Oak Ridge National Laboratory | Spallation Neutron Source

September 10, 2025 | Oak Ridge, TN

Artificial Intelligence for Scattering Experiments

Neutron Scattering School 2025

Thomas Proffen

Neutron Sciences

About me

- PhD in Crystallography from Ludwig Maximilians Univers
Munich, Germany
- Postdoc at the Australian National University in Canberra
Australia
- Postdoc at Michigan State University
- Instrument scientist at Los Alamos National Laboratory
- Diffraction Group Leader at Oak Ridge National Laboratory (SNS and HFIR)
- Director Neutron Data Analysis and Visualization at ORNL.
- Distinguished Staff Member and Director Science Initiative High Performance Computing, Modeling and Data Analytics.
- Founder of Oak Ridge Computer Science Girls.

My car

Forschungsreaktor München

NPDF at Lujan Center (LANL)

First Neutron Scattering Paper ..

Acta Cryst. (1993). **B49**, 599–604

Defect Structure and Diffuse Scattering of Zirconia Single Crystals Doped with 7 mol% CaO

BY TH. PROFFEN, R. B. NEDER AND F. FREY

Institut für Kristallographie und Mineralogie, Theresienstrasse 41, 8000 München 2, Germany

AND W. ASSMUS

Physikalisches Institut der Universität Frankfurt, Germany

(*Received* 21 *September* 1992; *accepted* 4 *January* 1993)

This layer of diffuse scattering took **several month** to collect – 180 x 120 points, ~10 min per point

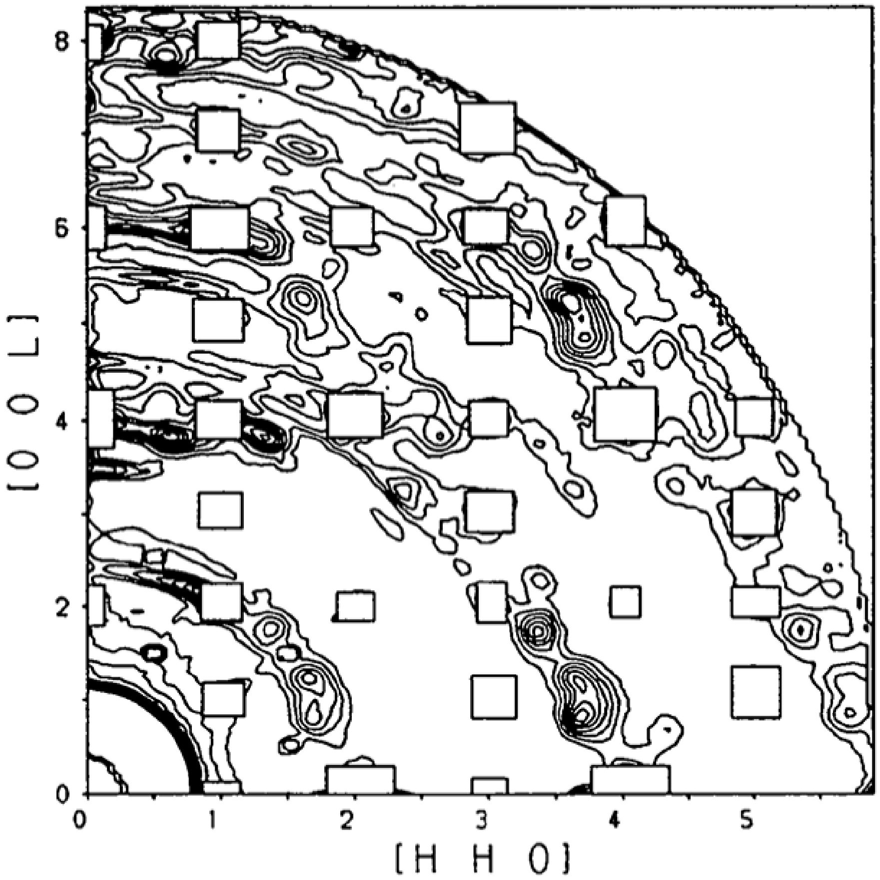
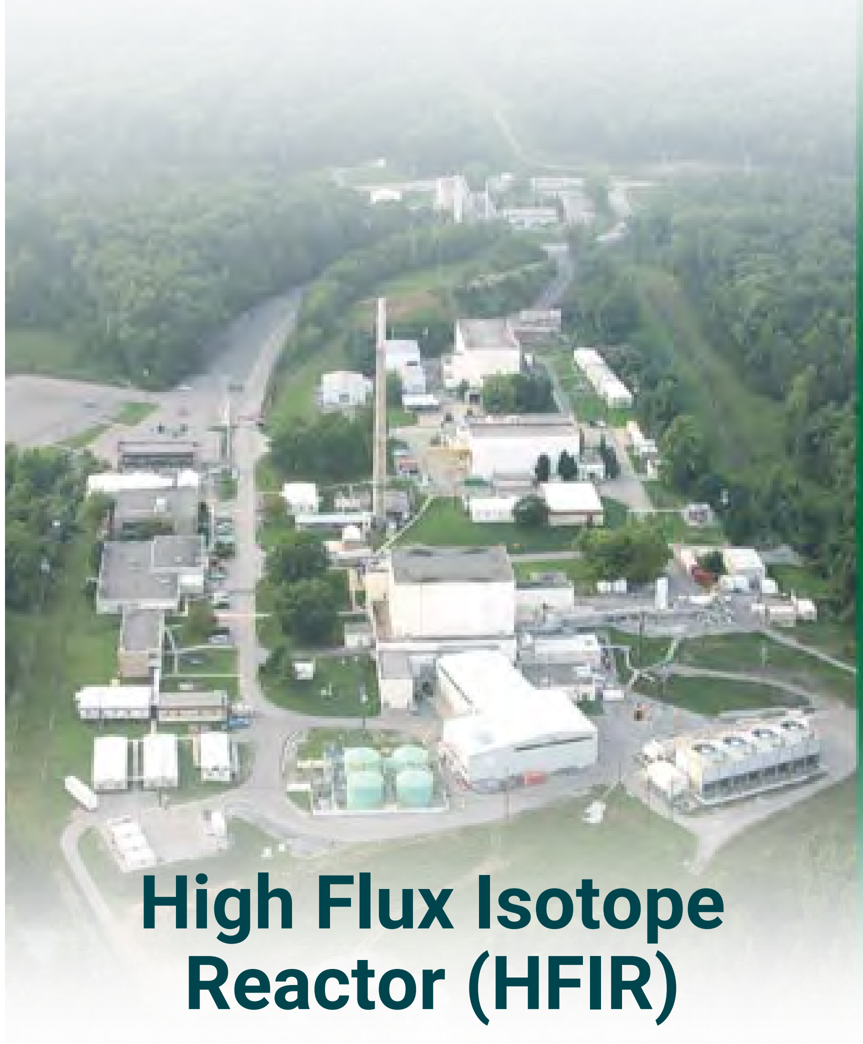
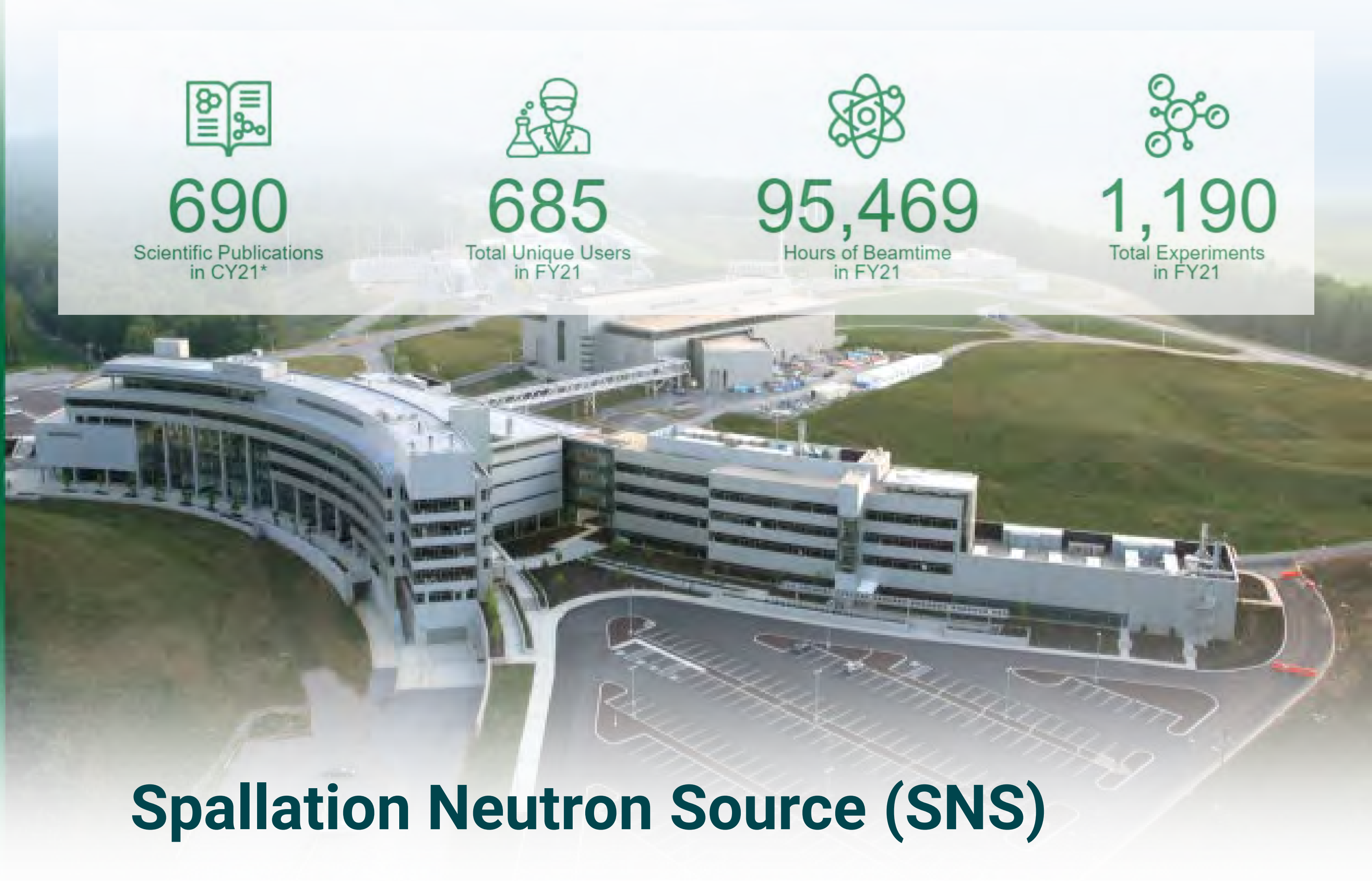


Fig. 1. Zero layer of the [1$\bar{1}$0] zone. The intensities are stepped with linear intervals of 25 counts, the lowest intensity represented is 125 counts.

