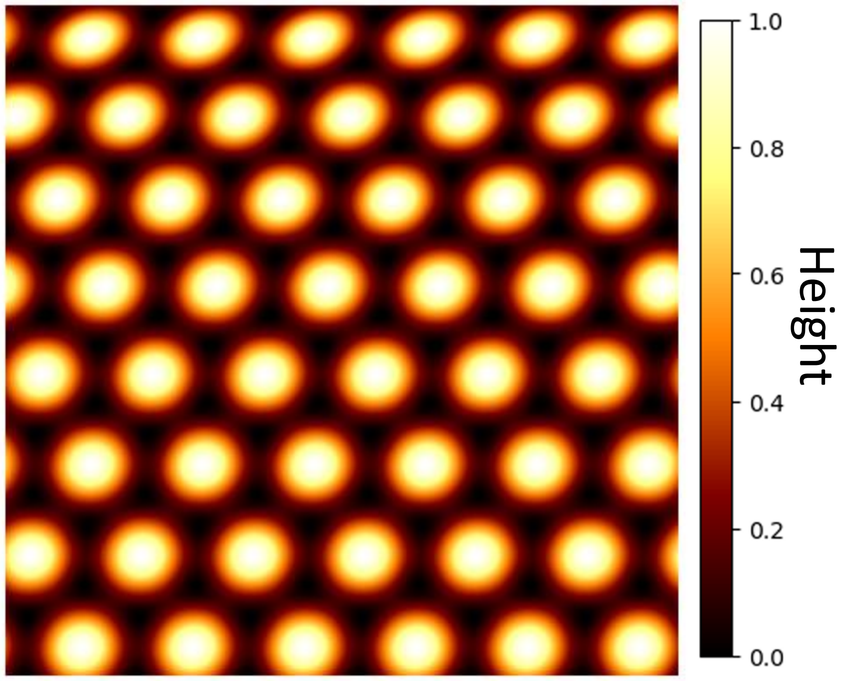


Hysteresis

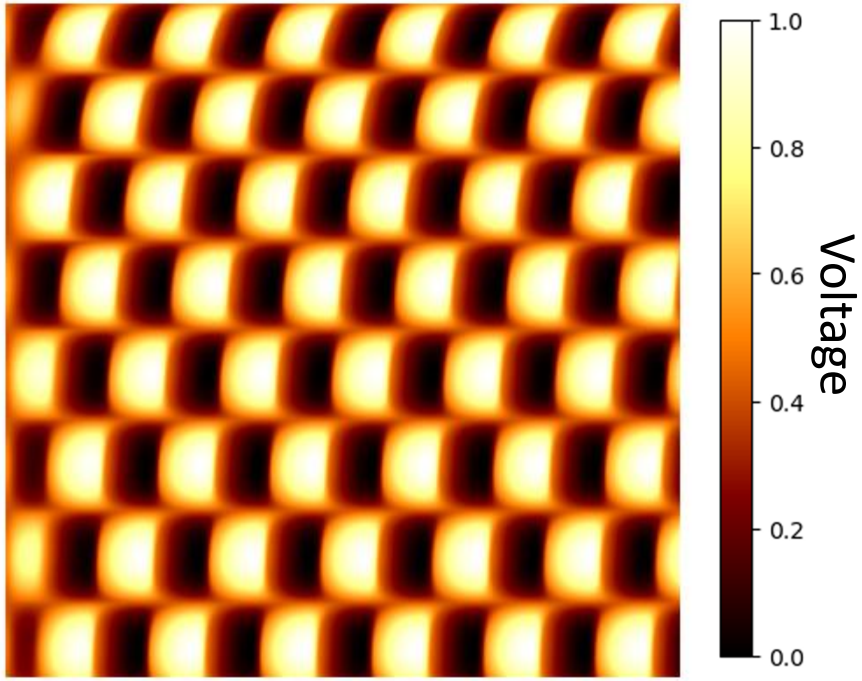


Hysteresis – Vertical Distortion

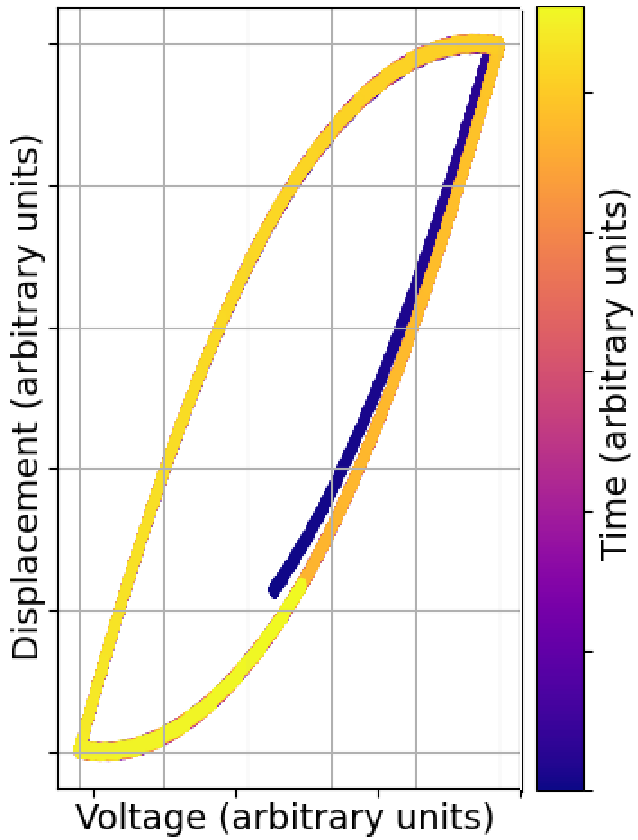
True Topography



Voltage Profile

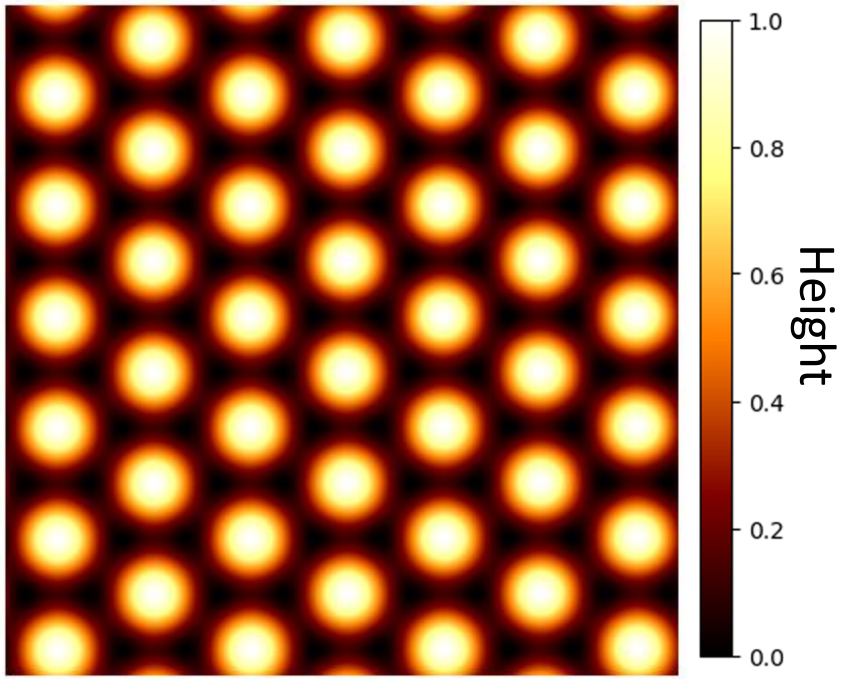


Hysteresis

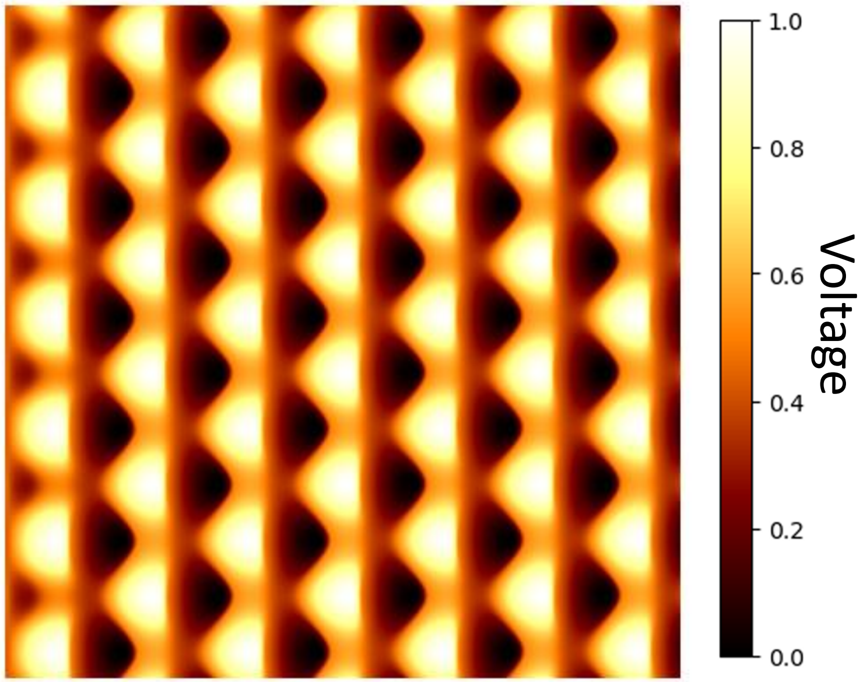


Hysteresis – Vertical Distortion

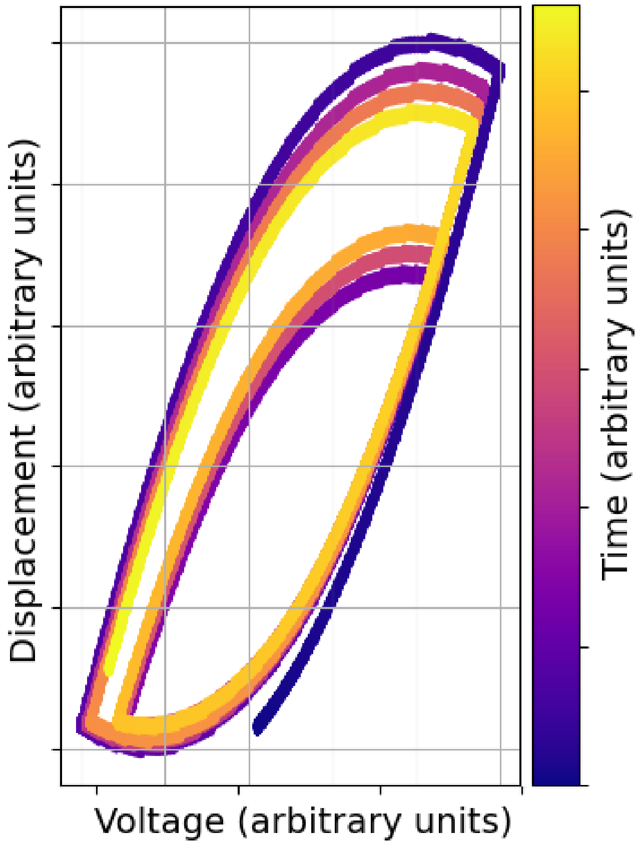
True Topography



Voltage Profile



Hysteresis



Simulation - Summary

