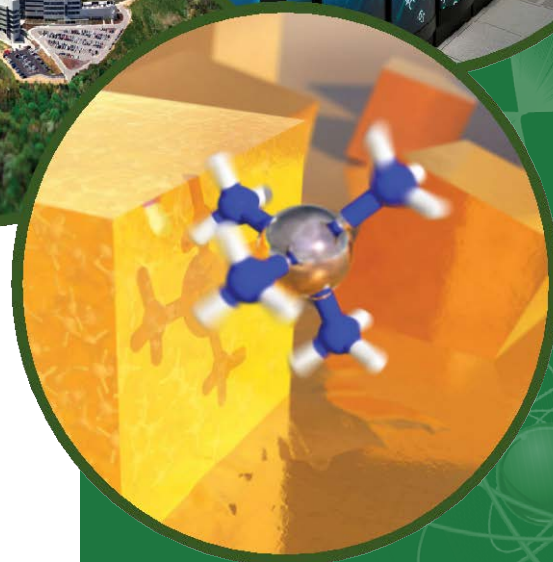


# Simulation of inelastic neutron scattering

2018 National School on  
Neutron and X-ray Scattering

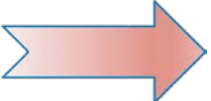
Yongqiang (YQ) Cheng

Spectroscopy Group  
Neutron Scattering Division  
Oak Ridge National Laboratory



# Why do we need simulations for inelastic neutron scattering (INS)?

- 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 *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

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

$V$ : potential describing the interaction between neutrons and the system

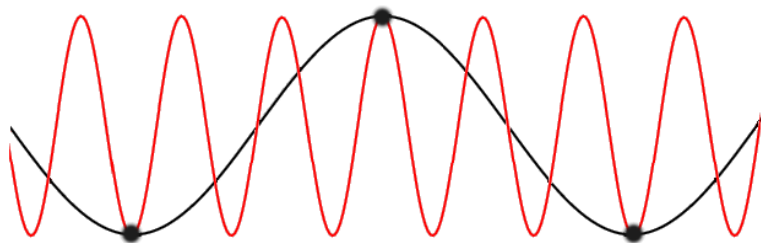
$\hbar\omega$  : fundamental excitation in the system

- 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.

# Two types of scattering

- Nuclear scattering: exchange of energy and momentum between neutrons and **phonons**
- Magnetic scattering: exchange of energy and momentum between neutrons and **magnons**

Phonons	Magnons
Fundamental excitation of atomic vibration	Fundamental excitation of spin wave
Energy vs atomic displacement	Energy vs spin orientation



- **INS** measures at what  $E$  and  $Q$  such excitation exists, as well as its magnitude.

# Inelastic nuclear scattering



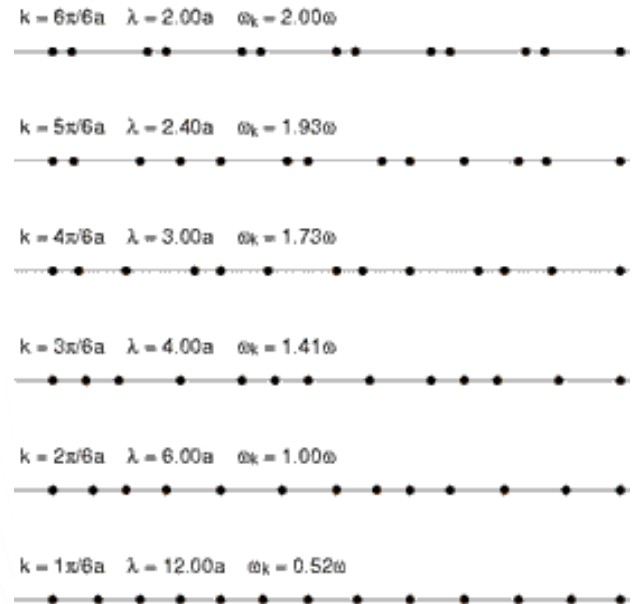
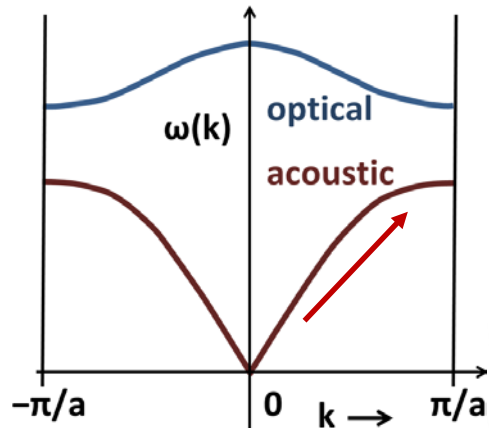
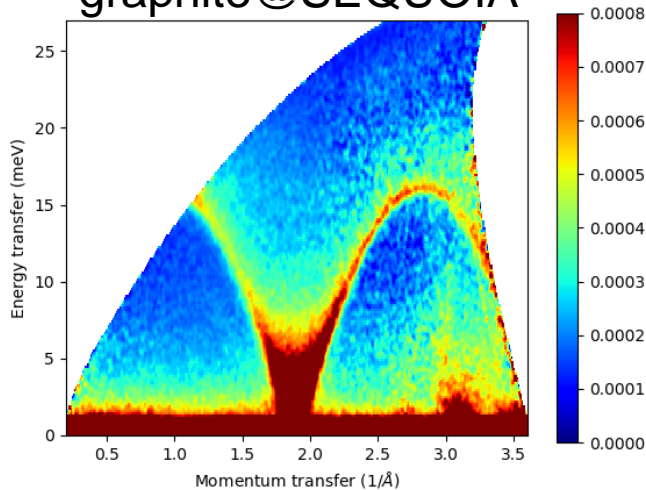


# Coherent inelastic scattering

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

$$S_{coh\pm 1}(\mathbf{Q}, \omega) = \frac{1}{2N} \sum_s \sum_{\tau} \frac{1}{\omega_s} \left| \sum_d \frac{\bar{b}_d}{\sqrt{m_d}} \exp(-W_d) \exp(i\mathbf{Q} \cdot \mathbf{r}_d) (\mathbf{Q} \cdot \mathbf{e}_{ds}) \right|^2 \times \langle n_s + \frac{1}{2} \pm \frac{1}{2} \rangle \delta(\omega \mp \omega_s) \delta(\mathbf{Q} \mp \mathbf{q} - \boldsymbol{\tau})$$

graphite@SEQUOIA



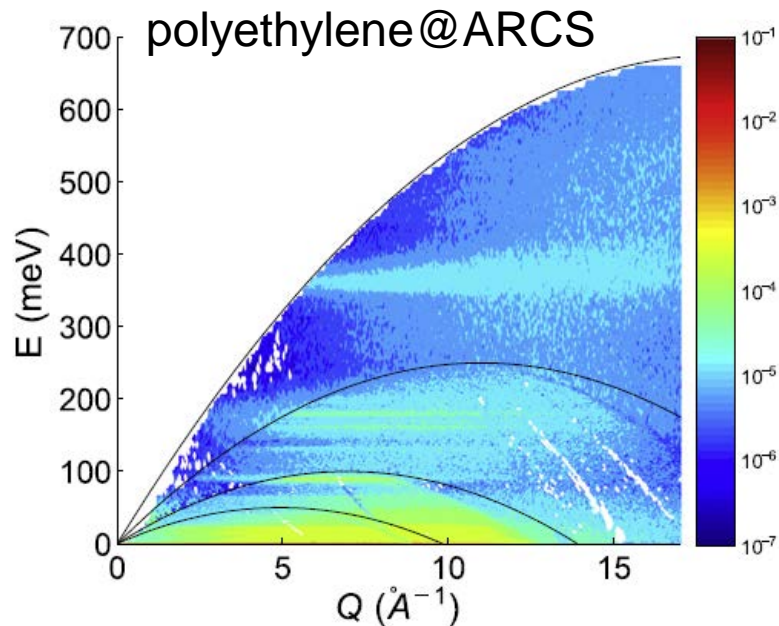
- **Frequency/energy depends on  $Q$ .**
- **Total intensity determined by not only how each atom moves, but also their relative phase.**