ORNL is home to two world class neutron sources



High Flux Isotope Reactor (HFIR)



690	685	95,469	1,190
Scientific Publications in CY21*	Total Unique Users in FY21	Hours of Beamtime in FY21	Total Experiments in FY21

Spallation Neutron Source (SNS)

OAK RIDGE National Laboratory | SPALLATION NEUTRON SOURCE

Materials research crosses facilities

Opportunities

- Multimodal analysis
- Applied Math. concepts
- Advanced Materials Modeling

FRONTIERS IN DATA, MODELING, AND SIMULATION

Workshop Report
Argonne National Laboratory
March 30-31, 2015

Organizers:
Peter Littlewood (Argonne National Laboratory)
Thomas Proffen (Oak Ridge National Laboratory)

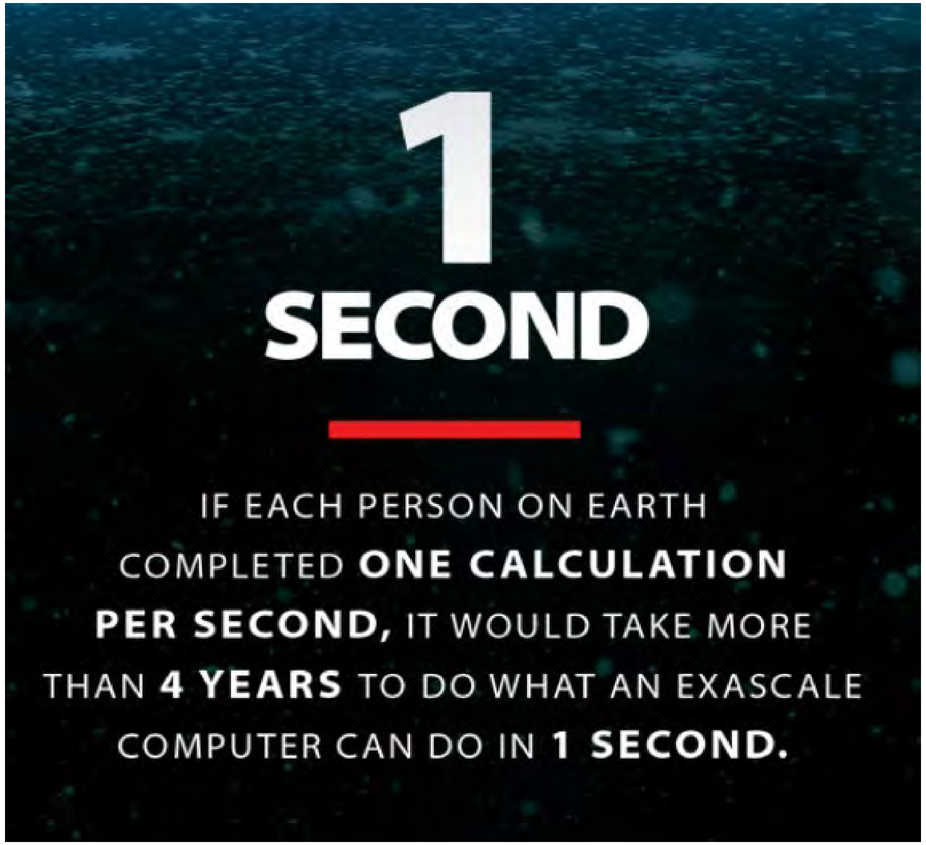
Sponsored by:
Oak Ridge National Laboratory

http://neutrons.ornl.gov/grand-challenge-workshops

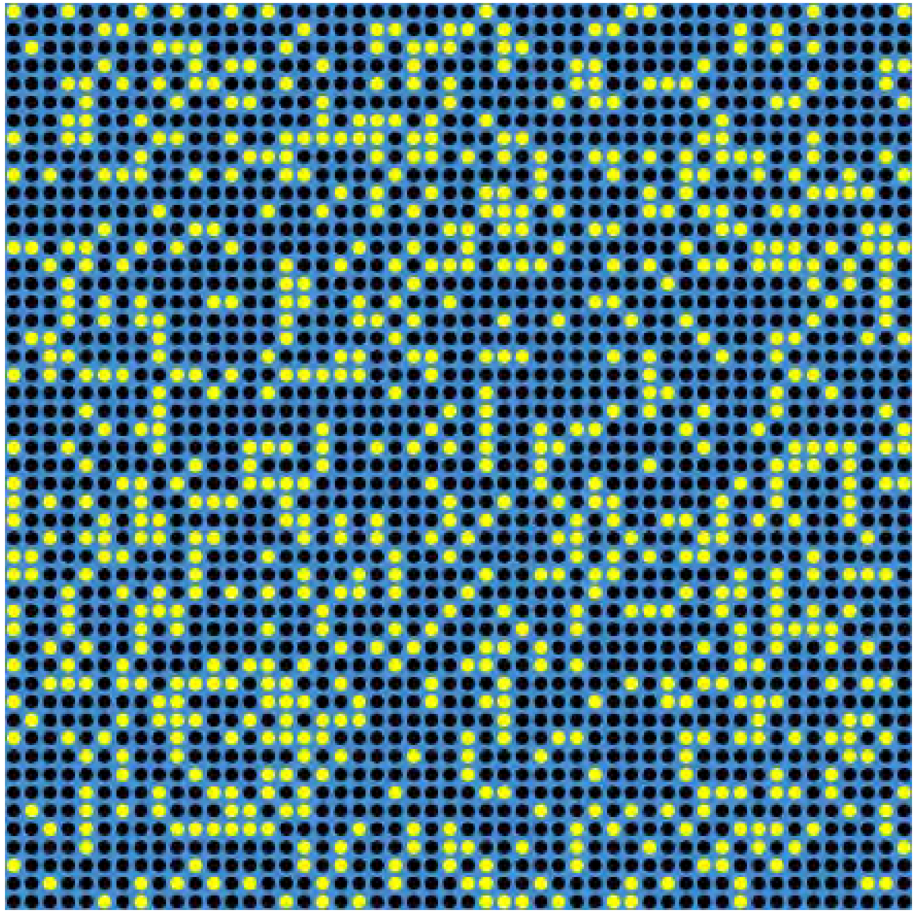
Exascale

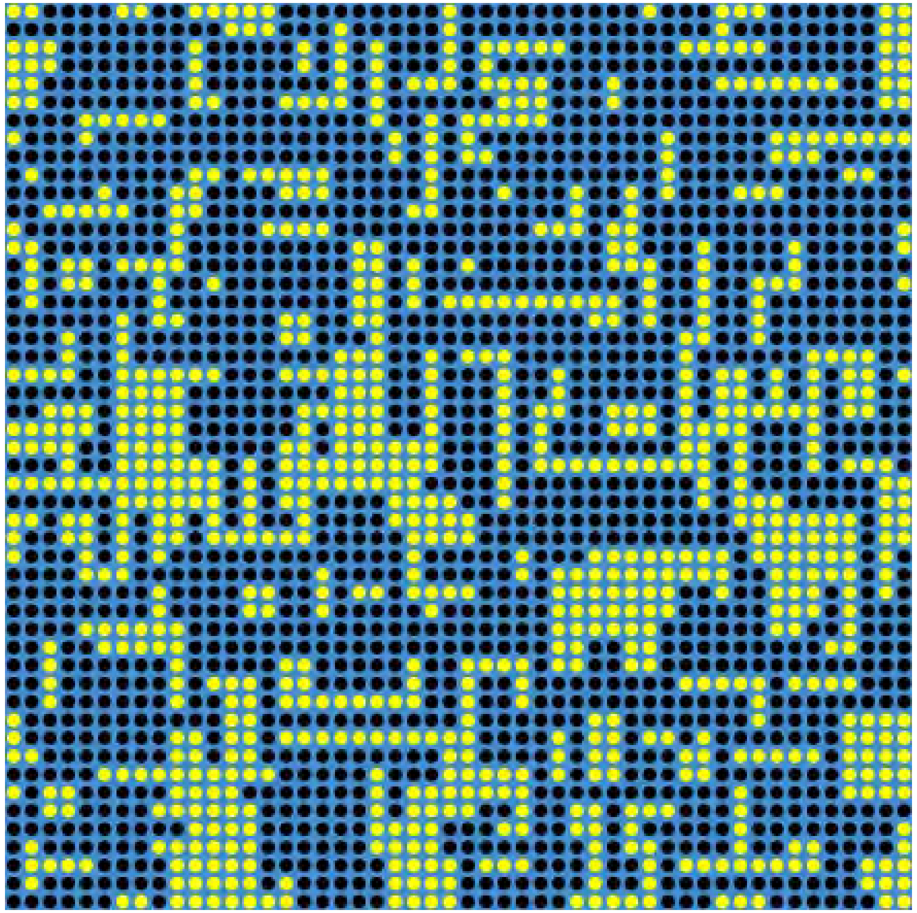

FRONTIER
FIRST TO BREAK THE
EXASCALE BARRIER AND
FASTEST COMPUTER
IN THE WORLD


FRONTIER
ENERGY
AMD


1
SECOND
IF EACH PERSON ON EARTH
COMPLETED ONE CALCULATION
PER SECOND, IT WOULD TAKE MORE
THAN 4 YEARS TO DO WHAT AN EXASCALE
COMPUTER CAN DO IN 1 SECOND.

Diffuse scattering ?