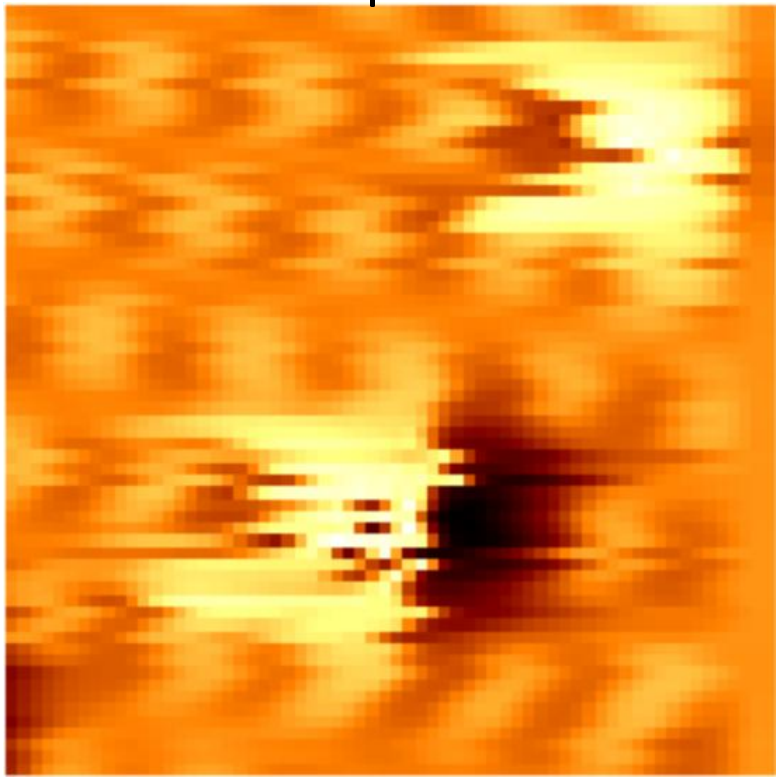
- Tunneling Current calculated using Tersoff-Hamann formula.
- Electronic structure approximated via Tight-Binding.
- Additional distortions are simulated:
  - Feedback Control
  - Statistical Noise
  - Piezoelectric Creep
  - Thermal Drift
  - Piezoelectric Hysteresis
  - Tilt (Not mentioned; trivial)

Application and Results

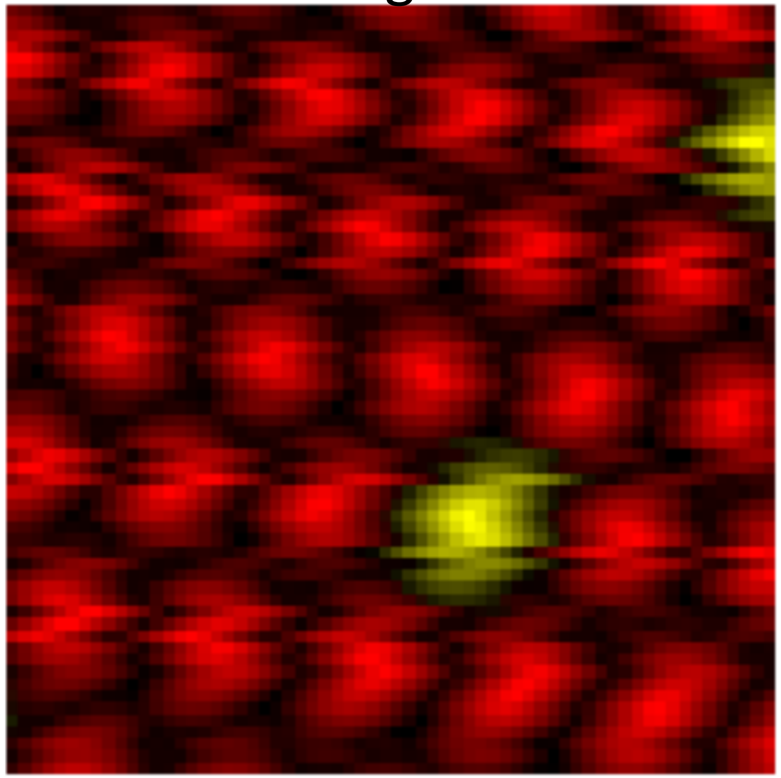
Task

- Defect localization for Tungsten Diselenide.

