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Decoding VO_x diffraction patterns using machine learning

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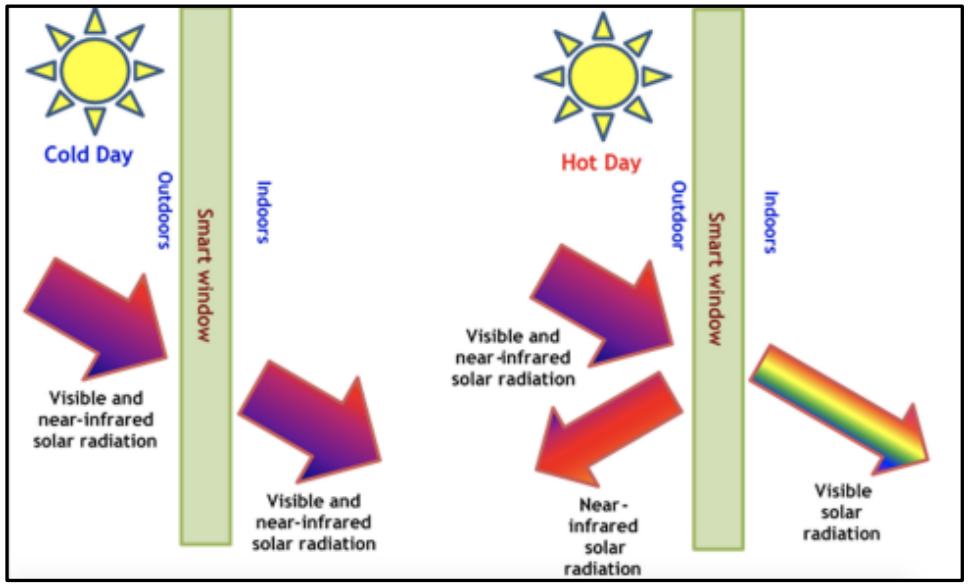
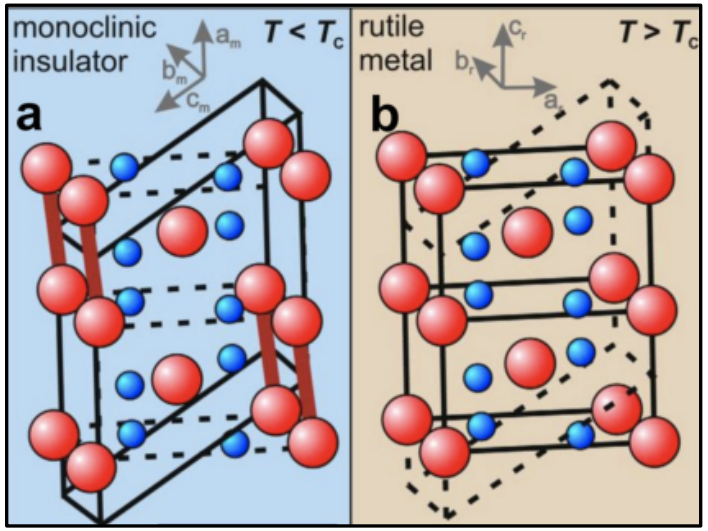
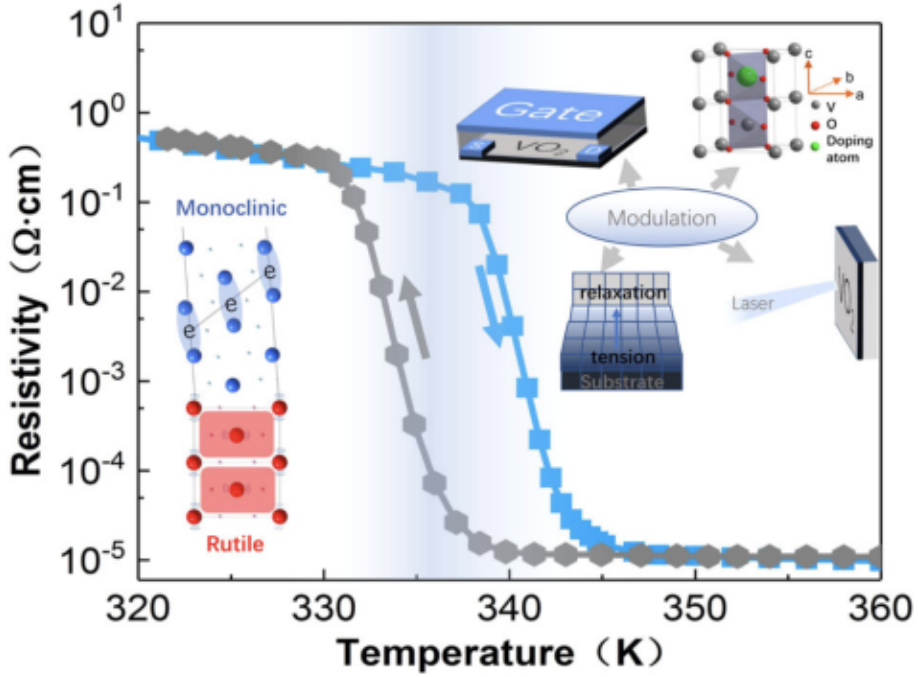
Metal to insulator transition in VO_x



Source: nist.gov

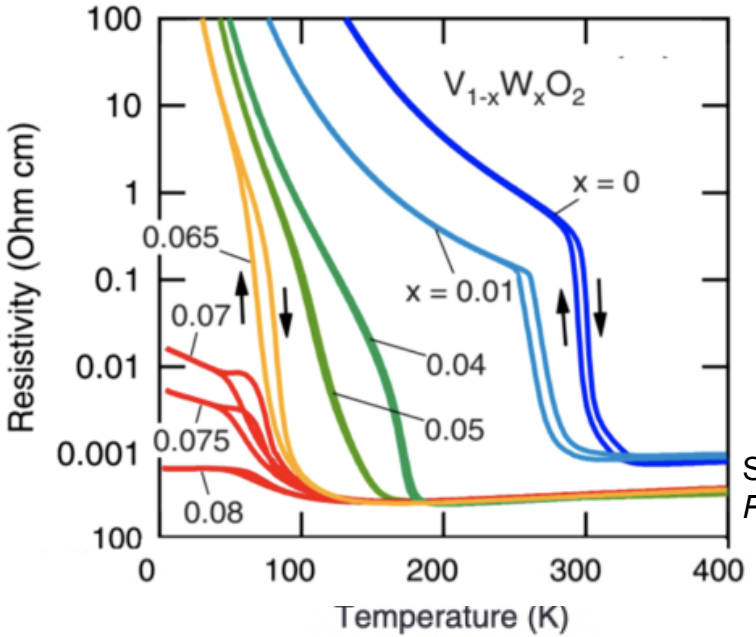




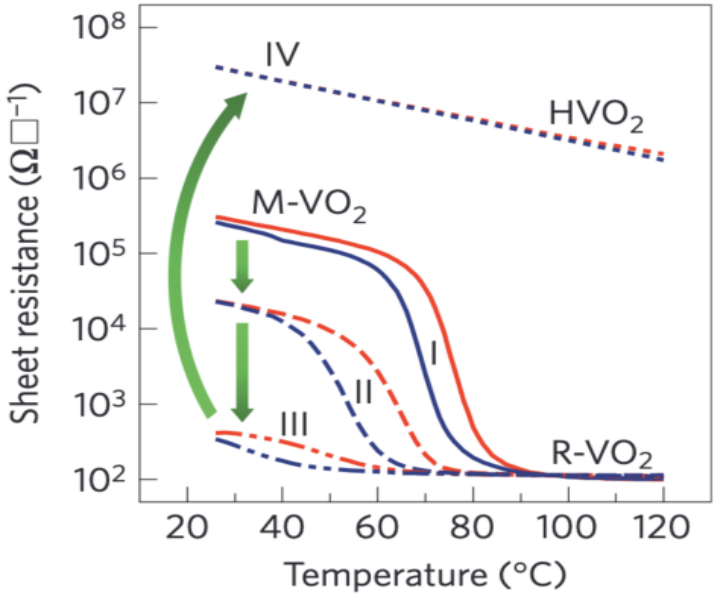
Shao et al. *NPG Asia Materials* (2018)

- VO_2 transforms from a metal to an insulator at 68°C
- Coincides with monoclinic to tetragonal transformation
- Applications: Smart window coatings, holographic memory, thermal IR imaging

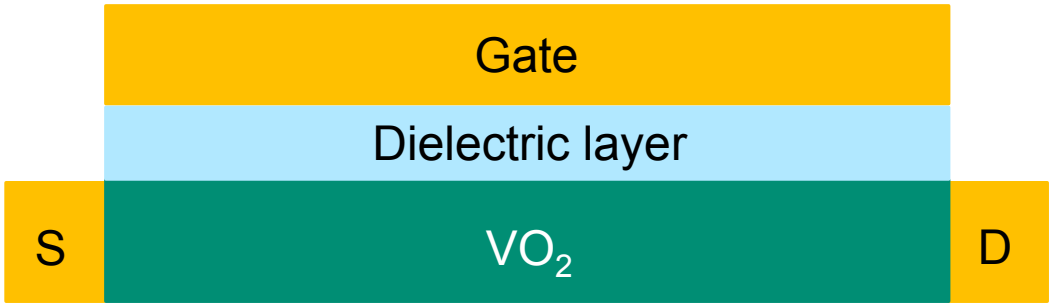
Controlling the metal to insulator transition in VOx

