OAK RIDGE National Laboratory

Chemical Spectroscopy Software

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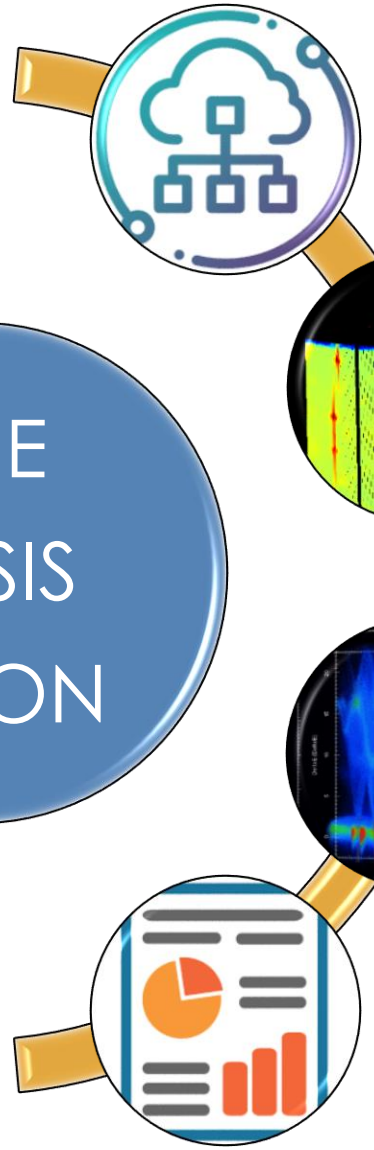
Neutron Scattering Division

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

U.S. DEPARTMENT OF ENERGY

2020 Review of the Instrument Suite for Spectroscopy
September 17-18, 2020

Overview



Data Acquisition → How to collect, convert, and save raw data

Data Reduction → How to translate detector data to the target output of the experiment

Data Analysis → How to process the reduced data for visualization and to extract physical properties

Data Interpretation (simulation) → How to understand the data, obtain atomistic insight, and tell the story

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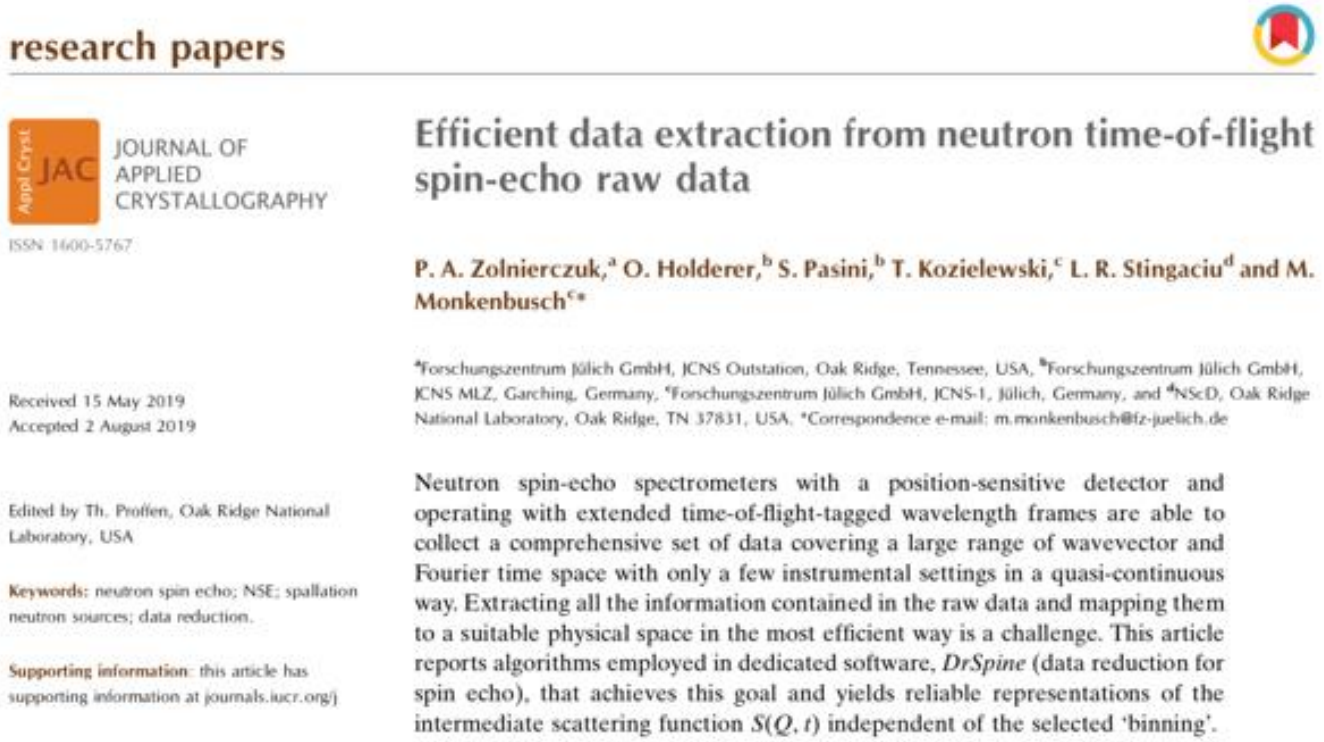
A brief summary of current status

- What is working
- What has been added recently
- What is needed in the near future

BL-15 (NSE)

- Data Acquisition
  - Jülich system
  - Transitioning to EPICS
- Data Reduction
  - DrSPINE (Data Reduction for SPIN Echo experiments)
- Data Analysis
  - Model fitting
- Data Interpretation
  - Molecular dynamics (MD) simulation



research papers

JOURNAL OF APPLIED CRYSTALLOGRAPHY

ISSN 1600-5767

Efficient data extraction from neutron time-of-flight spin-echo raw data

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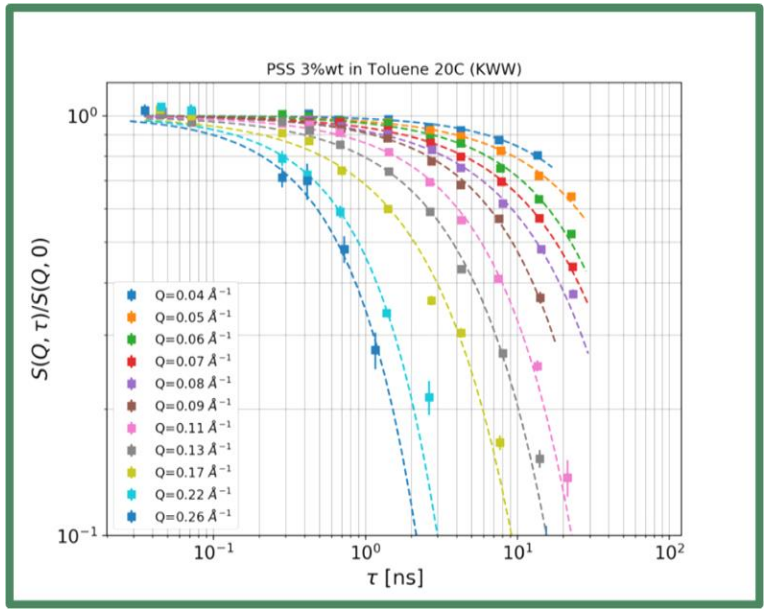
Keywords: neutron spin echo; NSE; spallation neutron sources; data reduction.

Supporting information: this article has supporting information at journals.iucr.org/j

Neutron spin-echo spectrometers with a position-sensitive detector and operating with extended time-of-flight-tagged wavelength frames are able to collect a comprehensive set of data covering a large range of wavevector and Fourier time space with only a few instrumental settings in a quasi-continuous way. Extracting all the information contained in the raw data and mapping them to a suitable physical space in the most efficient way is a challenge. This article reports algorithms employed in dedicated software, *DrSpine* (data reduction for spin echo), that achieves this goal and yields reliable representations of the intermediate scattering function $S(Q, t)$ independent of the selected 'binning'.

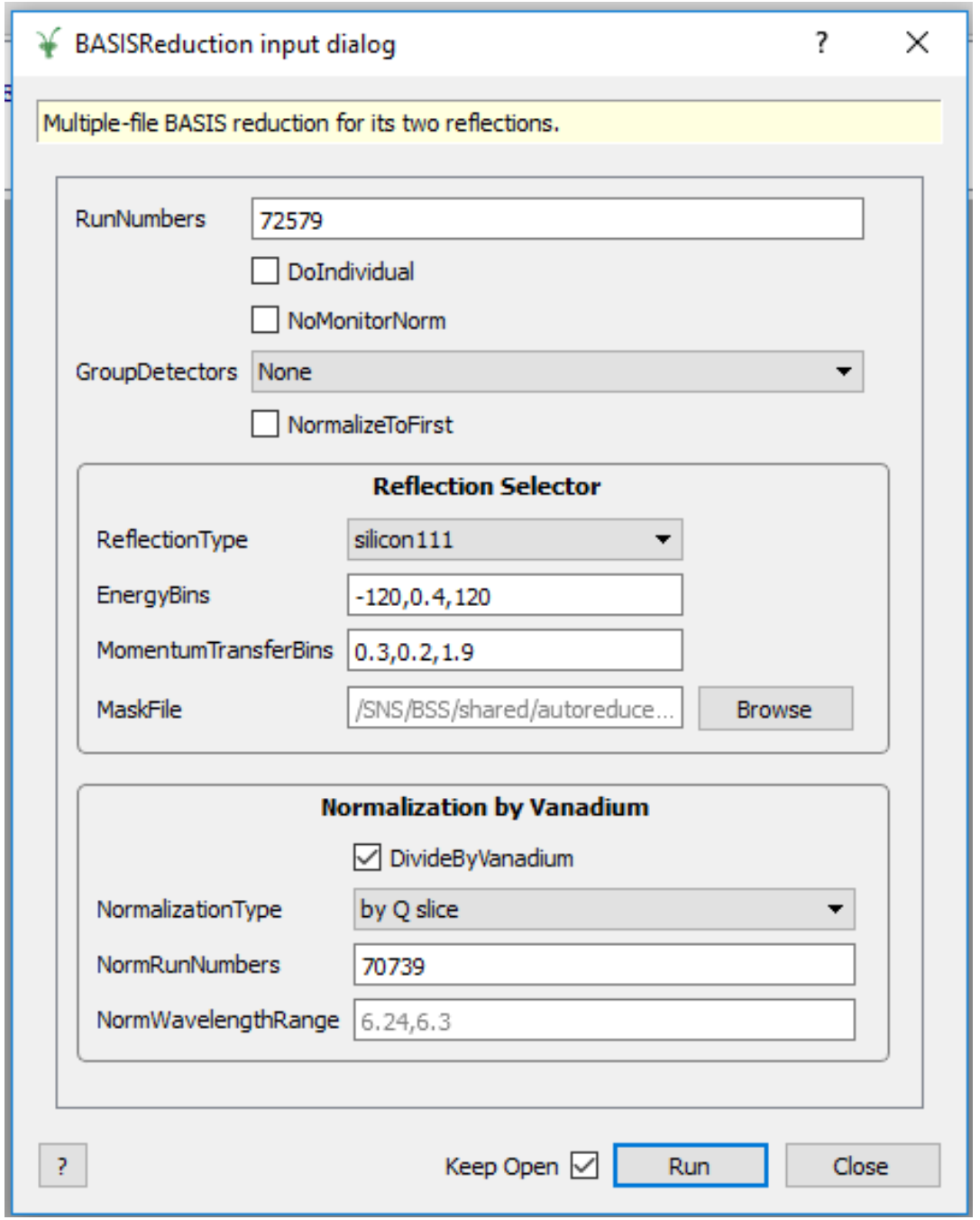
Zolnierczuk *et al.* J. Appl. Cryst. (2019). **52**,1022-1034 (DOI: 10.1107/S1600576719010847)

```
dr_macro_acns (~/Wrk/SpinEcho/ipts-19149) - VIM
macro

set a  1e-10
set ns 1e-09
bins  tof  custom 4 12 24 38

! resolution
read s8572.echo s8574.echo s8575.echo as res ! 8A
! buffer
read s8580.echo s8581.echo s8582.echo as bgr ! 8A
! PSS3 / Toluene 20C
read s8645.echo s8646.echo s8647.echo as sam tfac 0.8645 ! 8A

match all
fit   all

histo q custom 0.04/a 0.05/a 0.06/a 0.07/a 0.08/a 0.09/a 0.12/a 0.14/a 0.20/a 0.25/a 0.35/a
histo tau nbins 20  min 0.001*ns max 100*ns  log

collect bgr 1 volfrac 1.0
plot sqt model kww  summary

                                                        6,0-1         Top
```



BL-2 (BASIS)

- Data Acquisition
  - EPICS completed March 2019
- Data Reduction
  - Mantid (automatic and manual)
- Data Analysis
  - DAVE (PAN) or QCLIMAX
- Data Interpretation
  - MD simulation



MD simulations for NSE and BASIS

- Examples are few and the connections are indirect
- A bottleneck for instrument productivity and the impact of the publications
- Case-by-case (large-scale) MD models, challenges for automation and general user support
- Lack of proper tools to convert large-scale MD trajectories into neutron scattering functions (Sassena, nMOLDYN, LiquidLib)
- Software development priority for NSE and BASIS

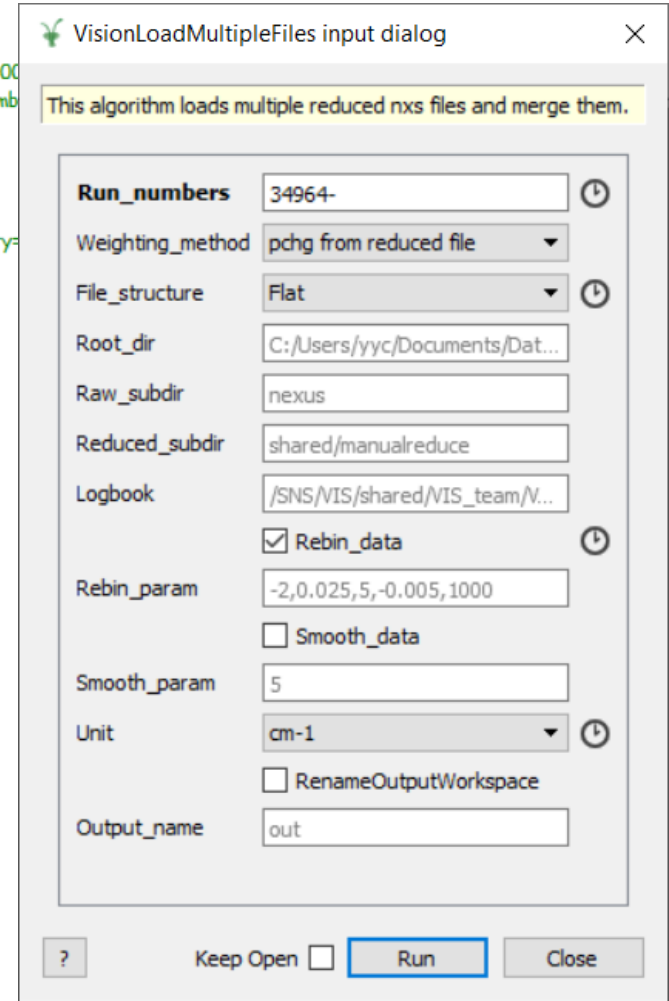
BL-16B (VISION)

- Data Acquisition
  - EPICS (since 2015)
- Data Reduction
  - Mantid (automatic)
- Data Analysis
  - Mantid scripts and algorithms
- Data Interpretation
  - Lattice dynamics (LD) or MD
  - OCLIMAX

```
#========control block================
file_structure = 'Flat'  # IPTS: IPTS subfolders. Flat: flat structure, all data under the root dir
weight = 1               # 0: from raw file. 1: from reduced file. 2: from log file. 3: from error
save_flag = 0            # 0: output not saved.  1: processed spectra saved in nxs and ascii
root_dir = 'C:/Users/yyc/Documents/Data/VISION' # root directory to the data files
save_dir = 'C:/Users/yyc/Documents/Data/Mantid/data/ascii'  # needed only for save_flag=1
#----------------------------
raw_subdir = 'nexus'    # needed only for file_structure='IPTS' and weight=0
red_subdir = 'shared/manualreduce'  # needed only for file_structure='IPTS'
log_file = 'E:/Data/VISION/VIS_logbook.csv' # needed only for weight=2
#========end of control block==========
#========global parameters============
scalex = 1.0; scaley = 1.0              # scaling parameters for x (energy transfer) and y (intensity)
shiftx = 0.0; shifty = 0.0              # shift along x and y (after scaling)
bin = '-2,0.025,5,-0.005,1000'          # binning parameters (meV)
smooth = 0                              # smooth parameter NPoints (0 for no smoothing)
align = '0,0,0,0'    # alignment params (meV): el_low,el_high,ins_low,ins_high (e.g. '-0.5,0.5,50,200
unit = 'meV'                            # unit for output workspaces/files, 'cm-1' for wavenumb
bose = False                            # False: no bose correction even temperature is given
#========end of global parameters======
#========data block====================
# the global parameters can be overwritten in the data block (redefined for a particular dataset)
# d.append( DataSet('16502-', 'b', temperature, bin='', smooth=, align=, scalex=, scaley=, shiftx=, shifty=

#  Cu-faujasite_2nd_blank_Al_conflat_can_slits_20x40_at_5K
d.append( DataSet('48292-', 'b', 5) )
```