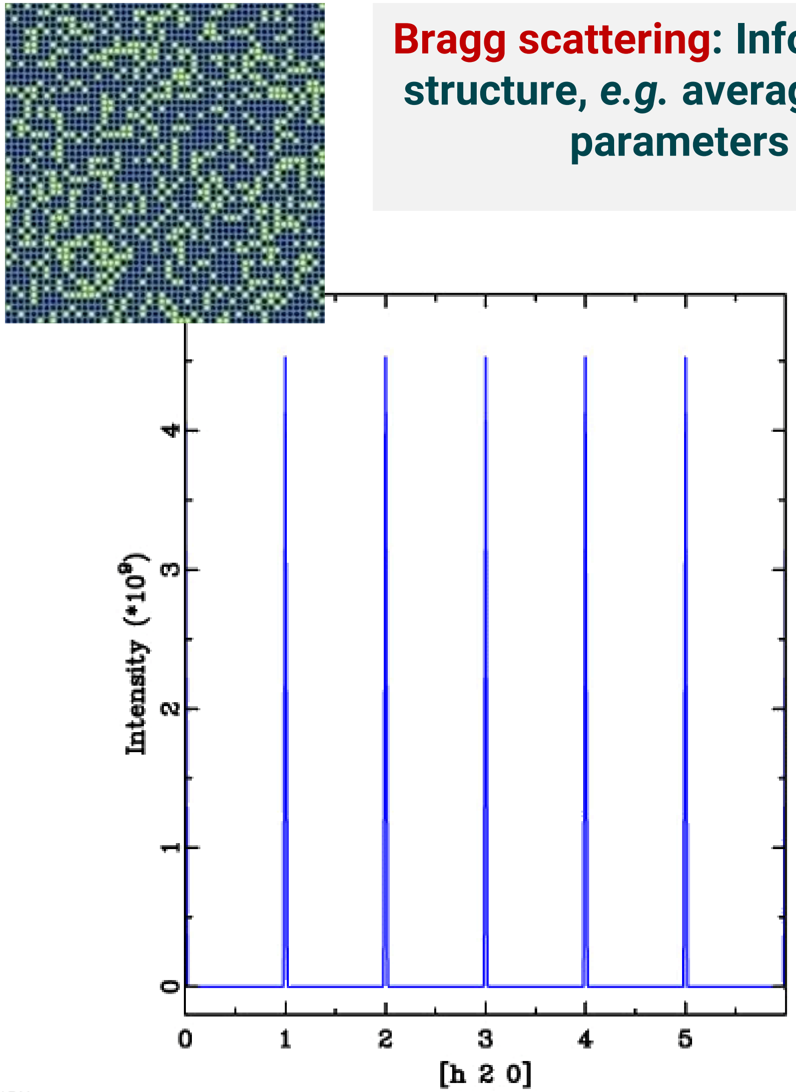


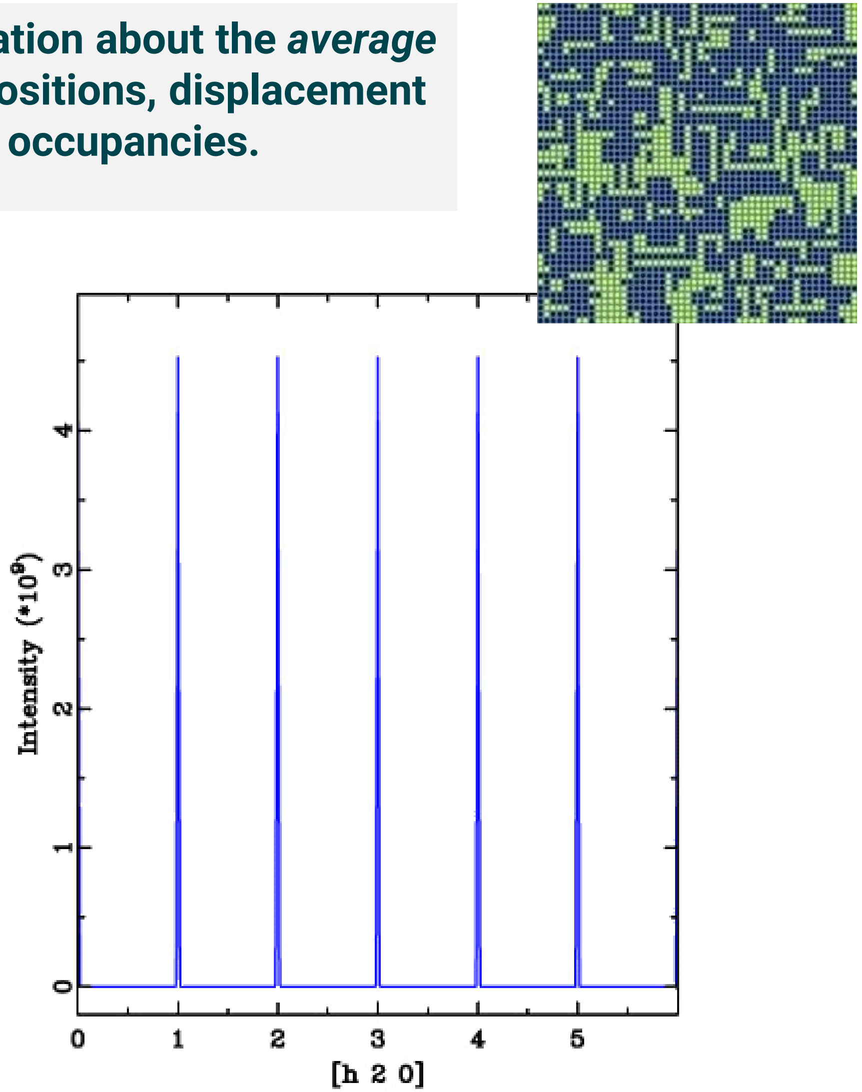


Cross section of 50x50x50 u.c. model crystal consisting of 70% black atoms and 30% *vacancies* !
Properties might depend on vacancy ordering !!

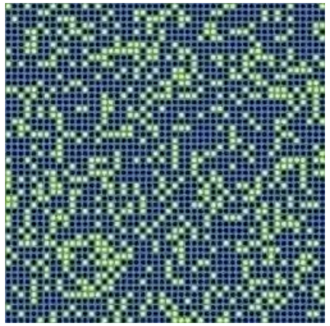
Bragg peaks are blind ..

Bragg scattering: Information about the *average* structure, *e.g.* average positions, displacement parameters and occupancies.

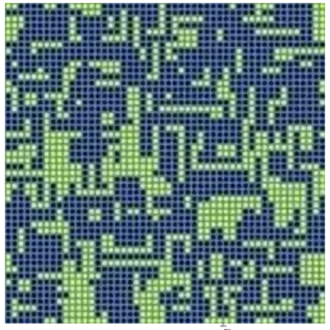


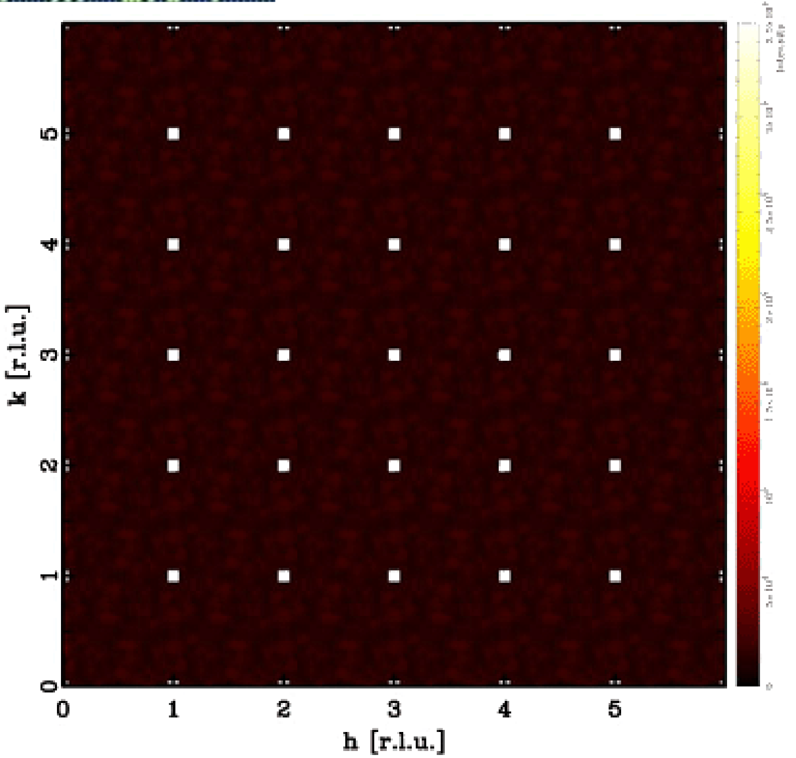


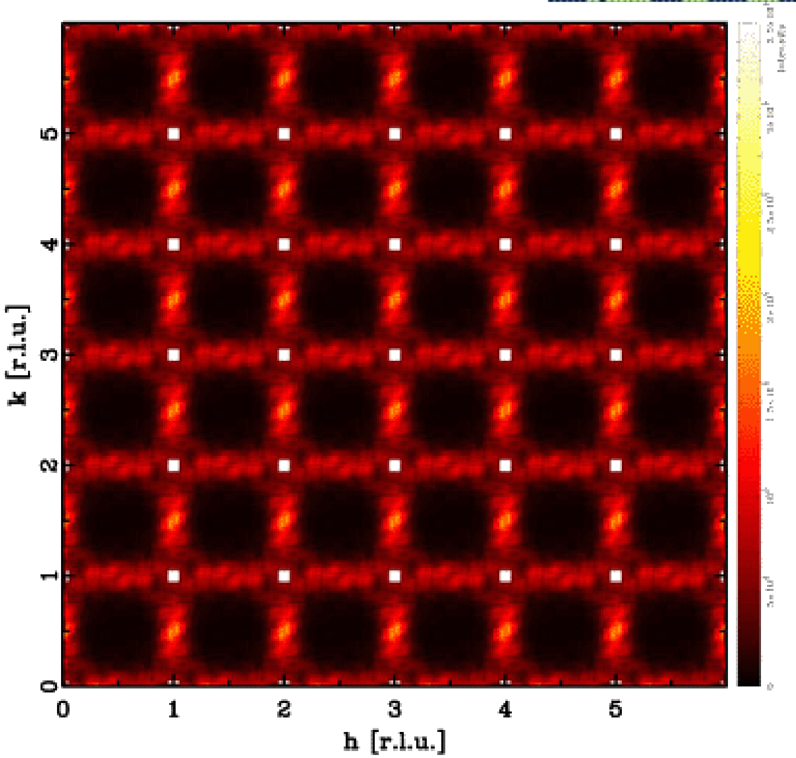
Diffuse scattering to the rescue ..



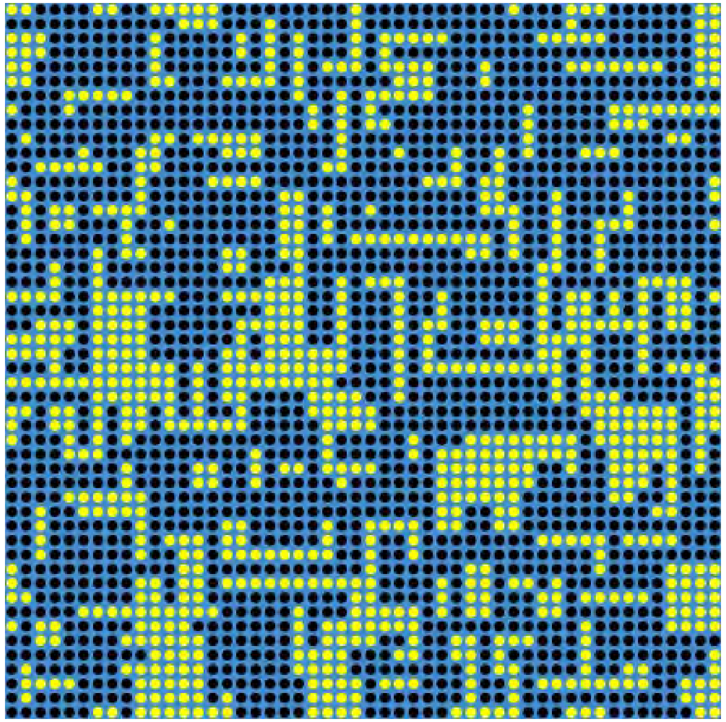
Diffuse scattering: Information about *two-body correlations*, *i.e.* chemical short-range order or local distortions.



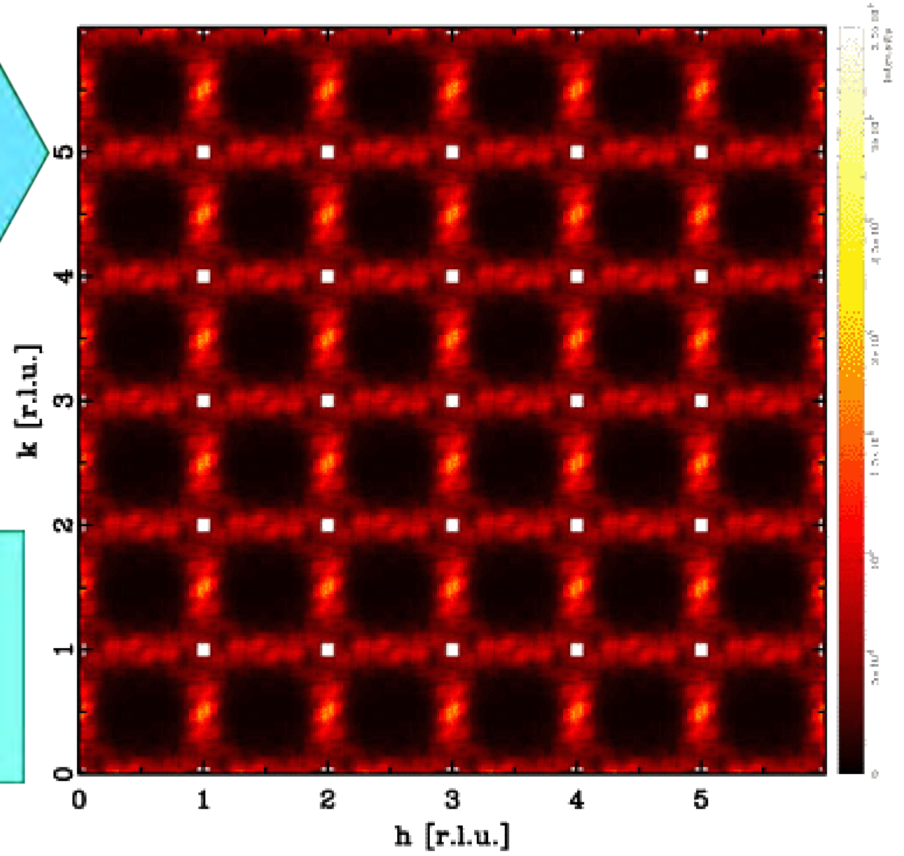




Inverse Problem *aka* Crystallographic Phase Problem



$$F(\mathbf{h}) = \sum_{i=1}^{N} f_i(\mathbf{h}) e^{2\pi i \mathbf{h}\mathbf{r}_i}$$



Intensities measured only give |**F**|and not the phase

☹

Analyzing diffuse scattering

- **Correlation approach**: Expansion of kinematic scattering equation in terms of displacement. Yields set of two-body correlations.