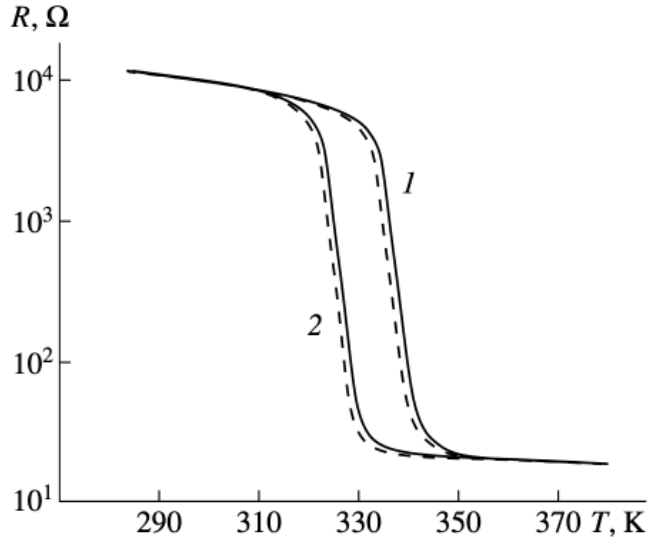


Shibuya et al. *Applied Physics Letters* (2010)



Yoon et al. *Nature Materials* (2016)



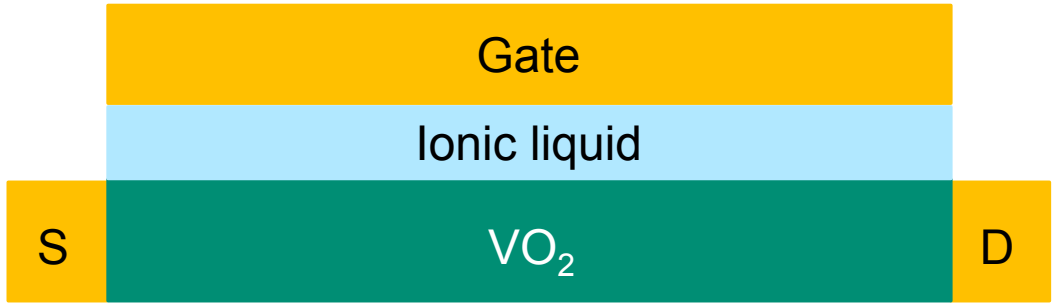


Boriskov *Technical Physics Letters* (2002)

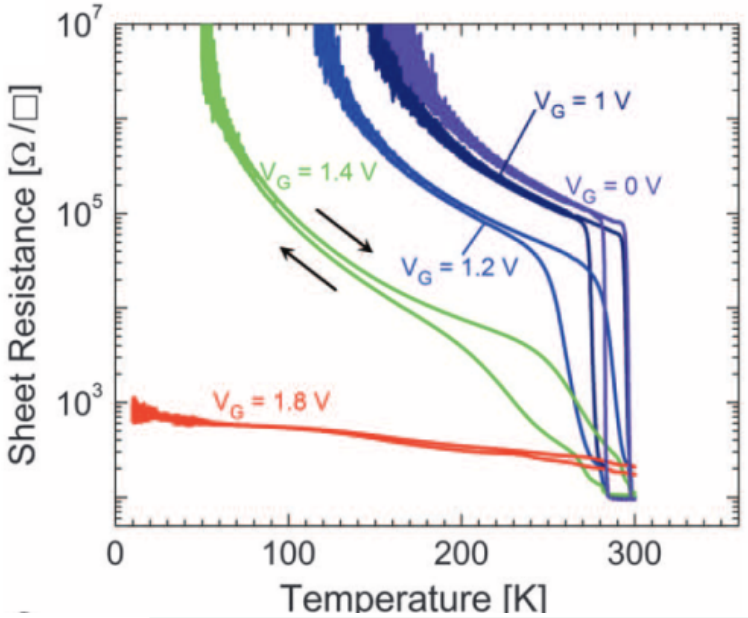
Doping elements change transformation

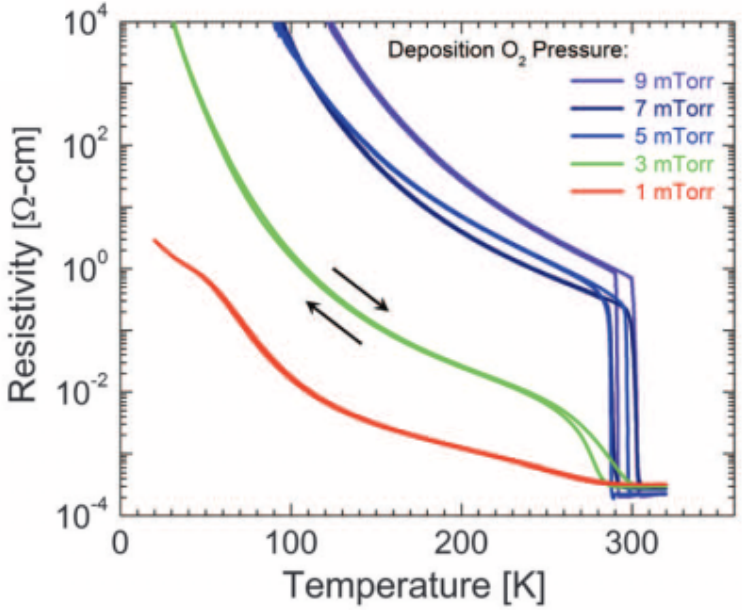
Gate voltage induces transformation

Vacancy induced suppression of MIT

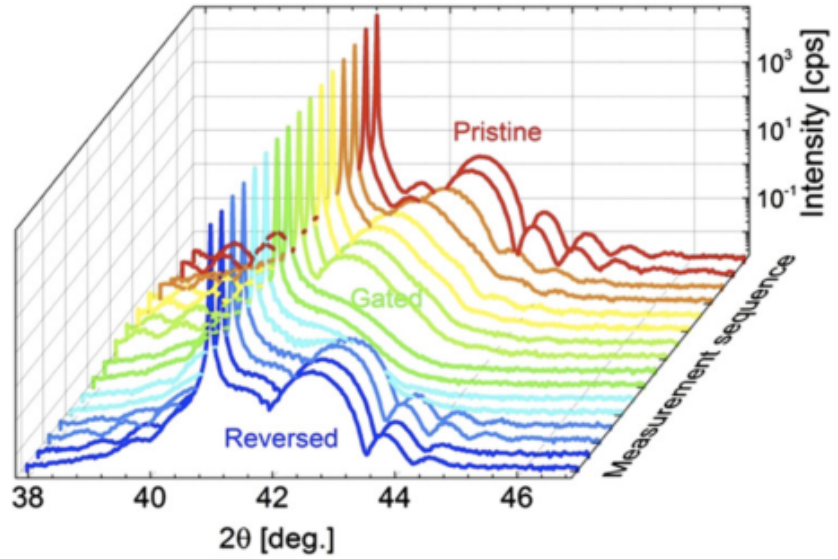


Jeong et al. *Science* (2013)





Jeong et al. *PNAS* (2015)



(002) peak shifts left on gating (opposite of PT)

Near neighbor distances [Å]		
Atom pair	Pristine	Gated
V-O	1.86	1.95
V-O	2.06	
V-V(intra dimer)	2.61	2.94
V-V(inter dimer)	3.03	3.16
V-V	3.49	3.53

Electrolyte gating induces oxygen vacancies in VO_2

Oxygen vacancies induce metallization in the monoclinic state while expanding lattice

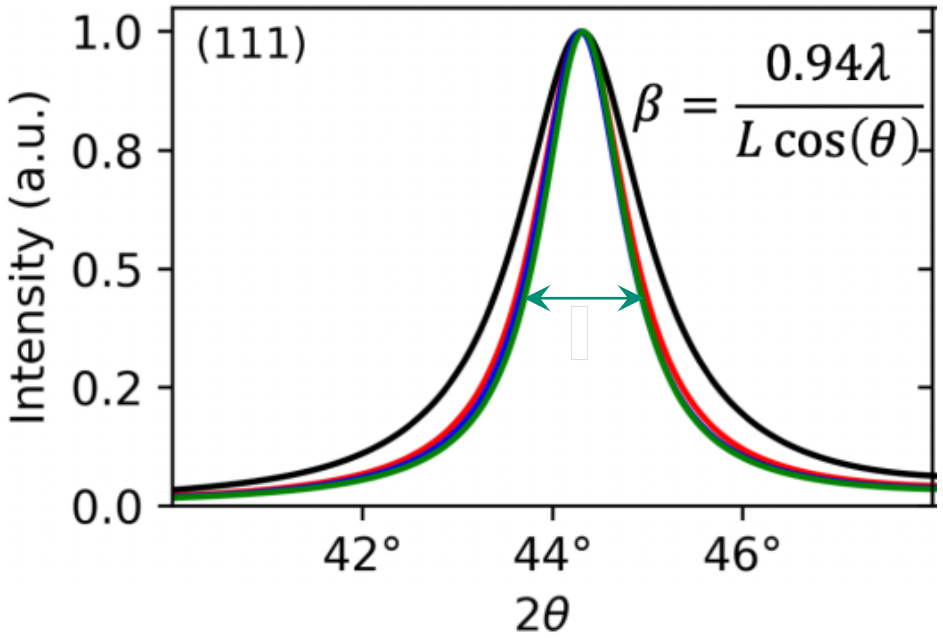
Decoding diffractograms to extract defect statistics

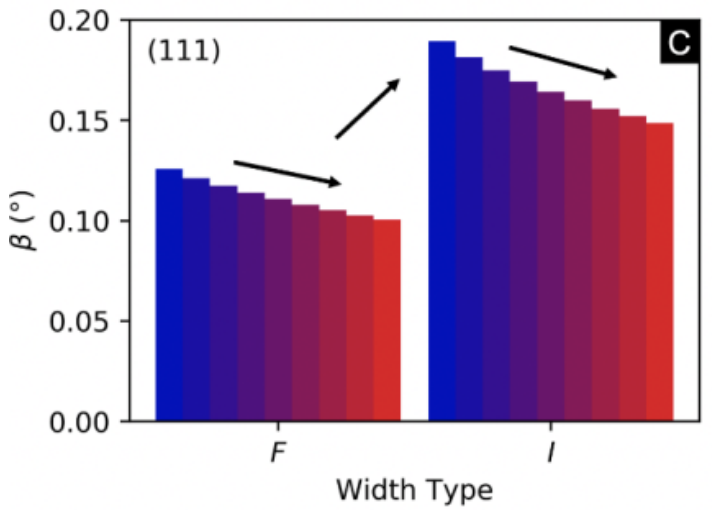
What vacancy concentrations resulted in the metallization of monoclinic VO_x?