Script (for batch processing with consistency and reproducibility)

GUI as Mantid algorithms (for ad hoc manual processing)



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Recent Development

- DrSPINE (Piotr Zolnierczuk)
- QCLIMAX (Timmy Ramirez-Cuesta)
- OCLIMAX (YQ Cheng)

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, error-prone
- Difficult to track the history of the data

QCLIMAX

Non-Linear Least-Squares Minimization and Curve-Fitting for Python

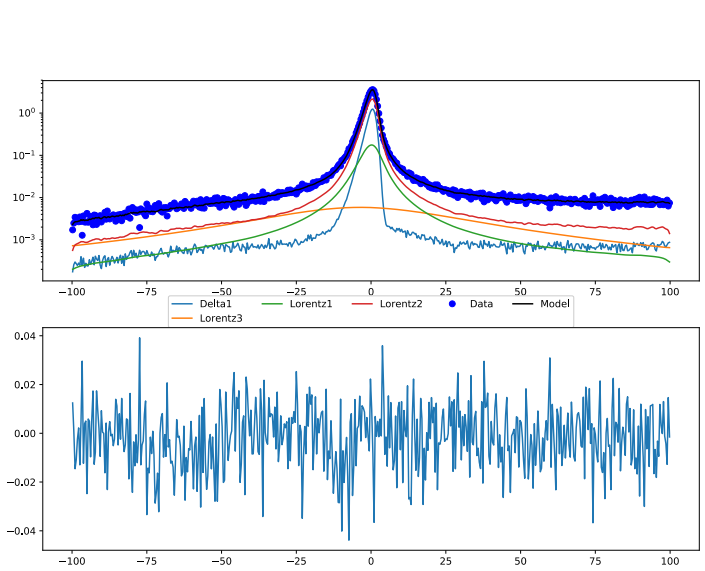
Release 0.9.6

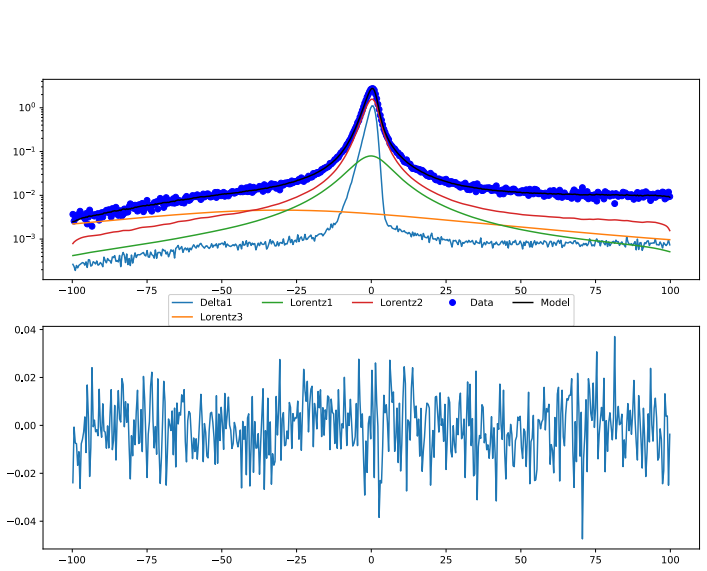
Matthew Newville, Till Stensitzki, and others

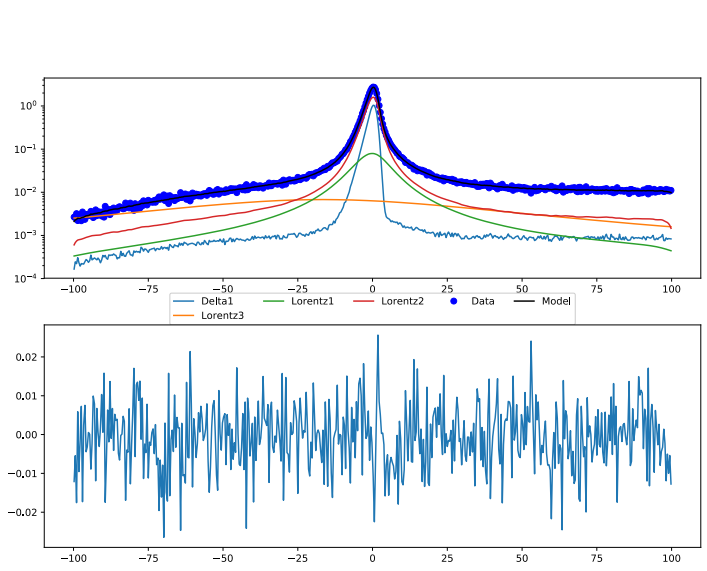
Mamontov E., Smith R.W., Billings J.J., Ramirez-Cuesta A.J., "Simple analytical model for fitting QENS data from liquids", Physica B: Condensed Matter, 566, 50-54 (2019).

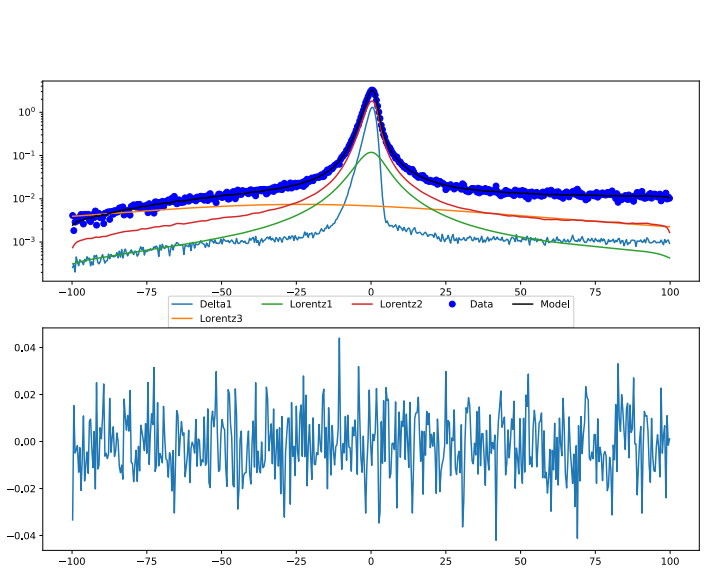
Courtesy: Timmy Ramirez-Cuesta

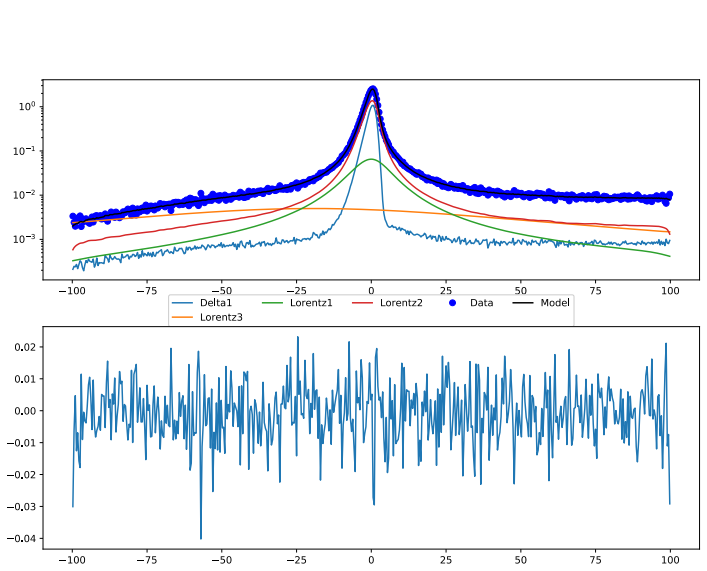
QCLIMAX example











Global fitting of the data

Fitting data using parameter while imposing constraints overt this parameter:

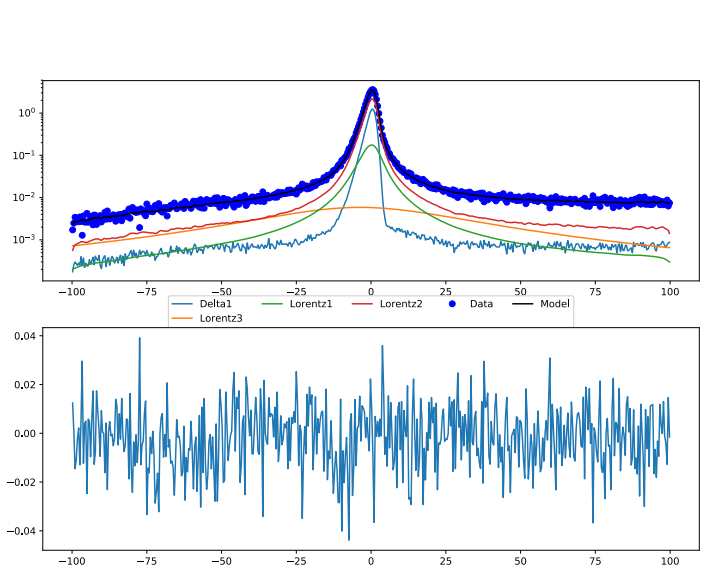
$$FWHM = f(\alpha, \beta, \gamma, \cdots, \omega, \boldsymbol{Q})$$

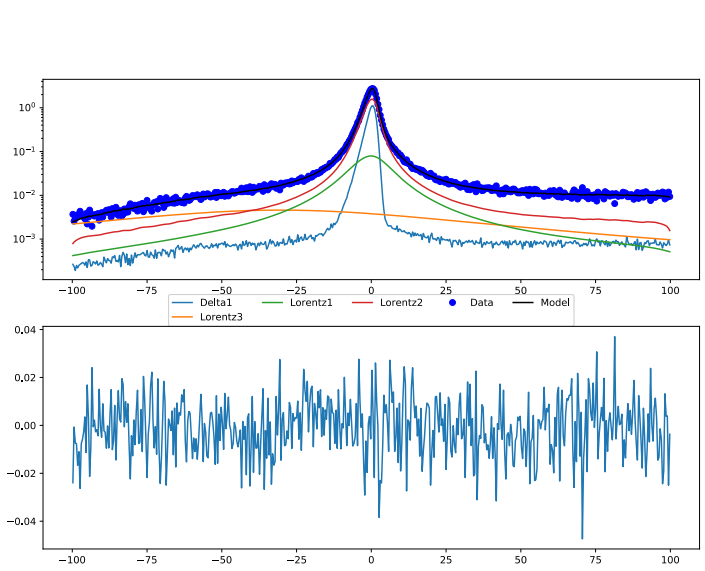
Returning best fit values for $\alpha, \beta, \gamma \cdots$ and so forth

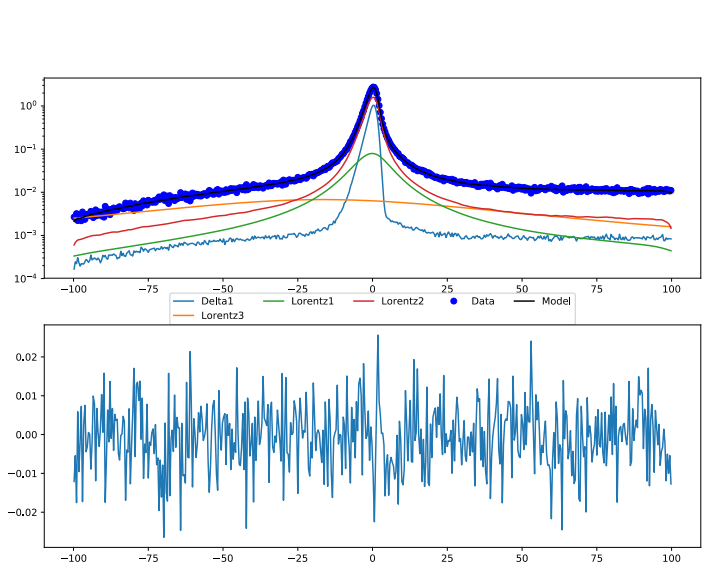
Chudley-Elliot $HWHM(Q) = \frac{\hbar}{\tau} \cdot \left(1 - \frac{sin(Ql)}{Ql}\right)$

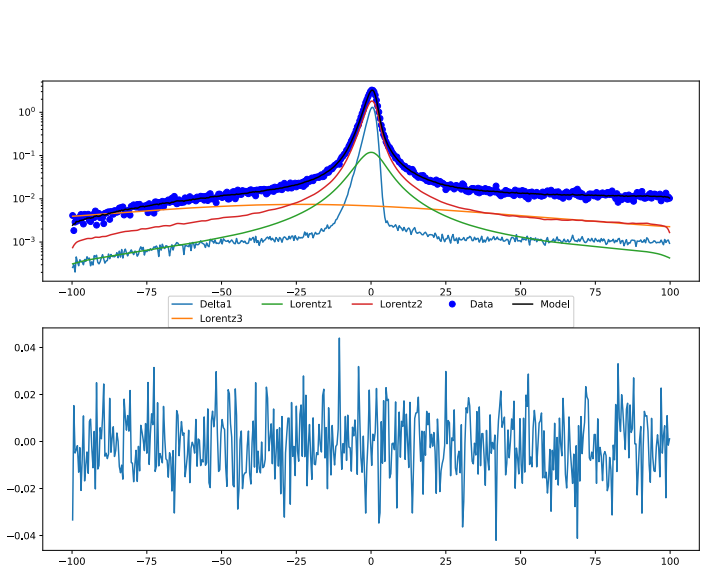
Courtesy: Timmy Ramirez-Cuesta

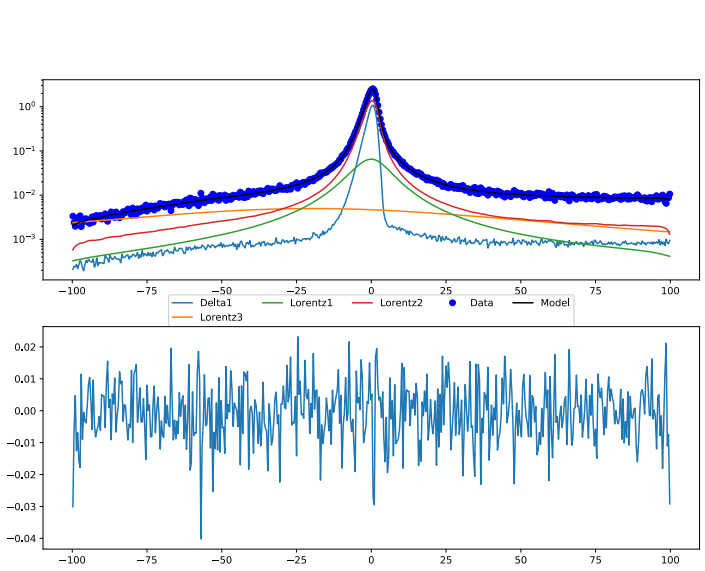
QCLIMAX example



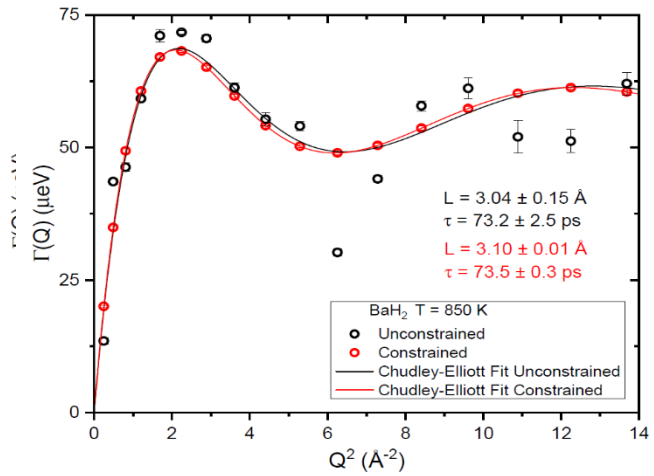








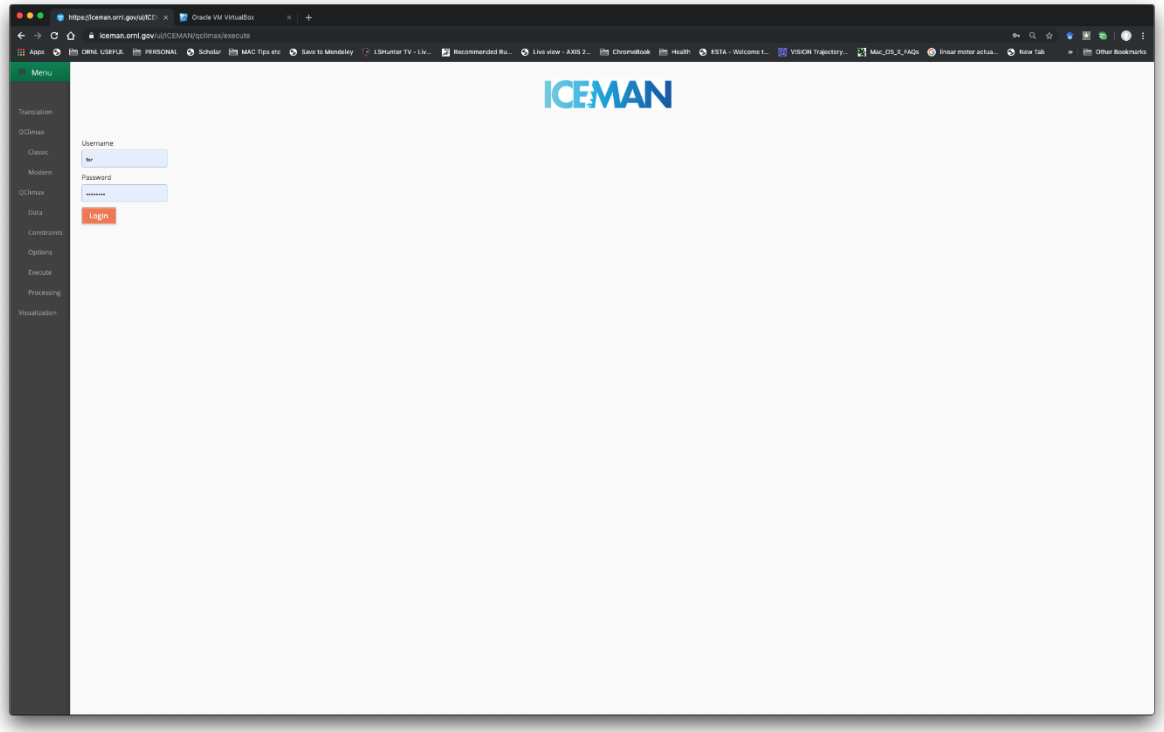
Example:
Chudley-Elliot fit to hydrogen in metal hydrides