- **Monte Carlo based computer simulations**: Scientist might "win" solution to the problem ..

  - Minimize total energy E: AMC
  - Minimize (observed – calculated)^2: RMC

- More: "Diffuse Neutron Scattering from Crystalline Materials" by Nield and Keen, Oxford University Press

Table 1. Summary of the properties of the different components of the diffuse intensity.

Term	$\mathbf{I_0}$	$\mathbf{I_1}$	$\mathbf{I_2}$	$\mathbf{I_3}$
Description	Short-range order (SRO) term	Warren Size-effect	Huang Scattering 1st order TDS	3rd order size term
Lattice averages involved	SRO parameters α^{ij}	$\langle X^{ij}\rangle, \langle Y^{ij}\rangle$ *etc.*	$\langle (X^{ij})^2\rangle$, $\langle X^{ij}Y^{ij}\rangle$ *etc.*	$\langle (X^{ij})^3\rangle$, $\langle (X^{ij})^2 Y^{ij}\rangle$ *etc.*
Type of Summation	cosine	sine	cosine	sine
Symmetry	symmetric	anti-symmetric	symmetric	anti-symmetric
Variation in $\boldsymbol{k}$–space	nil	linear, *i.e.* with h_1, h_2 *etc.*	quadratic, *i.e.* with h_1^2, h_1h_2 *etc.*	cubic, *i.e.* with $h_1^3, h_1^2h_2$ *etc.*
Dependence on f_A, f_B for binary	$(f_A - f_B)^2$	$f_A(f_A - f_B)$, $f_B(f_A - f_B)$	f_A^2, f_Af_B, f_B^2	f_A^2, f_Af_B, f_B^2
Number of components for binary	1	6	18	30

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The Automatic Monte Carlo Method

Input:

- Observed diffuse scattering
- Starting structure (e.g. average)
- Model for disorder in terms of interaction energies for MC simulation.

Result:

- Set of interaction energies for given model that best match the data.

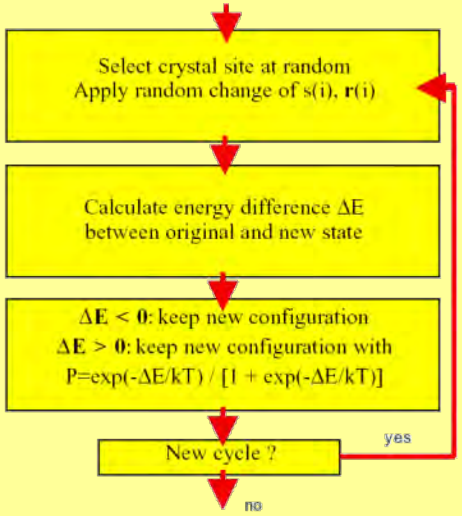
Questions:

- Finding the right model ..
- It is very slow ..

Least Squares

For each parameter in MC model

Run MC



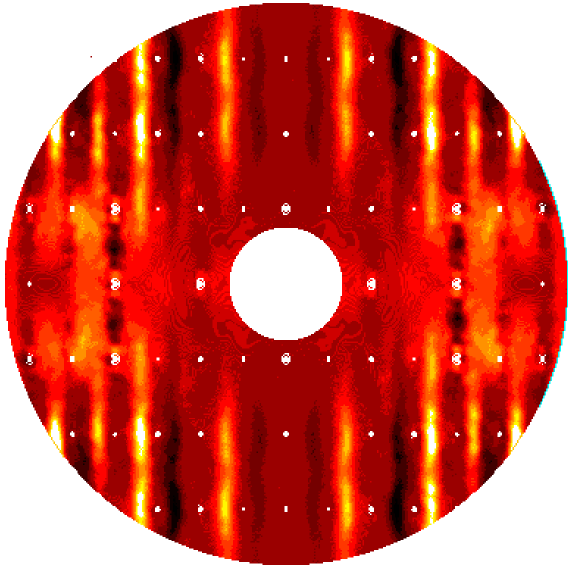
Calculate diffuse scattering

Obtain derivatives

Disorder in $Fe_3(CO)_{12}$ – AMC refinement

Numerical estimates of Differentials

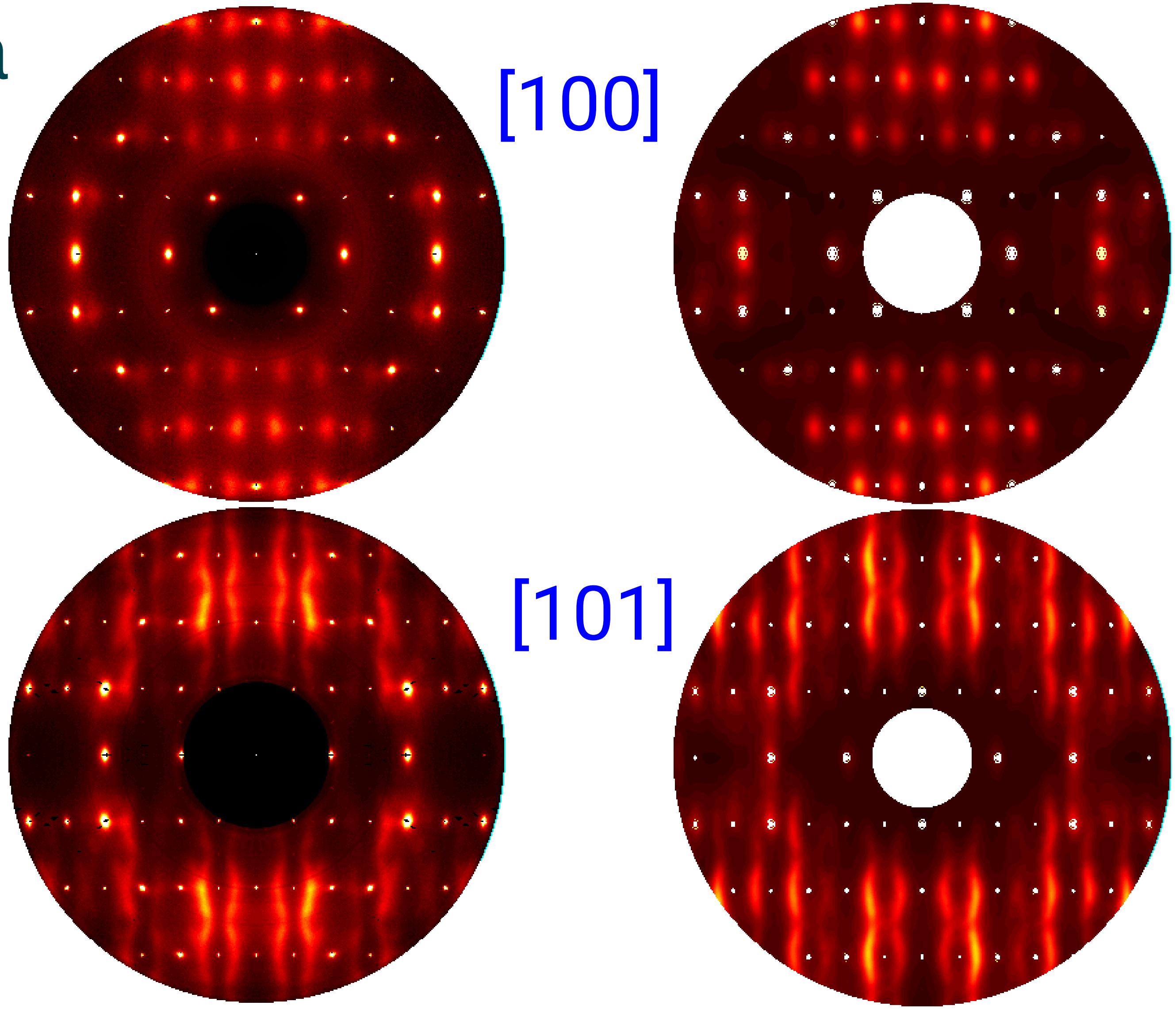
$$\frac{\partial \Delta I}{\partial p_i} = \sum_{hklm} \frac{\left(\Delta I_{p+} - \Delta I_{p-}\right)}{2\delta_i}$$



Difference between two calculated diffraction patterns

Data

calculated

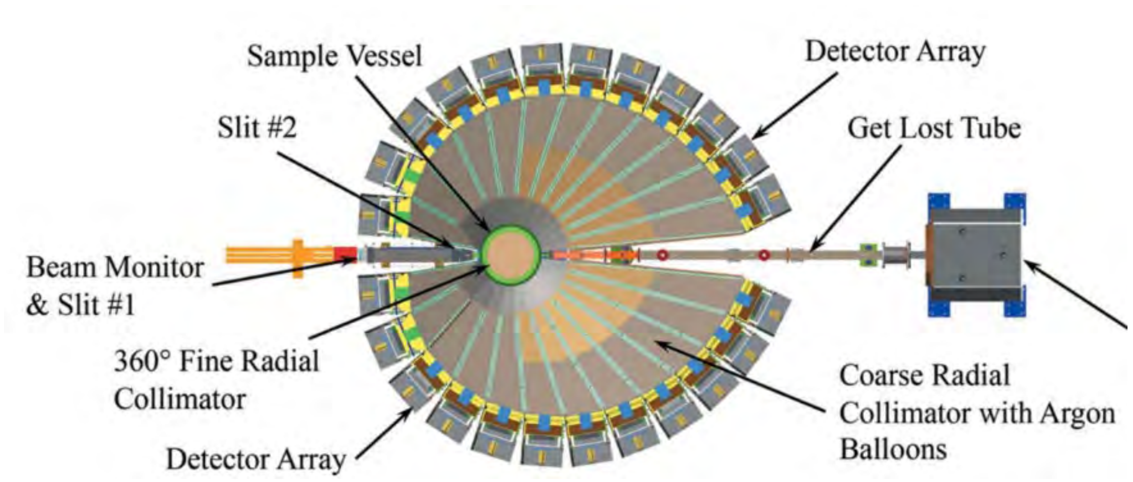


Opportunities using Machine Learning

AI is about how we use and process data. It will be, and is, transformative in knowledge-based disciplines. AI will not replace scientists, but scientists who use AI will replace those who don't.*

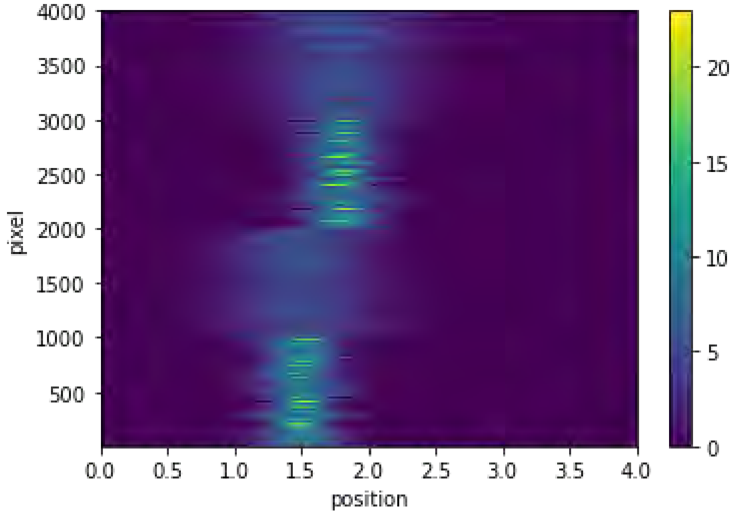
*Modified from a quote in the Microsoft report, "The Future Computed: Artificial Intelligence And Its Role In Society"