Input



Target

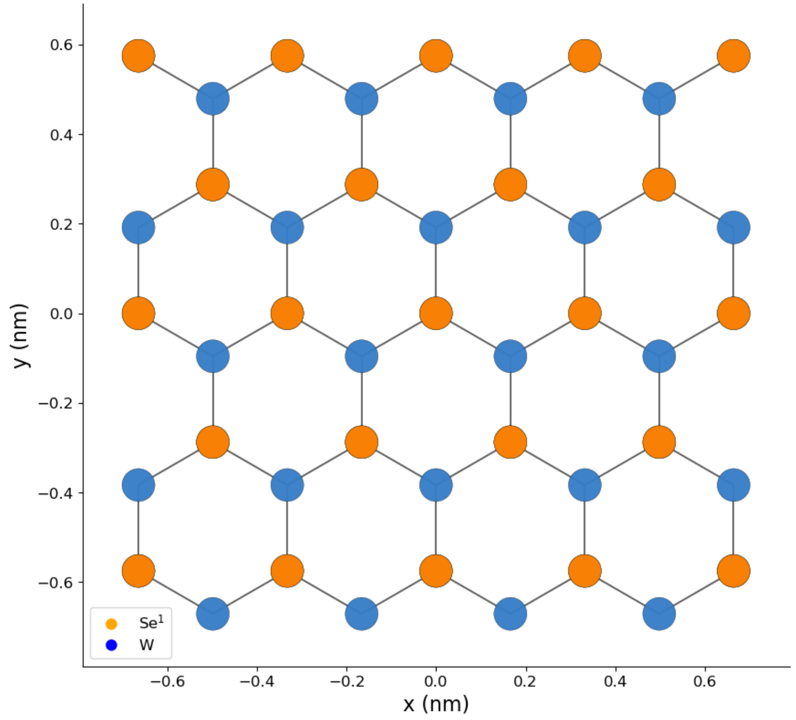


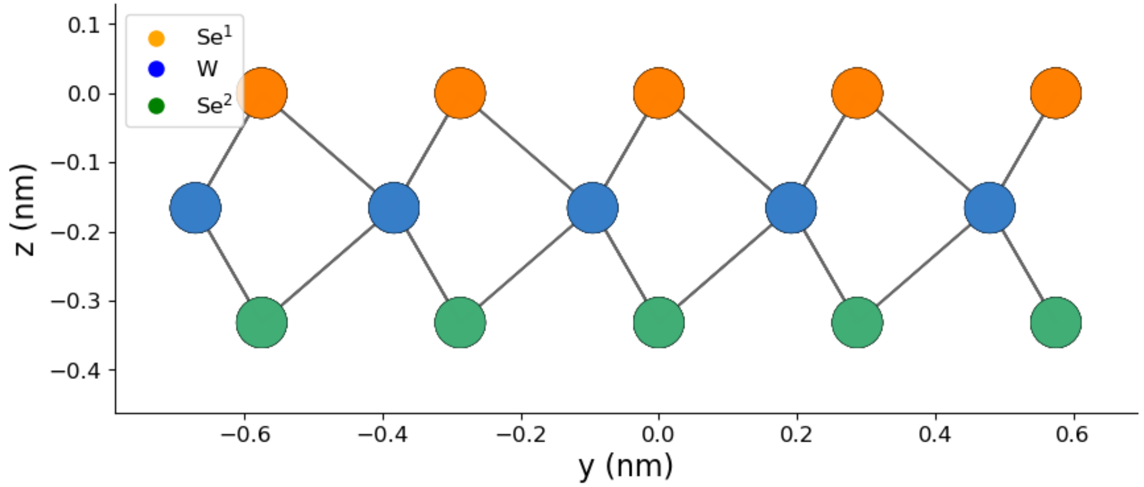
Application Outline

- Simulation Configuration
  - Tight-Binding Models
  - Dataset parameters
  - Dataset omissions
- Machine Learning Model
  - Architecture
  - Training
  - Validation
- Real Sample Evaluation

Tight-Binding Models - Sample

- Empirical Single-Orbital Model was used.
- Vacancies were simulated by directly removing an atom.
- Dopants were simulated indirectly as an additional onsite energy.



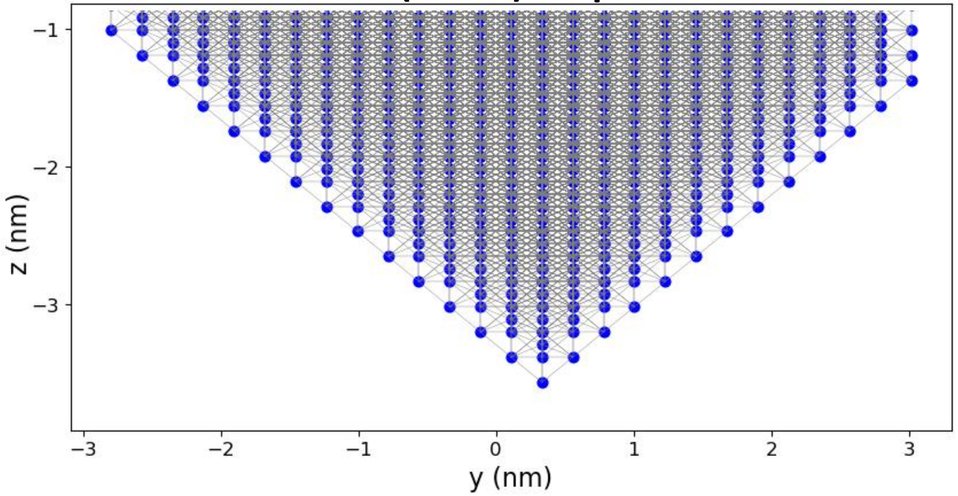


Tight-Binding Models - Tip

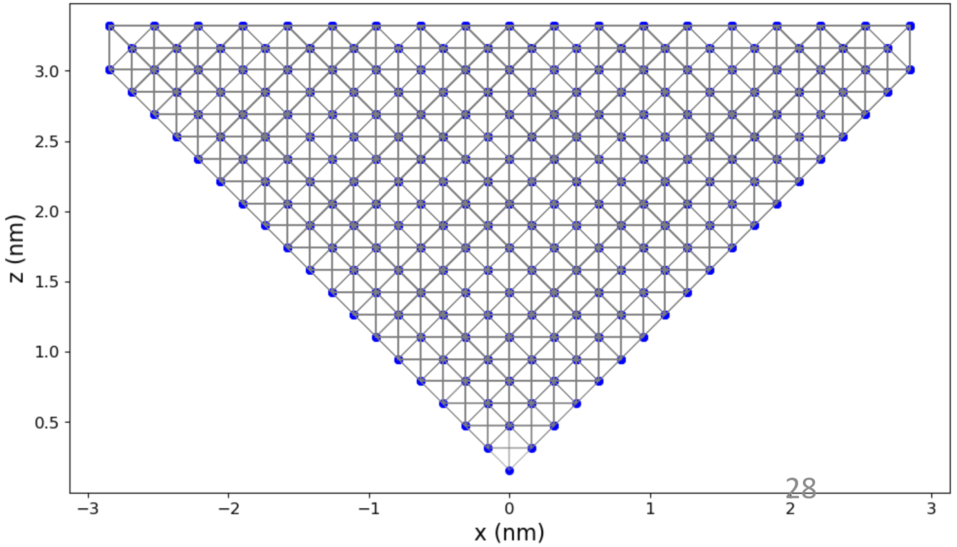
- The most common tip orientation is (111).
  - Represented as a rotated 4x4x4 nm cube.
- (001) Tip.
  - Represented as a 6x6x3 nm cone.

- Tips are progressively dulled to increase variety.