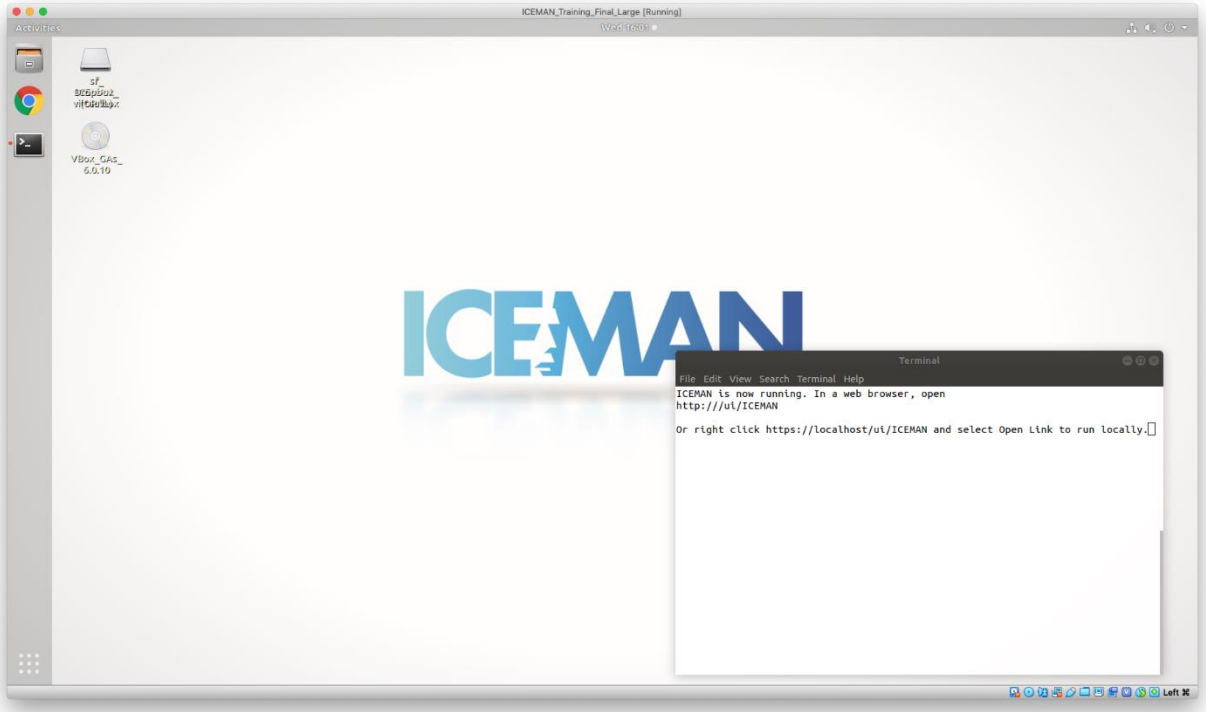


Courtesy: Timmy Ramirez-Cuesta

How to use QCLIMAX: many flavors

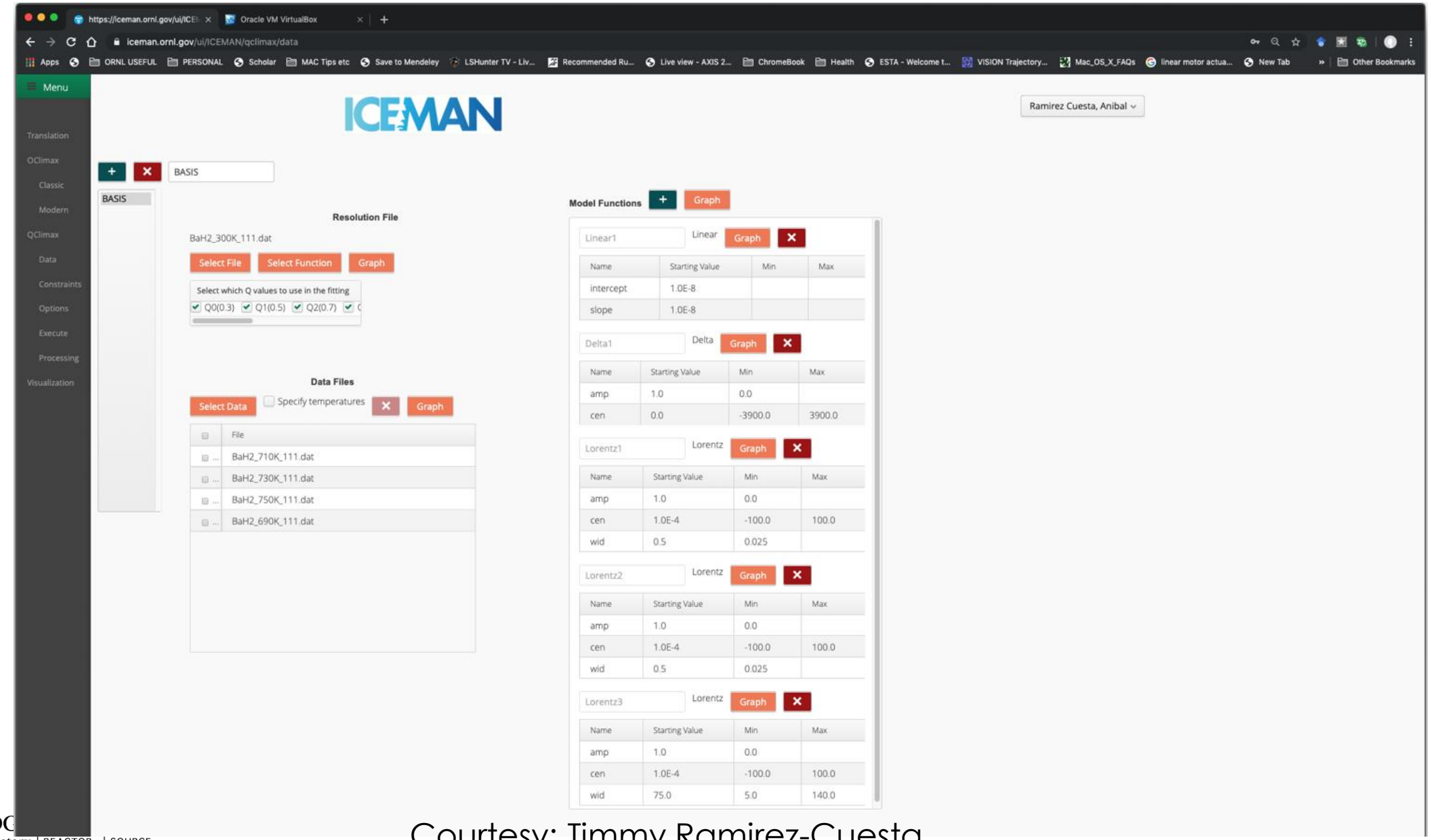
- Docker container
- Virtual machine
- Web access (hosted at ORNL, CADES)