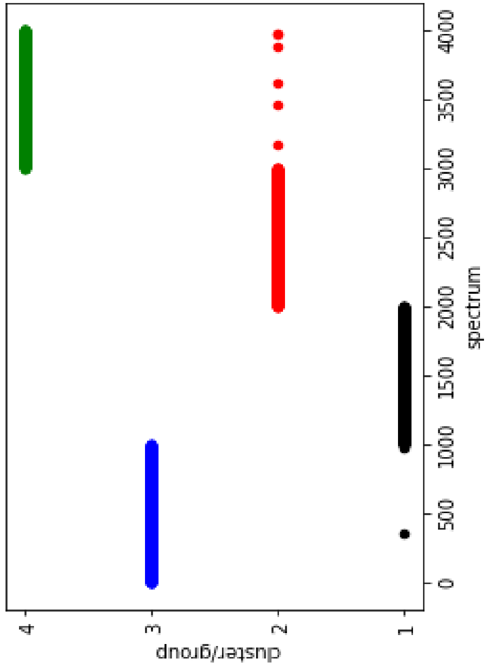
Unsupervised Machine Learning – Instrument calibration



Unsupervised clustering algorithm for Time focusing and selection of groups of detectors with 'similar' features, e.g. resolution

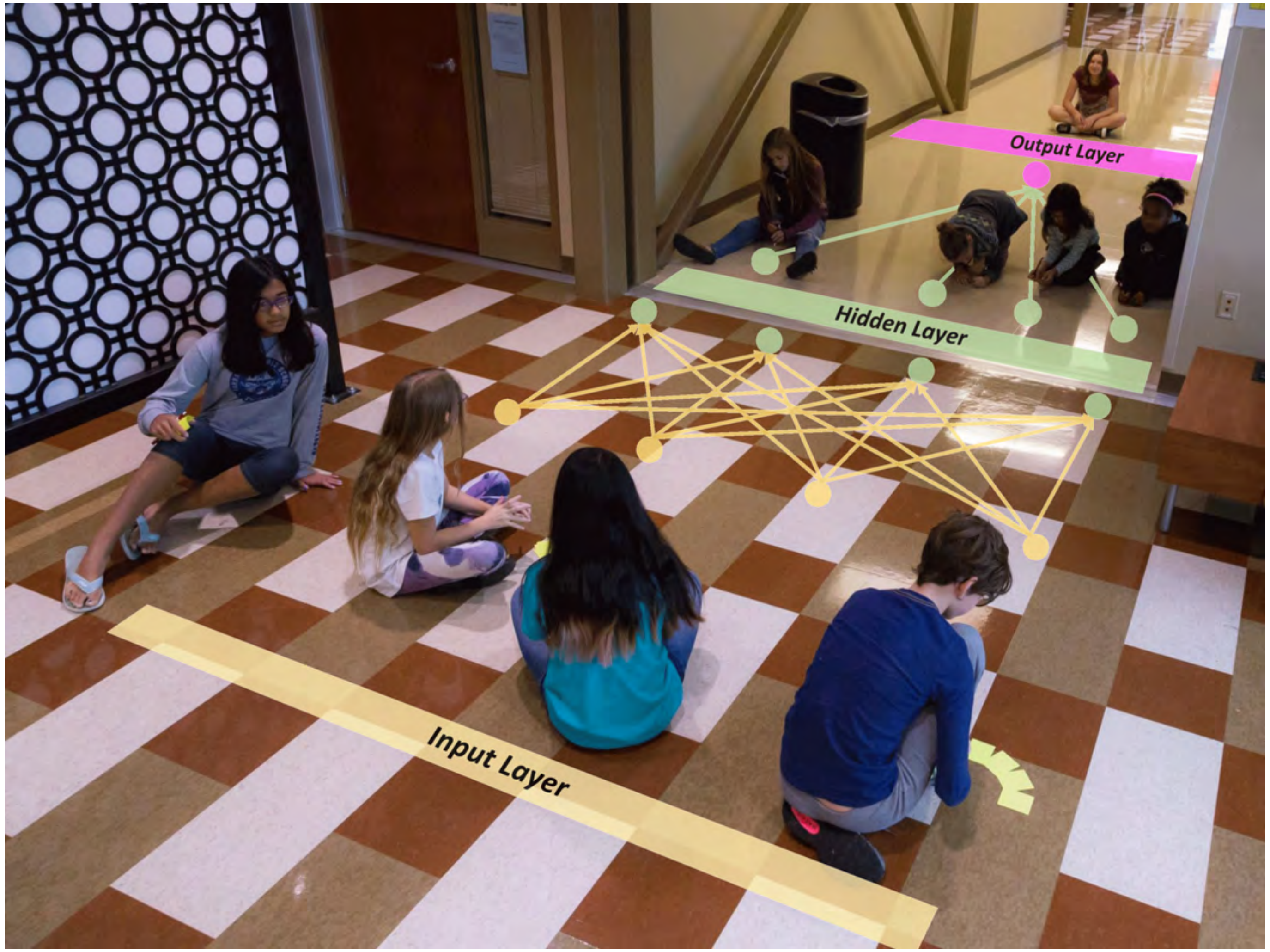
POWGEN





Work by Yuanpeng Zhang

(Supervised) Machine Learning



ORCS Girls
www.orcsgirls.org

Machine Learning

A machine learning method takes a bunch of data and "learns" from it!

Data

Label: Not a pig

Label: Pig

Label: Pig

Label: Not a pig

Machine Learning Method

Machine's rules for determining whether or not an image is a pig

Did it “learn” something?

Label: Not a pig

Label: Pig

Label: Pig

Label: Not a pig

Training Data

The data we give to the machine learning method to learn from

Label: Not a pig

Label: Pig

Testing Data

The data we hold out and use to check to see if the method actually learned something!

Deep Learning

Simulated scattering 'images'

- Small Angle Scattering
- Diffraction
- Diffuse Scattering
- Quasi Elastic Scattering

Labels

- Relate to model / parameters
- Related to topology
- Good/Bad

Training Data

The data we give to the machine learning method to learn from

Testing Data

The data we hold out and use to check to see if the method actually learned something!

Machine Learning for classification

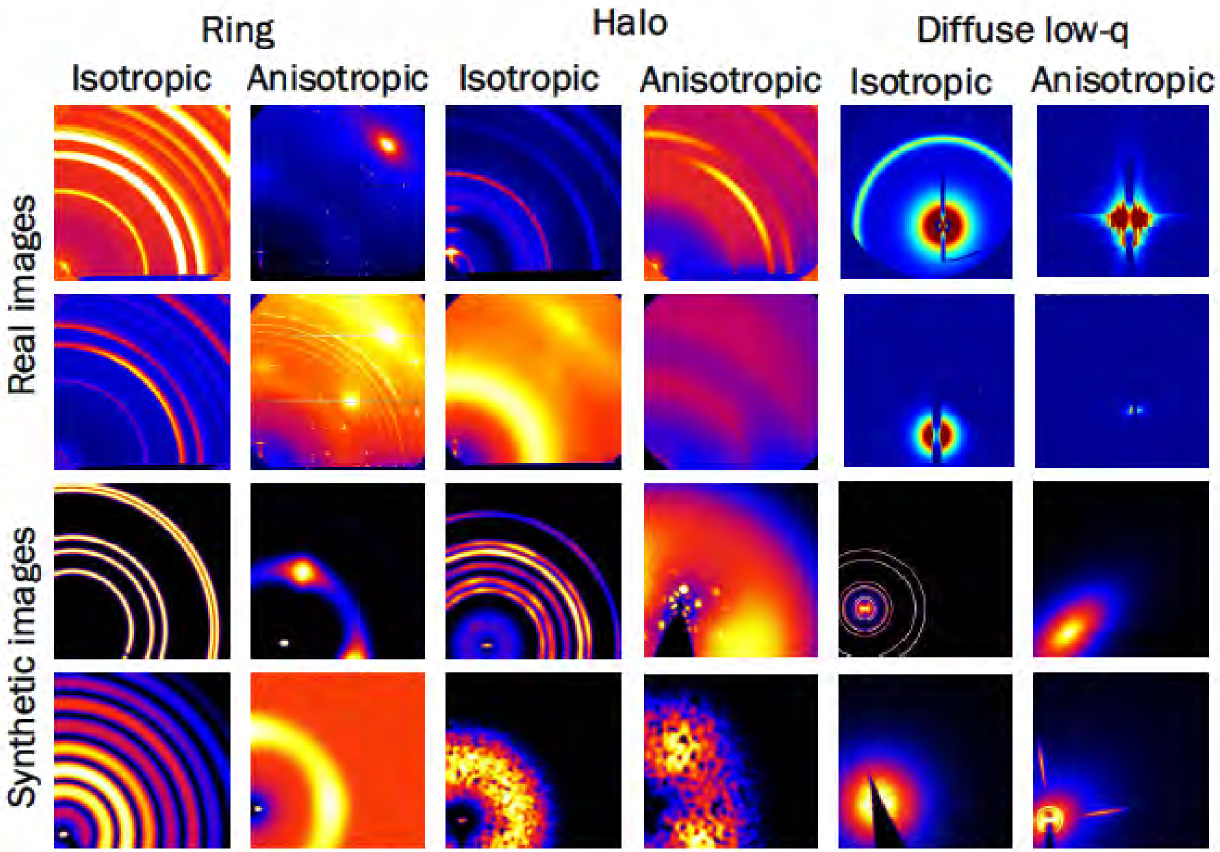


Figure 2: Comparison between synthetic images and real experimental images. The first and second rows are real experimental images, while the third and forth rows are synthetic images. Images in the same column have the same attribute. From left to right, the attributes are: Ring: Isotropic, Ring: Anisotropic, Halo: Isotropic, Halo: Anisotropic, Diffuse low q: Isotropic, and Diffuse low q: Anisotropic. Visually, synthetic and real images are indiscernible.

2017 IEEE Winter Conference on Applications of Computer Vision

X-ray Scattering Image Classification Using Deep Learning

Boyu Wang[1], Kevin Yager[2], Dantong Yu[2], and Minh Hoai[1]
[1]Stony Brook University, Stony Brook, NY, USA
{boywang, minhhoai}@cs.stonybrook.edu
[2]Brookhaven National Laboratory, Upton, NY, USA
{kyager,dtyu}@bnl.gov

XsymNet:
ML + Exhaustive Symmetry for Phase Transitions

Objectives with XsymNet

– Lower barrier for subtle or complex phase transition studies

– Identify SG, lattice parameters, and distortions modes from powder diffraction data

Exhaustive Symmetry - ISODISTORT

– Provides symmetry adapted distortion modes to model the phase transition

XsymNet Workflow

1) Generate Subgroup tree (SGT) with ISODISTORT Method 3

2) Create 250-1000 perturbations of each subgroup member by randomly choosing:

   Strain Mode Amplitudes

   » 1 to 6 modes depending on symmetry

   » Random(-0.01, 0.01)

   Displacement Mode Amplitudes

   » Gaussian(0, σ = 0.33)

   BEQ Intensity – Thermal Parameters

3) Simulate powder patterns of all perturbed structures

4) Train XsymNet to classify powder patterns by subgroup symmetry

5) Classify Experimental diffraction data

Slides thanks to Dayton G. Kizzire - kizziredg@ornl.gov

Slides thanks to Dayton G. Kizzire - kizziredg@ornl.gov

XsymNet: ML + Exhaustive Symmetry for Phase Transitions

XsymNet – Convolutional Neural Network

- Accurately classifies subgroup symmetry to powder patterns
- Automated Rietveld refinement on top 5 subgroups → scientist reviews results

Simulated Validation Data

Classification	Accuracy Metric	α phase	β phase
Subgroup (547 classes)	Top 1	89.2%	87.5%
	Top 5	99.5%	98.2%

Experimental Data – $Bi_2Sn_2O_7$

Confidence Rank	α phase	β phase
1	0176	**0152**
2	**0088**	0077
3	0236	0383
4	0544	0169
5	0183	0170