From: wikipedia

# Incoherent inelastic scattering

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

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



Coherent



Incoherent

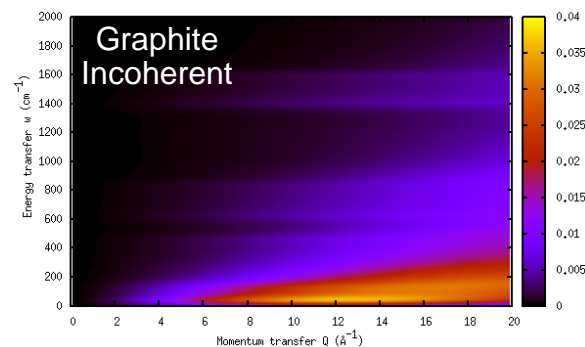
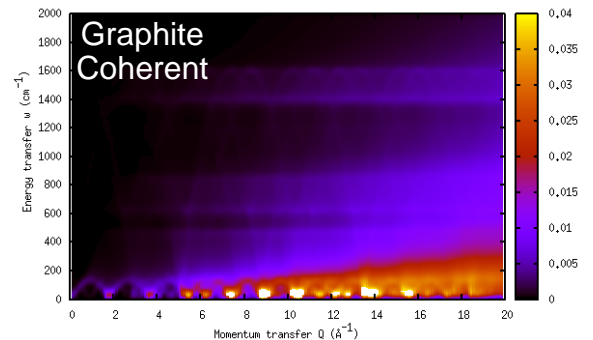
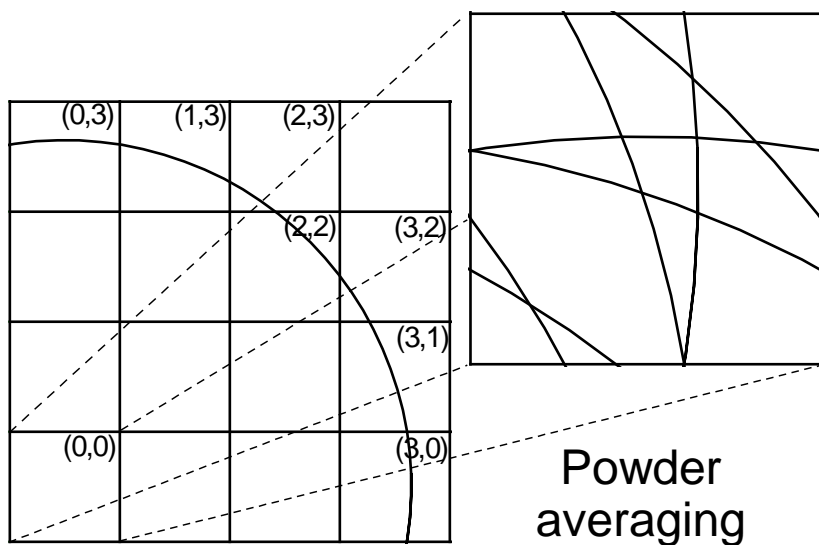
C.M. Lavelle et al. / Nuclear Instruments and Methods  
in Physics Research A 711 (2013) 166–179

- Frequency/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.**

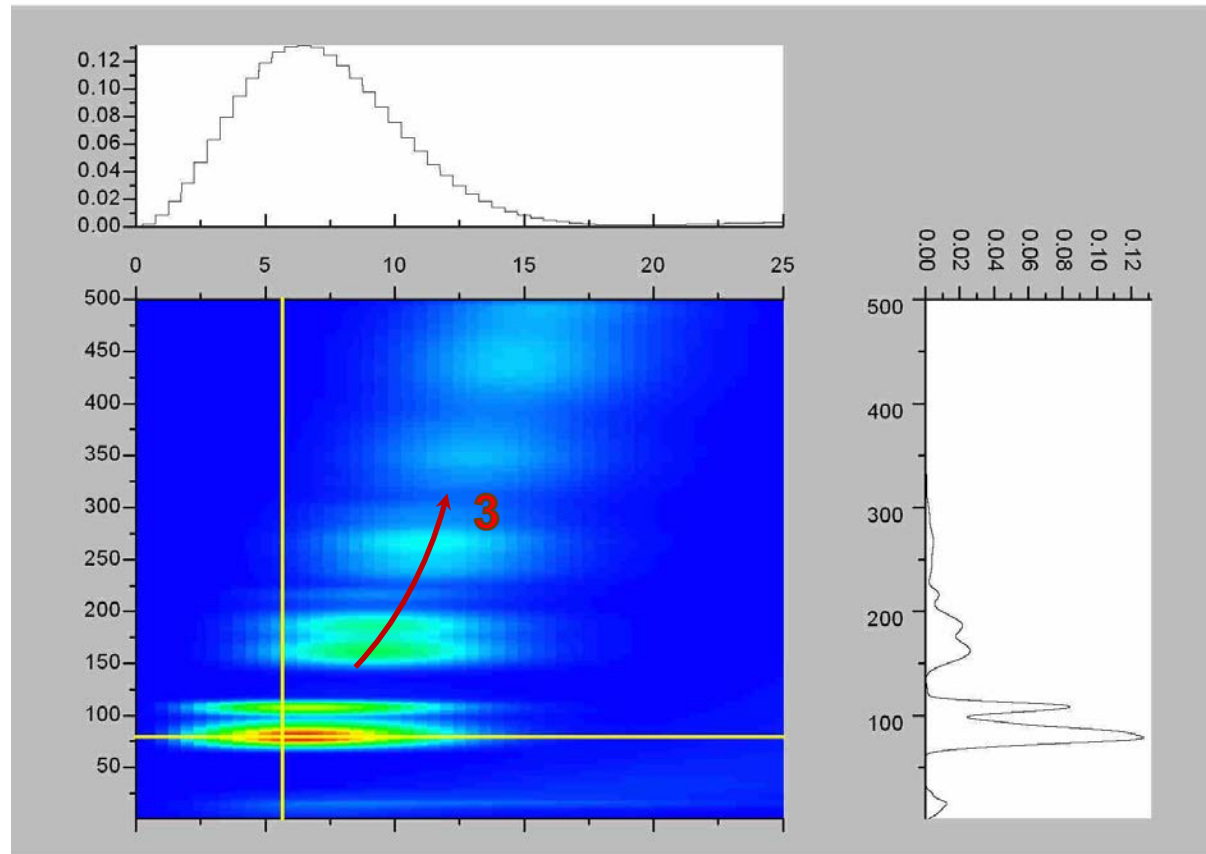
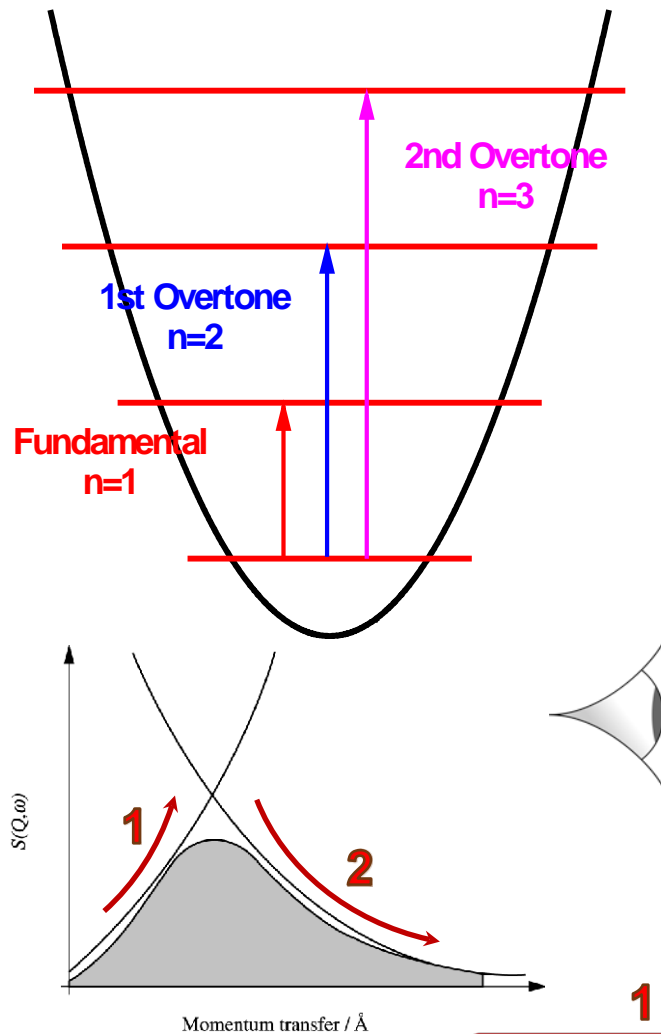


$$S(\mathbf{Q}, n\omega_s) = \frac{(\mathbf{Q} \cdot \mathbf{U}_s)^{2n}}{n!} \exp(-\mathbf{Q} \cdot \mathbf{U}_{total})^2$$