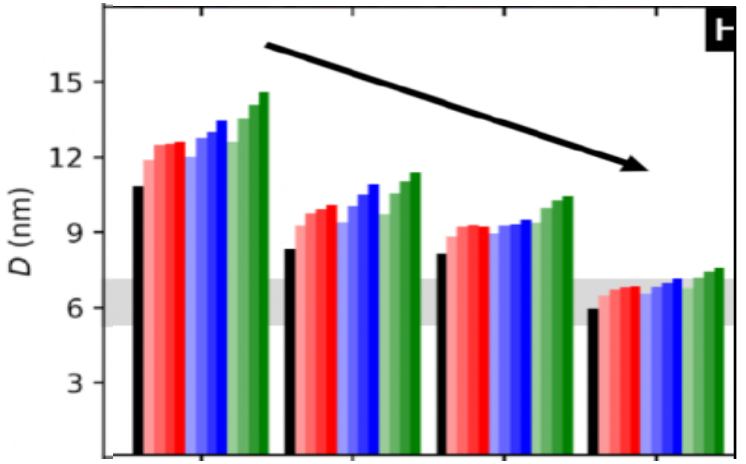
What defect statistics resulted in the metallization of monoclinic VO_x?

Diffractograms = non-intrusive way to proxy defect statistics

How do we decode VO_x diffractograms and relate them to defect statistics?





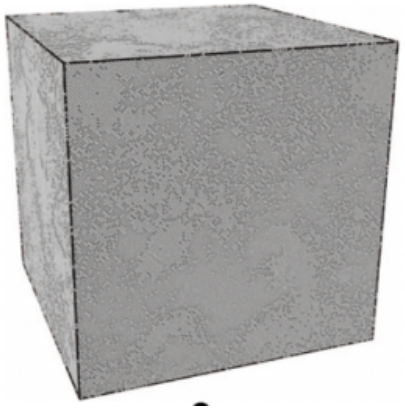


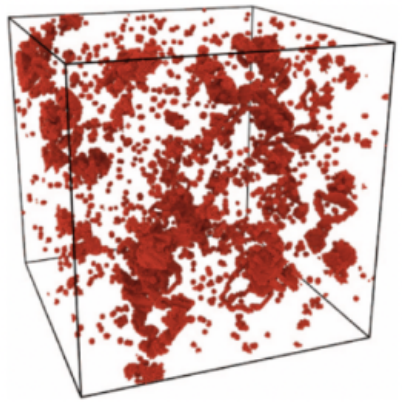
Manual decoding methods may not capture all information accurately

Kunka et al. *Nanoscale* (2019)

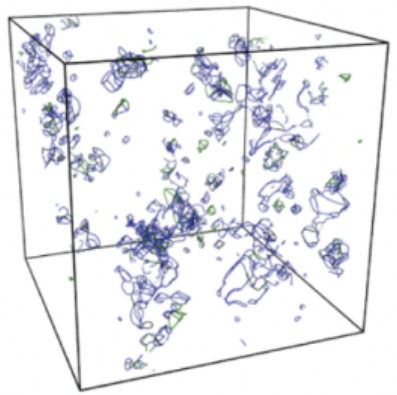
Using machine learning to decode diffraction patterns

Irradiated copper

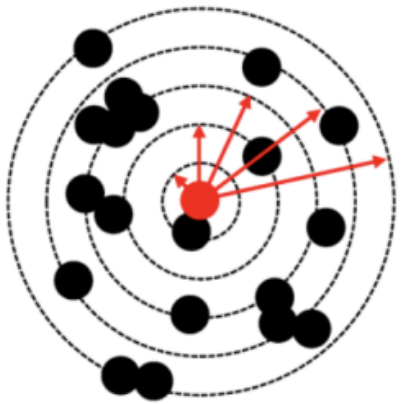




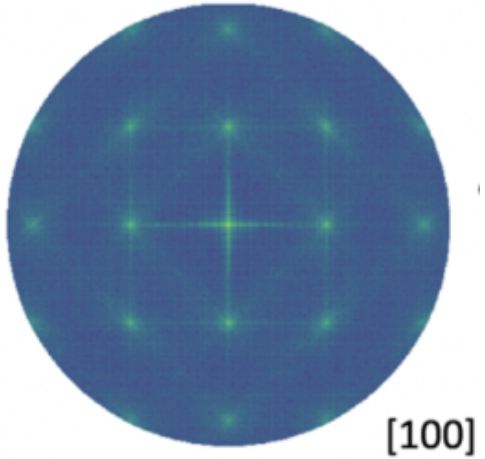
Point defect density



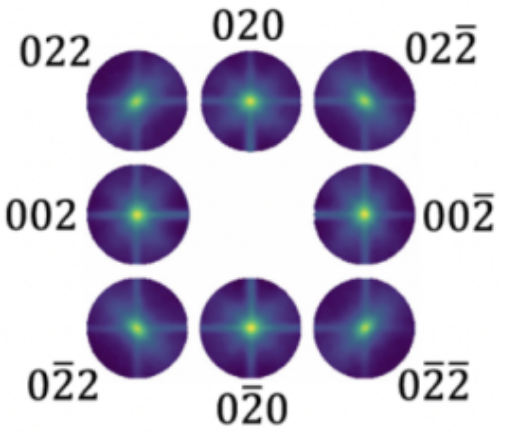
Dislocation density



Pair correlation function



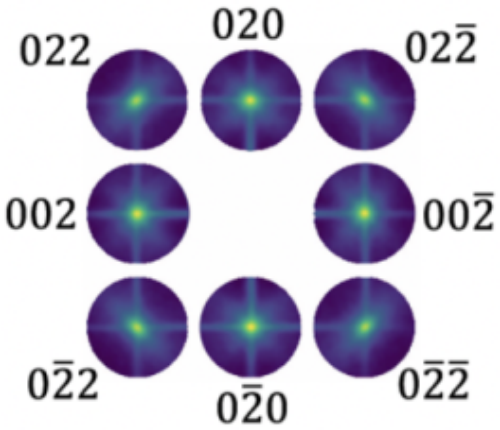
SAED pattern



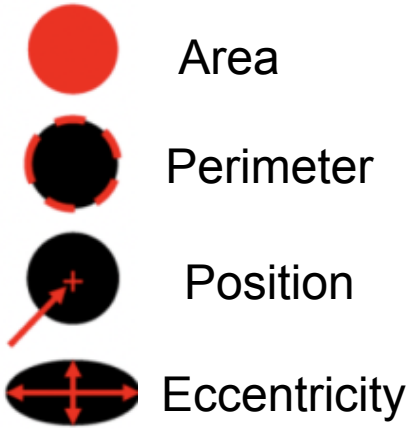
Processed pattern

Kunka et al. *npj Computational Materials* (2021)

Using machine learning to decode diffraction patterns



Processed pattern

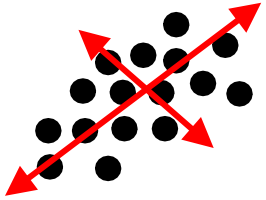


Gaussian Process Regression

Point defect density

Dislocation density

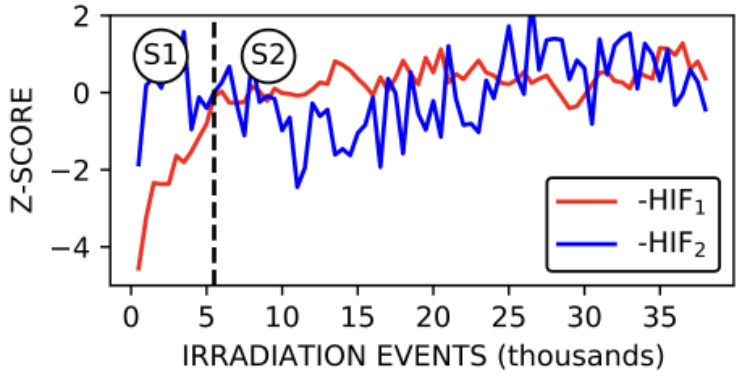
Pair correlation function

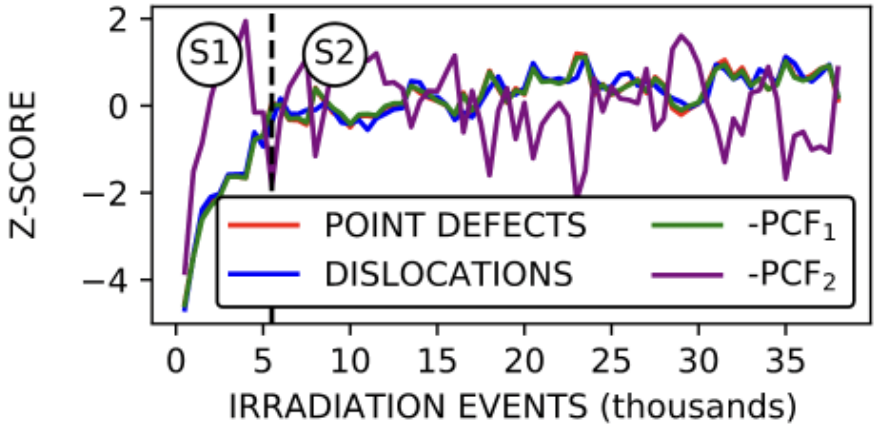


PCA of diffractogram

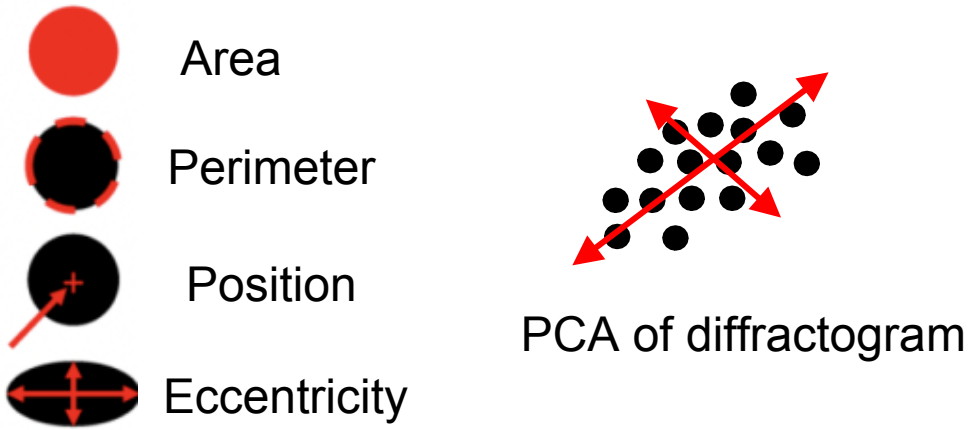
Gaussian Process Regression

Machine identified features capture defect densities and correlations more accurately than human identified features