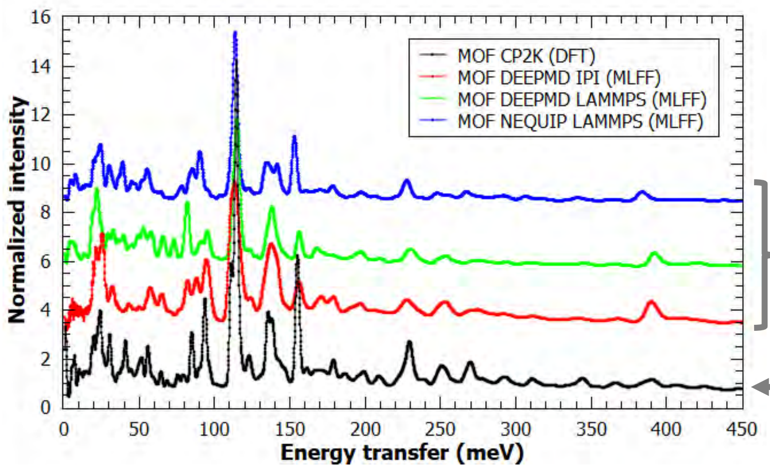
Parent Symmetry CIF → ISODISTORT Subgroup Tree → XsymNet Classification → Rietveld Refinement of Top 5 → Scientist Review

Machine learning force fields (MLFFs) for neutron scattering

Task:	DFT calculations to generate training datasets	Training of MLFFs	Atomistic modeling with MLFFs	Neutron scattering simulation	Analysis, visualization, and interpretation
Software:	VASP/CP2K/etc.	DeePMD/NequIP	LAMMPS/i-PI/etc.	OCLIMAX	Mantid/Dave/etc.
Hardware:	CADES/HPC	DGX	Analysis/PC	Analysis/PC	Analysis/PC

DeepMD: Zhang et al. Phys. Rev. Lett. 120, 143001 (2018)
NequIP: Batzner et al. https://arxiv.org/abs/2101.03164 (2021)

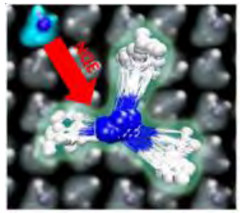
✓ Simulation of vibration and INS spectra of complex materials

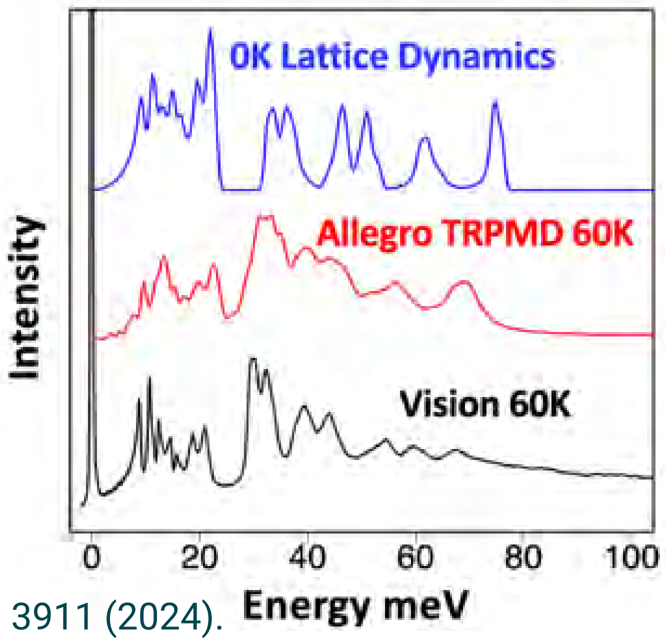


MLFF: Minutes on PC

DFT: Days on CADES

✓ Nuclear quantum effects in spectroscopy





10,000 speedup and linear scaling with size, while inheriting spectroscopic accuracy from DFT:

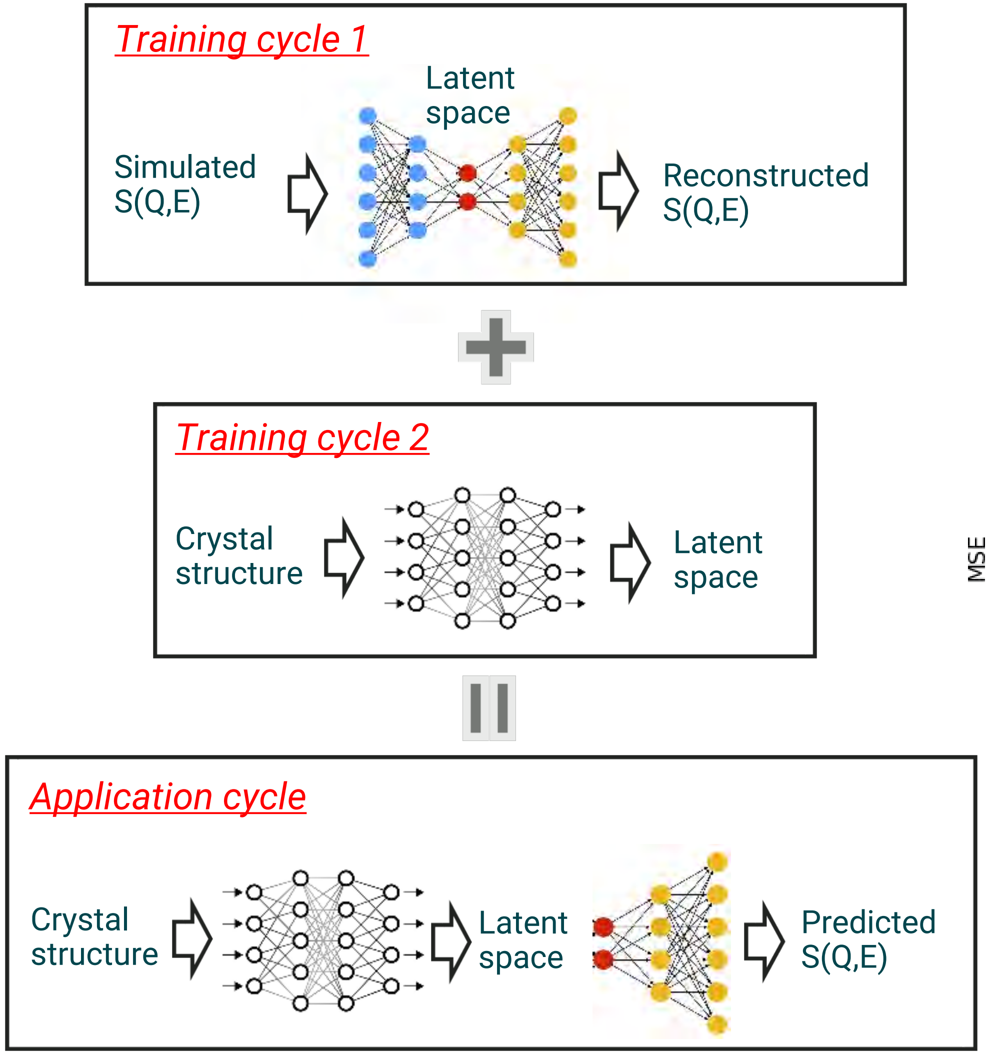
- Disordered, defective, or distorted crystals
- Heterogeneous structure (interface, boundary, guest-host systems)
- Long-range correlations
- Slow dynamics and rare events
- Nuclear quantum effects

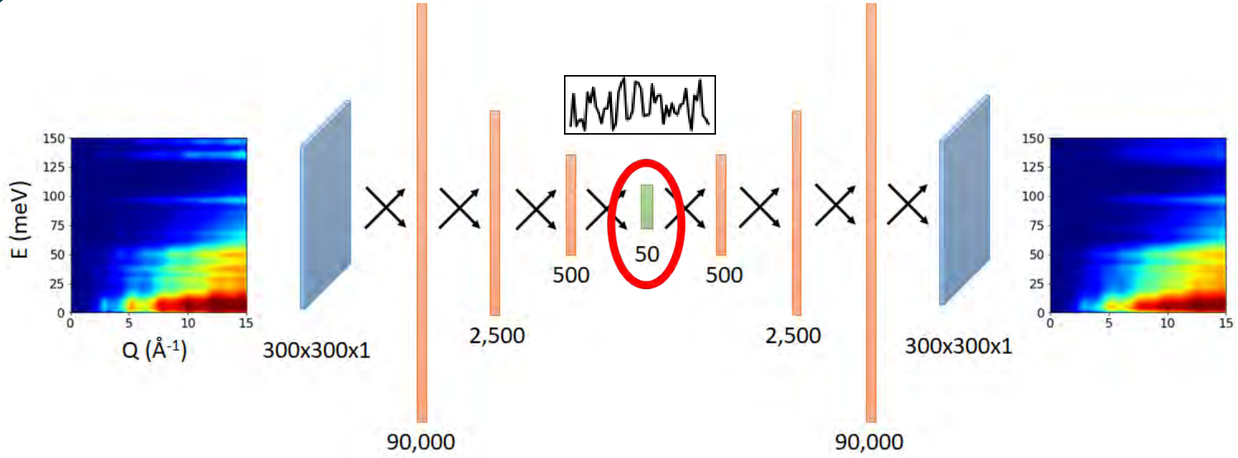
Linker, T.M. et al. Nat Commun 15, 3911 (2024).

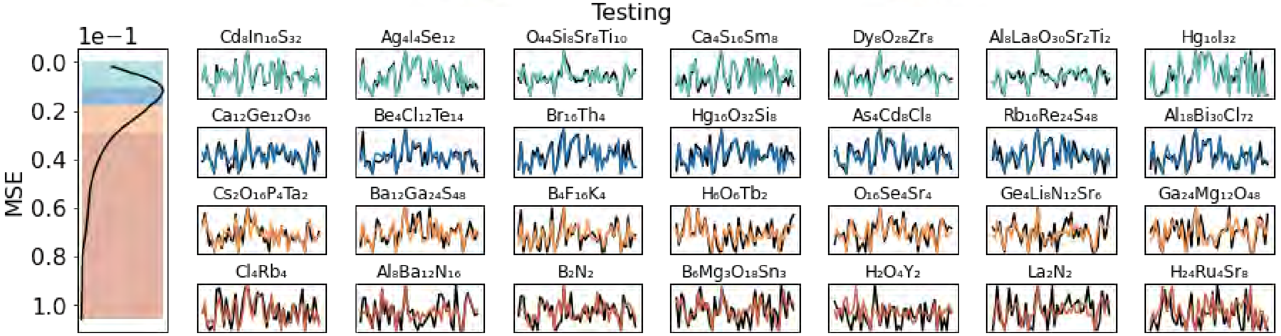
Slides thanks to Yongqiang Cheng - chengy@ornl.gov

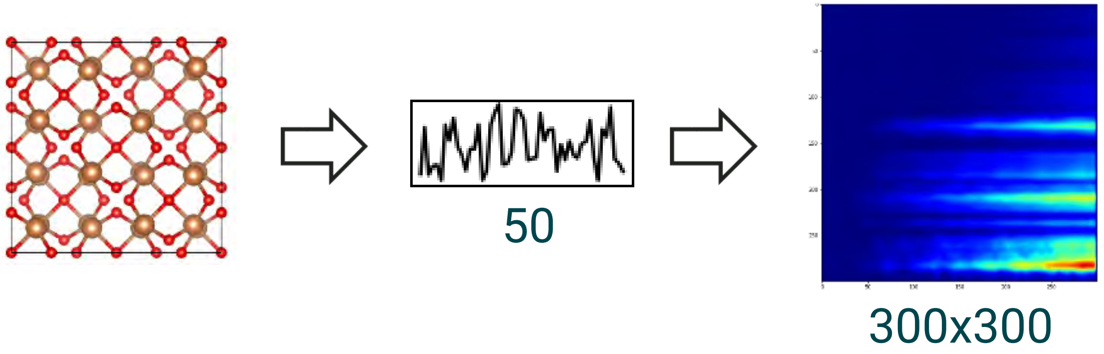
Direct prediction of powder S(Q,E)

Slides thanks to Yongqiang Cheng - chengy@ornl.gov









Cheng, Y., et al. Mach. Learn.: Sci. Technol. 4, 015010 (2023).

