ICE-MAN and VirtuES, the Integrated Computational Environment-Modeling & Analysis for Neutrons at ORNL

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ORNL is managed by UT-Battelle for the US Department of Energy
VISION

- Indirect geometry spectrometer optimized to study chemical systems
- High flux/throughput
- Broadband (-2 to 1000 meV)
- Constant dE/E (~1.5%)
- Elastic line HMFW ~150 μeV
- Simultaneous diffraction
Integrated modeling for data interpretation

Computer modeling is crucial to understand and interpret INS data.

• Dual 16 core Intel Haswell E5-2698v3 3.2 GHz Processors per node
• 50 compute nodes, 1600 (non-hyperthreaded) cores
• 128 GB memory/node, 6.4 TB Total memory
• Each node has 10Gbe and Infiniband networking for connectivity.
• Installed as part of the ORNL Compute and Data Environment for Science (CADES)
Integrated Computational Environment, for Modeling and Analysis of Neutron data

Acknowledgement: Laboratory Directed Research and Development program at ORNL
Databases

- DFT codes (RMG-DFT, CP2K, abinit Quantum Espresso)
- Molecular Mechanics, Dynamics (LAMMPS, GROMACS)
- Real Space Rietveld (PdfGUI)
- Reciprocal Space Rietveld (GSAS, FullProf)
Why do we need simulations for NVS (or INS in general)?

- Interpret neutron data
  - assigning peaks to vibrational modes
- Obtain insight on fundamental properties
  - understanding interatomic interactions, anharmonicity, complex excitations, phase transitions, chemical reactions
- Connect theory and experiment
  - simulation is a virtual experiment and an \textit{in silico} implementation of theory

We can measure it. We do understand it.
What to simulate for INS?

- Double differential cross-section

\[
\frac{d^2\sigma}{d\Omega dE'} = \text{(number of neutrons scattered per second into a small solid angle } d\Omega \text{ in the direction } \theta, \phi \text{ with final energy between } E' \text{ and } E' + dE')/\Phi d\Omega dE',
\]

- Fermi’s golden rule

\[
\left( \frac{d^2\sigma}{d\Omega dE'} \right)_{\lambda \rightarrow \lambda'} = k' \left( \frac{m}{2\pi\hbar^2} \right) |\langle k'\lambda' | V | k\lambda \rangle|^2 \delta(E_\lambda - E_{\lambda'} + \hbar\omega) \propto \frac{k'}{k} S(Q, \omega)
\]

\[V: \text{ potential describing the interaction between neutrons and the system}\]
\[\hbar\omega: \text{ fundamental excitation in the system}\]

- The goal is to formulate the interaction between neutrons and the system, so that \( S(Q, \omega) \) can be expressed by the excitations of interest.
Coherent inelastic scattering

• One-phonon $S(Q, \omega)$

$$S_{coh \pm 1}(Q, \omega) = \frac{1}{2N} \sum_s \sum_{\tau} \frac{1}{\omega_s} \left| \sum_d \frac{\bar{b}_d}{\sqrt{m_d}} \exp(-W_d) \exp(iQ \cdot r_d) (Q \cdot e_{ds}) \right|^2$$

$$\times \langle n_s + \frac{1}{2} \pm \frac{1}{2} \rangle \delta(\omega \mp \omega_s) \delta(Q \mp q - \tau)$$

- Peak position in energy depends on $Q$.
- Total intensity determined by not only how each atom moves, but also their relative phase.
Incoherent inelastic scattering

- One-phonon $S(Q,\omega)$

$$S_{inc\pm 1}(Q, \omega) = \sum_d \frac{1}{2m_d} \left\{ \vec{b}_d^2 - (\vec{b}_d)^2 \right\} \exp(-2W_d) \sum_s \frac{|\vec{q} \cdot e_d s|^2}{\omega_s} \langle n_s + \frac{1}{2} \pm \frac{1}{2} \rangle \delta(\omega \mp \omega_s)$$

- Peak position in energy does not depend on $Q$
- Each atom contributes to the total intensity independently.
Incoherent approximation

• When and why
  – Elements/isotopes with large incoherent scattering cross-section (e.g., hydrogen, vanadium) – The scattering itself is intrinsically incoherent.
  – High Q or large unit cell (small Brillouin zone), e.g. in low symmetry or disordered structure – The scattering may be coherent, but the ruler is too big for the pattern to be resolved.