(111) Tip



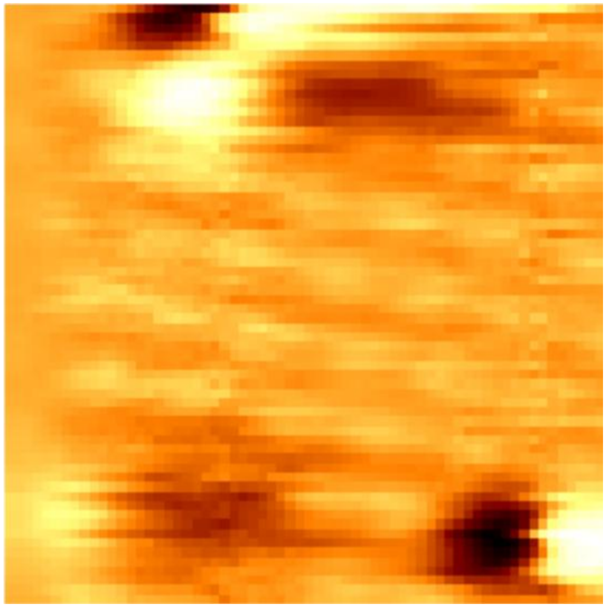
(001) Tip



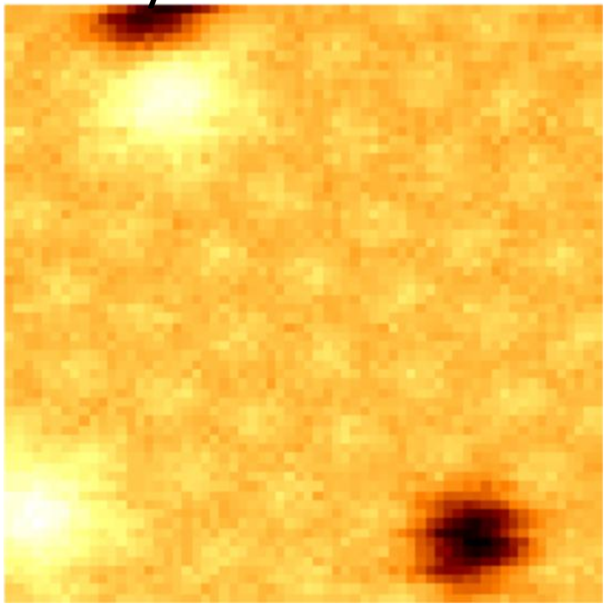
Dataset overview

- Simulation parameters sampled from a uniform random distribution.
- 100000 samples; 64x64 resolution; 1-4nm lateral dimension