Experiment Steering

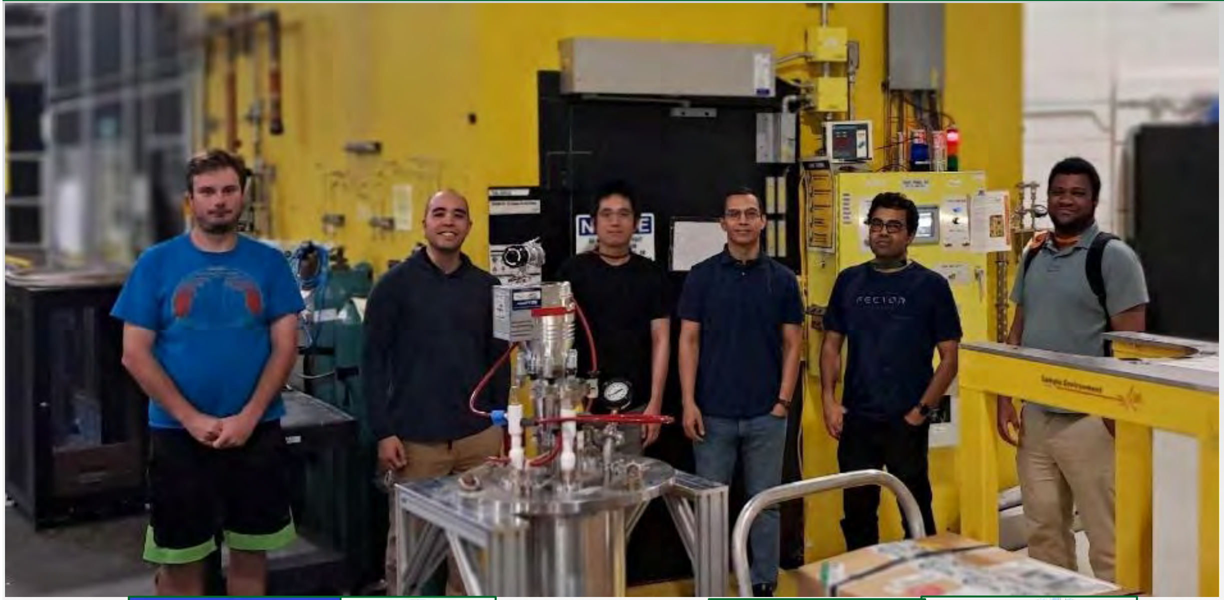
- **ESPD Team:**
  - Ray Gregory, Kaz Gofron, Bogdan Vacaliuc, Zach Thurman, Gregory Cage, Gavin Wiggins, Cody Stiner, Lance Drane, Jesse McGaha, Andrew Ayres, Robert Smith, Marshall McDonnell
- **ESPD Advisors:**
  - Greg Watson, Addi Malviya Thakur, Yuanpeng Zhang, Jue Liu
- **Other ORNL Neutrons Collaborators:**
  - Mathieu Doucet, Fahima Islam, Thomas Huegle, Sudip Seal, Maksudul Alam, Garrett Granroth, Matt Tucker, Anibal “Timmy” Ramirez Cuesta, Emily R Van Auken, Luke Daemen
- **Other ORNL INTERSECT Collaborators:**
  - Stephen DeWitt, Ankit Shrivastava, Paul Laiu, Craig Bridges
- **NIST Collaborators:**
  - Austin McDannald, Gilad Kusne, William Ratcliff
- **NSDF Collaborators:**
  - Michela Taufer, Jack Marquez, Kin Hong NG, Valerio Pascucci, Giorgio Scorzelli, Amy Gooch

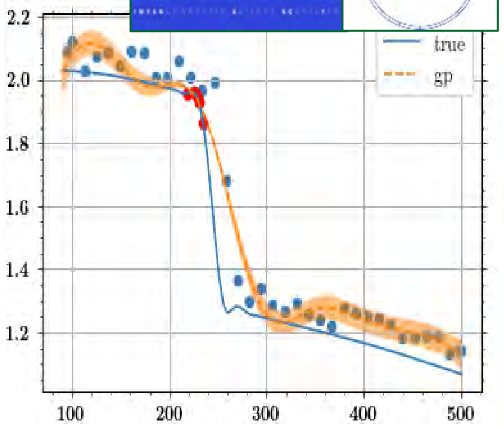
ESPD Highlight: NOMAD experiment steering for alpha-Fe2O3

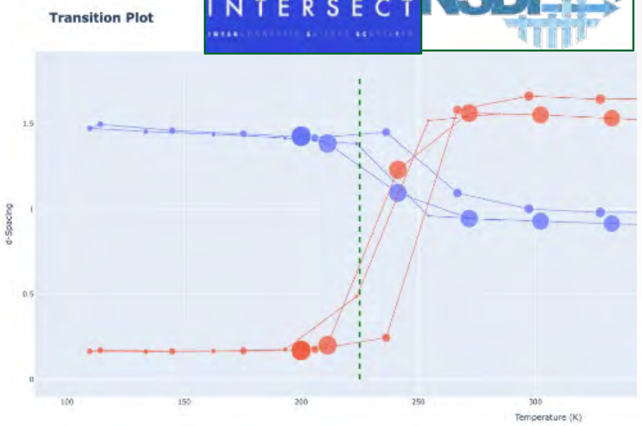
PI: Marshall McDonnell

Research Objective	Steer experiment for exploring alpha-Fe2O3 magnetic phase transition leveraging multiple autonomous science software platforms
Scientific Achievement	• Steered NOMAD experiment using External Instrument Control (EIC), Interconnected Science Ecosystem (INTERSECT), Distributed INTERSECT Active Learning for Experimental Design (DIALED), and National Science Data Fabric (NSDF) • Measured bulk Fe2O3, Fe2O3 + NIST silicon for calibrant, and 100nm 200nm Fe2O3 nanoparticles using Gaussian Process in DIALED • Measured bulk Fe2O3 using NIST team ANDiE algorithm in DIALED
Significance and Impact	Commissioned reusable experiment steering for other neutron scattering instruments across ORNL

Team @ NOMAD and Fe2O3 Phase Transition







Autonomous Phase Transition Exploration of Fe2O3 on NOMAD @ SNS

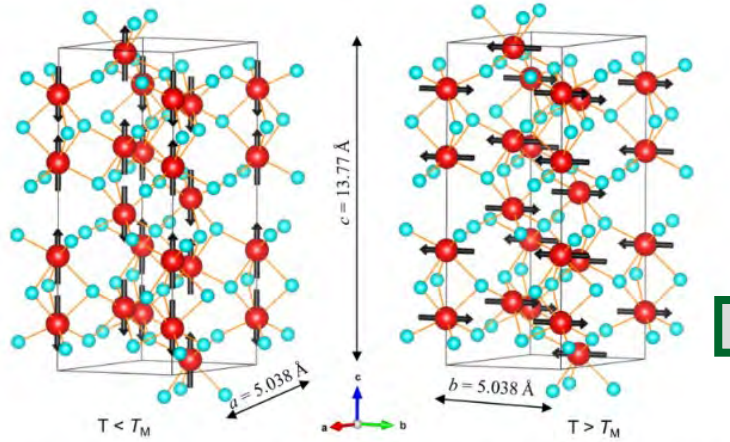
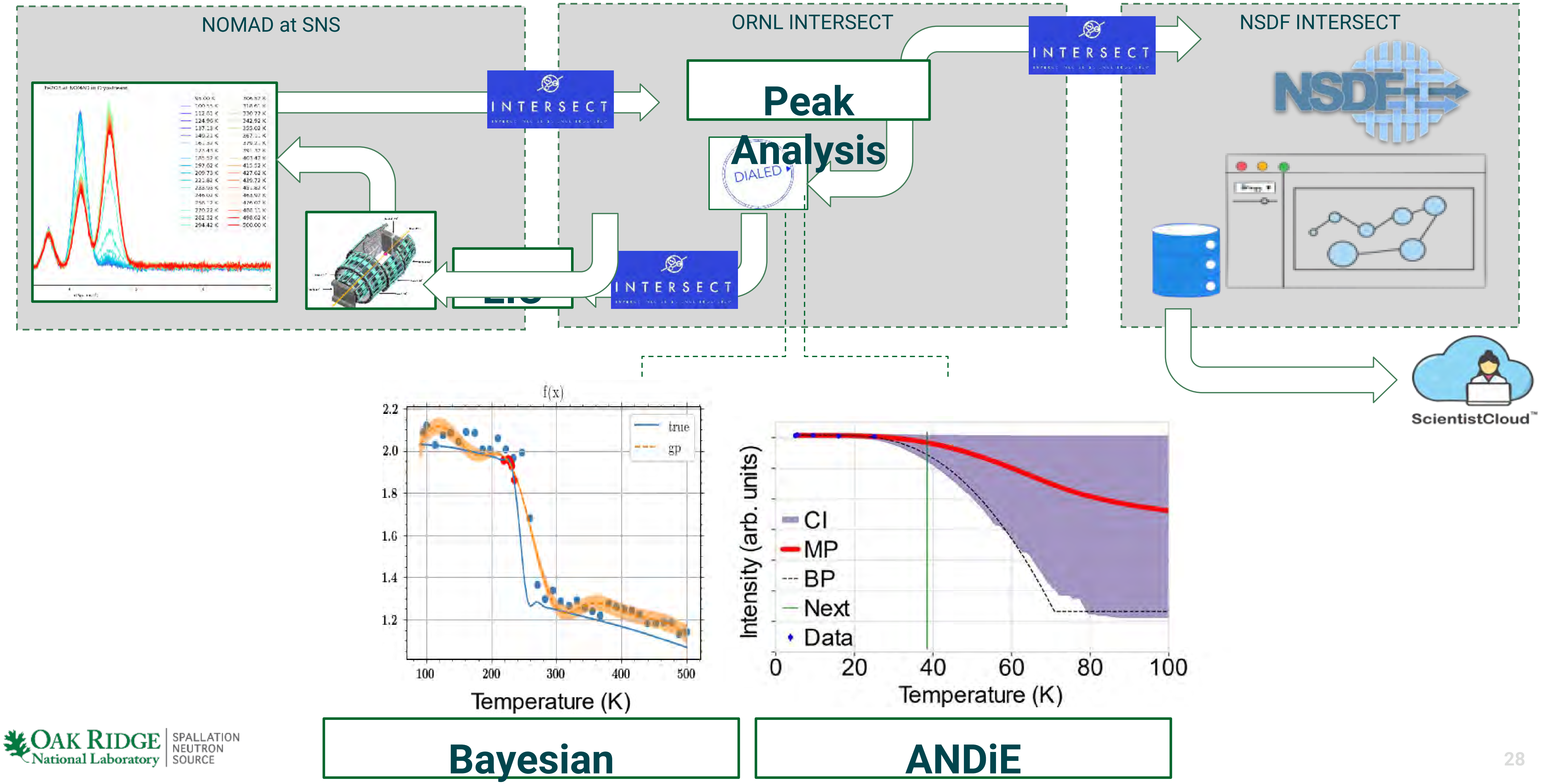


Figure 6. Crystallographic and magnetic structure of hematite below ($T < T_M$) and above ($T > T_M$) the Morin transition temperature (T_M). The unit cell is hexagonal with $a = b = 5.038$ Å and $c = 13.77$ Å (Blake et al., 1966). Spins are aligned parallel to the c axis [0 0 1] below T_M and lie in the (0 0 1) plane; spins lie in the (1 1 1) plane above T_M.