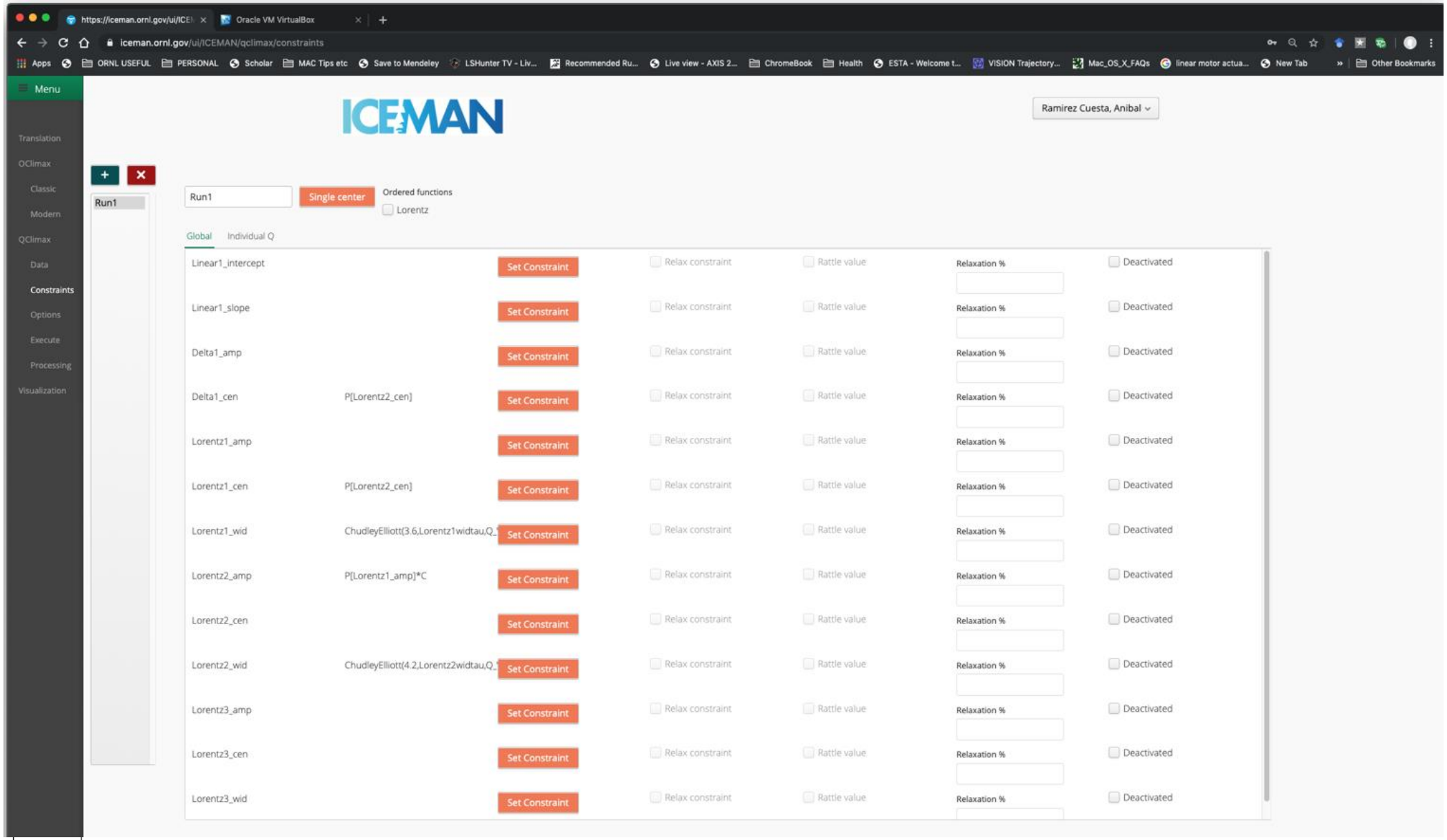
Courtesy: Timmy Ramirez-Cuesta

Simple interface



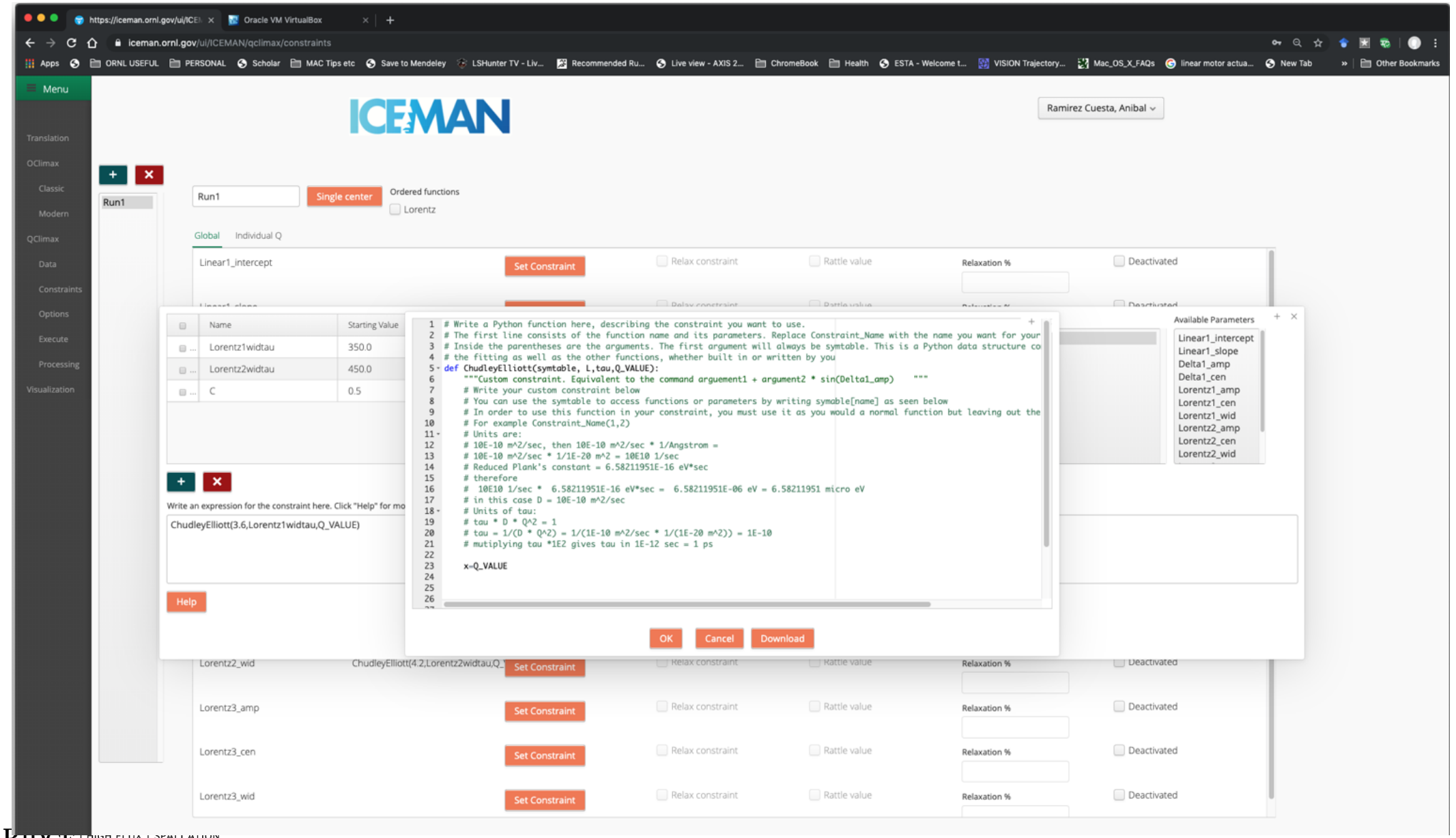
Courtesy: Timmy Ramirez-Cuesta

Use your constraints



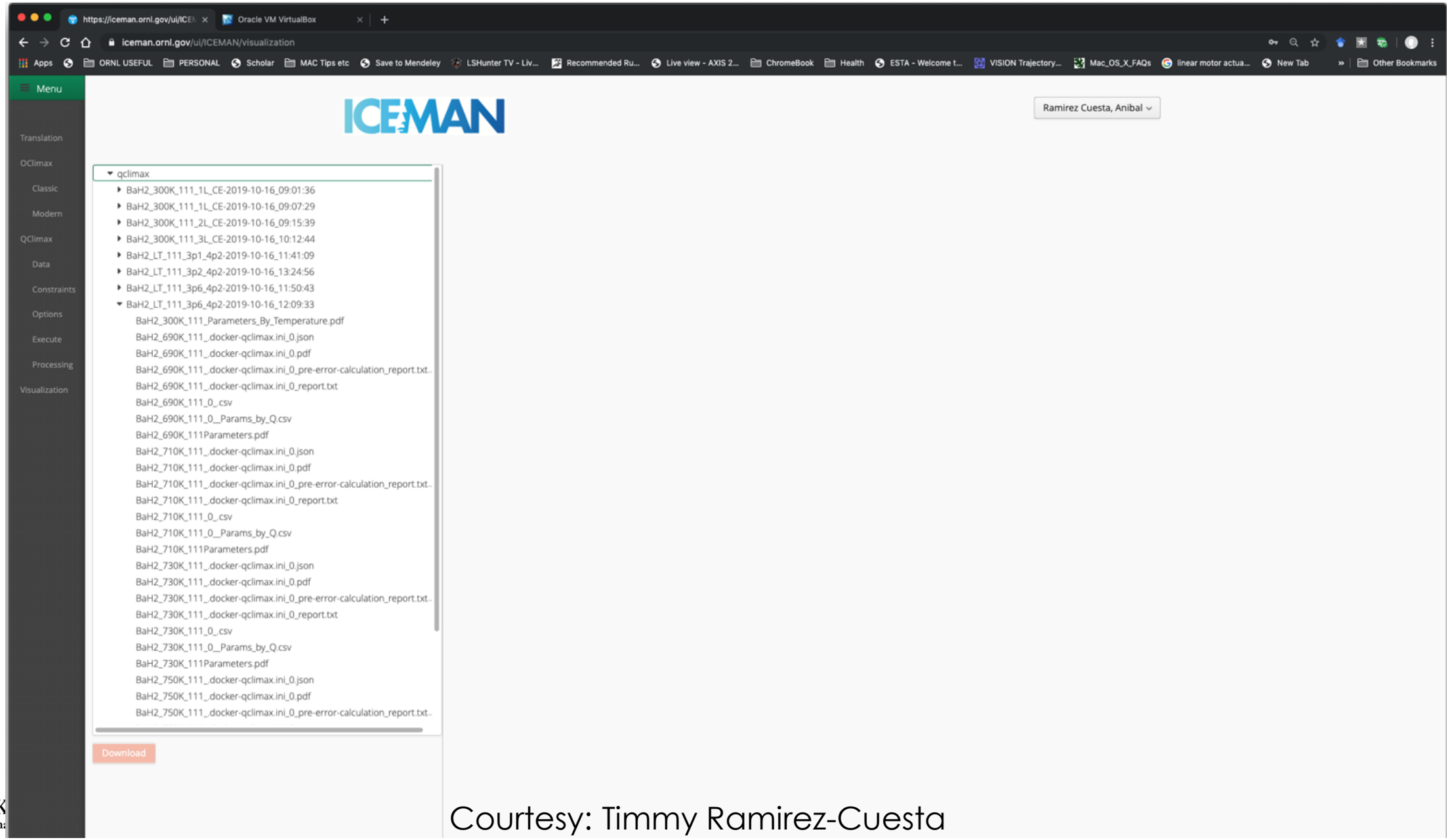
Courtesy: Timmy Ramirez-Cuesta

Constraints not there? No problem, write them in Python!



Courtesy: Timmy Ramirez-Cuesta

Visualize, edit results, restart, download etc.



Courtesy: Timmy Ramirez-Cuesta

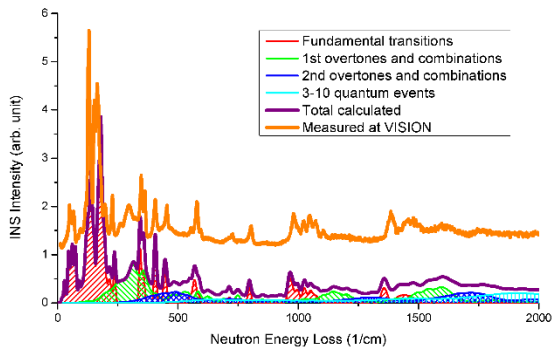
OAK RIDGE National Laboratory

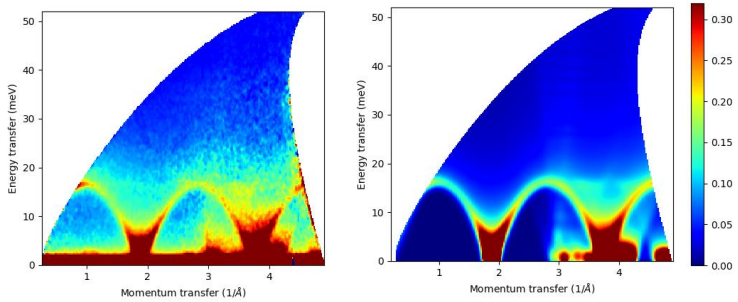
Recent Development

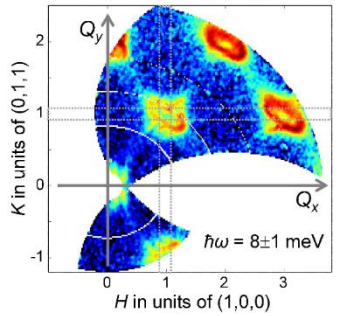
- DrSPINE (Piotr Zolnierczuk)
- QCLIMAX (Timmy Ramirez-Cuesta)
- OCLIMAX (YQ Cheng)

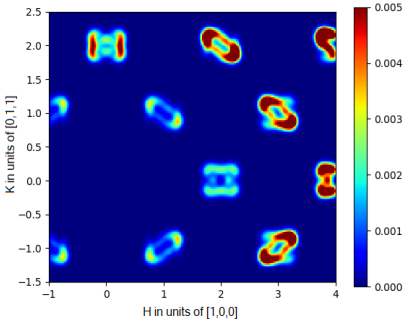
OCLIMAX bridges theory and INS experiments

Normal modes or phonons → **OCLIMAX** ← Molecular dynamics trajectories









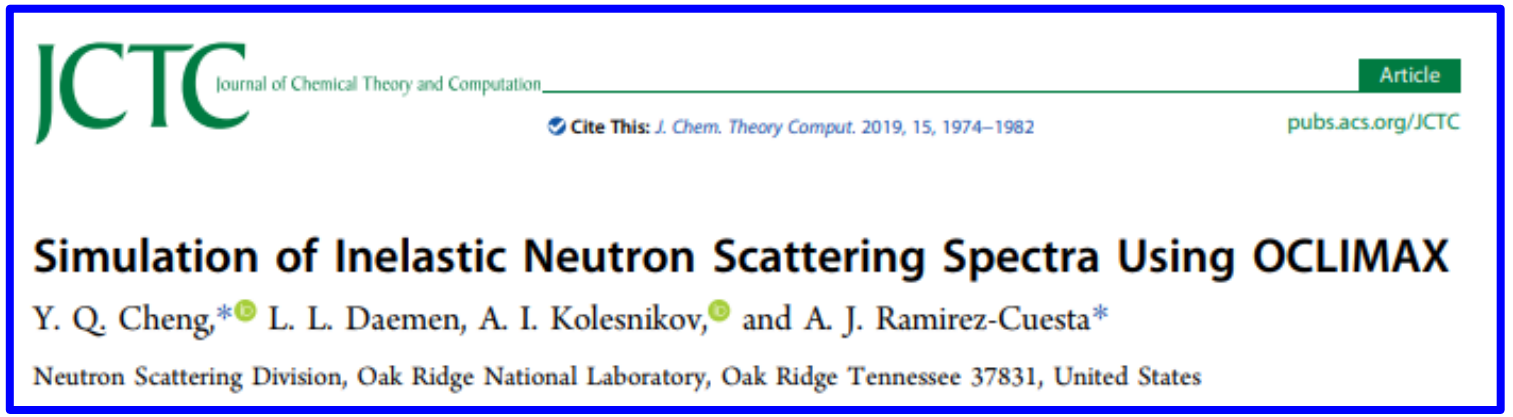
S(Q,E) including elastic/inelastic/coherent/incoherent contributions

Development of OCLIMAX

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

Features:

- ❖ Full scattering (coherent, incoherent, elastic, inelastic)
- ❖ 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)
- ❖ Parameter-free thermal neutron scattering cross-sections
- ❖ Molecular dynamics trajectories to INS spectra


JCTC Journal of Chemical Theory and Computation
Article
Cite This: J. Chem. Theory Comput. 2019, 15, 1974−1982
pubs.acs.org/JCTC

Simulation of Inelastic Neutron Scattering Spectra Using OCLIMAX

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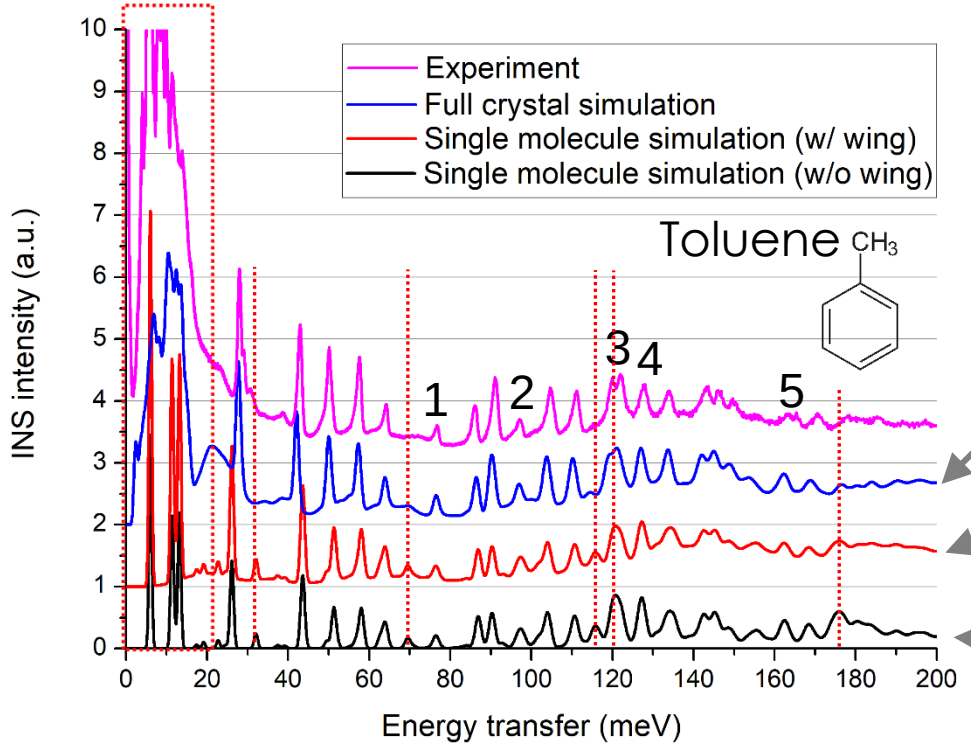
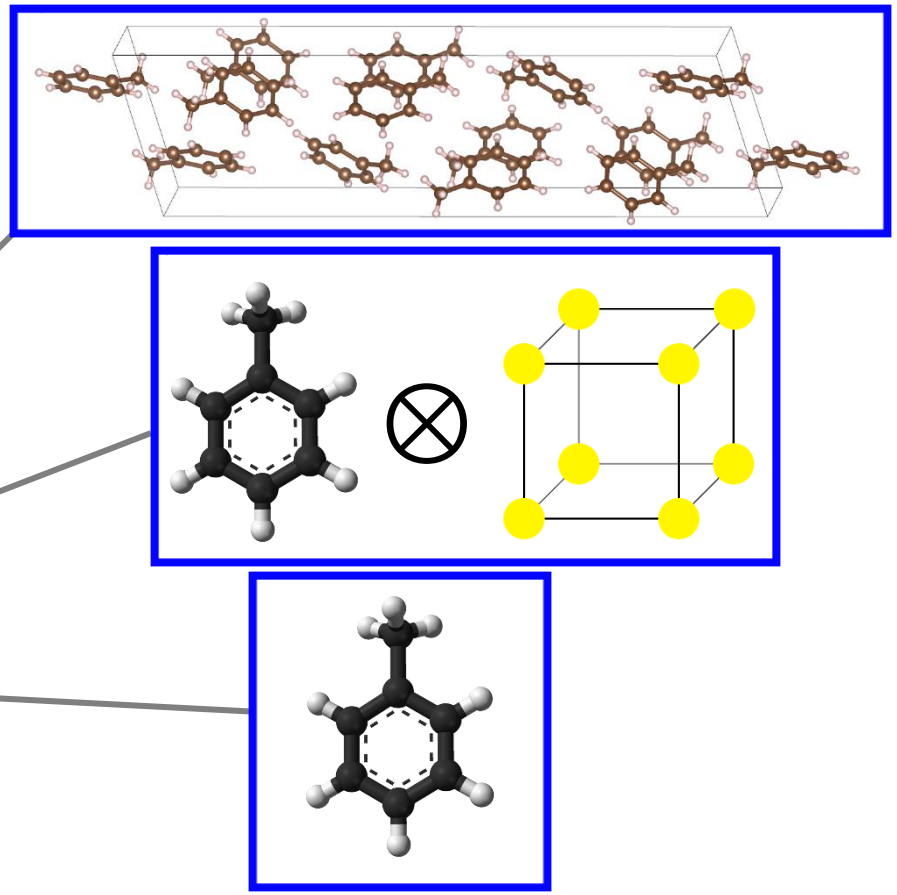
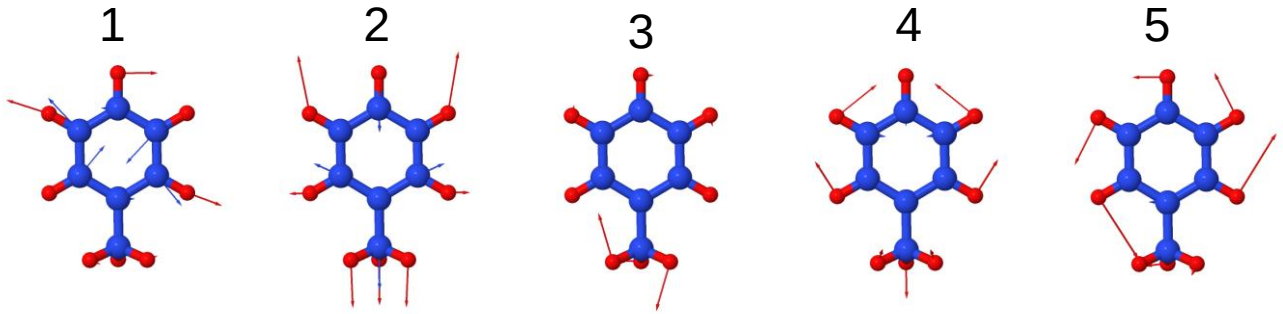
Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge Tennessee 37831, United States