$$\mathbf{U}_s = \sqrt{\frac{\hbar}{2m\omega_s}} \mathbf{e}_{ds}$$



# The $S(Q, \omega)$ map: what to expect



Courtesy of Timmy Ramirez-Cuesta

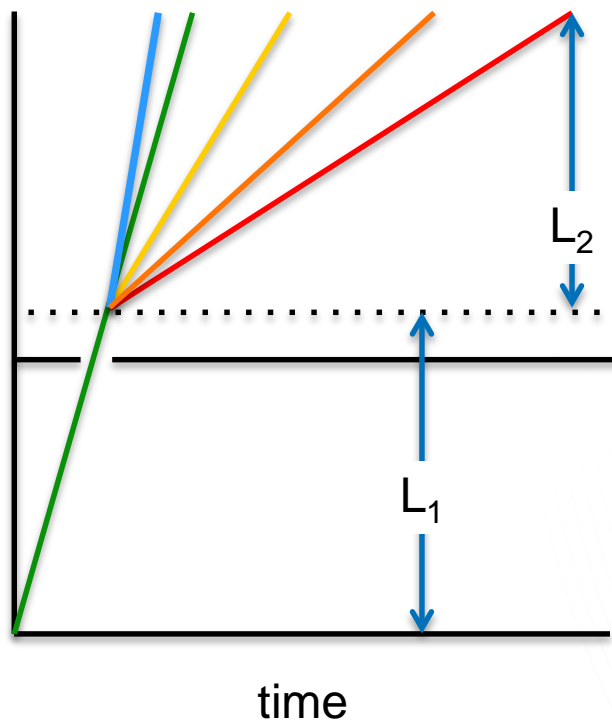
$$S(\mathbf{Q}, n\omega_s) = \frac{(\mathbf{Q} \cdot \mathbf{U}_s)^{2n}}{n!} \exp(-\mathbf{Q} \cdot \mathbf{U}_{total})^2$$

$$\mathbf{U}_s = \sqrt{\frac{\hbar}{2m\omega_s}} e^{ds}$$

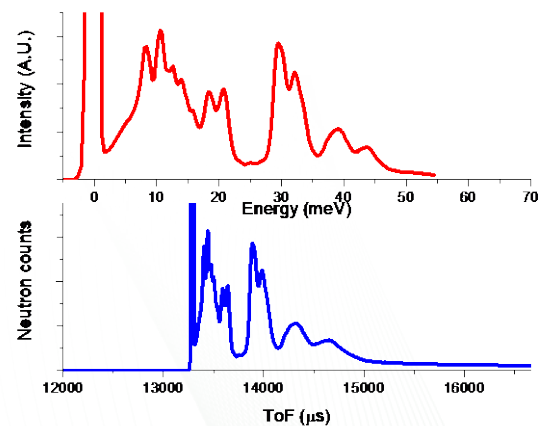
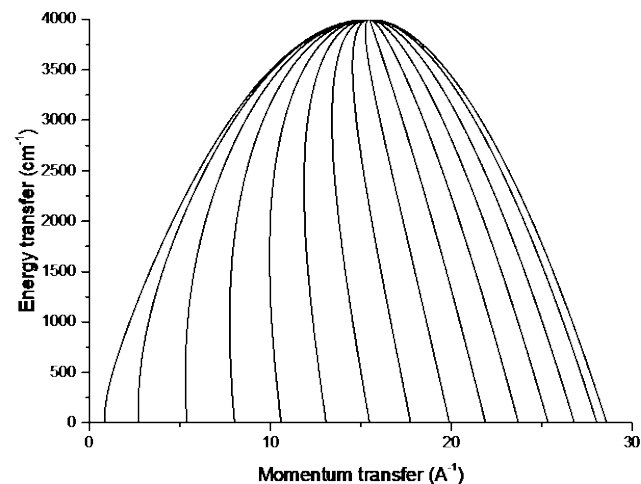
# Instrument geometry: direct



Examples: ARCS, CNCS, HYSPEC, SEQUIOA

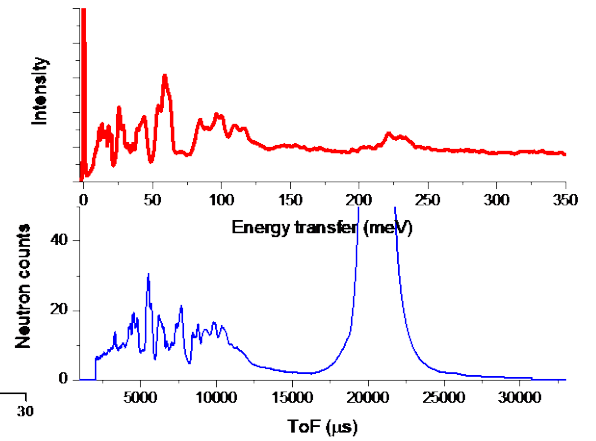
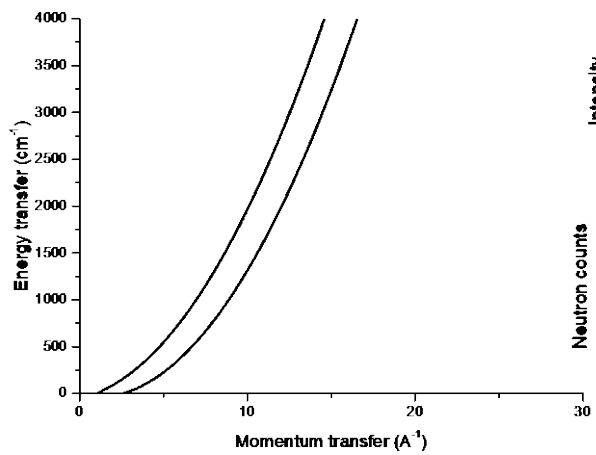
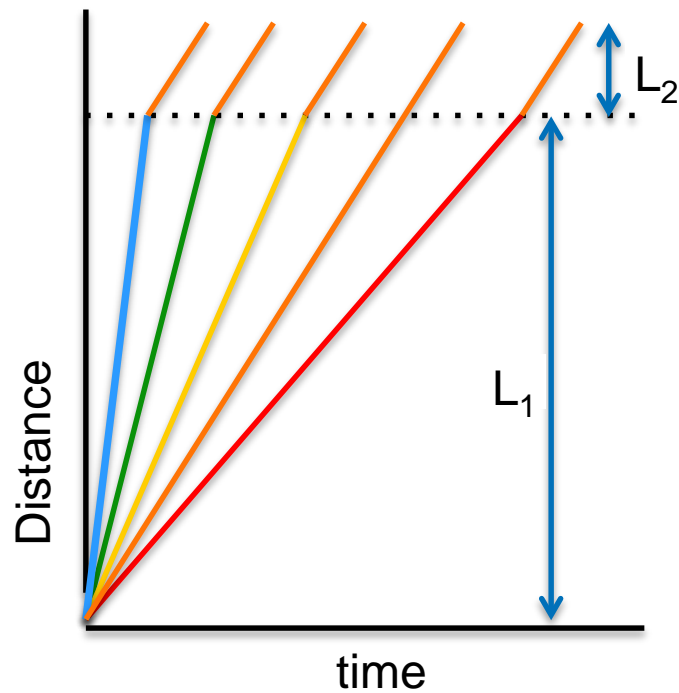


Fixed incident energy, measure final energy.



Resolution is almost a constant fraction of incident energy

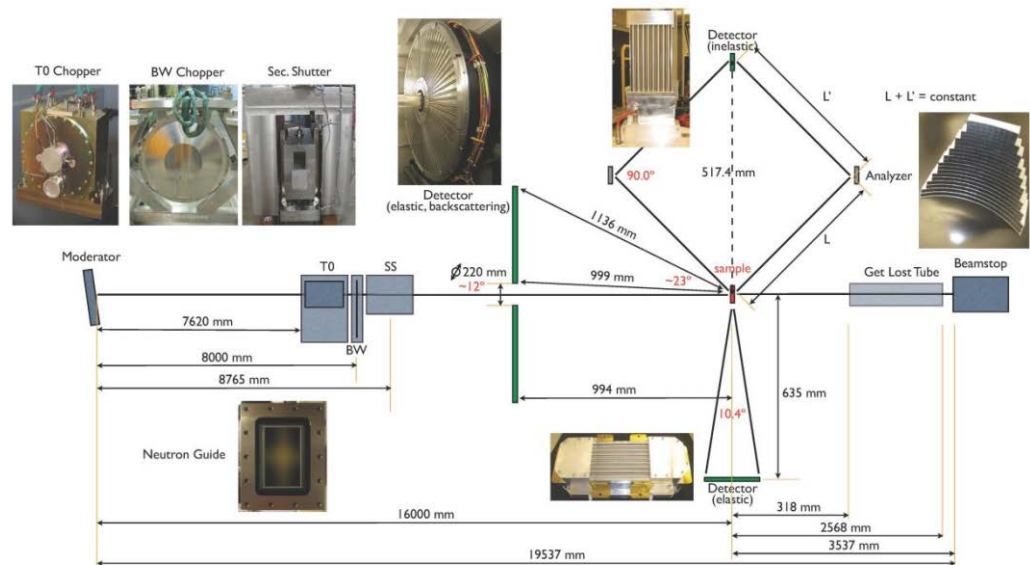
# Instrument geometry: indirect



Resolution is almost a constant fraction of energy transfer

White incident beam,  
fixed final energy.