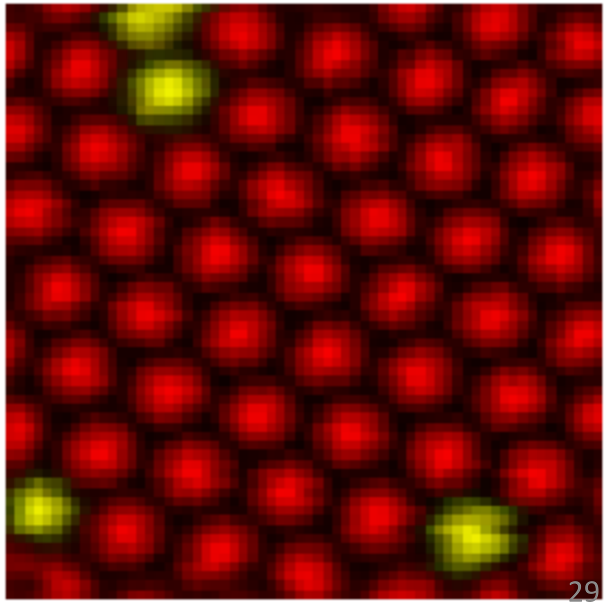
Main dataset
- Full PID Simulation



Control dataset
- Only statistical noise

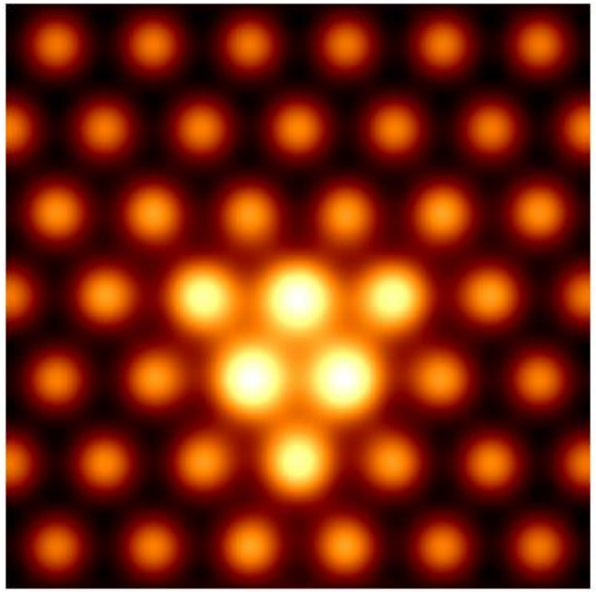


Common Target

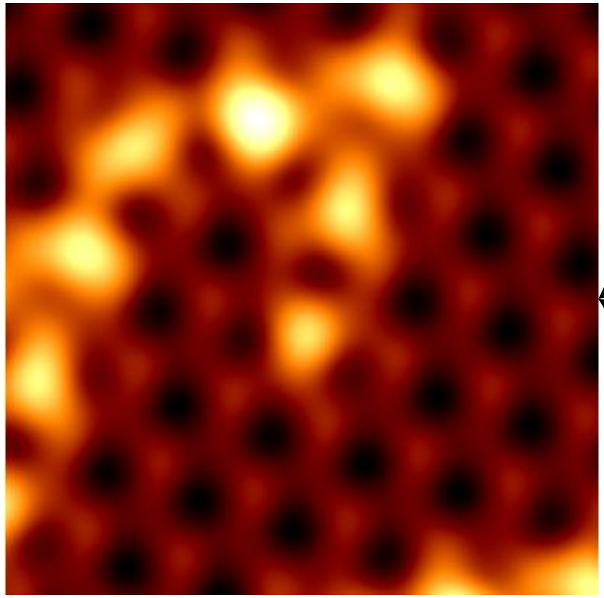


Dataset overview - Omissions

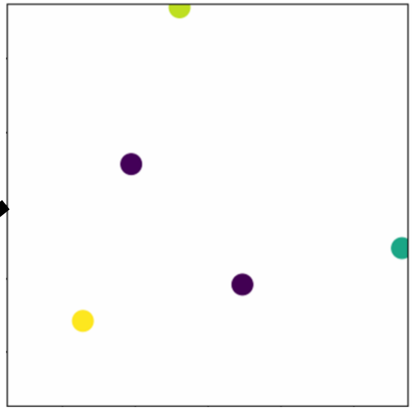
Tungsten defects



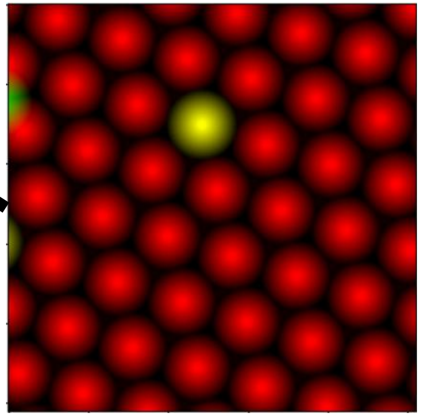
Multi-atom tip distortion



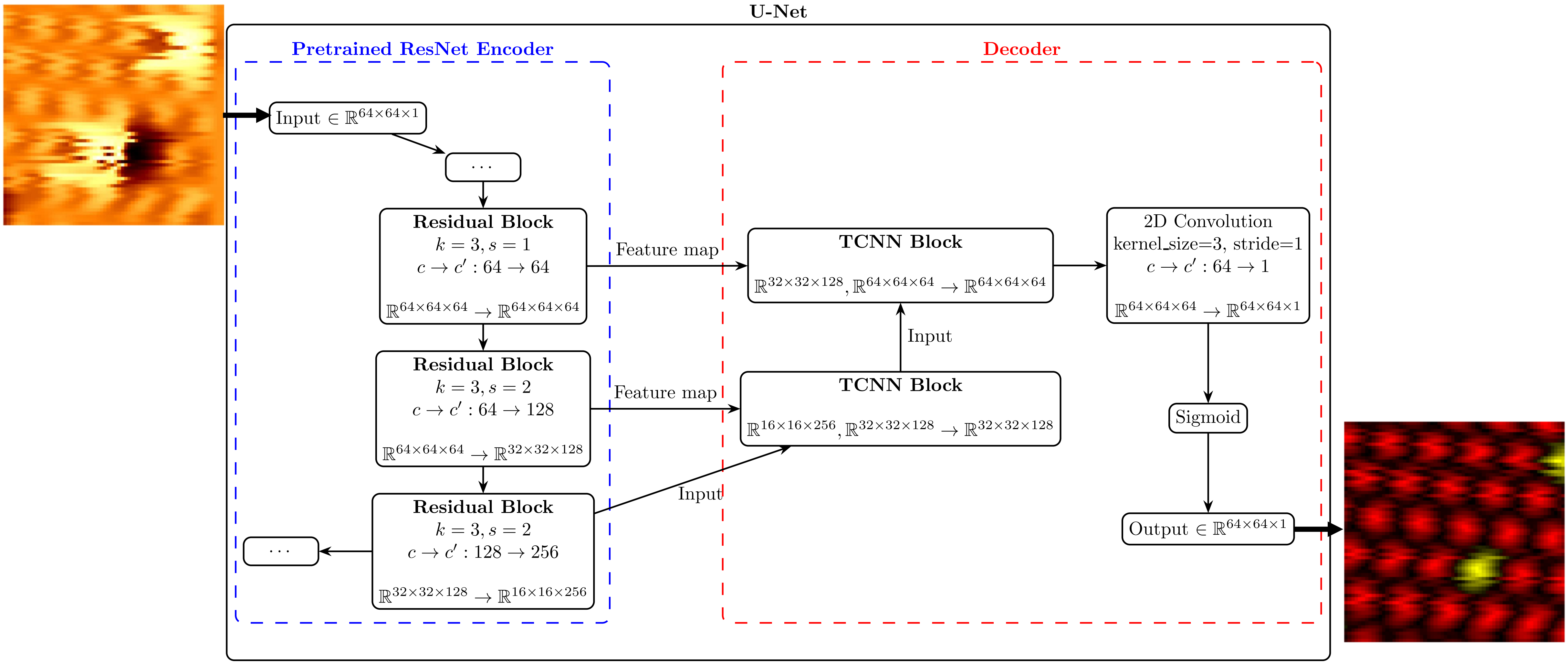
Multi-atom tip apex



Lattice



U-Net architecture



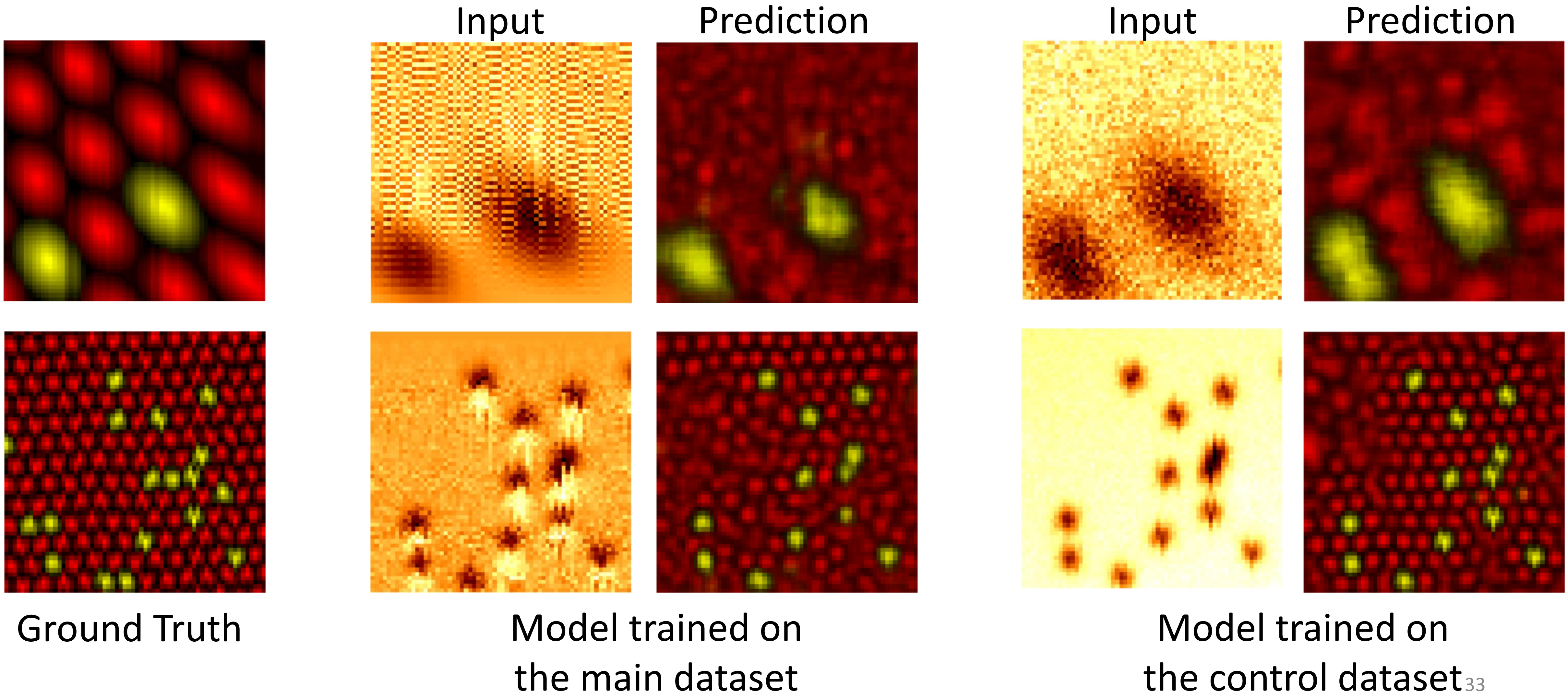
Training

- 40 epochs over 3 hours
- Batch size 16
- Learning Rate Range Test was used

- Ground truth pixel-wise target variance: 0.07096

Model	Dataset	MSE	Smooth L1 Loss
Main U-Net	Validation	0.0173	0.00864
	Training	0.0165	0.00824
Control U-Net	Validation	0.0137	0.00686
	Training	0.0131	0.00655