OCLIMAX example: From single molecule to solid

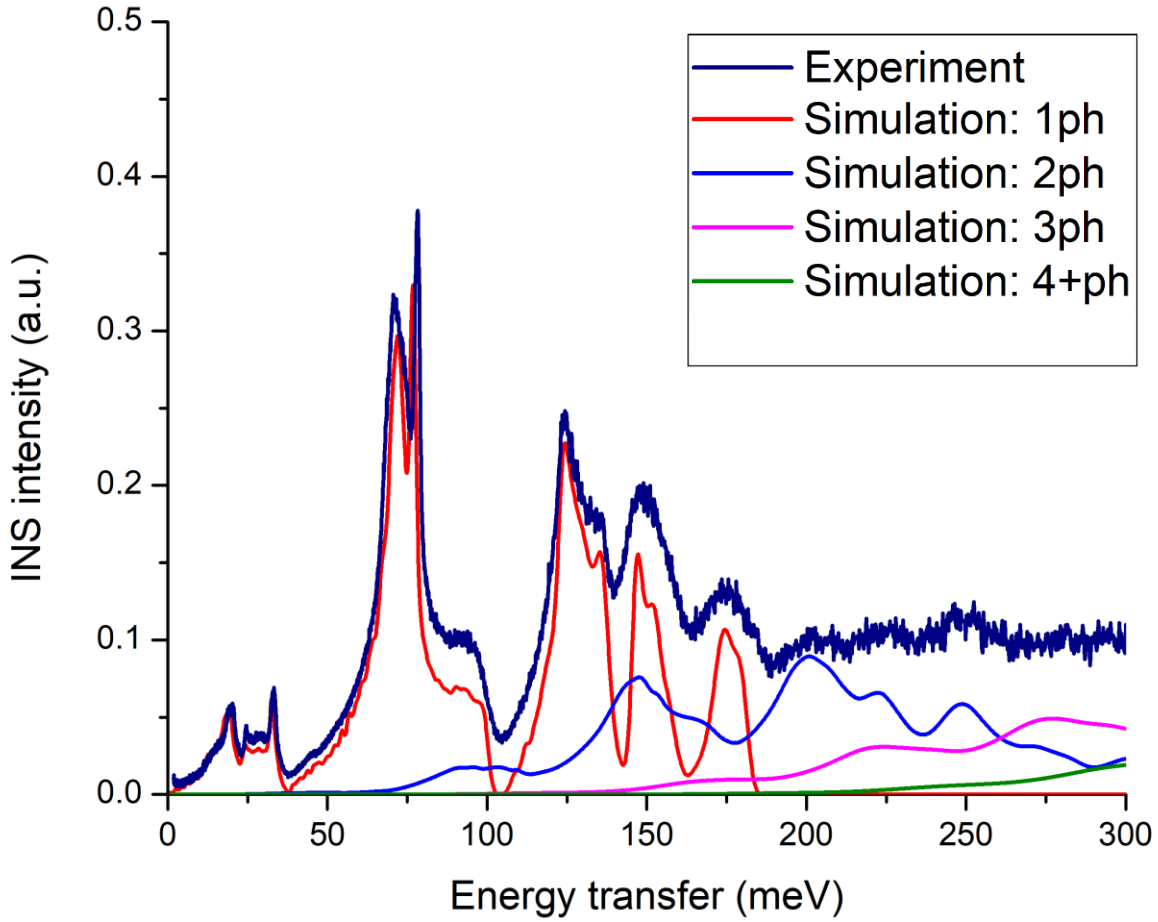


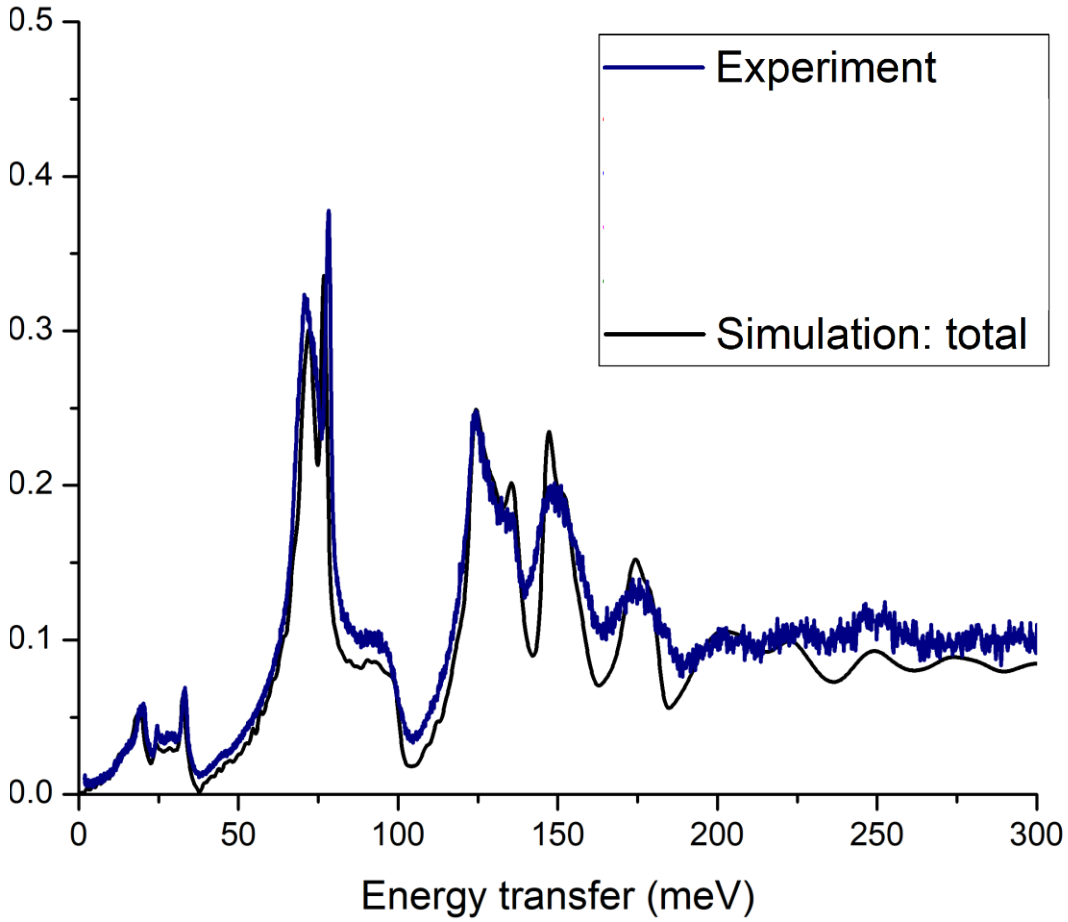




- ✓ Understanding intermolecular interactions (van der Waals forces, hydrogen bonding, charge transfer)

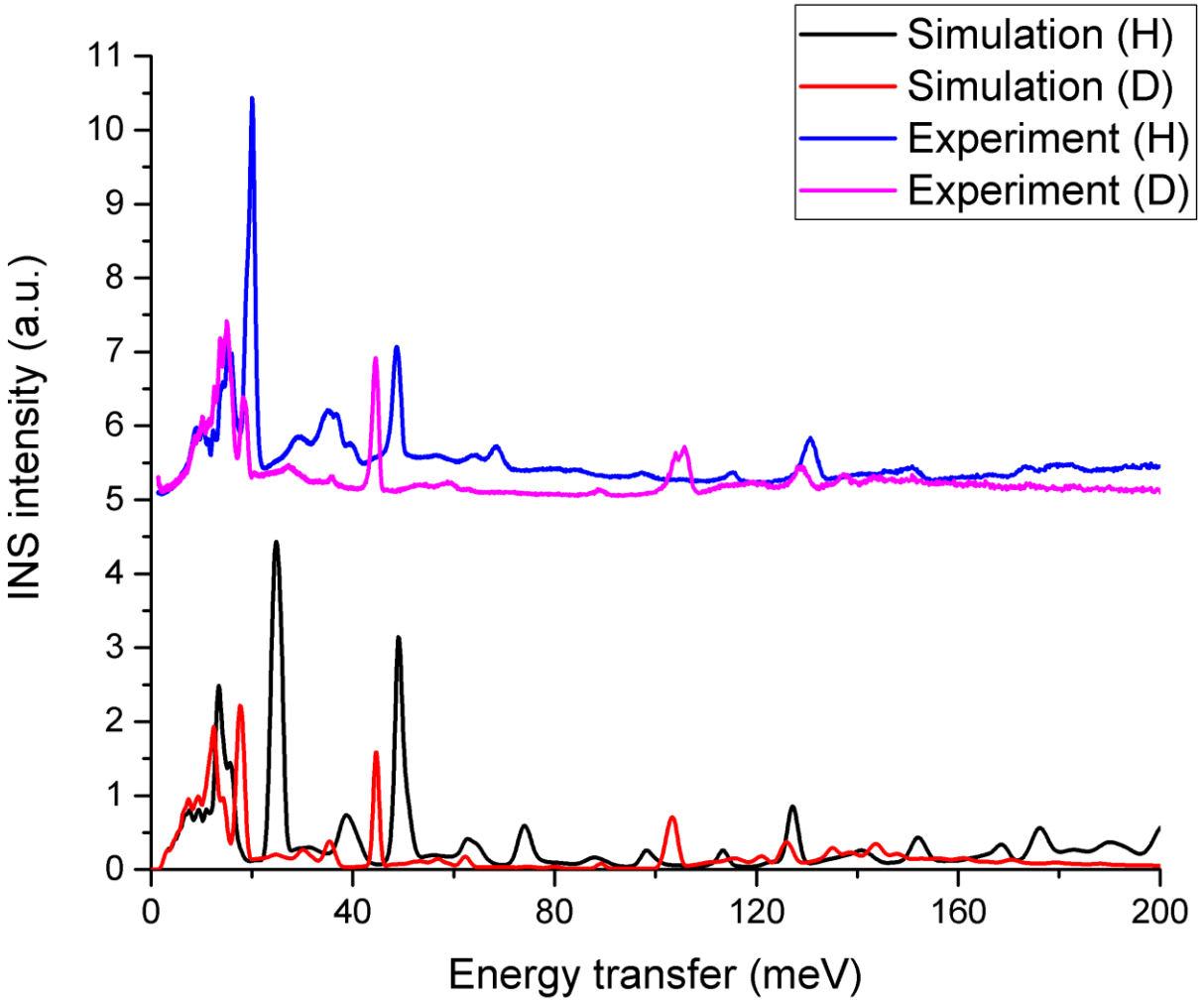
OCLIMAX example: Multiphonon excitations

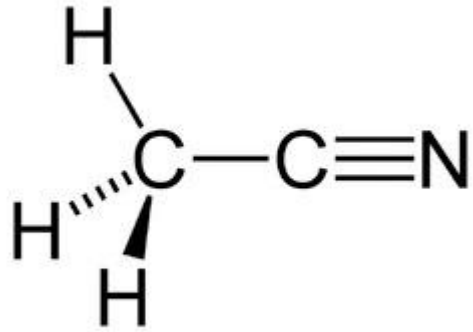




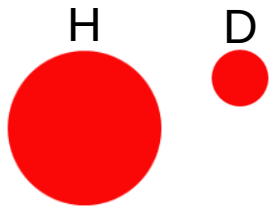
- ✓ Solving phonon density of states
- ✓ Understanding anharmonicity and potential energy landscape

OCLIMAX example: Isotope substitution





$$\omega = \sqrt{\frac{k}{m}}$$



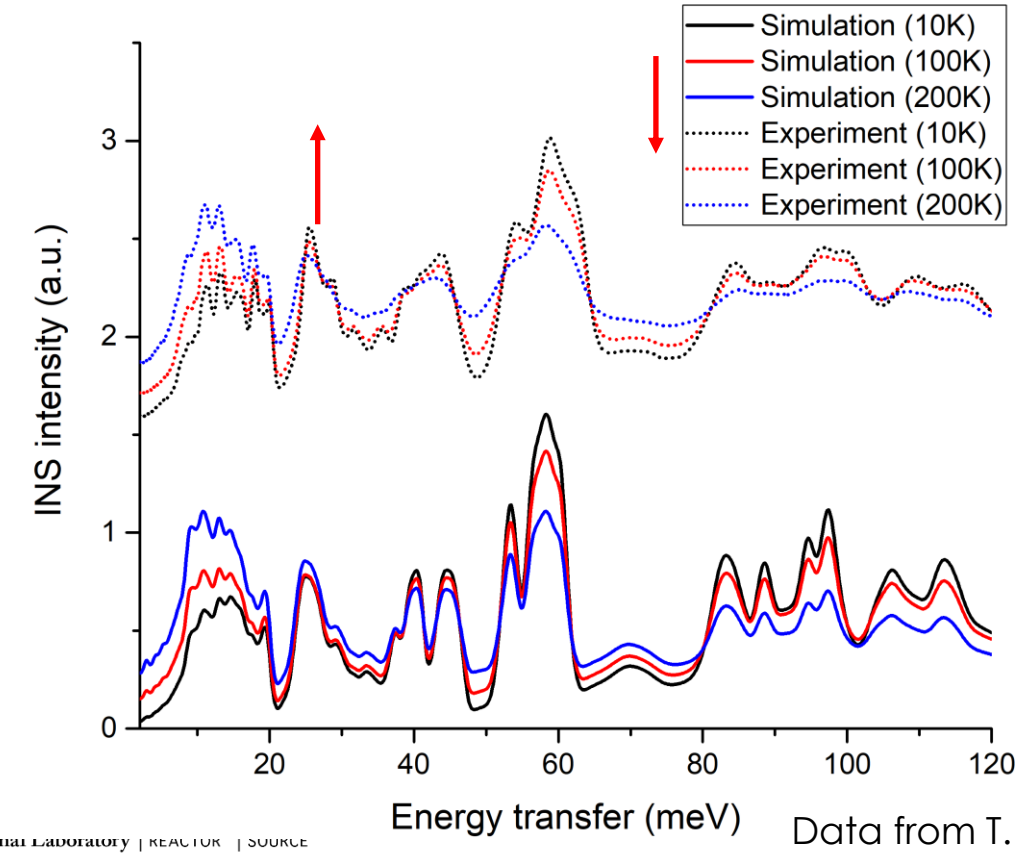
- ✓ Virtual experiment for doping effects and isotope labeling
- ✓ Breaking down the total intensity into partial contributions from individual species or atoms

OCLIMAX example: Temperature effects

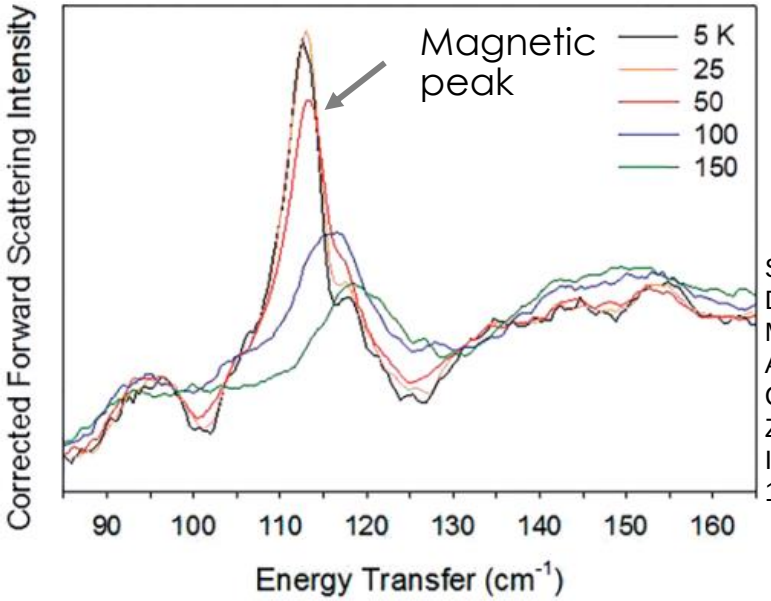
- Phonon population
- Debye-Waller factor

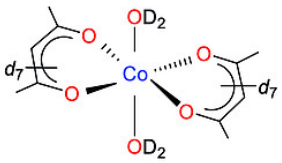
$$W_d = \frac{\hbar}{4m_d N_q}\sum_s \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_{ds})^2}{\omega_s}(2n_s+1)$$

$$S_{inc\pm1}(\boldsymbol{Q},\omega) = \sum_d \frac{1}{2m_d}\left\{\bar{b}_d^2 - (\bar{b}_d)^2\right\}\exp(-2W_d)\sum_s \frac{|\boldsymbol{Q}\cdot\boldsymbol{e}_{ds}|^2}{\omega_s}\left\langle n_s + \frac{1}{2} \pm \frac{1}{2}\right\rangle \delta(\omega \mp \omega_s)$$