Using machine learning to decode diffraction patterns



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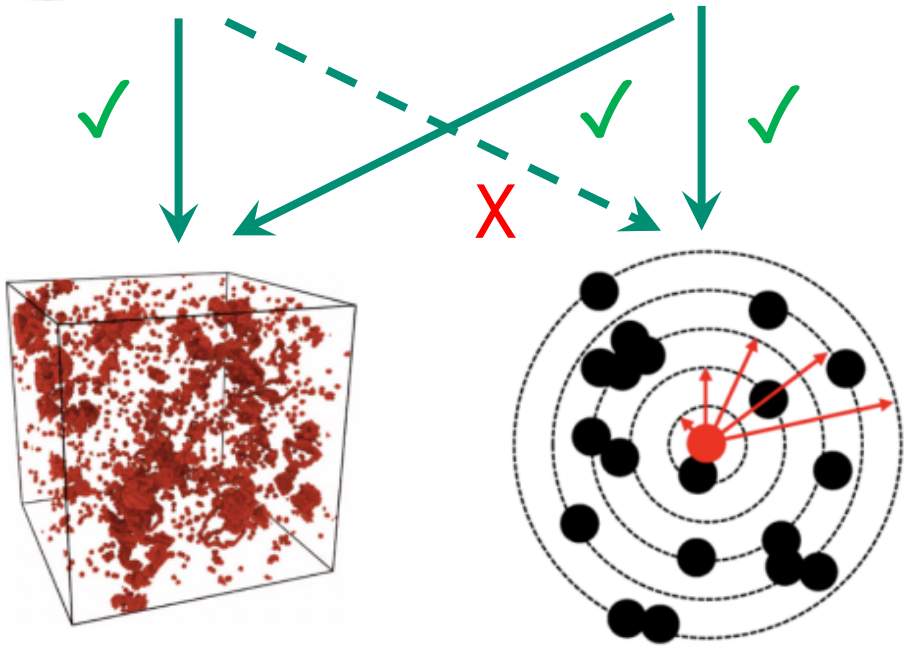
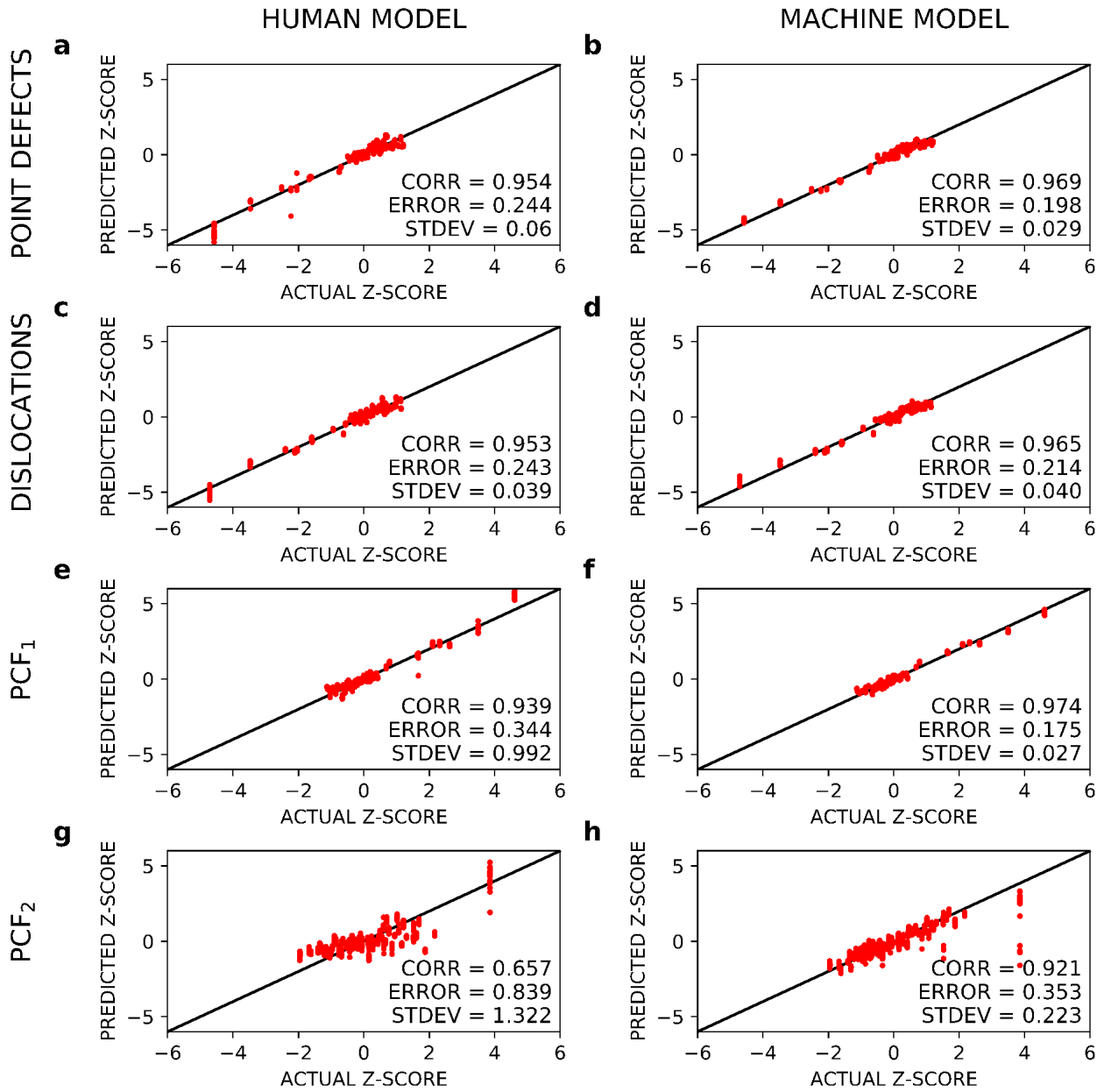
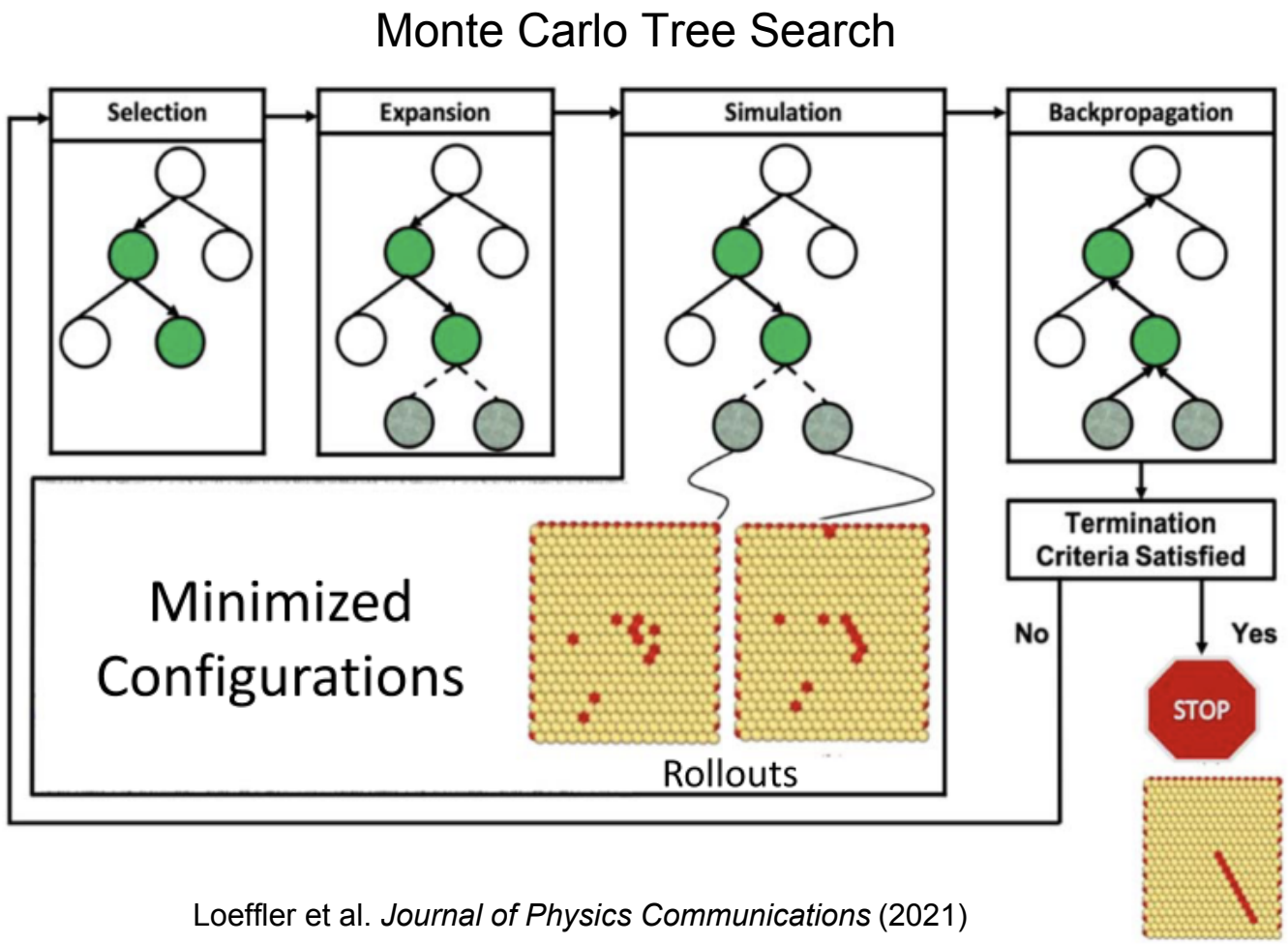