Examples: VISION, TOSCA



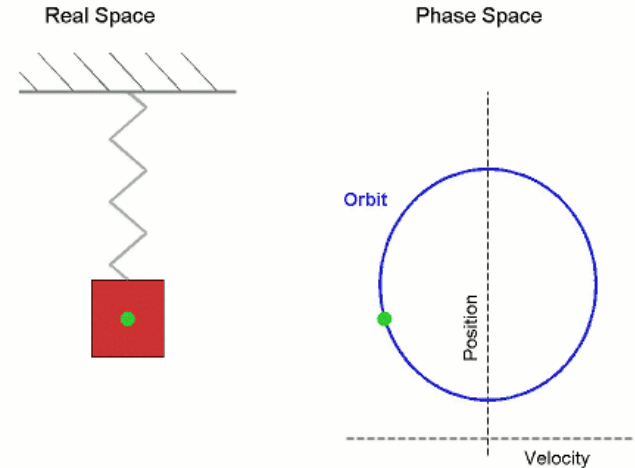
# Calculation of phonons



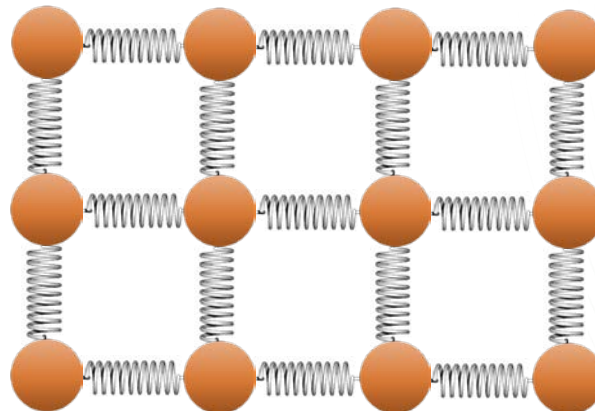
# Harmonic approximation

- Potential energy/force as a function of displacement
- Harmonic oscillator:

$$F = -kx = m\ddot{x}$$
$$x = Ax^{i\omega t} \quad \omega = \sqrt{\frac{k}{m}}$$



- Harmonic oscillators are non-interacting
- How to describe the vibration of atoms in a solid? A network of harmonic oscillators (harmonic approximation)





# Force constants and dynamical matrix

- Expansion of potential energy

$$E = \Phi_0 + \sum_a \sum_i \Phi_{ai} U_{ai} + \frac{1}{2!} \sum_{ab} \sum_{ij} \Phi_{aibj} U_{ai} U_{bj} + \frac{1}{3!} \sum_{abc} \sum_{ijk} \Phi_{aibjck} U_{ai} U_{bj} U_{ck} + \dots$$

$$\Phi_{aibj} = \frac{\partial^2 E}{\partial U_{ai} \partial U_{bj}} \quad \text{force constants}$$

$a, b, c$ : atom labels  
 $i, j, k$ : cartesian components  
 $U$ : displacement

Plane-wave solution: 
$$U_{ai} = \frac{1}{\sqrt{m_a}} e_{ai}(\mathbf{q}) \exp(i\mathbf{q} \cdot \mathbf{R}_{ai} - \omega t)$$

$$\sum_{bj} \Phi_{aibj} U_{bj} = F_{ai} = m_a \ddot{U}_{ai}$$

$$\text{Dynamical matrix: } D_{aibj}(\mathbf{q}) = \frac{\Phi_{aibj}}{\sqrt{m_a m_b}} \exp[i\mathbf{q} \cdot (\mathbf{R}_{bj} - \mathbf{R}_{ai})]$$

# Frequencies and polarization vectors

- Diagonalization of dynamical matrix

$$D(\mathbf{q})\mathbf{e}_s(\mathbf{q}) = \omega_s^2(\mathbf{q})\mathbf{e}_s(\mathbf{q})$$

$$\mathbf{e}_s(\mathbf{q}) = [\sqrt{m_a}U_{ai}, \sqrt{m_a}U_{aj}, \sqrt{m_a}U_{ak}, \dots, \sqrt{m_b}U_{bi}, \sqrt{m_b}U_{bj}, \sqrt{m_b}U_{bk} \dots]^T$$

$$\sum_{ai} e_{ais}(\mathbf{q})^* e_{ais'}(\mathbf{q}) = \delta_{ss'}$$

$$\sum_s e_{ais}(\mathbf{q})^* e_{bjs}(\mathbf{q}) = \delta_{ai,bj}$$

- Solving the  $S(\mathbf{Q}, \omega)$

$$S_{coh\pm 1}(\mathbf{Q}, \omega)$$

$$= \frac{1}{2N} \sum_s \sum_{\tau} \frac{1}{\omega_s} \left| \sum_d \frac{\bar{b}_d}{\sqrt{m_d}} \exp(-W_d) \exp(i\mathbf{Q} \cdot \mathbf{r}_d) (\mathbf{Q} \cdot \mathbf{e}_{ds}) \right|^2$$

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

Population of mode  $s$

$$n_s = \frac{1}{\exp\left(\frac{\hbar\omega_s}{k_B T}\right) - 1}$$

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

$\exp(-2W)$  Debye-Waller factor

MSD of atom  $d$

$$W_d^{iso} = \frac{1}{6} Q^2 u_d^2$$

# How to obtain force constant matrix or dynamical matrix – method 1

- Finite displacement  $\sum_{bj} \Phi_{aibj} U_{bj} = F_{ai}$

$$\Delta_{bj} = [0, 0, 0, \dots, 0, U_{bj}, 0 \dots]^T$$

$$\Phi_{aibj} = \frac{F_{ai}}{U_{bj}}$$

- Force can be determined by classical or quantum methods

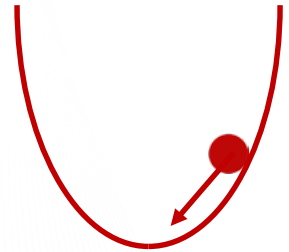
$$\mathbf{F}_I = -\frac{\partial E(\mathbf{R})}{\partial \mathbf{R}_I}$$

Hellman-Feynman Theorem  $\mathbf{F}_I = -\left\langle \Psi \left| \frac{\partial H(\mathbf{R})}{\partial \mathbf{R}_I} \right| \Psi \right\rangle$

$$\mathbf{F}_I = -\int n_{\mathbf{R}}(\mathbf{r}) \frac{\partial V_{e-n}(\mathbf{r})}{\partial \mathbf{R}_I} d\mathbf{r} - \frac{\partial V_{n-n}(\mathbf{R})}{\partial \mathbf{R}_I}$$

$$V_{n-n}(\mathbf{R}) = \frac{e^2}{2} \sum_{I \neq J} \frac{Z_I Z_J}{|\mathbf{R}_I - \mathbf{R}_J|}$$

$$V_{e-n}(\mathbf{R}) = -\sum_{il} \frac{Z_I e^2}{|\mathbf{r}_i - \mathbf{R}_I|}$$



# How to obtain force constant matrix or dynamical matrix – method 2

- Linear response (DFPT)

$$\Phi_{IJ} = \frac{\partial^2 E(\mathbf{R})}{\partial \mathbf{R}_I \partial \mathbf{R}_J} = -\frac{\partial \mathbf{F}_I}{\partial \mathbf{R}_J} = \int \frac{\partial n_{\mathbf{R}}(\mathbf{r})}{\partial \mathbf{R}_J} \frac{\partial V_{e-n}(\mathbf{r})}{\partial \mathbf{R}_I} d\mathbf{r} + \int n_{\mathbf{R}}(\mathbf{r}) \frac{\partial^2 V_{n-n}(\mathbf{R})}{\partial \mathbf{R}_I \partial \mathbf{R}_J} d\mathbf{r} + \frac{\partial^2 V_{n-n}(\mathbf{R})}{\partial \mathbf{R}_I \partial \mathbf{R}_J}$$