Validation on Simulated Samples

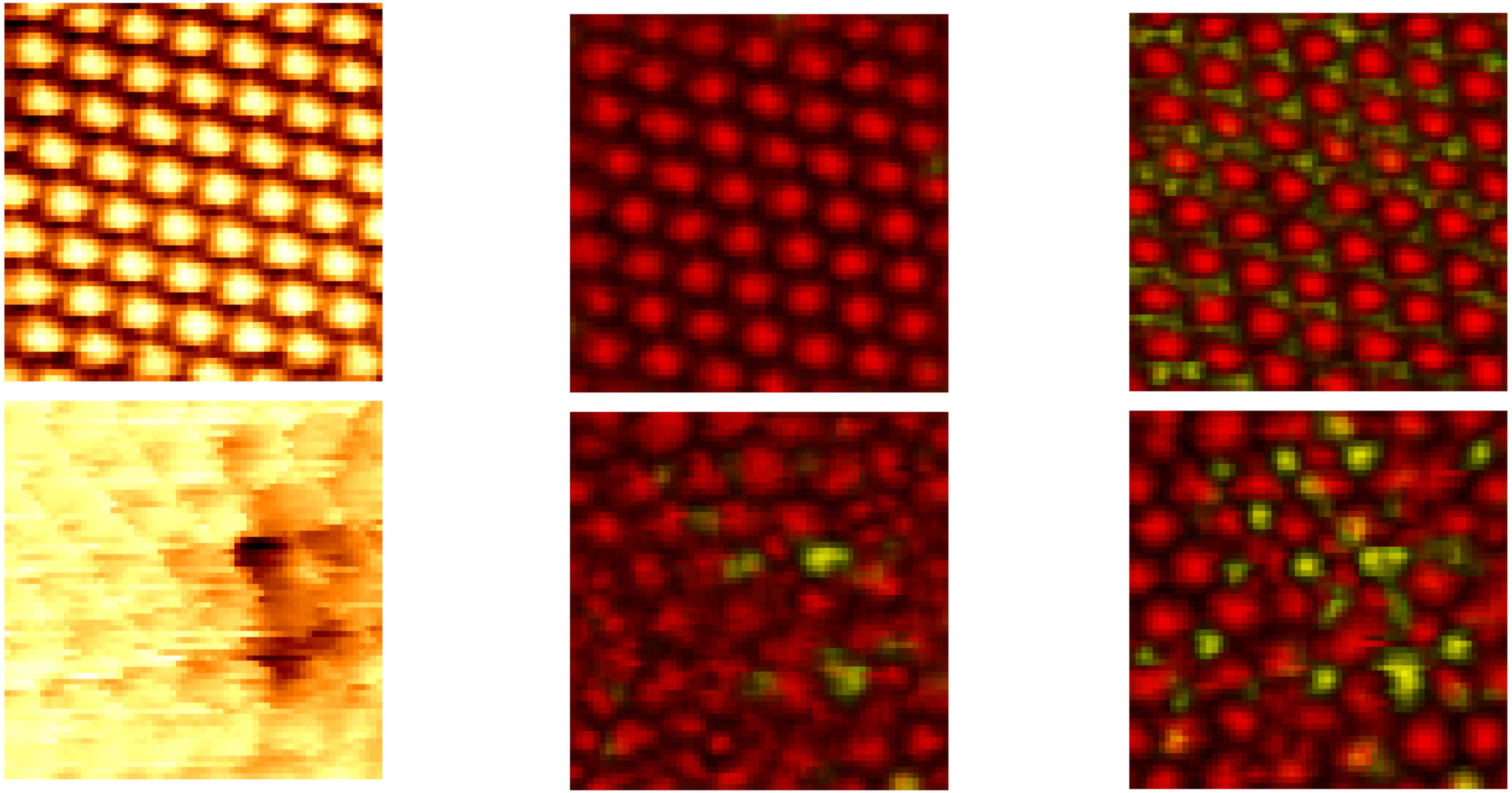


Real Sample Evaluation

Real measurements

Model trained on the main dataset

Model trained on the control dataset



Conclusion

Conclusion – Summary of contributions

- Simulation framework of STM imaging was developed.

- This framework can be used for the training of machine learning models that show better generalization than previous approaches.
- This was demonstrated through a simple machine learning task of defect localization.

Conclusion – Limitations and Future Work

- GPU parallelization could greatly speed up dataset creation.

- More accurate modelling needed for more complex tasks.

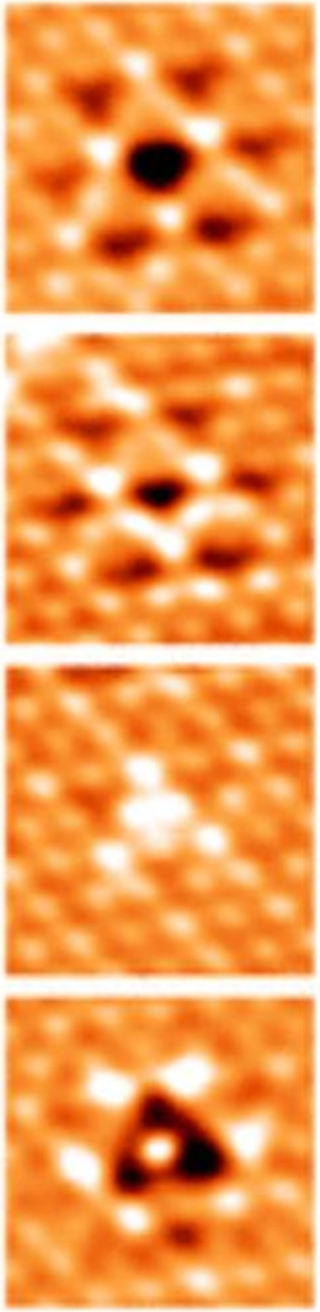
- No ground truth in real STM images for quantitative evaluation.

Thank you!

Q&A

Supplement

- MoS_2 defects [5]:



[5] H.-Y. Chen, H.-C. Hsu, J.-Y. Liang, B.-H. Wu, Y.-F. Chen, C.-C. Huang, M.-Y. Li, I. P. Radu, and Y.-P. Chiu. Atomically resolved defect-engineering scattering potential in 2d semiconductors. ACS Nano, 18(27):17622–17629, June 2024.

Supplement

- (111) tip apex dull layers

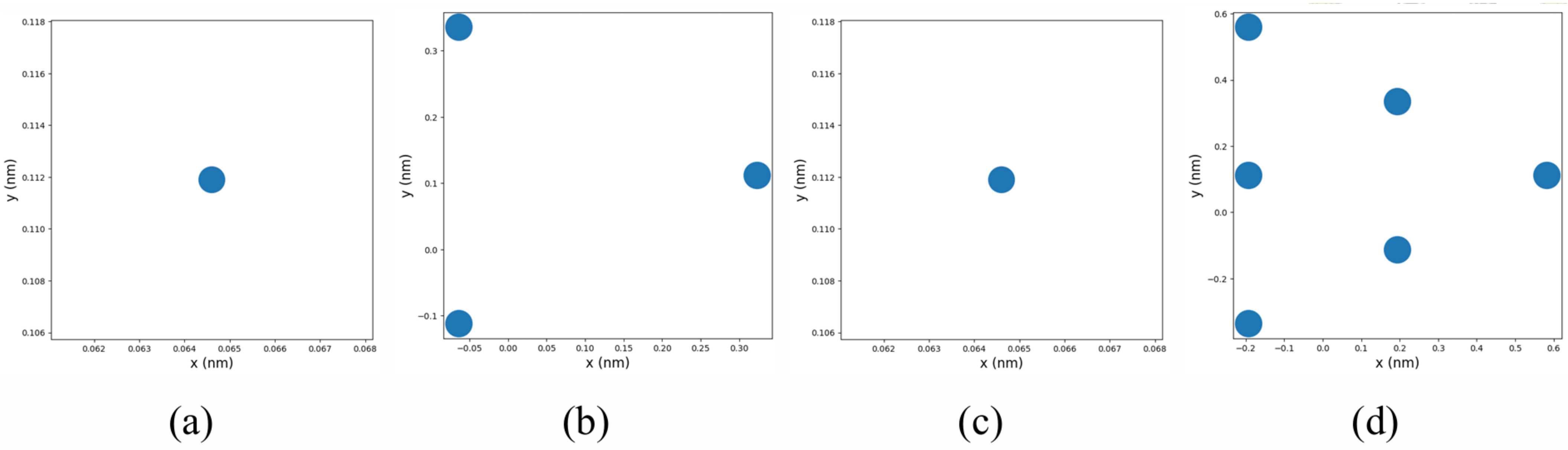
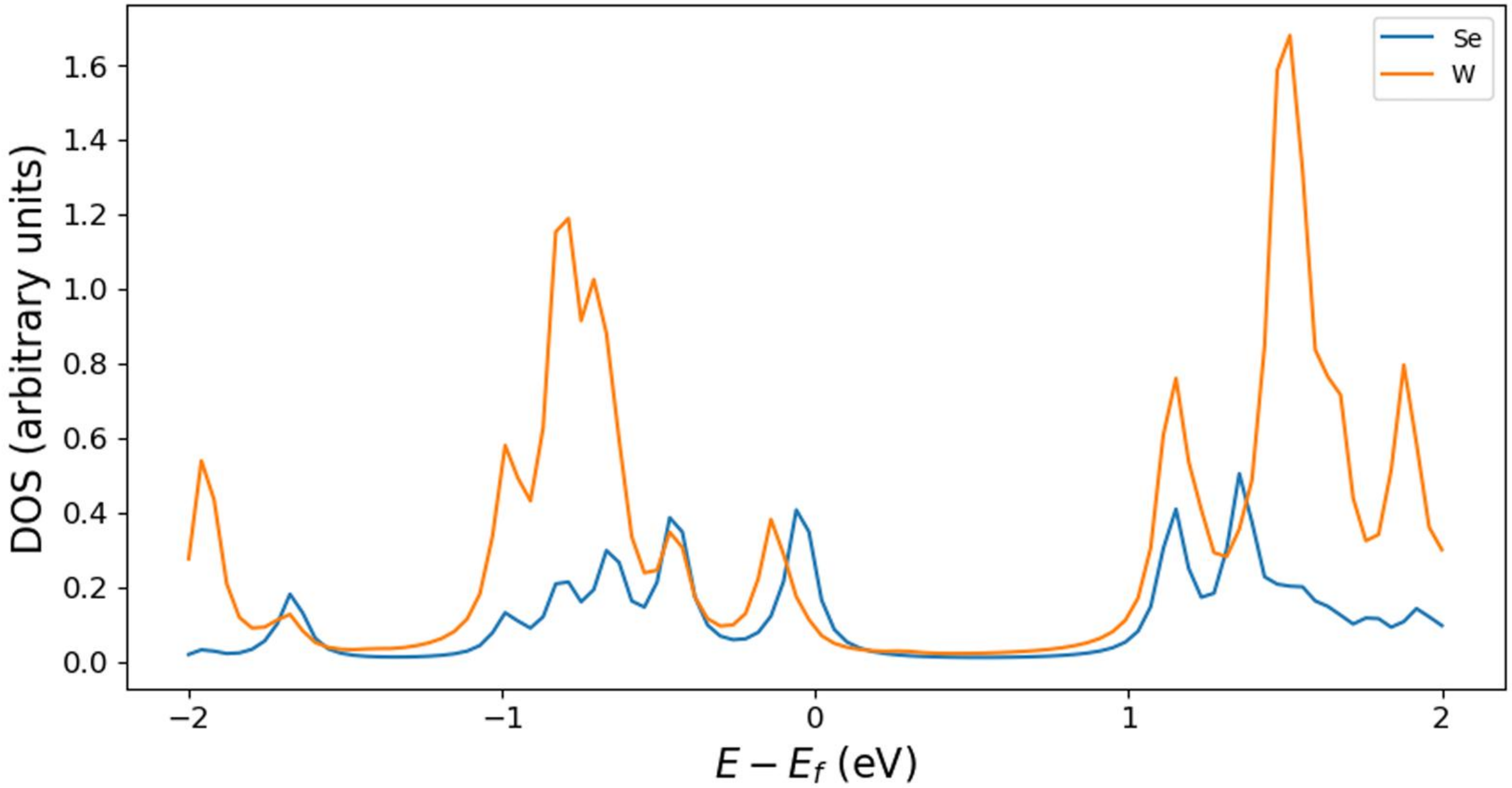