Data from T. Sato

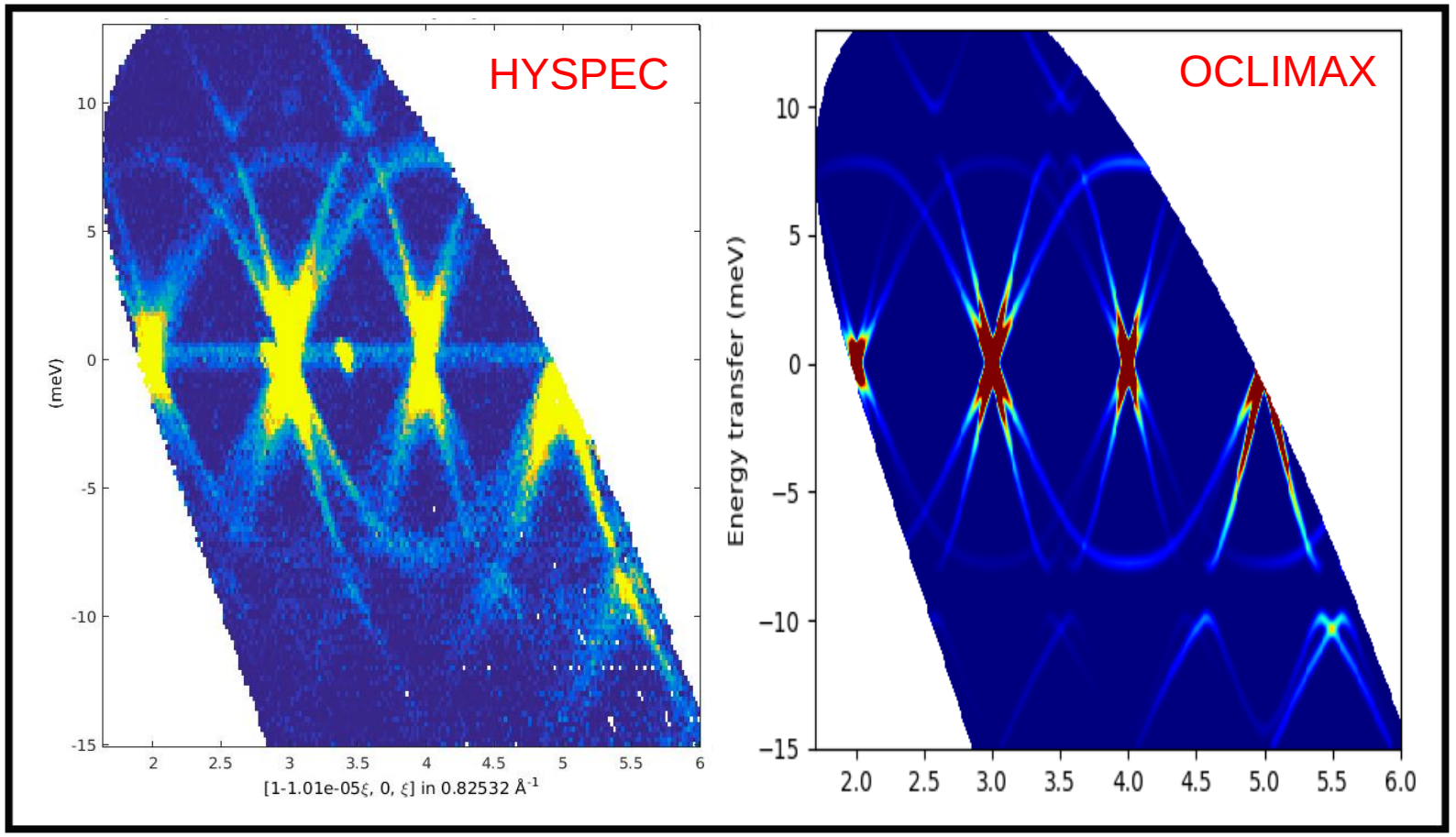


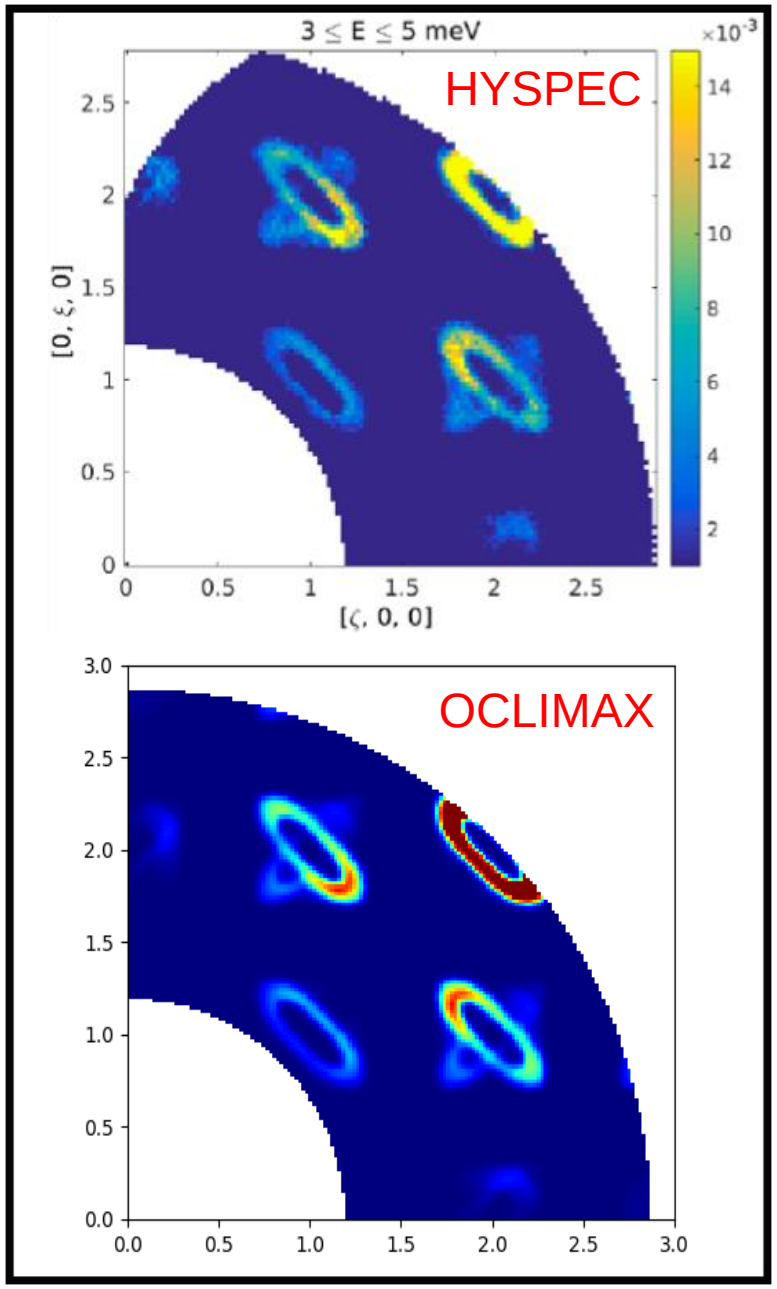


Stavretis S.E., Cheng Y.Q., Daemen L.L., Brown C.M., Moseley D.H., Bill E., Atanasov M., Ramirez-Cuesta A.J., Neese F., Xue Z., European Journal of Inorganic Chemistry, 2019, 8, 1119-1127 (2019).

- ✓ Separating different excitations based on temperature dependence
- ✓ Benchmarking atomic displacement parameters

OCLIMAX example: Single crystal cuts in 4-D Q-E space



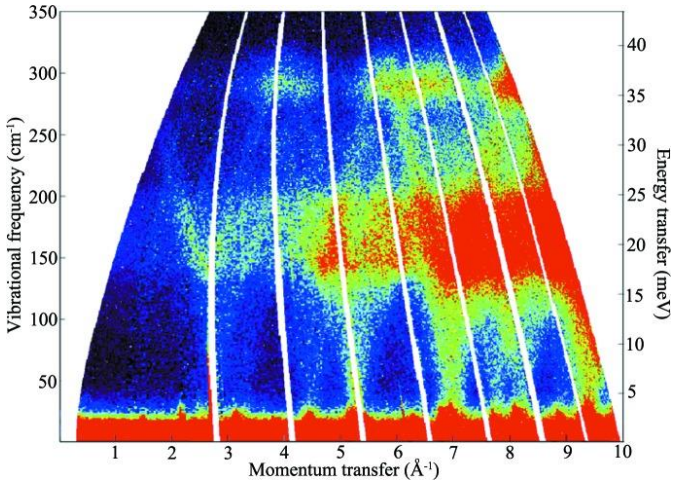


- ✓ Validating phonon frequencies, polarization vectors, and force constants
- ✓ Understanding phonon anomalies.

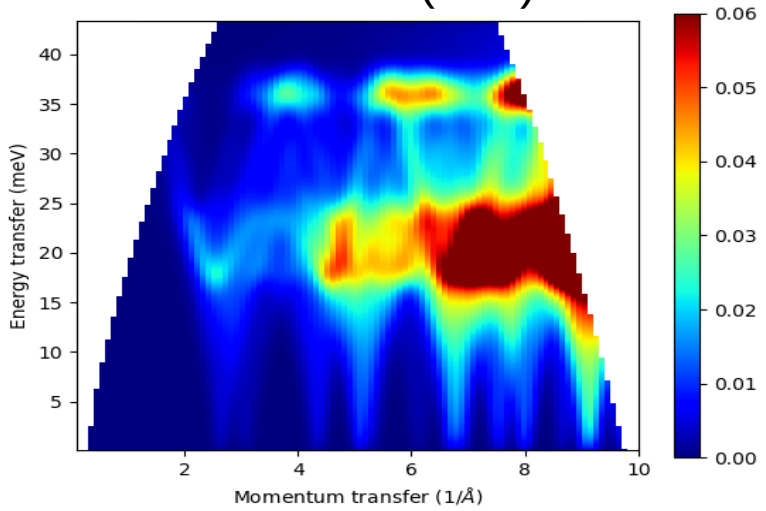
Unpublished INS data from R. Hermann. DFT simulations by L. Lindsay and T. Pandey @ORNL

OCLIMAX example: Coherent powder INS (aluminum)

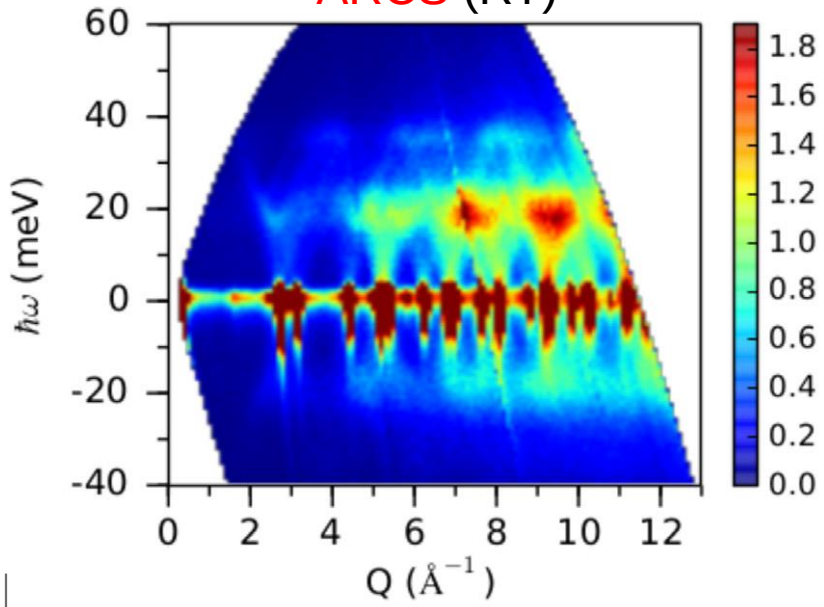
MARI (10K)



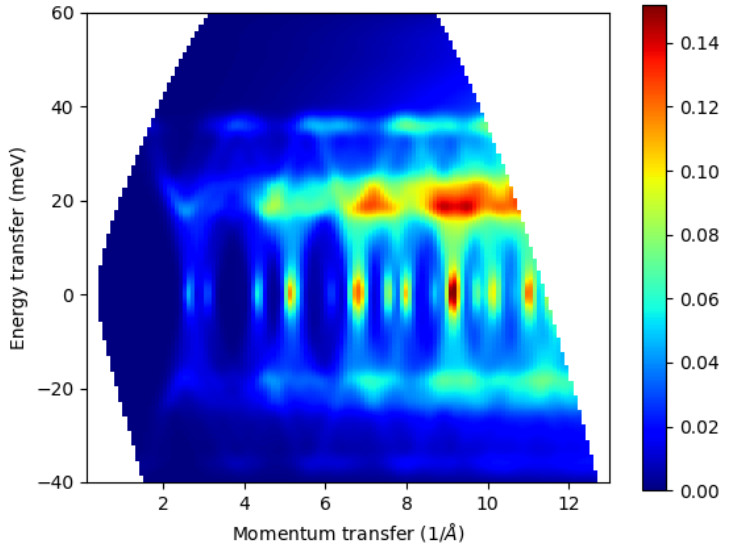
OCLIMAX (10K)



ARCS (RT)

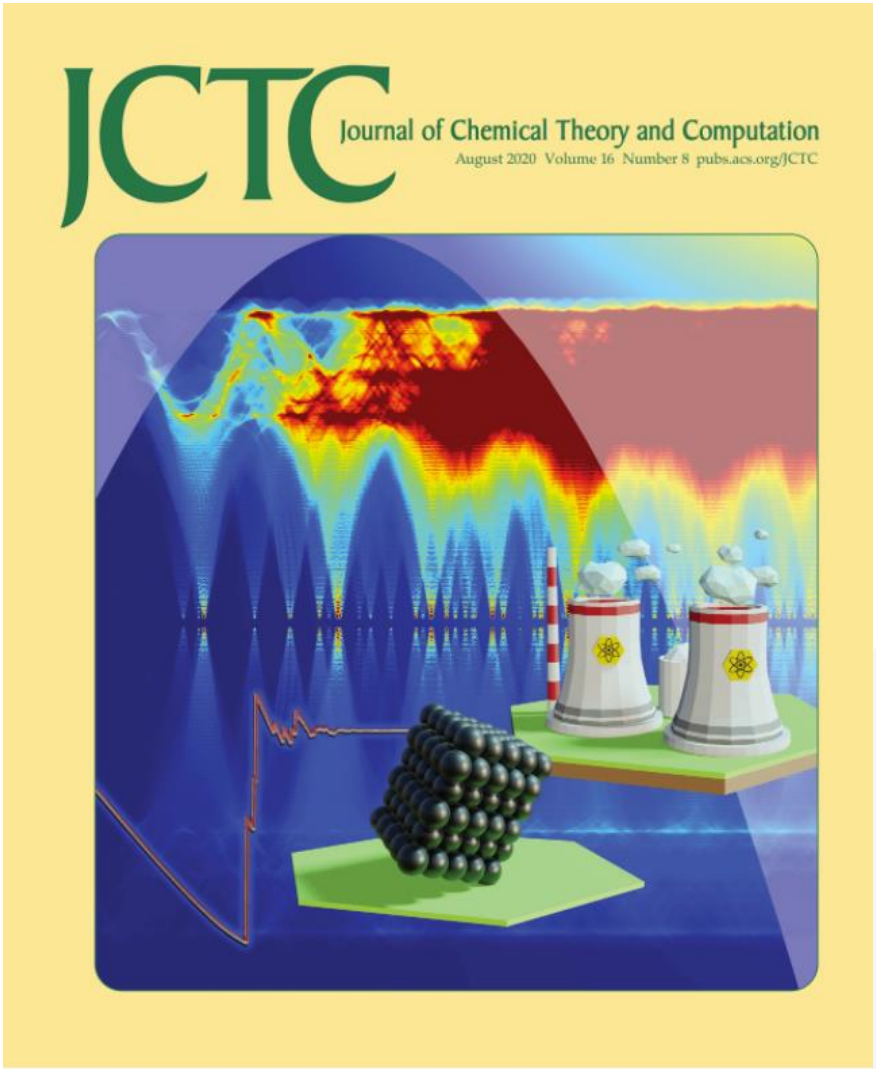


OCLIMAX (RT)



MARI data from: D. L. Roach et al. J. Appl. Cryst. **46**, 1755-1770 (2013).
ARCS data from: Lin et al. Nucl. Instrum. Methods Phys. Res., Sect. A 2016, 810, 86–99.

Ab initio thermal neutron scattering cross-section of solids



JCTC Journal of Chemical Theory and Computation

pubs.acs.org/JCTC Article

Calculation of the Thermal Neutron Scattering Cross-Section of Solids Using OCLIMAX

Y. Q. Cheng* and A. J. Ramirez-Cuesta

Cite This: *J. Chem. Theory Comput.* 2020, 16, 5212−5217 Read Online

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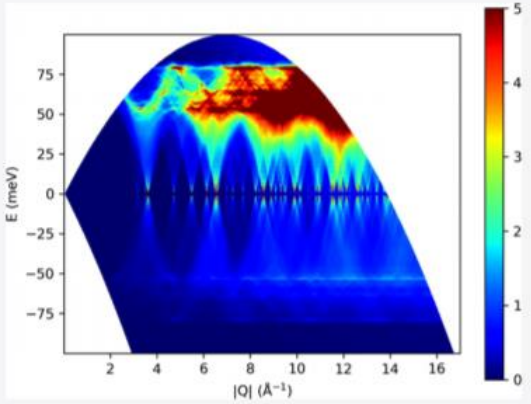
ABSTRACT: The thermal neutron scattering cross-section of a solid depends on the energy (or wavelength) of the incident neutrons. Devising a method to calculate the energy dependence from first principles, without the approximations built in the scattering theory, has been a major undertaking in nuclear engineering. Here, we demonstrate such a calculation method using the program OCLIMAX. Our approach eliminates various approximations and limitations involved in a regular calculation with the LEAPR module of NJOY code, and the results are compared with available experimental and theoretical data. It is also demonstrated how additional insight can be obtained from the calculated full dynamical structure factor. The results reported here show the great potential and excellent platform provided by OCLIMAX for future development in the study of neutron thermalization in solid materials for different applications.