- Linearization

$$n(\mathbf{r}) = \sum_i |\psi_i(\mathbf{r})|^2 \quad \Delta n(\mathbf{r}) = 2\text{Re} \sum_i \psi_i(\mathbf{r})^* \Delta \psi_i(\mathbf{r})$$

$$\left[ -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial \mathbf{r}^2} + V_{KS}(\mathbf{r}) - \epsilon_i \right] |\Delta \psi_i\rangle = [-\Delta V_{KS}(\mathbf{r}) - \Delta \epsilon_i] |\psi_i\rangle$$

$$\Delta V_{KS}(\mathbf{r}) = \Delta V(\mathbf{r}) + e^2 \int \frac{\Delta n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' + \left. \frac{dv_{xc}(n)}{dn} \right|_{n=n(\mathbf{r})} \Delta n(\mathbf{r}) \quad \Delta \epsilon_i = \langle \psi_i | \Delta V_{KS}(\mathbf{r}) | \psi_i \rangle$$

Self-consistent solution for  $\frac{\partial n(\mathbf{r})}{\partial \mathbf{R}}$

# How to obtain force constant matrix or dynamical matrix – method 3

- Minimization of the residual

$$\chi^2 = \sum_t \sum_i |F_i(t) - \tilde{F}_i(t)|^2$$

For atom  $i$  in a series of configurations indexed by  $t$ :

$F_i(t)$  : force determined from potential energy

$\tilde{F}_i(t)$  : force determined from displacement using the trial force constants

- Effective force constants to (partially) describe anharmonicity.
- Finite displacement method is a special case.
- Multiple implementations: Alamode<sup>[1]</sup>, TDEP<sup>[2]</sup>, CS<sup>[3]</sup>

1. <http://alamode.readthedocs.io/en/latest/index.html>

2. <http://ollehellman.github.io/index.html>

3. <https://arxiv.org/pdf/1404.5923.pdf>



# Phonon density of states without dynamical matrix

- Velocity autocorrelation

$$\rho(\omega) = \frac{1}{3NTk_B} \int \sum_i \langle \mathbf{v}_i(t) \cdot \mathbf{v}_i(0) \rangle e^{i\omega t} dt$$

- Partial (atomic) density of states

$$S_{inc\pm 1}(Q, \omega) = \sum_d \frac{\sigma_d}{6m_d} Q^2 \exp(-2W_d) \frac{\rho_d(\omega)}{\omega} \left( n + \frac{1}{2} \pm \frac{1}{2} \right)$$

$$W_d = \frac{1}{6} Q^2 u_d^2 \quad n = \frac{1}{\exp\left(\frac{\hbar\omega}{k_B T}\right) - 1}$$

- Incoherent only, isotropic approximation
- Anharmonic effect included
- **Key parameters for simulation: system size, time step, total time, temperature control**

# Inelastic magnetic scattering



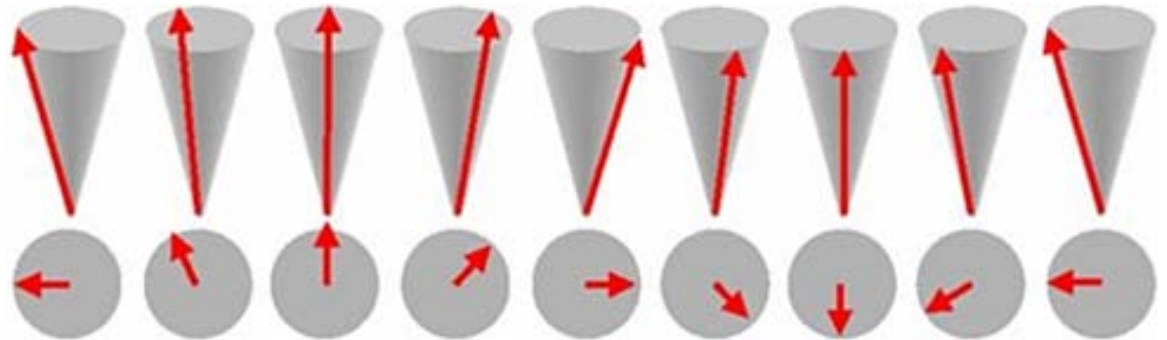
# Magnetic excitation (spin wave)

- Energy as a function of spin orientation

$$H = - \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \quad J(\mathbf{q}) = \sum_{\mathbf{r}} J(\mathbf{r}) \exp(i\mathbf{q} \cdot \mathbf{r})$$

$$H_{\mathbf{q}}|n\rangle = n\hbar\omega_{\mathbf{q}}|n\rangle \quad \hbar\omega_{\mathbf{q}} = 2S[J(0) - J(\mathbf{q})]$$

- Local coupling: low dispersion (softer)
- Long-range coupling: high dispersion (stiffer)



[https://www.uni-muenster.de/imperia/md/images/physik\\_ap/demokritov/research/becforonphysicists/magnon.png](https://www.uni-muenster.de/imperia/md/images/physik_ap/demokritov/research/becforonphysicists/magnon.png)

## Phonons

Fundamental excitation of atomic vibration

Energy vs atomic displacement

## Magnons

Fundamental excitation of spin wave

Energy vs spin orientation

# Inelastic magnetic scattering

- One-magnon processes

$$S_{mag\pm 1}(\mathbf{Q}, \omega)$$

$$\propto S(1 + \hat{Q}_z^2) \left[ \frac{1}{2} gF(\mathbf{Q}) \right]^2 \exp(-2W) \sum_{\tau, \mathbf{q}} \langle n_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2} \rangle \delta(\omega \mp \omega_{\mathbf{q}}) \delta(\mathbf{Q} \mp \mathbf{q} - \boldsymbol{\tau})$$

$$F(\mathbf{Q}) = \int s(\mathbf{r}) \exp(i\mathbf{Q} \cdot \mathbf{r}) d\mathbf{r}$$

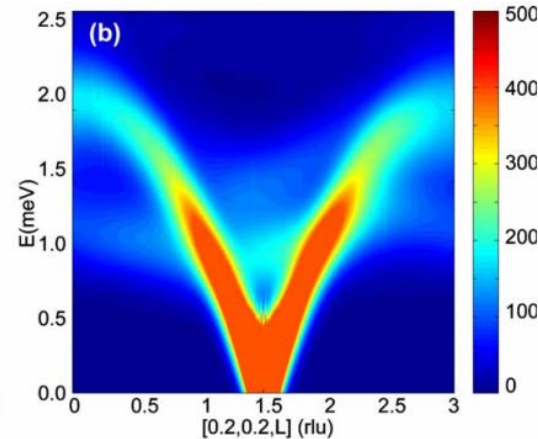
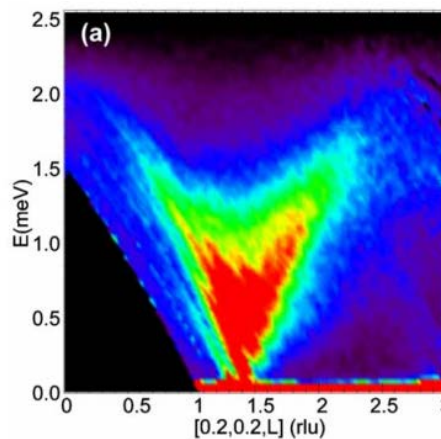
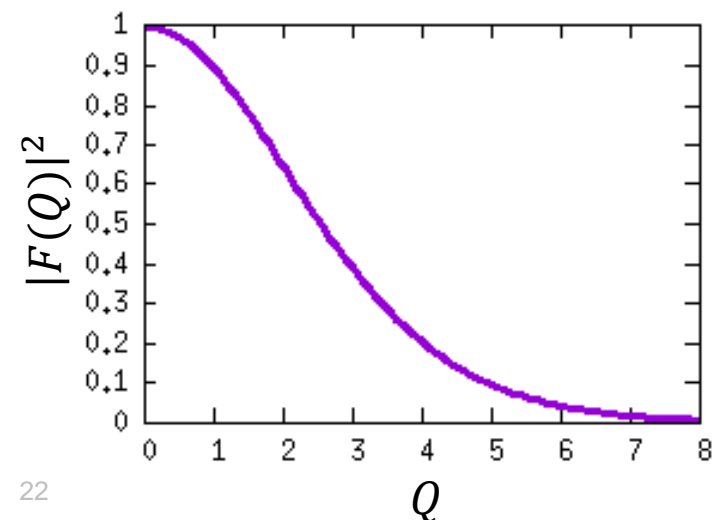
$F(\mathbf{Q})$ : magnetic form factor