Figure 3.7: Atom positions in the 4 layers modelling the tip's apex.

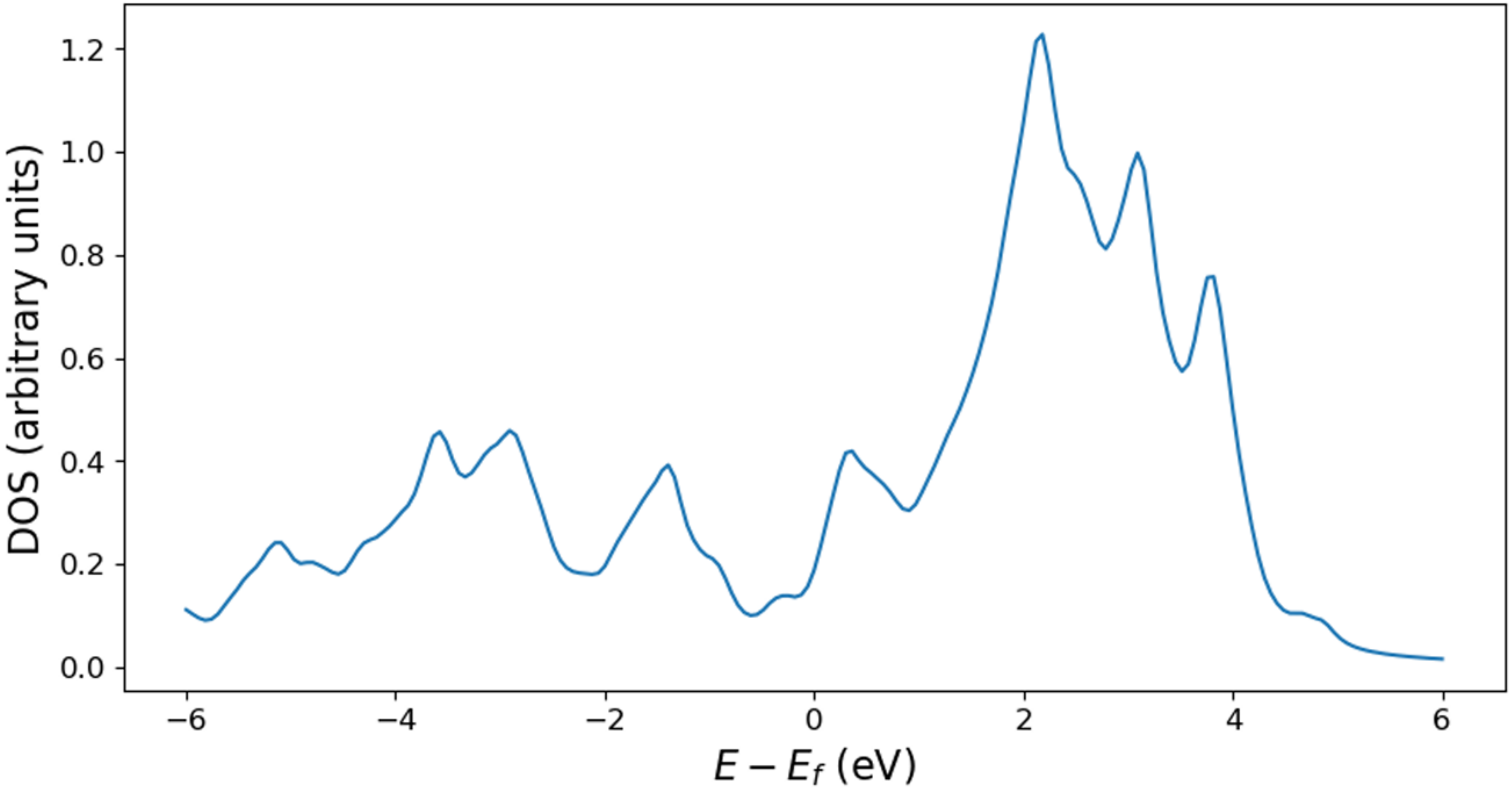
Supplement

- Multi-orbital WSe_2 Model LDOS



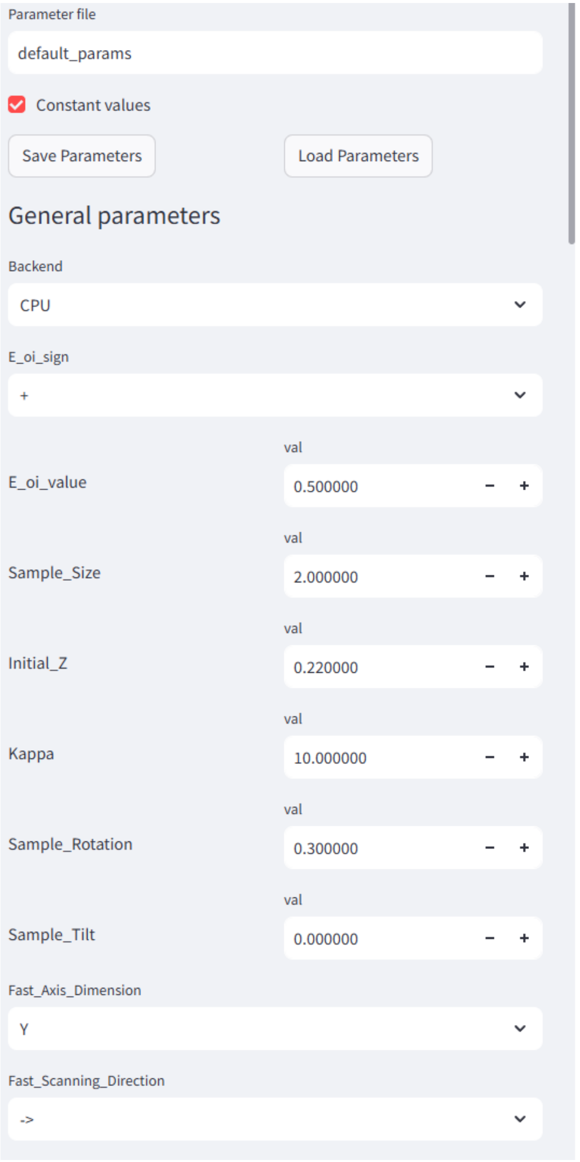
Supplement

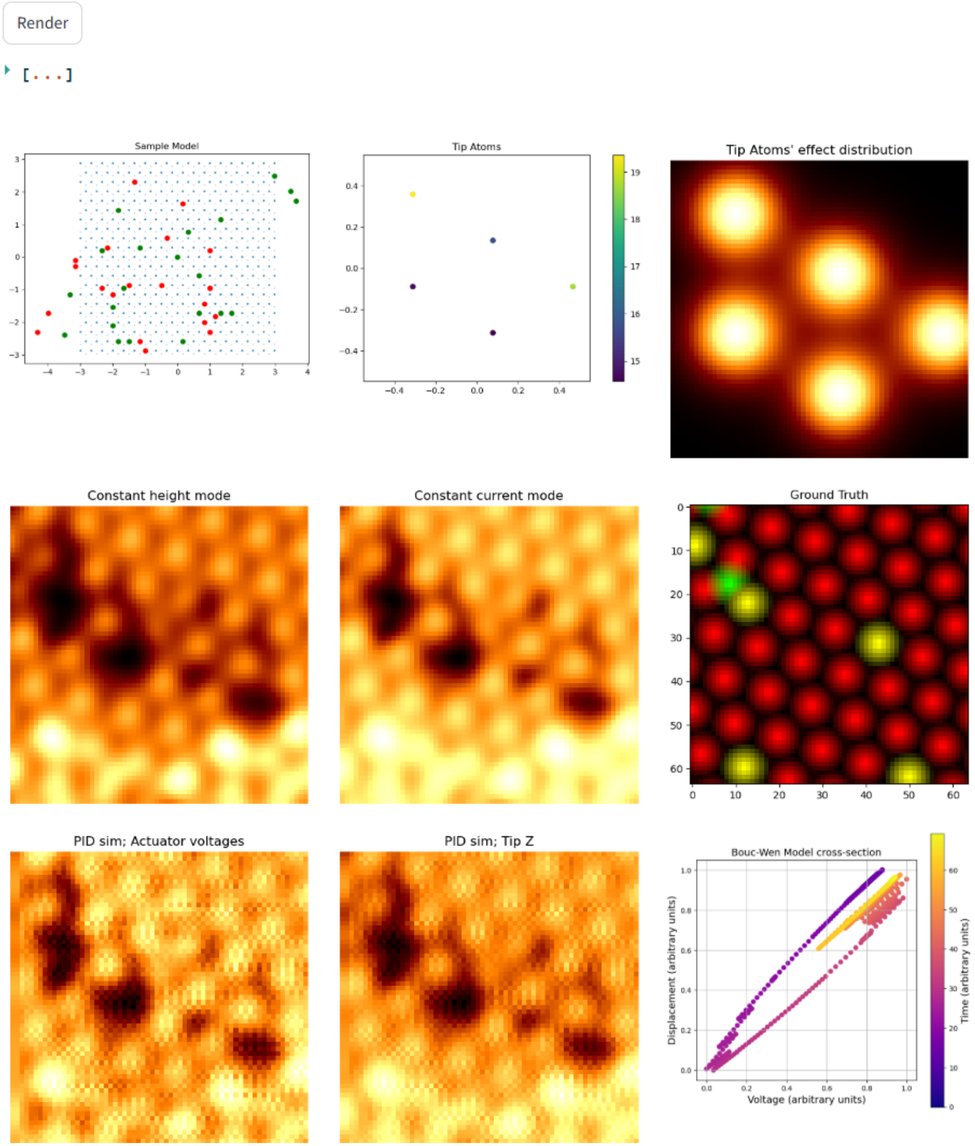
- Multi-orbital W Model LDOS



Supplement

- GUI
- Parameter selection





Supplement

- Classical Runge-Kutta Method

$$\hat{y}_0 \equiv y(t_0) = \alpha$$