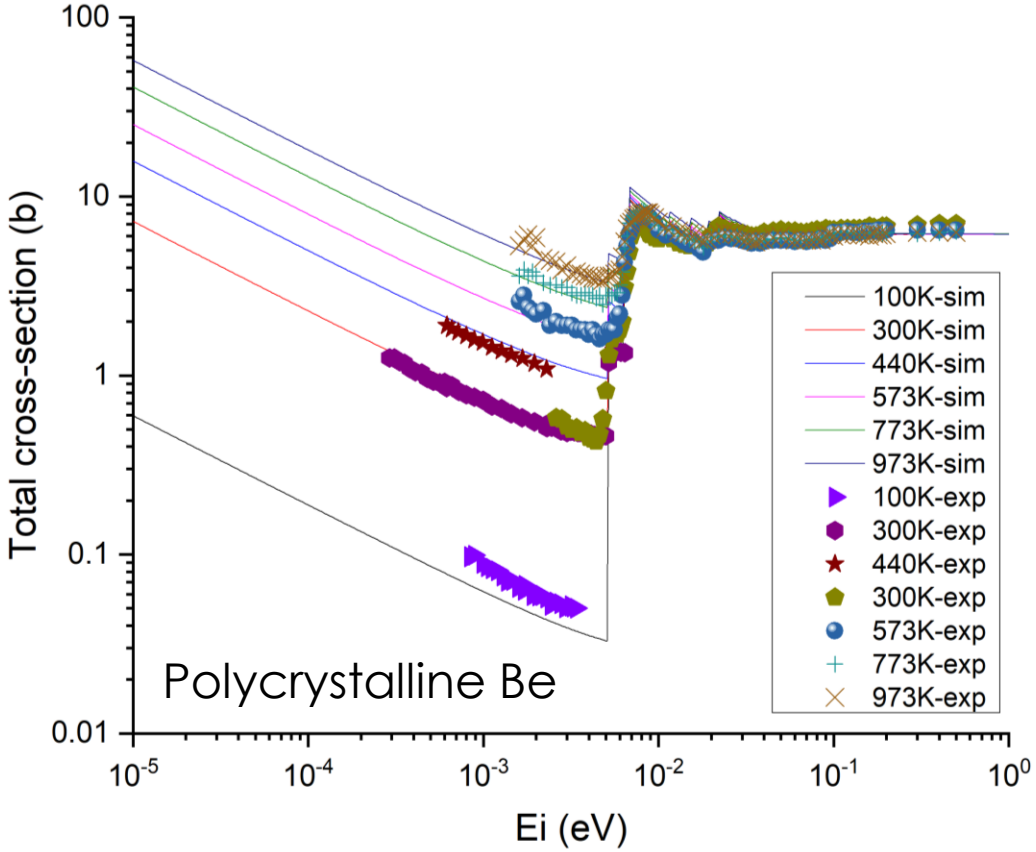


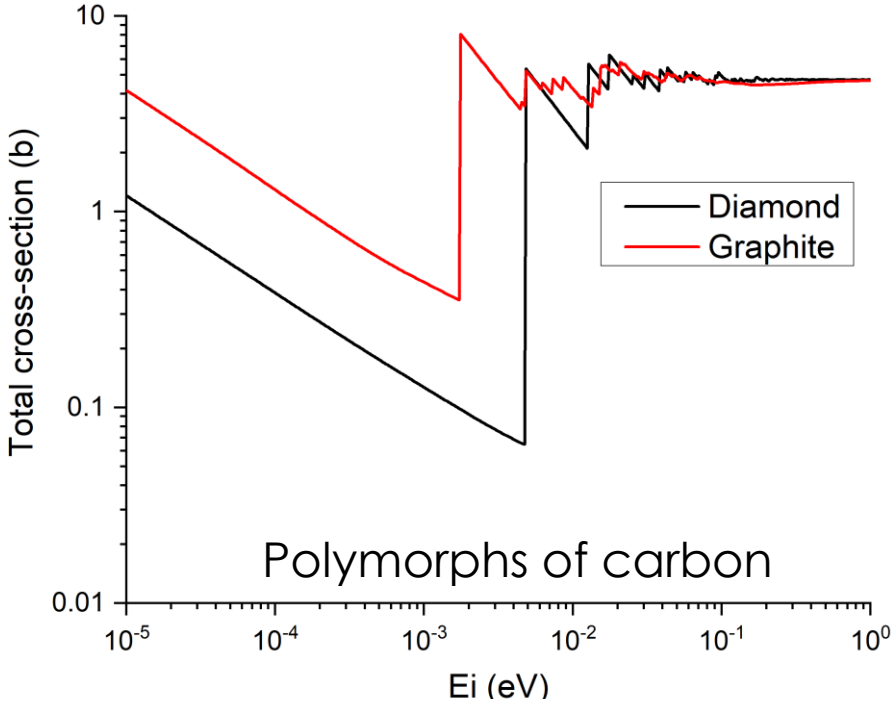
Cover image:
Jill Hemman

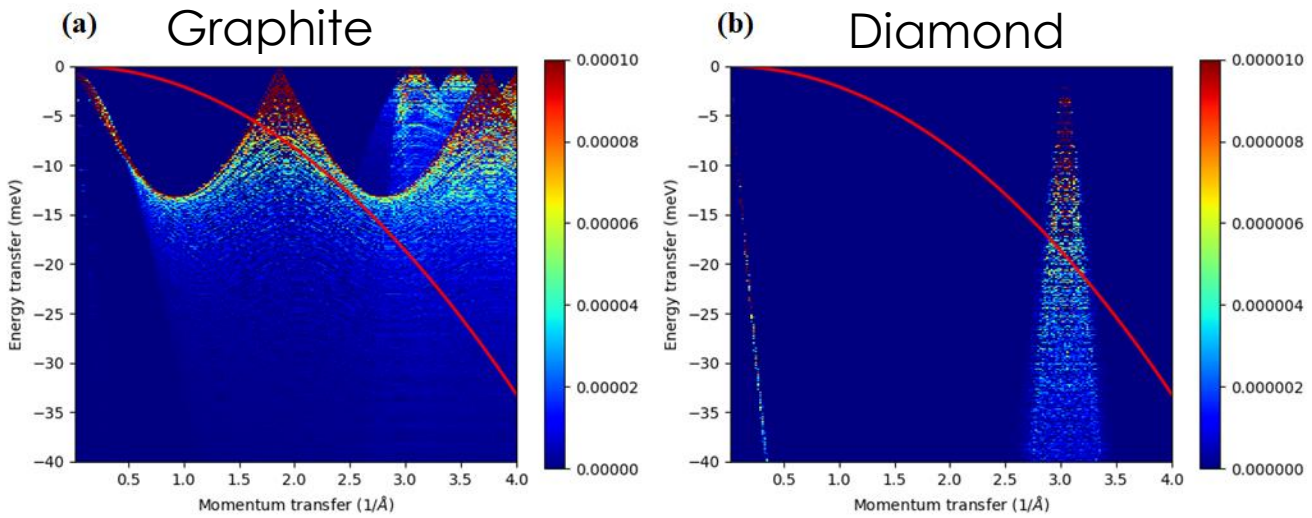
OAK RIDGE National Laboratory | HIGH FLUX ISOTOPE REACTOR | SPALLATION NEUTRON SOURCE

Ab initio thermal neutron scattering cross-section of solids

- ✓ DFT → XS, no intermediate model/parameter
- ✓ Incoherent approximation lifted
- ✓ Computational screening and nuclear material database
- ✓ Insight on dynamical mechanism



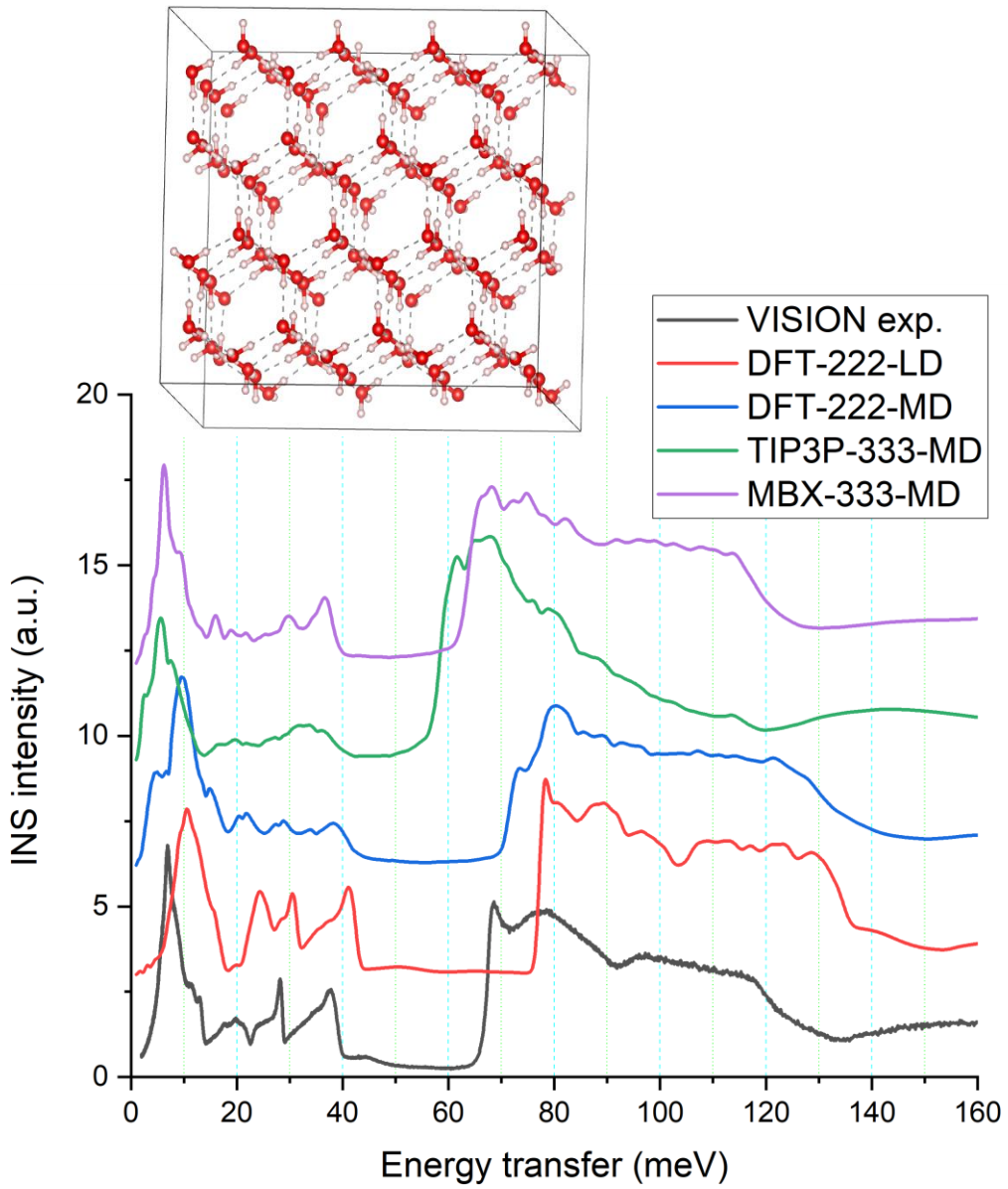




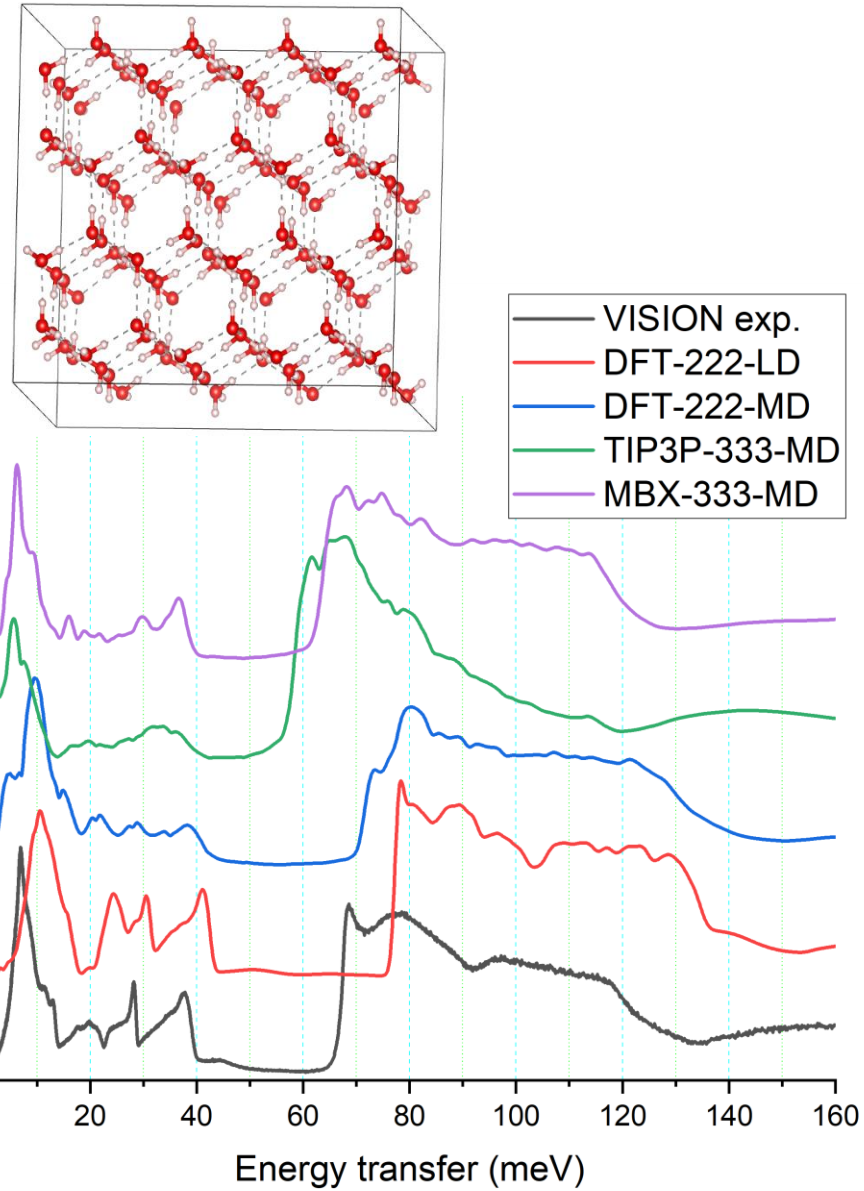
MD trajectories to INS: Motivations

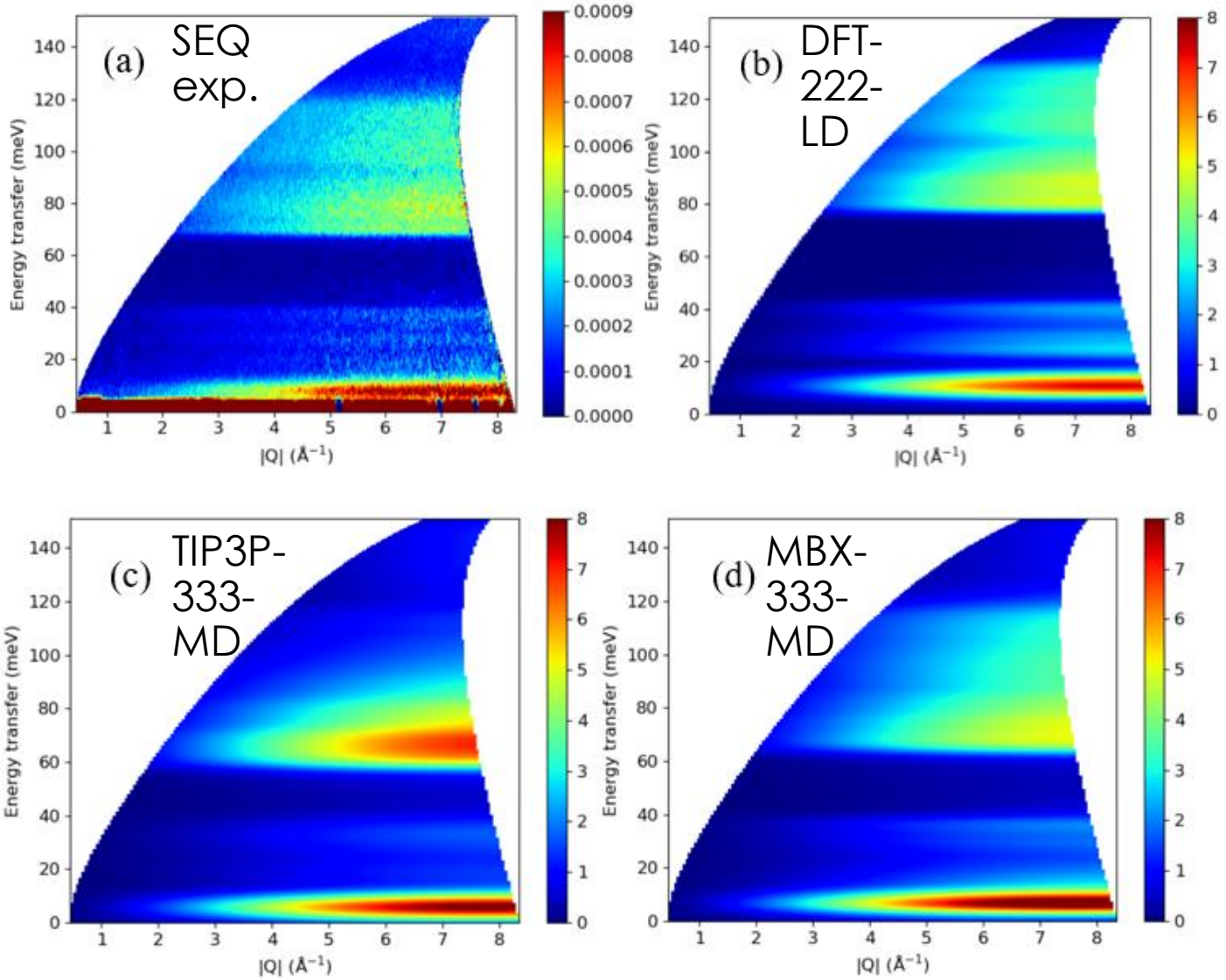
- MD simulations are more routinely and efficiently done than LD simulations, especially for large systems (defective, disordered, amorphous, polymeric, biological, or simply complex).
- An incomplete LD simulation is useless, but an "incomplete" MD simulation can still provide useful information.
- MD simulation requires much less accuracy (in terms of convergence criteria for energy and force evaluation) to achieve comparable quality of INS spectra.
- MD simulation naturally includes temperature-dependent anharmonicity