$s(\mathbf{r})$ : normalized density of unpaired electrons



<https://www.psi.ch/spinw/spinw>

S. Toth and B. Lake, J. Phys.:  
Condens. Matter 27, 166002 (2015).



J. Haraldsen et al. Phys. Rev. B 82, 020404(R) (2010).

# OCLIMAX: a program for the calculation of inelastic nuclear scattering



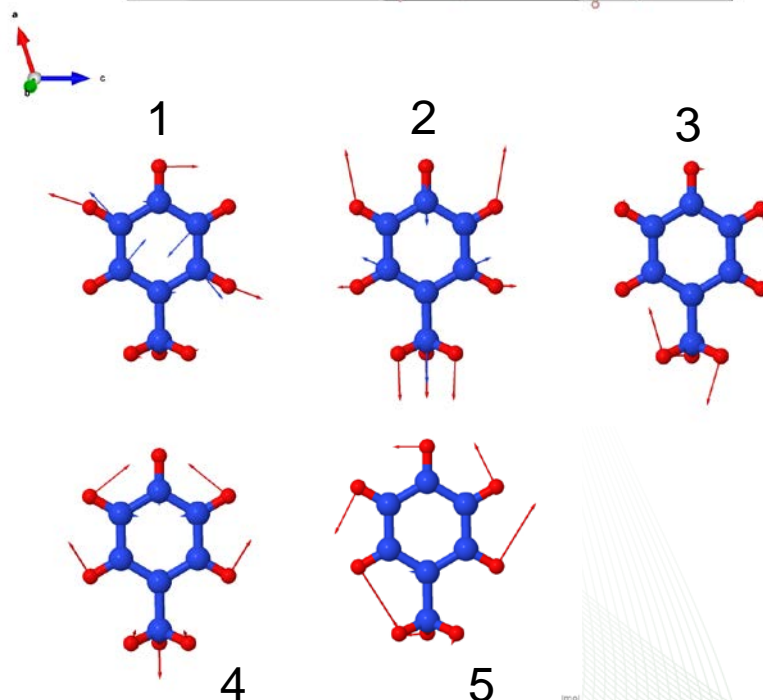
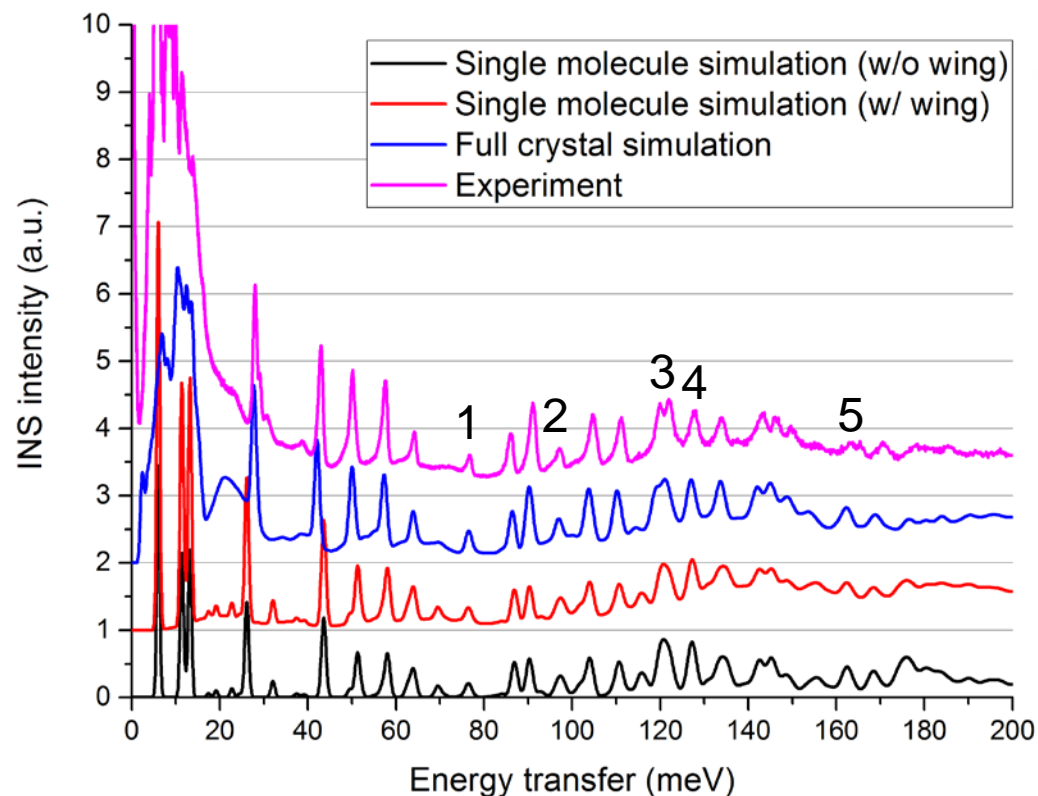
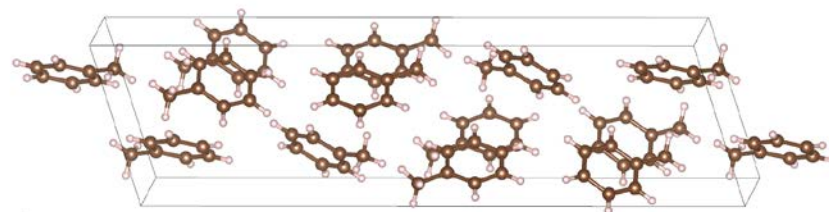
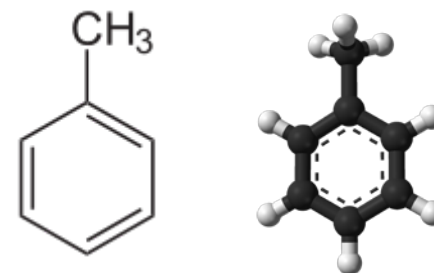


# OCLIMAX: introduction

- INS calculation of powder samples
- Full calculation (including coherent effects) and incoherent approximation
- Combinations and overtones
- Temperature effect
- Phonon wing calculation for single molecules
- Sampling trajectories in  $Q$ - $\omega$  space for indirect and direct geometry instruments
- Flexible ways to determine resolution
- Easy interface with common DFT programs
- Released as a Docker image: no system dependence (supporting Linux, Mac, and Windows), self-contained, easy to install, run, and update

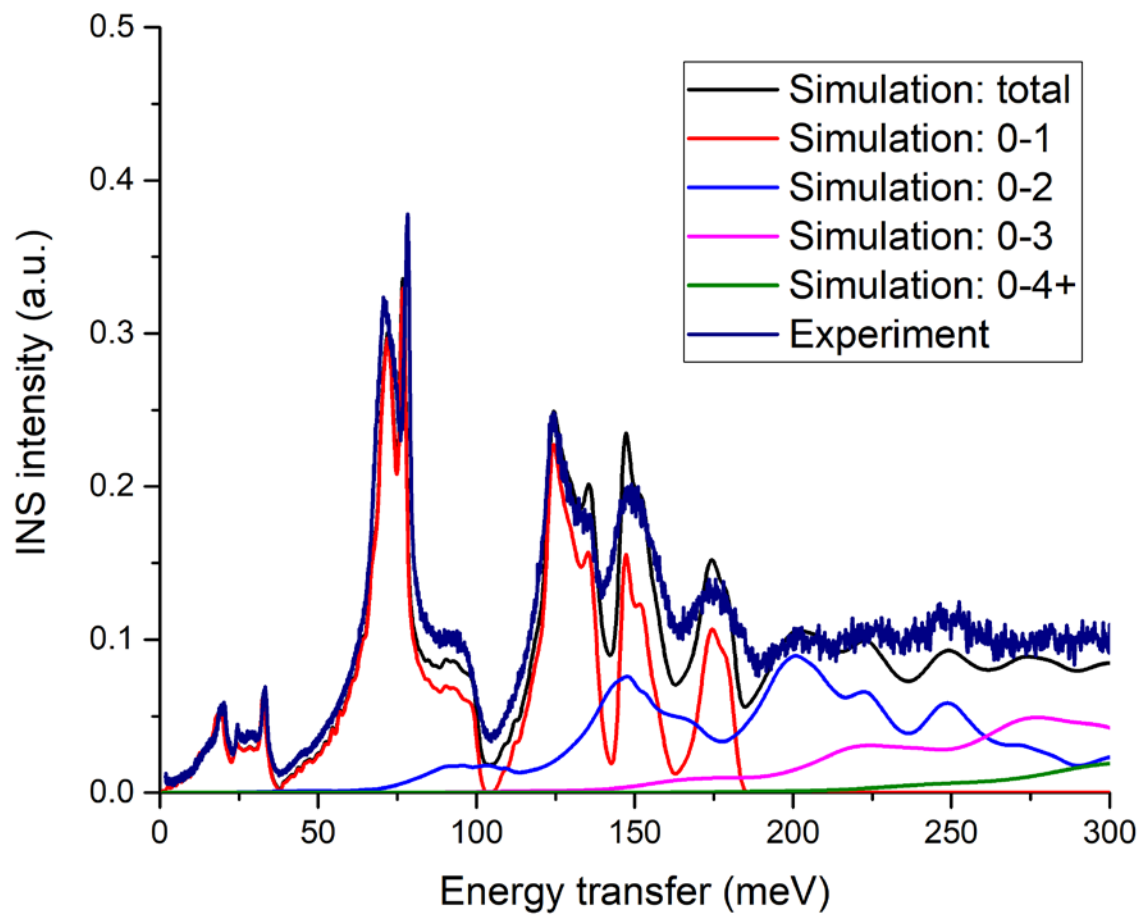
# OCLIMAX example: toluene

- Single molecule
- Wing calculation
- Full crystal calculation
- Role of intermolecular interactions