1 point statistics

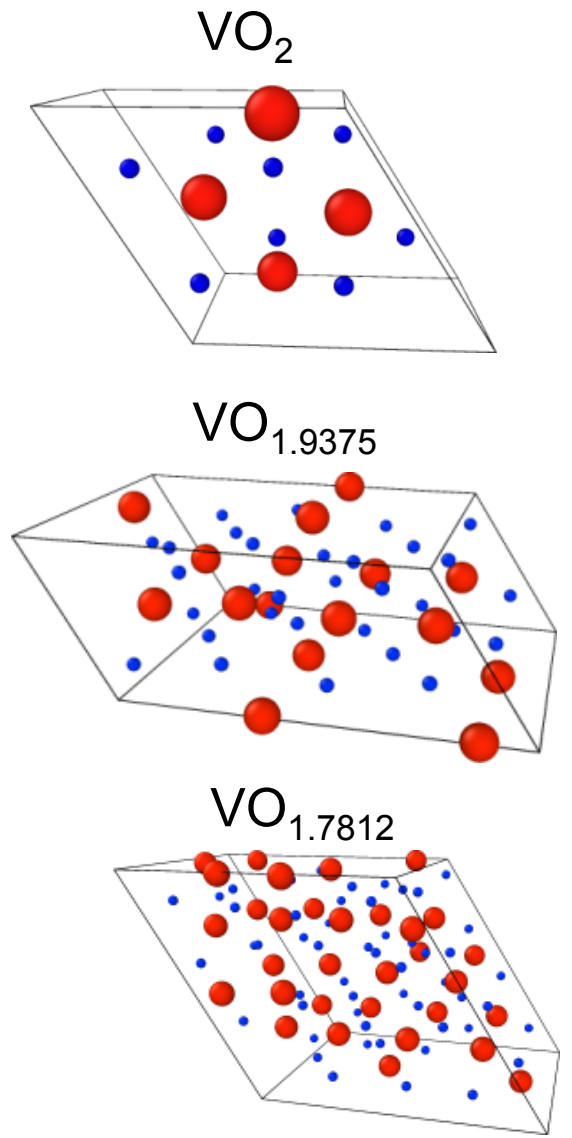
2 point statistics



Generating VO_x structures



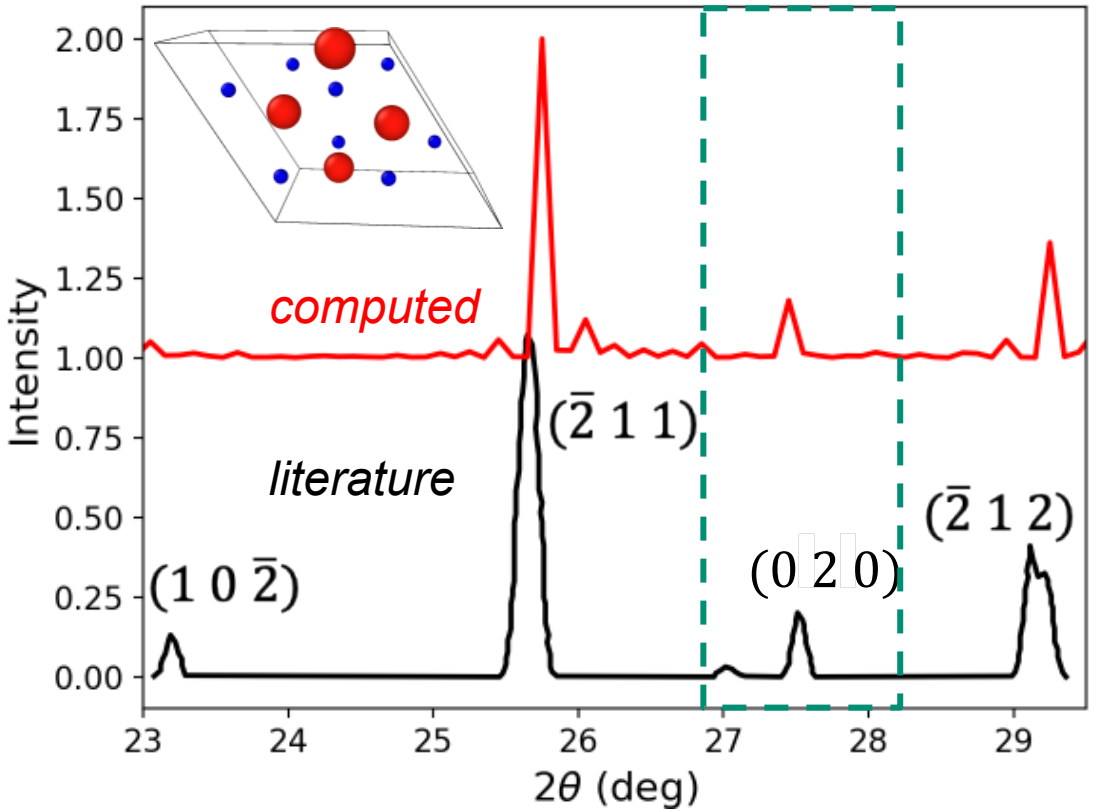
Loeffler et al. *Journal of Physics Communications* (2021)



Machine identified features capture defect densities and correlations more accurately than human identified

Calibrating XRD calculations

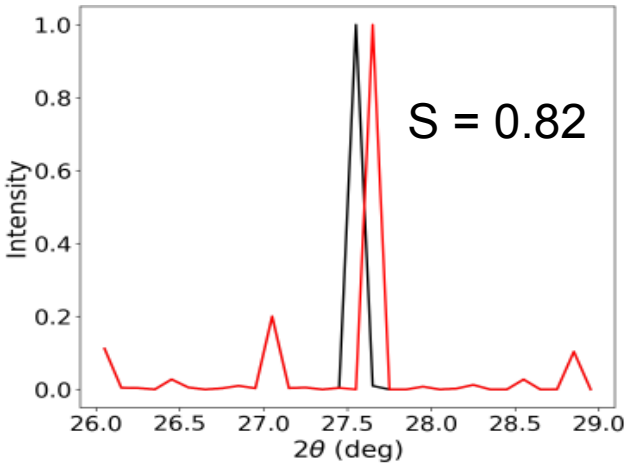
VO_2 structure

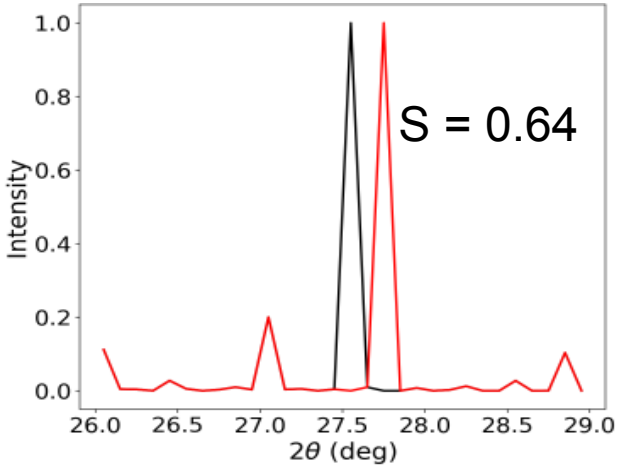


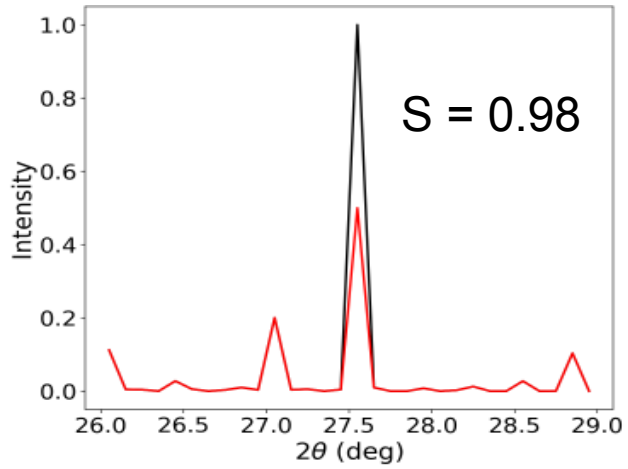
Simulated XRD patterns captures important peak locations accurately