\[
S_{coh\pm1}(Q,\omega) = \frac{1}{2N} \sum_s \sum_{\omega_s} \frac{1}{\sqrt{m_d}} \sum_d \frac{d_d}{\exp(-W_d)} \exp(iQ \cdot r_d) (Q \cdot e_{ds})^2 \\
\times (n_s + \frac{1}{2} \pm \frac{1}{2}) \delta(\omega \mp \omega_s) \delta(Q \mp q - \tau)
\]

\[
S(Q, n\omega_s) = \frac{(Q \cdot U_s)^{2n}}{n!} \exp[-(Q \cdot U_{total})^2]
\]

\[
U_s = \sqrt{\frac{\hbar m_s e_n}{2m_s \omega_s}}
\]
Development of OCLIMAX

- Started 2016
- First version released 2017
- Paper published 2019
- Used to analyze data from VISION and multiple other neutron spectrometers

Features:
- Incoherent and coherent scattering
- Powders and single crystals
- Temperature effects
- Multiphonon excitations
- Arbitrary instrument geometry and resolution
- Arbitrary cuts in 4-dimensional Q-E space
- Interface with atomistic modeling tools (e.g. DFT codes)
- Interface with INS data analysis tools (e.g. DAVE and Mantid)
- User-friendly (multiple platform, easy to use, fast on PCs)
OCLIMAX example: graphite

- Coherent scattering
  - Powders
  - Single crystal
- Kinematics
  - Option to generate masks in the map

Full calculation versus incoherent approximation
Calculated $S(Q,\omega)$ map and various sampling trajectories

OCLIMAX example: single crystal

Validating phonon frequencies, polarization vectors, and force constants
Understanding phonon anomalies.

$\hbar \omega = 8 \pm 1 \text{ meV}$

Total cross sections for solids from first principles calculations
OCLIMAX bridges theory and INS experiments

VISION, CNCS, HYSPEC, SEQUOIA, ARCS and many other neutron spectrometers.
Traditional methods

- Minimization, least squares algorithms
- Sequentially fitting QENS functions to data
- Parameters collected
- Plot parameters vs Q
- Fit parameters to functionality
- Laborious, time consuming
- Tedious, error prone method
- It is very difficult to track what’s been done to the data

\[
I(Q, E) = \left[ f_{eisf}(Q) \frac{1}{\pi} \frac{\Gamma_n(Q)}{\Gamma_n(Q)^2 + E^2} + (1 - f_{eisf}(Q)) \frac{1}{\pi} \frac{\Gamma_b(Q)}{\Gamma_b(Q)^2 + E^2} \right] \otimes R(Q, E) + (C_1(Q)E + C_2(Q))
\]
Global fitting of the data

Fitting data using parameter while imposing constraints overt this parameter:

\[ FWHM = f(\alpha, \beta, \gamma, \ldots, \omega, Q) \]

Returning best fit values for \(\alpha, \beta, \gamma \ldots\) and so forth
Global fitting of the data

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$$FWHM = f(\alpha, \beta, \gamma, \ldots, \omega, Q)$$

Returning best fit values for \(\alpha, \beta, \gamma \ldots\) and so forth
Example: Chudley-Elliot fit to hydrogen in metal hydrides
Bayesian behaviour

\[ L = 3.04 \pm 0.15 \text{ Å} \]
\[ \tau = 73.2 \pm 2.5 \text{ ps} \]
\[ L = 3.10 \pm 0.01 \text{ Å} \]
\[ \tau = 73.5 \pm 0.3 \text{ ps} \]
Many flavours

- Docker container
Many flavours

- Virtual machine
Many flavours

- Web access (hosted at ORNL, CADES)
Simple interface
Simple interface
Simple interface
Simple interface
Use your constraints
Constraints not there? No problem write them in python!
Select methods
Repeat old runs
Modify old runs, make expert changes
Run fittings
Visualize, edit results, get new starting configurations, download etc.
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Structure of heterogeneous software

User computer Linux, mac, PC
Structure of heterogeneous software

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VirtualBox

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User computer Linux, mac, PC

Docker Container Translations PYTHON
Docker Container Translations PERL
Docker Container Oclimax FORTRAN
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Conclusion

• Instrumental needs in VISION at SNS require respectable computer resources (1600+ cores cluster today) to help interpret the data
• The VirtuES cluster is available for users to run mostly DFT
• Software to generate inelastic neutron spectra from calculations to directly compare with experimental data has been developed (Oclimax)
• Software to analyze QENS data using global fitting and easy interface and extensibility in the fitting functions and constraints is also available.
• Virtual machine technologies, VirtualBox and Docker are used to produce software that is easy to maintain, expand and that can be operated from the command line and a web interface if desired.
• Secondary objective for software is to automate processes by exchanging file formats and generate a number of input/output files for different codes
• And much more…
• These codes provide the basis for a different approach to automation and reproducibility of neutron data analysis as well as routine integration of computer modeling in neutron scattering.
• Questions?