- Fe2O3
- Fe2O3 + calibrant
- Fe2O3 nano

Image from Jiang, Z., Liu, Q., Roberts, A. P., Dekkers, M. J., Barrón, V., Torrent, J., & Li, S. (2022). The magnetic and color reflectance properties of hematite: From Earth to Mars. Reviews of Geophysics, 60, e2020RG000698. https://doi.org/10.1029/2020RG000698

Recent: Autonomous Phase Transition Exploration on NOMAD @ SNS



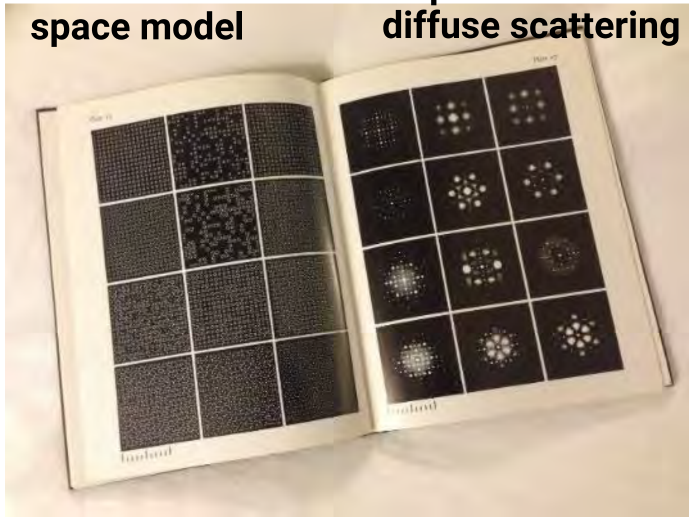
Analysis and feature detection in large volumes of diffuse x-ray and neutron scattering from complex materials

Thomas Proffen, Ray Osborn, Rick Archibald, Stuart Campbell, Ian Foster, Scott Klasky, Tashin Kurc, Dave Pugmire, Michael Reuter, Galen Shipman, Chad Steed, Chris Symons, Ross Whitfield, Doug Fuller, Guru Kora, Mike Wilde, Justin Wozniak

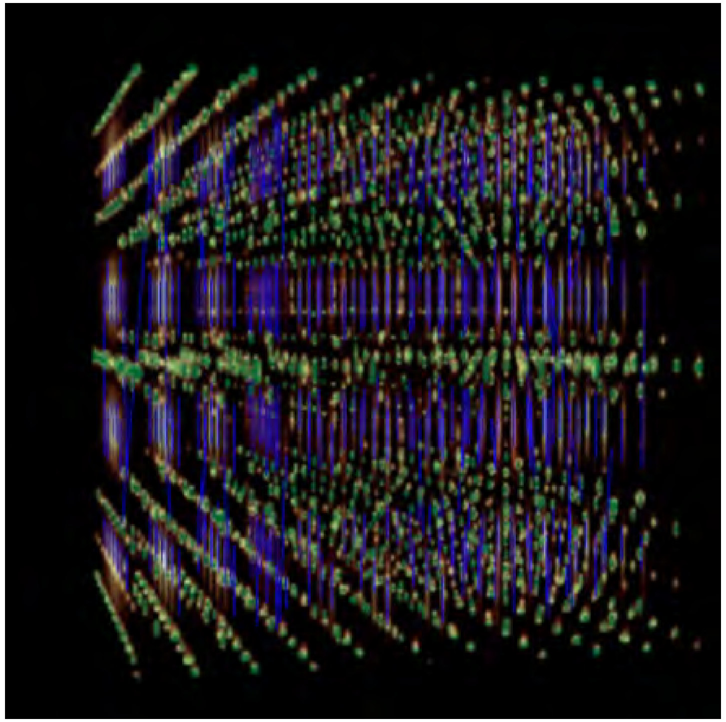
Facilities/Resources
SNS, APS, ALCF; OLCF; and CADES at ORNL

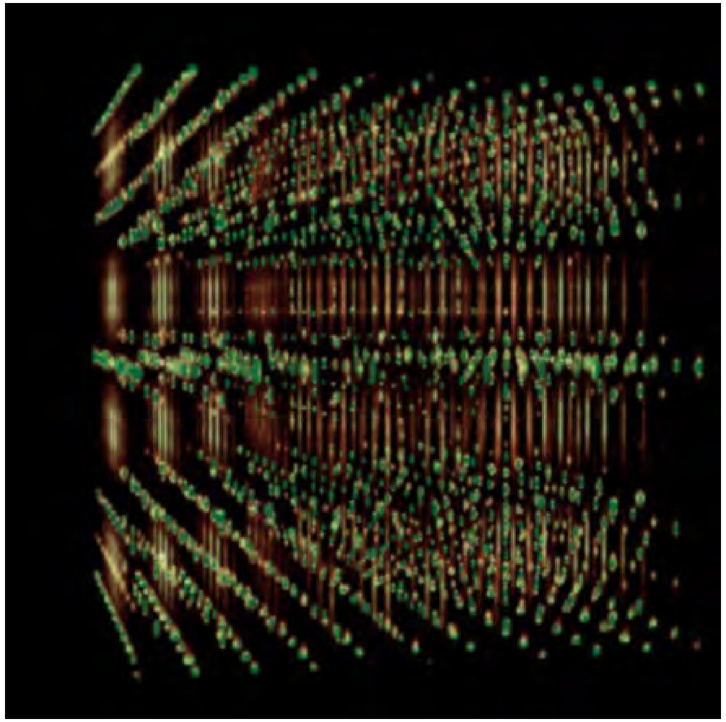
DOE Science Data Pilot Project

- **Diffuse scattering** contains information about **disorder in materials** which is critical to understand function.
- **Novel approach using pattern recognition and machine learning**.
- Aligned with science needs of CORELLI and TOPAZ.

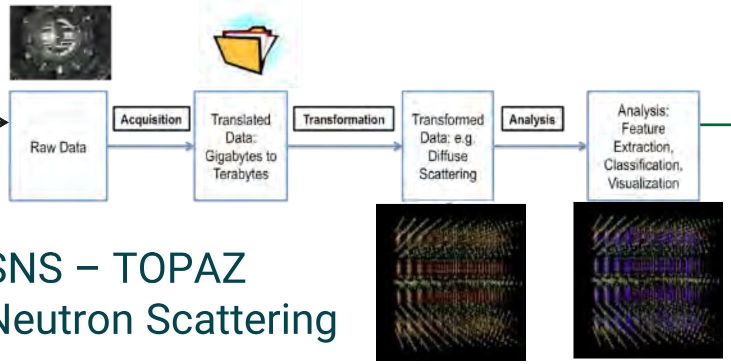


Atlas of Optical Transforms, Harburn, Taylor and Welberry (1975)



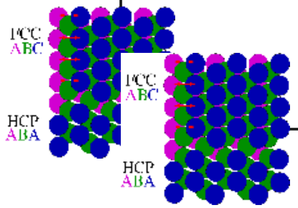


High Level Demonstration Workflow

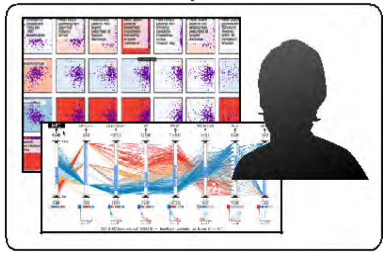


SNS – TOPAZ
Neutron Scattering

Feature detection "Streaks" in Diffuse scattering data



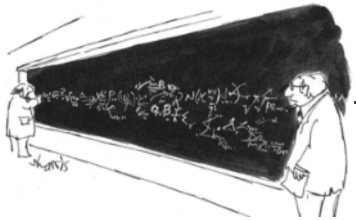
Classification of "streaks" into candidate stacking faults



Scientific visualization and visual analytics interface presenting detected features and candidate stacking faults based on classifiers

APS - X-ray Scattering coming soon

Diffuse scattering simulations over parameter space



Knowledge base of experiments and simulations

Data sharing and catalog of results

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swift

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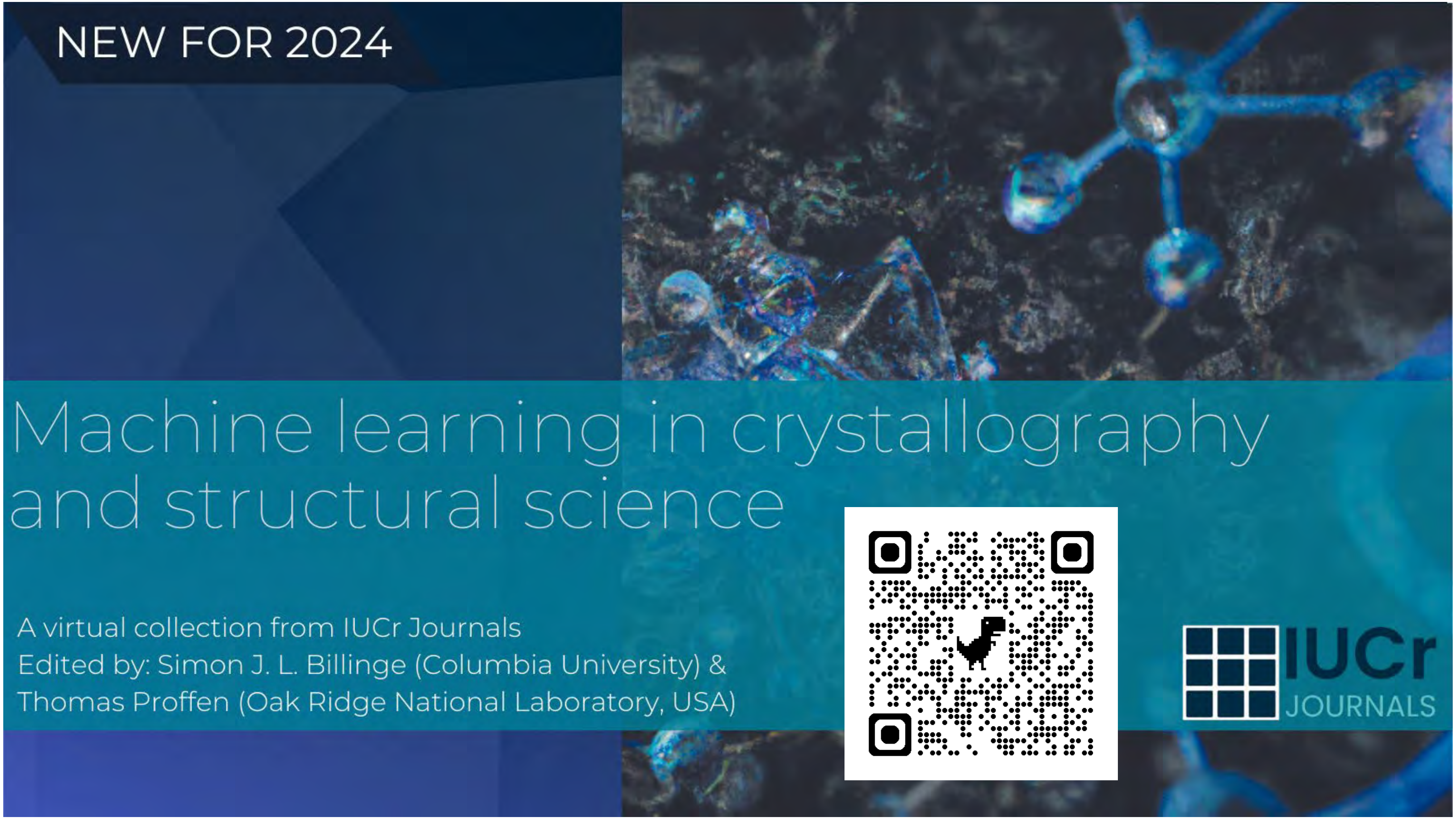
ADIOS

Challenges

- What are the correct labels?
- Sparse data.
- Data management and ‘ML friendly’ metadata.
- Correct normalization for scientific data.



https://journals.iucr.org/special_issues/2024/ML/

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Thank you

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