Artificial Intelligence for Scattering Experiments

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ORNL is managed by UT-Battelle, LLC for the US Department of Energy
ORNL is home to two world class neutron sources

High Flux Isotope Reactor (HFIR)

Spallation Neutron Source (SNS)
Materials research crosses facilities

Opportunities

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

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

\[ F(h) = \sum_{i=1}^{N} f_i(h) e^{2\pi i h r_i} \]

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 $(\text{observed} - \text{calculated})^2$: RMC


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

<table>
<thead>
<tr>
<th>Term</th>
<th>$I_0$</th>
<th>$I_1$</th>
<th>$I_2$</th>
<th>$I_3$</th>
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</thead>
<tbody>
<tr>
<td>Description</td>
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<tr>
<td>Short-range order (SRO) term</td>
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<td>Warren Size-effect</td>
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<td>Huang Scattering</td>
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<td>1st order TDS</td>
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<td>3rd order size term</td>
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<tr>
<td>Lattice averages involved</td>
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<tr>
<td>SRO parameters $\alpha^{ij}$</td>
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<tr>
<td>$\langle x^{ij} \rangle$, $\langle y^{ij} \rangle$, etc.</td>
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<tr>
<td>$\langle x^{ij} \rangle^2$, $\langle y^{ij} \rangle^2$, etc.</td>
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<tr>
<td>Type of Summation</td>
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<td>cosine</td>
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<tr>
<td>sine</td>
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<td>cosine</td>
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<td>sine</td>
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<td>Symmetry</td>
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<td>symmetric</td>
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<td>anti-symmetric</td>
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<td>symmetric</td>
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<tr>
<td>anti-symmetric</td>
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<td>Variation in \textit{k}-space</td>
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<td>nil</td>
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<tr>
<td>linear, \textit{i.e.} with $h_1$, $h_2$ etc.</td>
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<tr>
<td>quadratic, \textit{i.e.} with $h_1^2$, $h_1 h_2$ etc.</td>
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<td>cubic, \textit{i.e.} with $h_1^3$, $h_1^2 h_2$ etc.</td>
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<td>Dependence on $f_A$, $f_B$ for binary</td>
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<tr>
<td>$(f_A - f_B)^2$</td>
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<td>$f_A \left( f_A - f_B \right)$</td>
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<td>$f_B \left( f_A - f_B \right)$</td>
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<tr>
<td>$f_A^2 + f_A f_B + f_B^2$</td>
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<tr>
<td>$f_A^2 + f_A f_B + f_B^2$</td>
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<td>$f_A^2 + f_A f_B + f_B^2$</td>
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<tr>
<td>Number of components for binary</td>
<td>1</td>
<td>6</td>
<td>18</td>
<td>30</td>
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</table>
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 ..

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**Least Squares**

For each parameter in MC model:
- Run MC
- Select crystal site at random
  - Apply random change of $s(i), r(i)$
- Calculate energy difference $\Delta E$
  - between original and new state
  - if $\Delta E < 0$ keep new configuration
  - if $\Delta E > 0$ keep new configuration with
    $P_{\text{new}} = \exp(-\Delta E/kT) / [1 + \exp(-\Delta E/kT)]$
- Calculate diffuse scattering
- Obtain derivatives
- New cycle?
Disorder in $\text{Fe}_3(\text{CO})_{12}$ – AMC refinement

Numerical estimates of Differentials

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

Data

[100] calculated

Difference between two calculated diffraction patterns

[101]
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”
MACHINE LEARNING
A machine learning method takes a bunch of data and “learns” from it!
DID IT “LEARN” SOMETHING?

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

Label: Not a pig
Label: Pig

Label: Pig
Label: Not a pig

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

Label: Not a pig
Label: Pig
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.
## AI accelerating neutron scattering research

### Automatic model selection for neutron reflectivity
- Prototype allows to automatically detect and refine multi-layer models from experimental neutron reflectivity data.
- Training data set was calculated using **refl1d** for 1, 2- and 3-layer models.
- Future:
  - Expand to more models and deploy for users.
  - Integrate in automatic reduction and (initial) analysis workflow.

### Machine learning insight into spin ice
- Model Hamiltonians for spin ice were selected from experimental neutron scattering data.
- Approach used machine learning and training data were calculated using forward models.

![Graphical representation](image)

(a) Scattering data and (b) simulated data of Dy$_2$Ti$_2$O$_7$ [arXiv:1906.11275](https://arxiv.org/abs/1906.11275)

### Future opportunities
- Machine learning generated meta-data enabling automation (e.g. marking data from misaligned samples)
- Feature identification in elastic and inelastic neutron scattering data allowing automation and selecting modeling approaches
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.

High Level Demonstration Workflow

SNS – TOPAZ Neutron Scattering

Feature detection “Streaks” in Diffuse scattering data

APS - X-ray Scattering coming soon

Diffuse scattering simulations over parameter space

Knowledge base of experiments and simulations

Classification of “streaks” into candidate stacking faults

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

Data sharing and catalog of results
Challenges

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

Cristina Garcia Cardona (LANL), Ramakrishnan Kannan (ORNL), Thomas Proffen (ORNL), Travis Johnston (ORNL), Katherine Page (ORNL/UTK), David Womble (ORNL), Sudip K Seal (ORNL, POC)
Current workflow

Refine structural parameters

Create structural model

ExaLearn workflow

Refine structural Parameters (optional)

Predict model from ExaLearn trained model

Scientist input

Diffraction Data

Atomic structure
Thank you

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