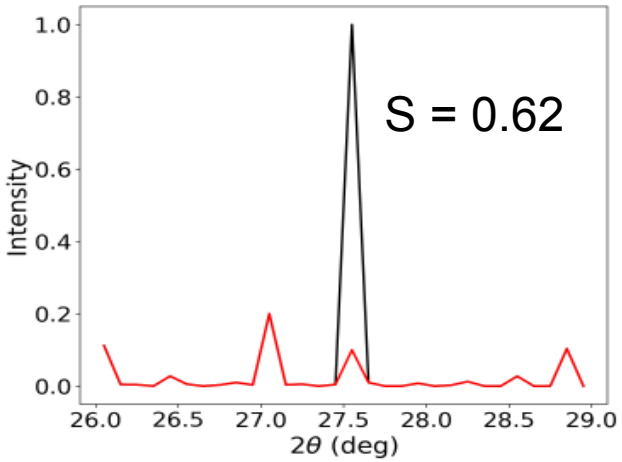
Cross-correlation metric

$$S_{fg} = \frac{\Sigma_{r=-t}^{r=t} w(r) \Sigma_{i=1}^{n} (f_i * g_{i+r})}{\left(\Sigma_{r=-t}^{r=t} w(r) \Sigma_{i=1}^{n} (f_i * f_{i+r})\right)^{1/2} \left(\Sigma_{r=-t}^{r=t} w(r) \Sigma_{i=1}^{n} (g_i * g_{i+r})\right)^{1/2}}$$

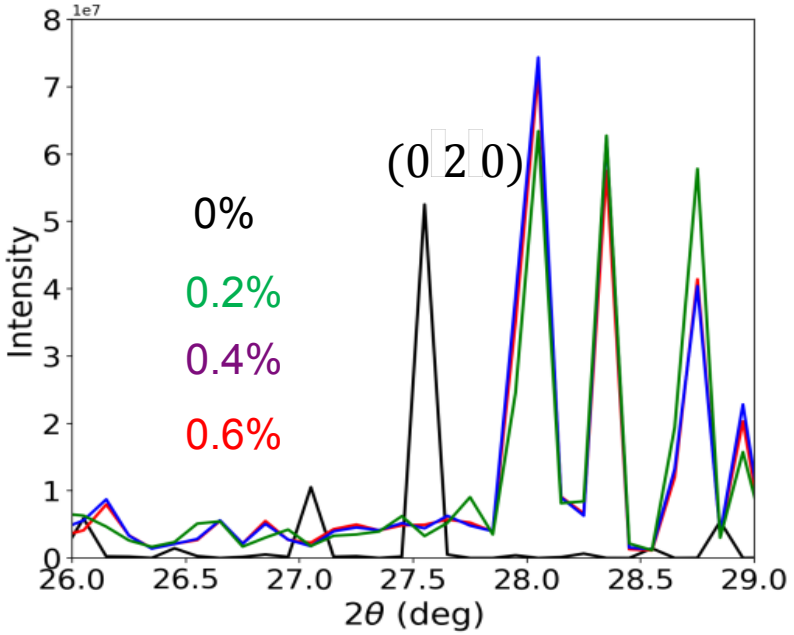




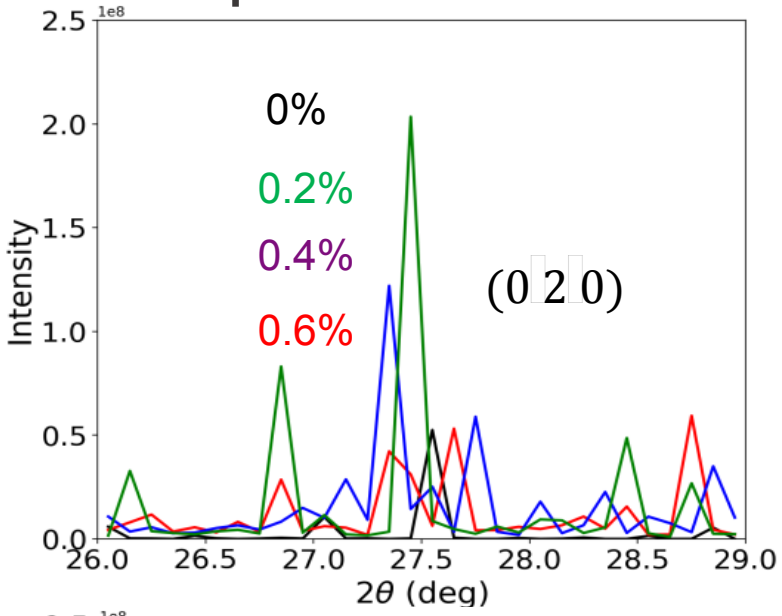


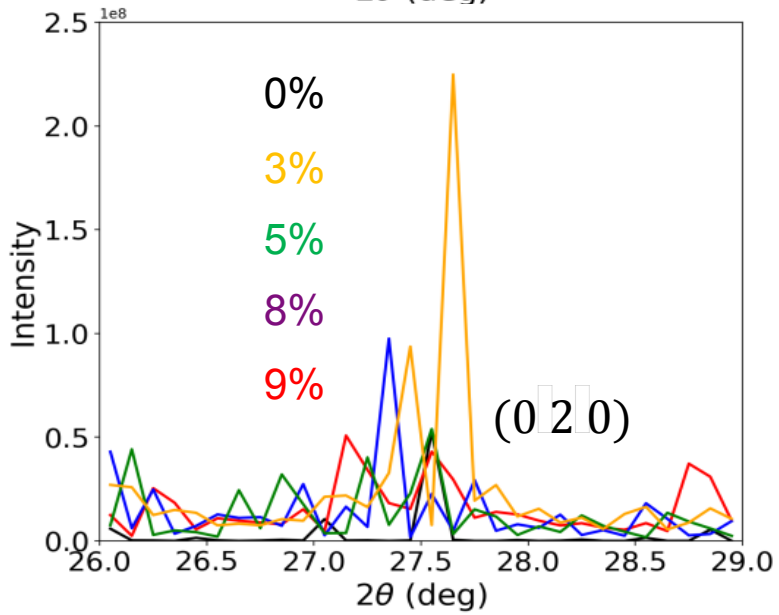


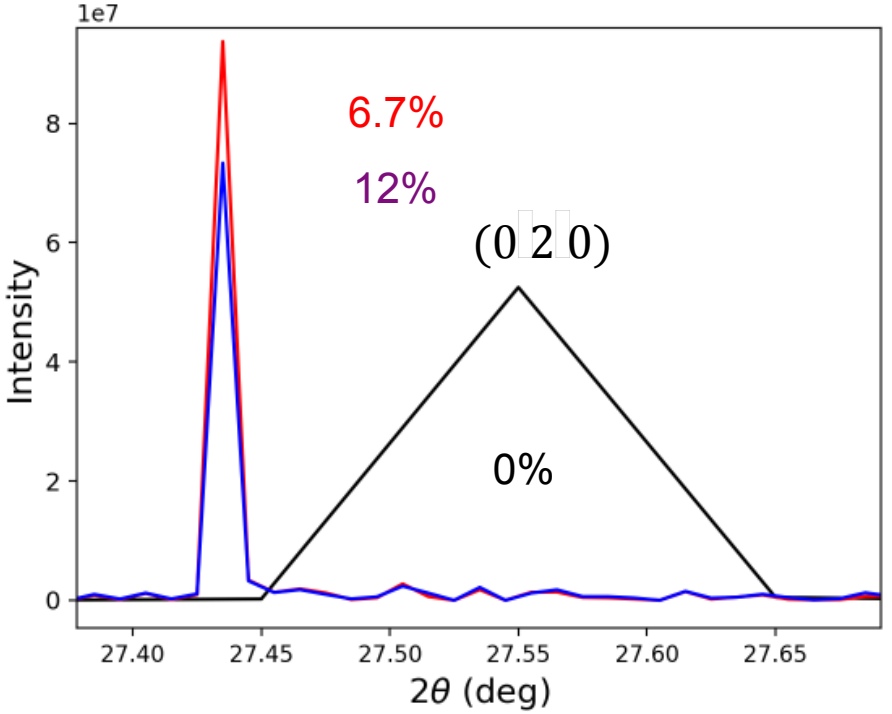
Decoding VO_x diffraction patterns



Lowest energy structure for each composition gives significantly different XRD patterns





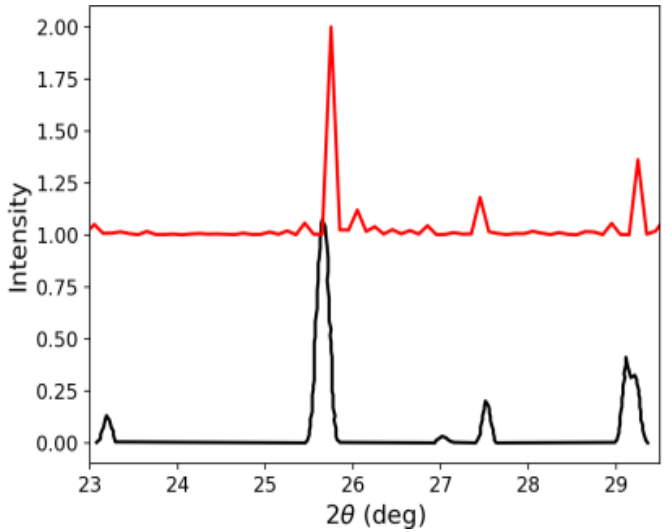


Large vacancy concentrations result in peak shift to left but also result in sharper peaks

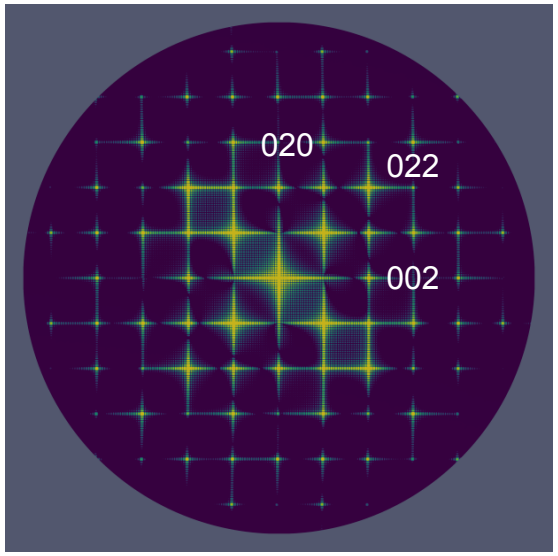
High similarity score structures do not give a consistent trend