$$k_1 = hf(t_i, \hat{y}_i)$$

$$k_2 = hf\left(t_i + \tfrac{h}{2}, \hat{y}_i + \tfrac{1}{2}k_1\right)$$

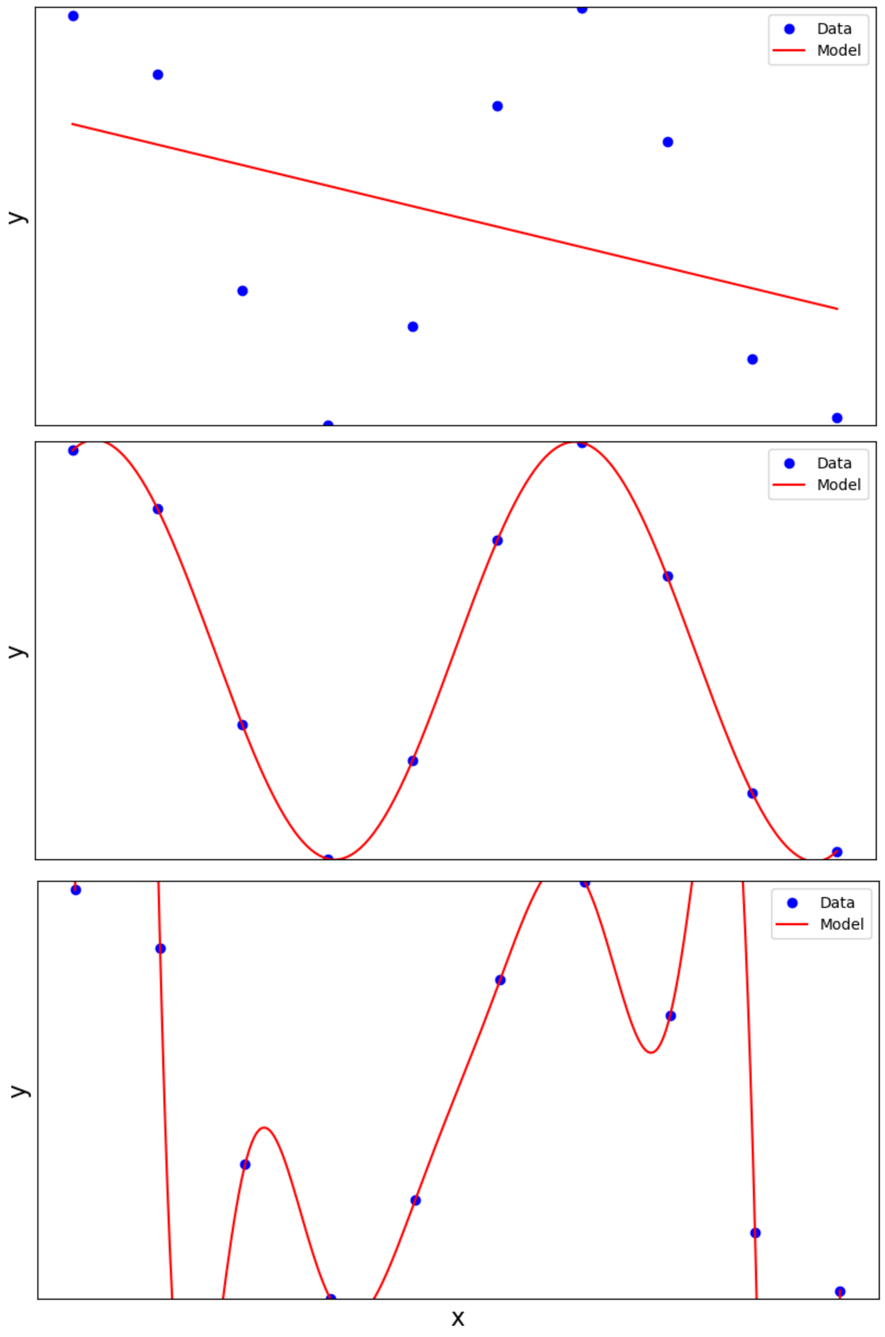
$$k_3 = hf\left(t_i + \tfrac{h}{2}, \hat{y}_i + \tfrac{1}{2}k_2\right)$$

$$k_4 = hf\left(t_{i+1}, \hat{y}_i + k_3\right)$$

$$\hat{y}_{i+1} = \hat{y}_i + \tfrac{1}{6}(k_1 + k_2 + k_3 + k_4)$$

Supplement

- Underfitting
- Appropriate capacity
- Overfitting



Supplement

- Convolutional layer