MD trajectories to INS: Example (ice Ih)



MD trajectories to INS: Example (ice Ih)





Summary and future work

Summary and future work

- Data acquisition and reduction
  - NSE: Recently developed DrSPINE addressed previous issues
  - NSE: Upcoming transition to EPICS
- Data analysis
  - BASIS: QCLIMAX is a major new development with many desired features such as global fitting and constraints.
- Data interpretation (simulation)
  - NSE/BASIS: MD simulation is the most relevant modeling technique; significant development is needed in order to better integrate MD simulations with NSE/BASIS experiments.
  - VISION: OCLIMAX has evolved to become a powerful tool for general vibrational INS simulations.

Questions?

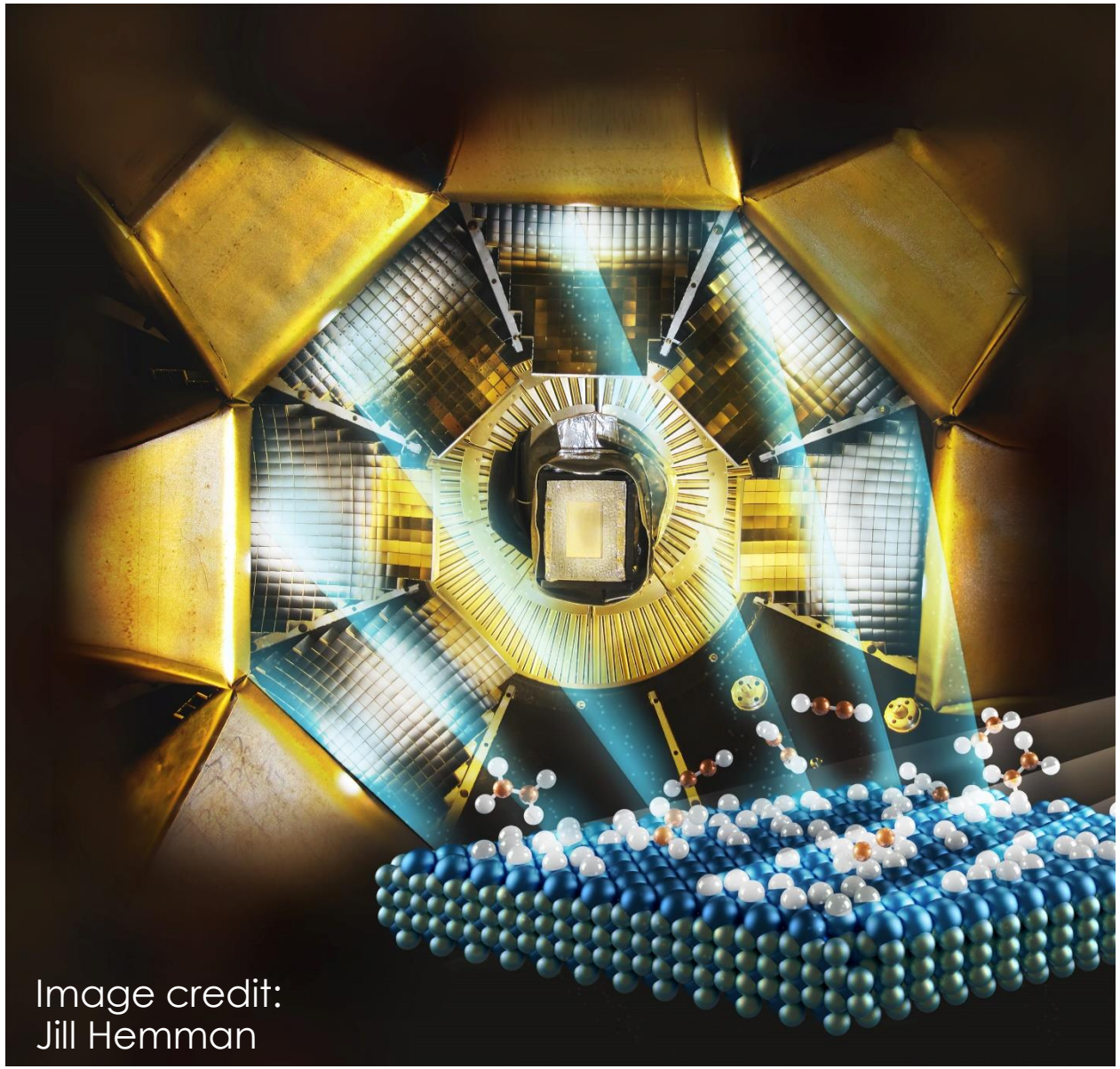


Image credit:
Jill Hemman

chengy@ornl.gov

How to obtain OCLIMAX

- Install Docker (https://www.docker.com/)
- Install OCLIMAX
  - For Linux/Mac (or Virtual Box on Windows)

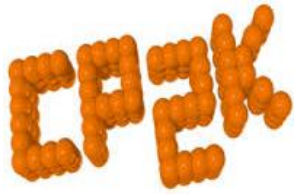
    Open a terminal, run:
    $ curl -sL https://sites.google.com/site/ornliceman/getoclimax | bash
    $ oclimax pull
  - For Windows (Native Windows 10)

    Visit https://sites.google.com/site/ornliceman/download
    Download oclimax.bat to your working directory
    Open the cmd prompt or Powershell prompt, go to the working directory, run:
    $ oclimax.bat pull
- For more information

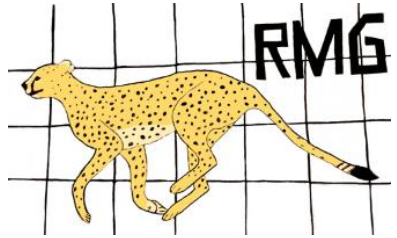
  Download the user manual at
  https://sites.google.com/site/ornliceman/download

Convert your files to OCLIMAX input file

- Automatically extract phonon frequencies and polarization vectors (or MD trajectories) from your DFT output files and generate the input file for OCLIMAX
- Currently support







CASTEP, VASP, Phonopy, CP2K, Quantum Espresso, Crystal, Gaussian, ORCA, NWChem, DMol3, RMG, LAMMPS

e.g., $ oclimax convert -c yourfile.phonon

How to run OCLIMAX

- Input file options for OCLIMAX:

  *.oclimax (full crystal information) *.aclimax (xyz, can be molecules) *.tclimax (MD trajectories)
  *.params (parameters that define the calculation)

- By default, OCLIMAX calculates VISION spectra with standard parameters. To do this, run:

  $ oclimax run yourfile.oclimax

- The output files of this run include:

  yourfile*.csv: The simulated INS spectra or other results
  yourfile.params: The (default) parameters used for this calculation

- To run the simulation with different parameters, you may edit the parameter file, and then run

  $ oclimax run yourfile.oclimax yourfile.params

Parameters for OCLIMAX calculation

```
## General parameters
TASK    =        0  # 0:inc approx. 1:coh+inc. 2:s-xtal Q-E. 3:s-xtal Q-Q
INSTR   =        0  # 0:VISION 1:indirect 2:direct 3:S(Q,E) or S(Qx,Qy) map
TEMP    =     0.00  # Temperature [K]
E_UNIT  =        0  # Energy unit [eu] (0:cm-1,1:meV,2:THz)
OUTPUT  =   0  # 0:standard, 1:restart, 2:SPE, 3:full, 4:DOS, 5:modes, 6:S(Q)

## Additional general parameters
MAXO    =       10  # Maximum order of excitation
CONV    =        2  # Start convolution from order=CONV (2 or 3)
PHASE   =        0  # Phase factor of polarization vector (0 or 1)
MASK    =        0  # Set 1 to apply mask on S(Q,E)/S(Qx,Qy) map (INSTR=3)
ELASTIC =  -0.10E+01 -0.10E+01  # e1>0:add el in S(Q,E),e2>0:S(Q) res

## E parameters
MINE    =     8.00  # Energy range (minimum) to calculate [eu]
MAXE    =   5000.00  # Energy range (maximum) to calculate [eu]
dE      =    1.000  # Energy bin size [eu]
ECUT    =    8.000  # Exclude modes below this cutoff energy [eu]
ERES    =   0.25E+01  0.50E-02  0.10E-06  # E resolution coeff

## Q parameters
MINQ    =     0.50  # Q range (minimum) to calculate
MAXQ    =    20.00  # Q range (maximum) to calculate
dQ      =     0.50  # Q bin size
QRES    =  -0.10E+01 # Q resolution coeff

## Instrument parameters
THETA   =  135.0  45.0  # List of scattering angles [degree]
Ef      =    32.00  # Final energy [eu] (INSTR=1)
Ei      =   5000.00  # Incident energy [eu] (INSTR=2)
L1      =    11.60  # L1 [m] for DGS (INSTR=2 or 3, ERES=0)
L2      =     2.00  # L2 [m] for DGS (INSTR=2 or 3, ERES=0)
L3      =     3.00  # L3 [m] for DGS (INSTR=2 or 3, ERES=0)
dt_m    =     3.91  # dt_m [us] for DGS (INSTR=2 or 3, ERES=0)
dt_ch   =     5.95  # dt_ch [us] for DGS (INSTR=2 or 3, ERES=0)
dL3     =     3.50  # dL3 [cm] for DGS (INSTR=2 or 3, ERES=0)

## Single crystal parameters
HKL     =    0.0   0.0   0.0  # HKL (TASK=2 or 3)
Q_vec   =    0.0   0.0   1.0  # Q vector dir (TASK=2 or 3)
Q_vec_y =    1.0   0.0   0.0  # Q vector dir y-axis (TASK=3)
MINQ_y  =     1.00  # Q range (minimum) y-axis (TASK=3)
MAXQ_y  =     2.00  # Q range (maximum) y-axis (TASK=3)
dQ_y    =     0.02  # Q bin size y-axis (TASK=3)

## Wing parameters
WING    =        0  # Wing calculation (0:no wing,1:isotropic,2:ST tensor)
A_ISO   =   0.0350  # Isotropic A_external for wing calculation
W_WIDTH =    150.0  # Energy width [eu] of initial wing
```

- **Powder and single crystal**
- **Coherent and incoherent**
- **Temperature effect**
- **Wing calculation for single molecules**
- **Instrument geometry and resolution**
- **Various output formats**

Ab initio thermal neutron scattering cross-section of solids

$$\sigma_{total}(E_i) = \sigma_{sc}(E_i) + \sigma_{abs}(E_i)$$

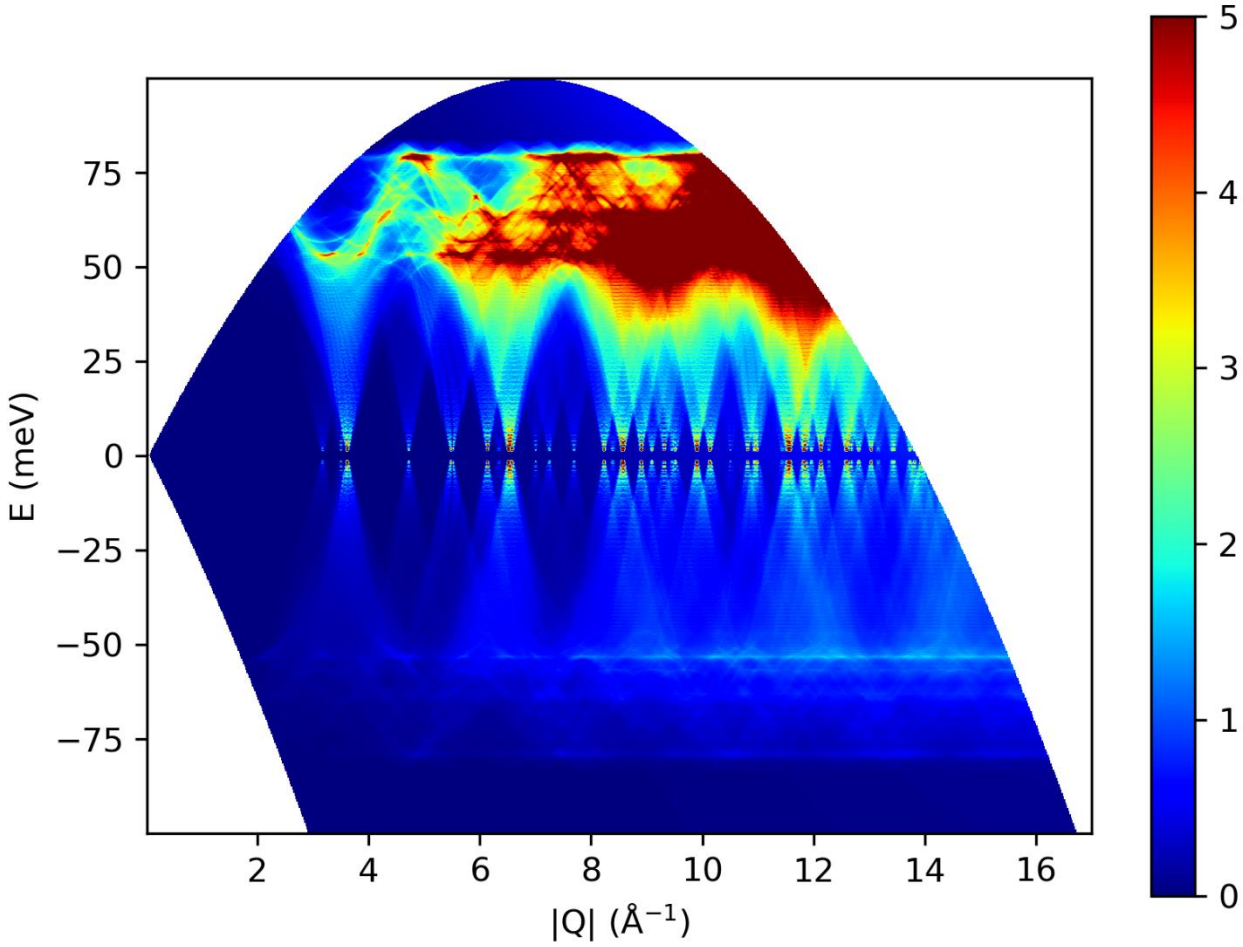
$$\sigma_{abs}(E_i) = \sigma_{abs}(25.30meV)\sqrt{\frac{25.30meV}{E_i}}$$

$$\sigma_{sc}(E_i) = \sigma_{coh}^{el}(E_i) + \sigma_{inc}^{el}(E_i) + \sigma_{coh}^{inel}(E_i) + \sigma_{inc}^{inel}(E_i)$$

$$\sigma_{sc}(E_i) = \frac{N}{2k_0^2}\int_0^{\infty} QdQ \int_{\omega_{min}}^{\omega_{max}} S(Q,\omega)d\omega$$

$$\omega_{max}(Q,k_0) = \frac{\hbar k_0 Q}{m}(1 + \frac{Q}{2k_0})$$

$$\omega_{min}(Q,k_0) = \frac{\hbar k_0 Q}{m}(1 - \frac{Q}{2k_0})$$



MD trajectories to INS: More examples