Path forward

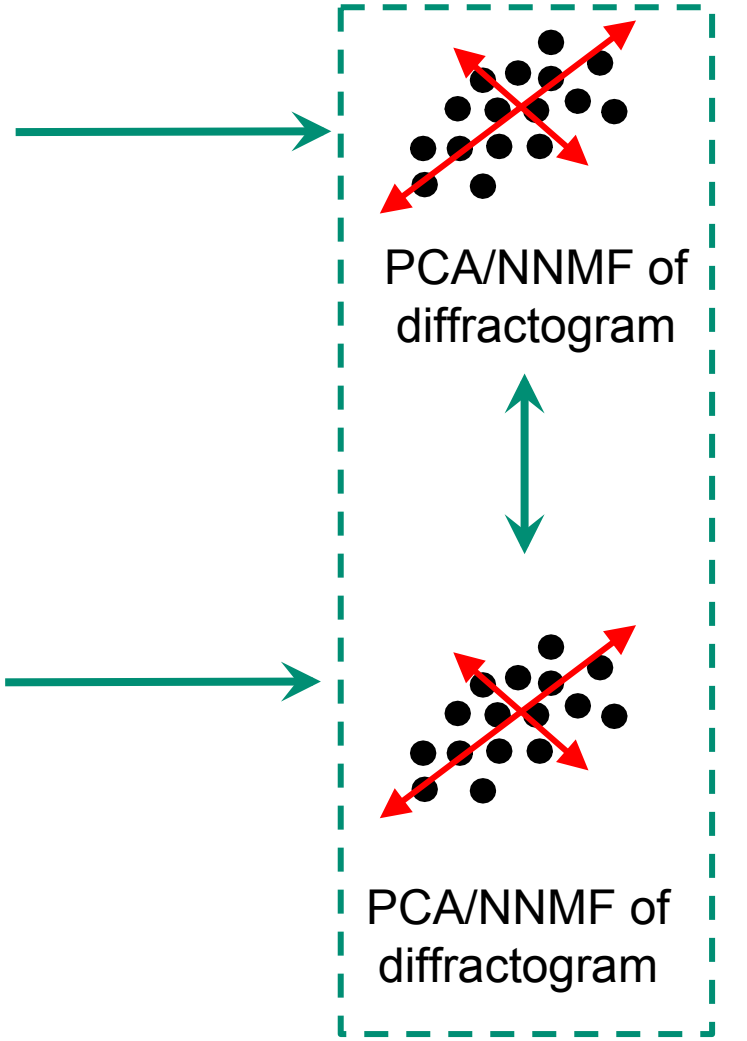
XRD



SAED

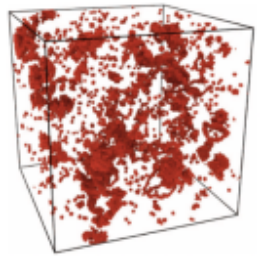


Multimodal data as inputs to ML models



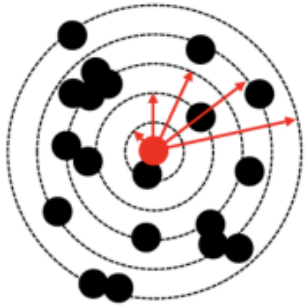
Gaussian Process Regression

Gaussian Process Regression



Point defect density

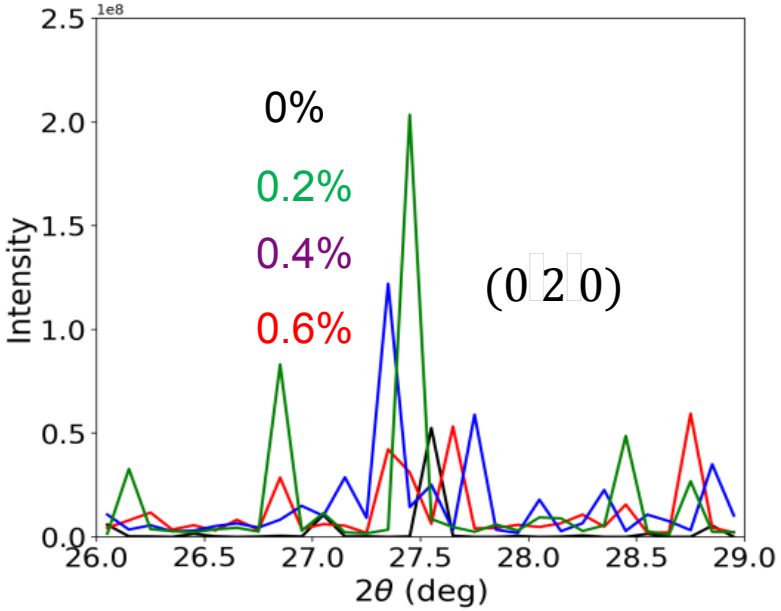
Pair correlation function

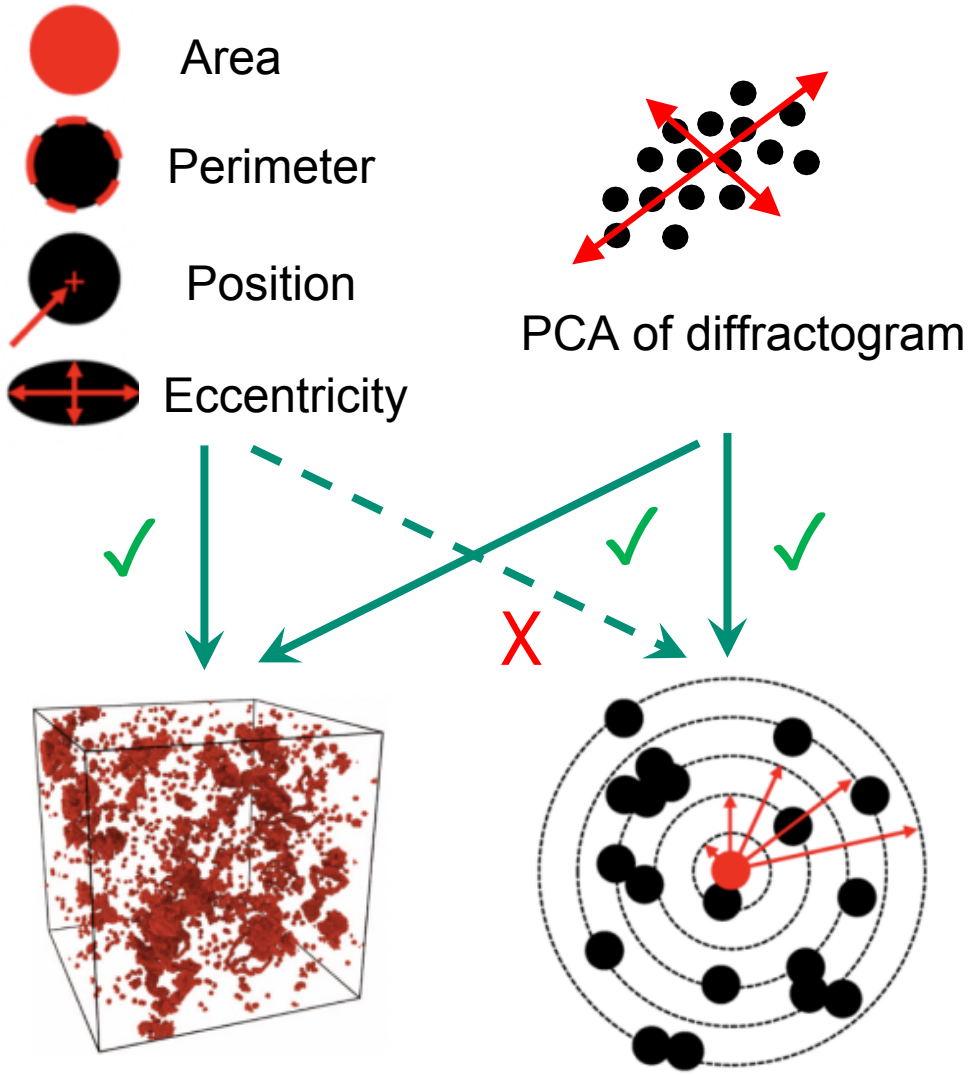


Take home message

- Correlating observable diffraction patterns to underlying vacancy statistics is critical to controlling metal to insulator transition

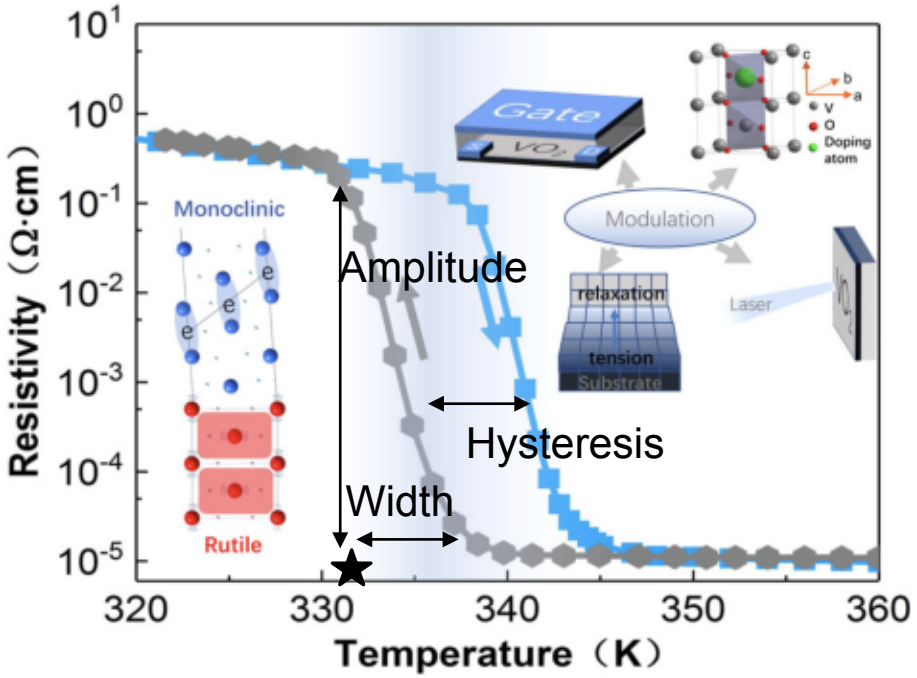
- Machine learning based approaches can extract more information from a diffractogram than human-intuition