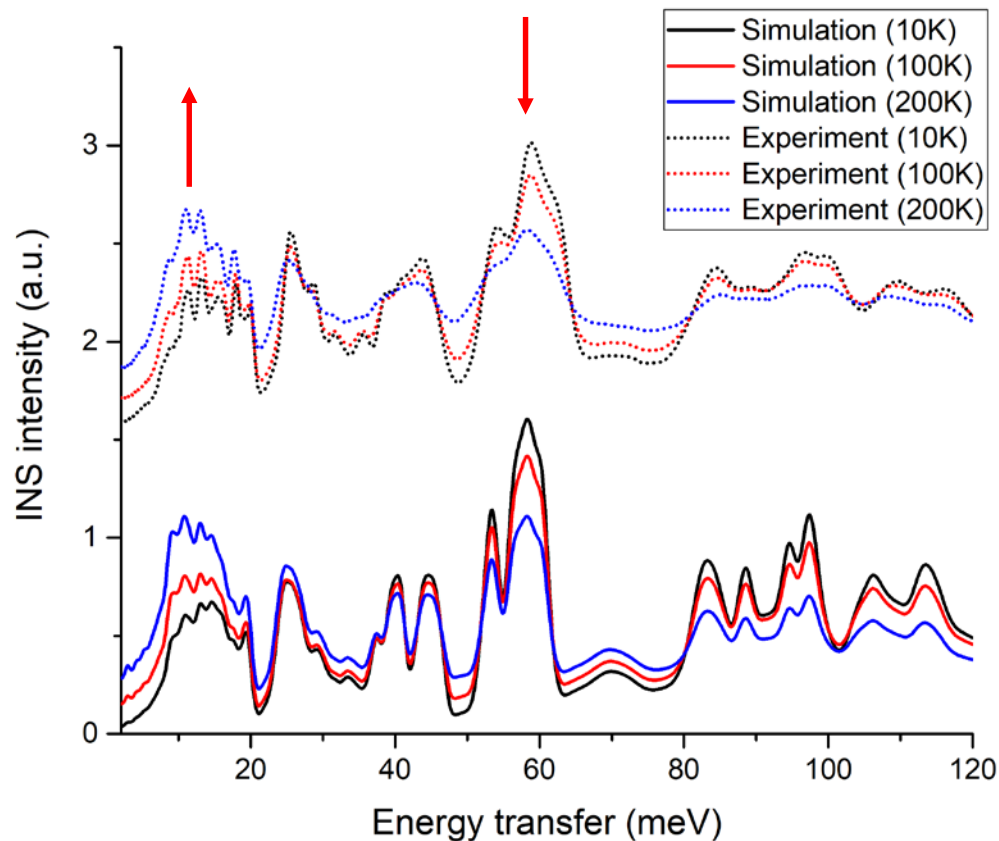
# OCLIMAX example: MgH<sub>2</sub>

- Higher order excitations



# OCLIMAX example: alanate

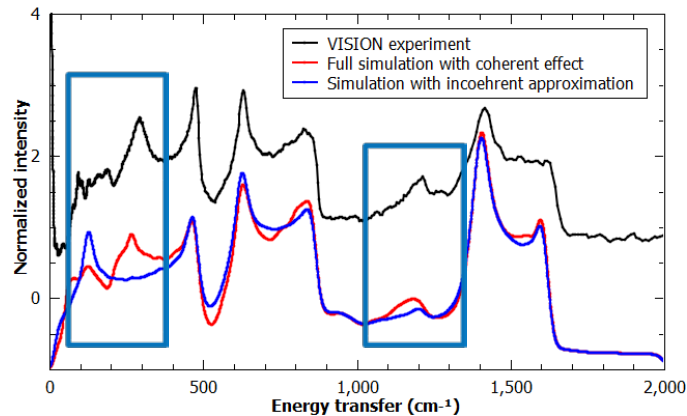
- Temperature effects
  - Phonon population
  - Debye-Waller factor



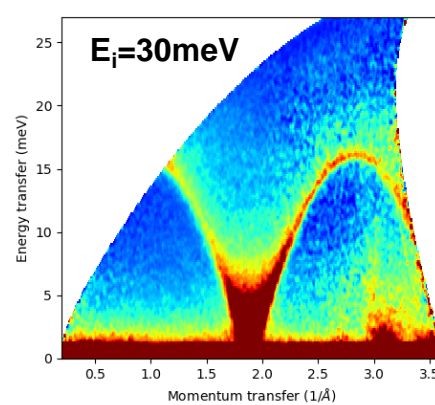
# OCLIMAX example: graphite

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

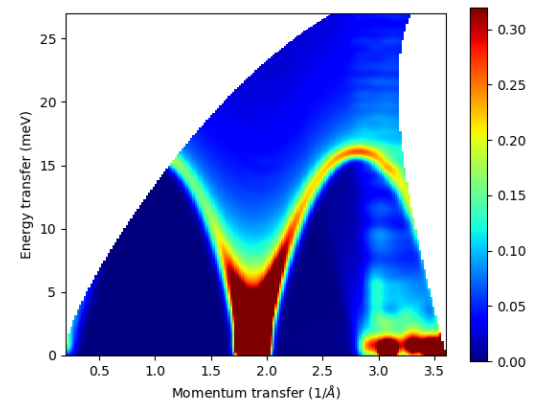
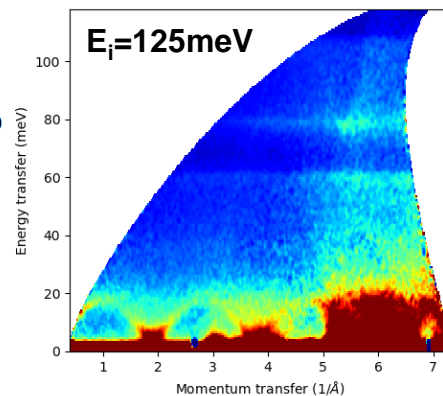
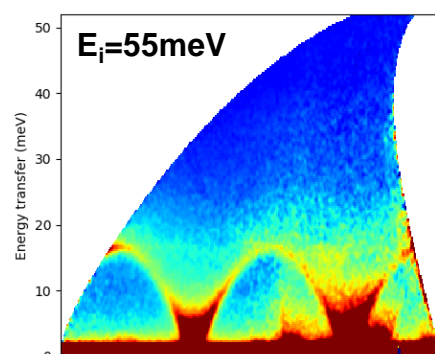
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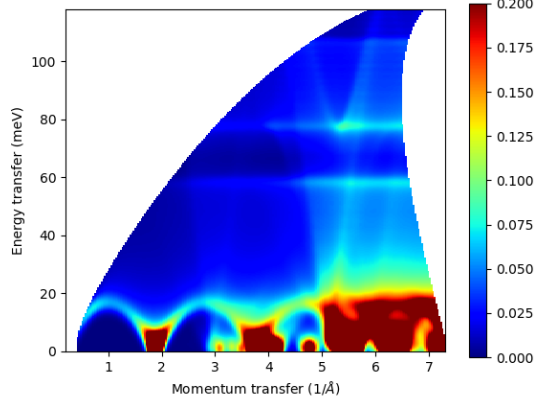
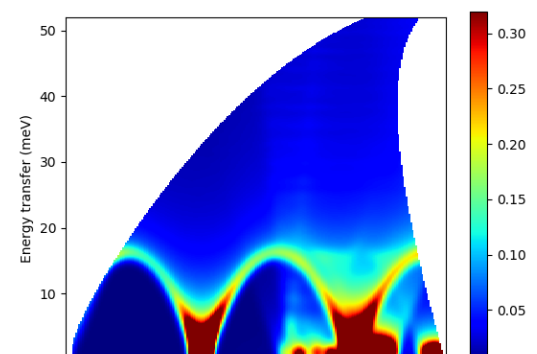
Full calculation versus incoherent approximation