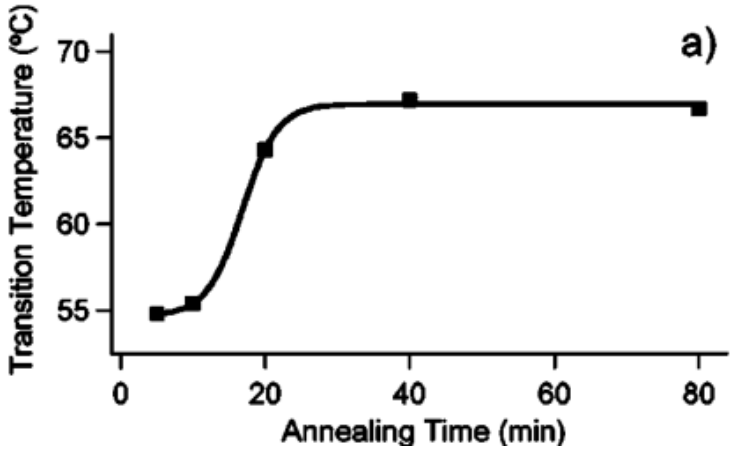
Backup slides

Controlling the metal to insulator transition in VOx

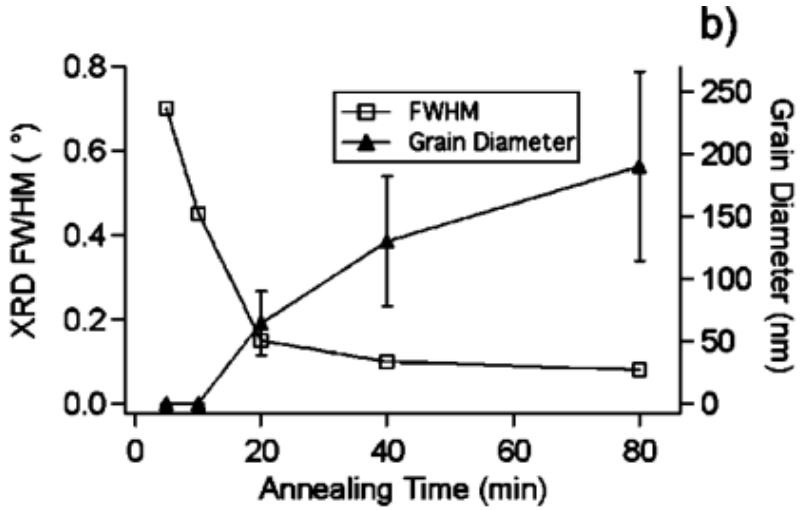


Shao et al. *NPG Asia Materials* (2018)

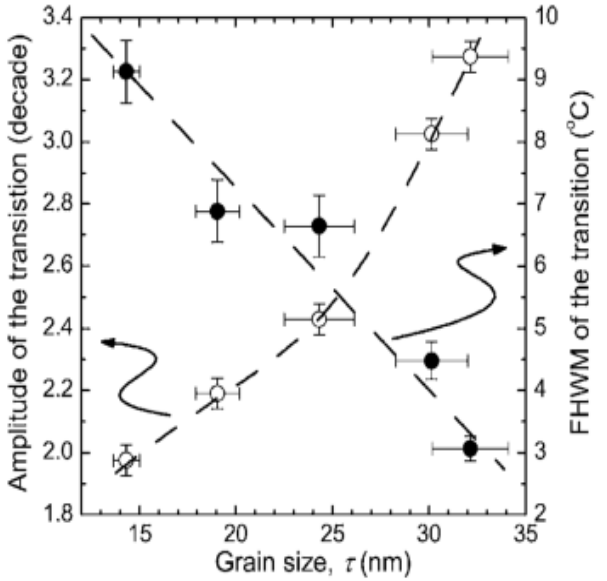
Width ∝ defect concentration
Amplitude ∝ 1/defect concentration
Hysteresis ∝ interfacial energy



Suh et al. *Journal of Applied Physics* (2004)

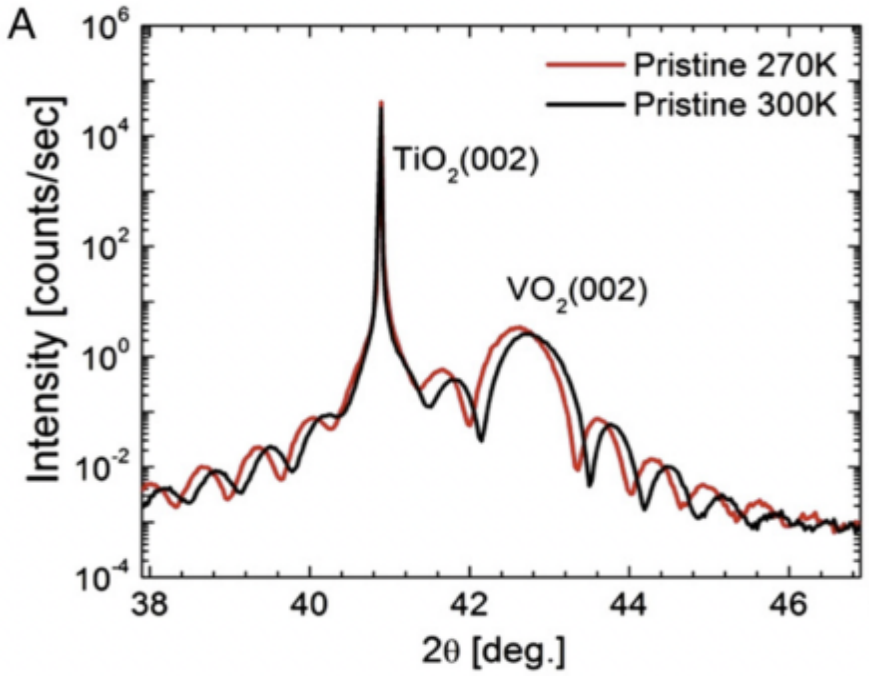


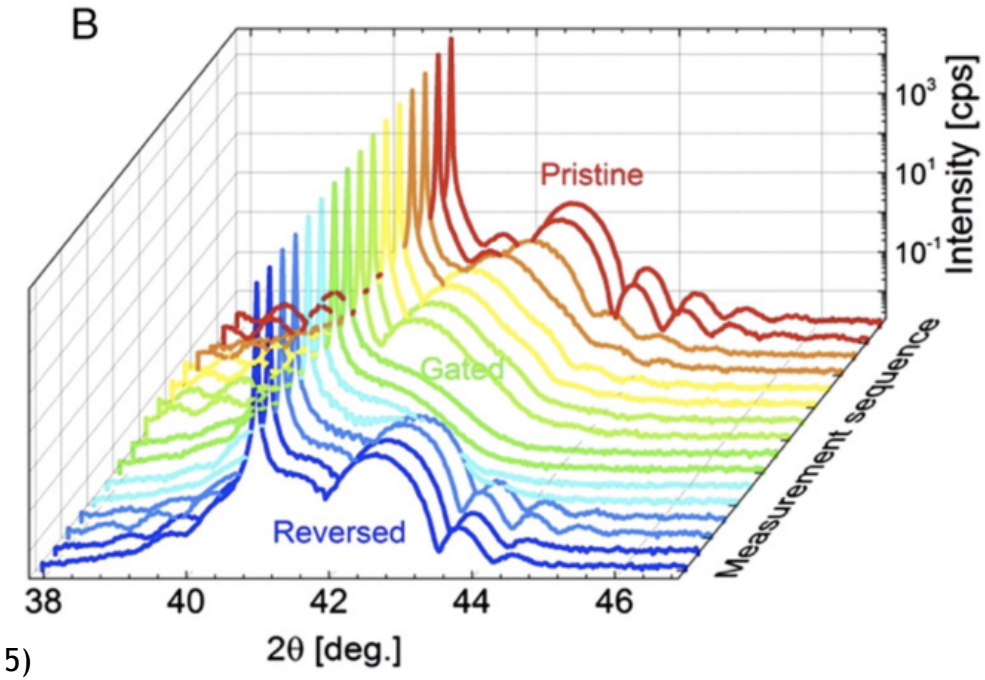
Narayan and Bhosle *Journal of Applied Physics* (2006)



Brassard et al. *Applied Physics Letters* (2005)

Vacancy induced metallization





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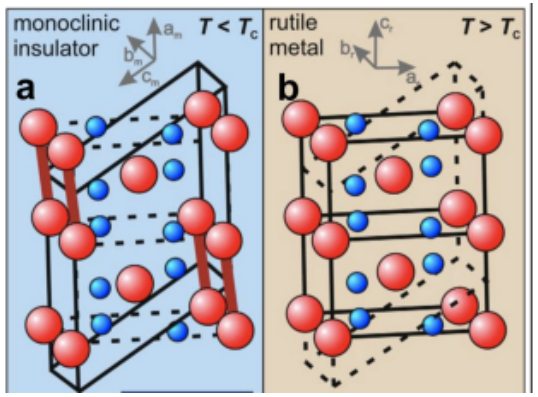

Near neighbor distances [Å]		
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V-O	1.86	1.95
V-O	2.06	
V-V(intra dimer)	2.61	2.94
V-V(inter dimer)	3.03	3.16
V-V	3.49	3.53

- (002) peak shifts right for monoclinic to rutile transformation
- (002) peak shifts left on gating
- Oxygen vacancies induce metallization in the monoclinic state while expanding lattice