SEQUOIA



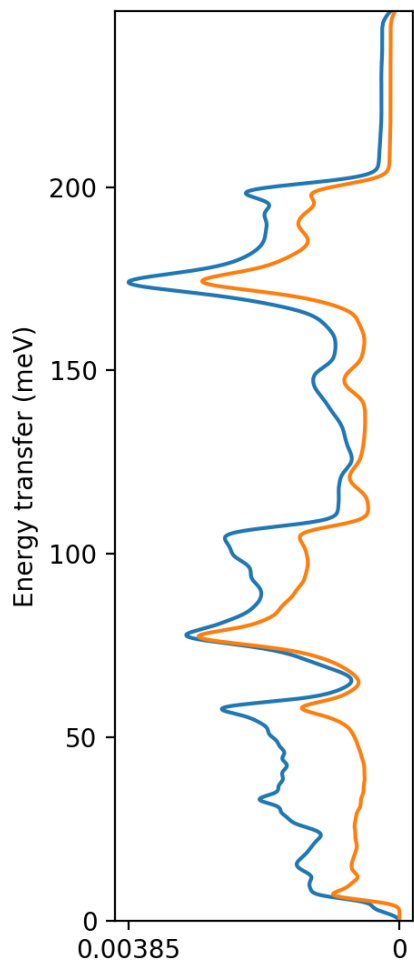
OCLIMAX



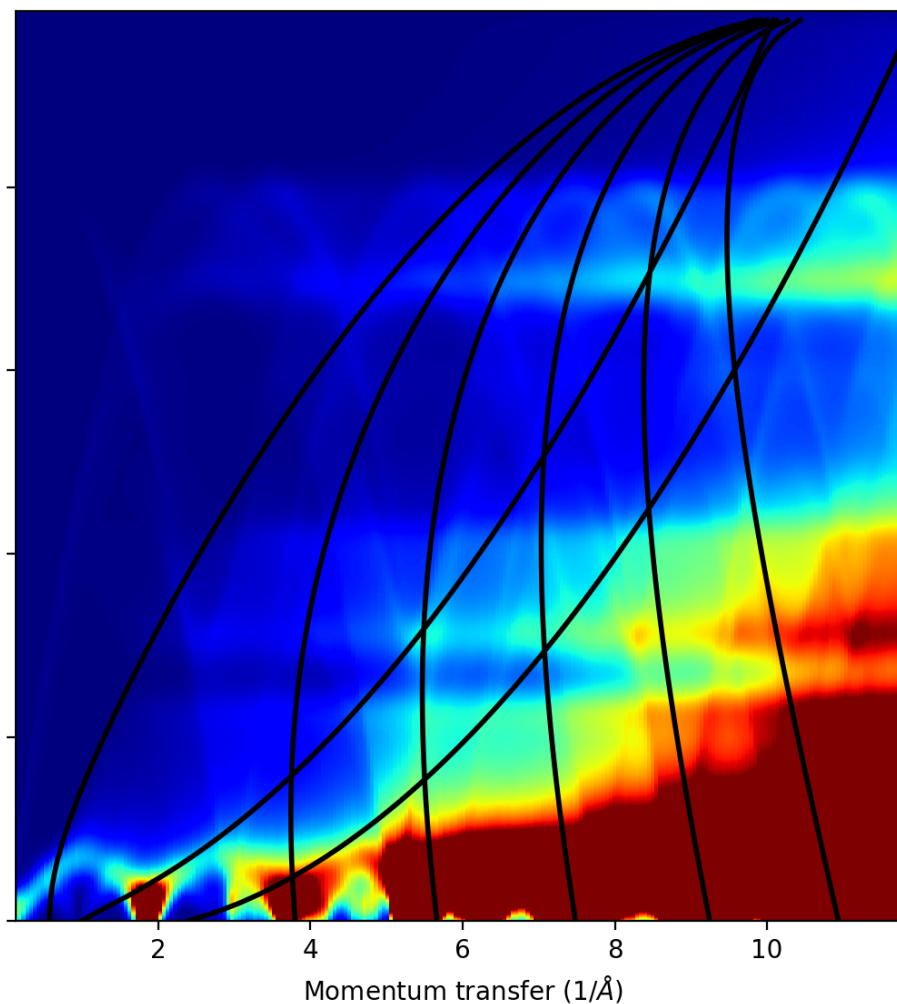


# Calculated $S(Q, \omega)$ map and various sampling trajectories

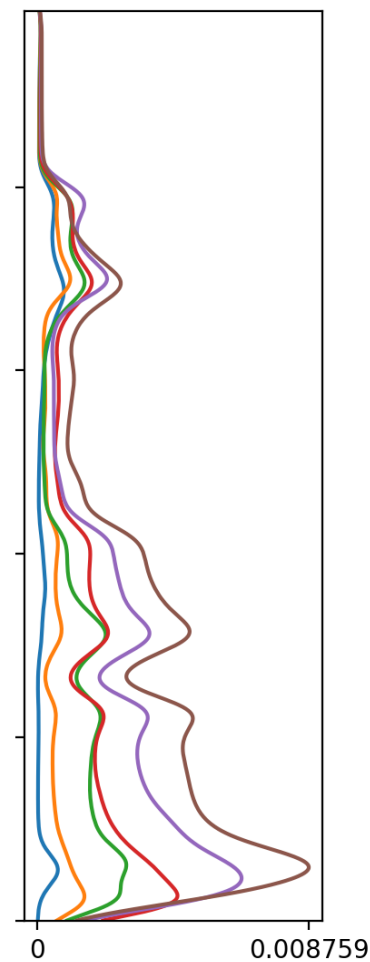
VISION



$S(Q, \omega)$  Map



SEQUOIA  
ARCS  
etc



# How to obtain OCLIMAX

- Install Docker (<https://www.docker.com/>)
- 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 Command Prompt “cmd”, go to the working directory, run:

```
$ oclimax.bat pull
```

- For more information

**Download the user manual at**

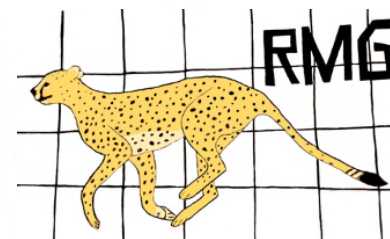
**<https://sites.google.com/site/ornliceman/download>**

# Convert your files to OCLIMAX input file

- Automatically extract phonon frequencies and polarization vectors from your DFT program output files and generate the input file for OCLIMAX
- Currently support



**NWCHEM**  
HIGH-PERFORMANCE COMPUTATIONAL  
CHEMISTRY SOFTWARE



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

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

# How to run OCLIMAX

- 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 for VISION

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
```

- The output are standard csv files. You may use your favorite software to visualize the data.
- You may also use the provided script (pclimax.py) to generate a quick plot.

# Parameters for OCLIMAX calculation

```
# All parameters for OCLIMAX calculation
# General parameters
TASK = 1 # 0:inc approx. 1:full coh+inc. 2: single crystal coh
INSTR = 3 # 0:VISION 1:general indirect 2:general direct 3:Q-omega mesh
TEMP = 293.00 # Temperature [K]
E_UNIT = 1 # Energy unit [eu] (0:cm-1,1:meV,2:THz)
OUTPUT = 0 1 # 0:standard, 1:restart, 2:SPE, 3:full, 4:DOS, 5:modes
```

```
# E parameters
MINE = 0.000 # Energy range (minimum) to calculate [eu]
MAXE = 30.00 # Energy range (maximum) to calculate [eu]
dE = 0.010 # Energy bin size [eu]
ECUT = 0.010 # Exclude modes below this cutoff energy [eu]
ERES = 0.5751 -0.018 0.0002 # E resolution coeff
```

```
# Q parameters
MINQ = 0.02 # Q [1/Ang] range (minimum) to calculate
MAXQ = 4.00 # Q [1/Ang] range (maximum) to calculate
dQ = 0.02 # Q [1/Ang] bin size
QRES = 0.50E-01 # Q resolution coeff (INSTR=3)
```

```
# Instrument parameters
THETA = 2.9 56.7 # List of scattering angles [degree]
Ef = 32.00 # Final energy [eu] (INSTR=1)
Ei = 30.00 # 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)
```

```
# Additional parameters
MAXO = 10 # Maximum order of excitation (up to 10)
CONV = 2 # Start convolution from order=CONV (2 or 3)
MASK = 1 # Set 1 to apply mask on Q-w map (INSTR=3)
ELASTIC = -0.10E+01 -0.10E+01 # E Q, <0:no EL,0:cal res,>0:given res
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
NS = 6.0 # Number of sigma in resolution function
HKL = 0 0 0 # HKL (TASK=2 and INSTR=3)
Q_vec = 0 0 1 # Q vector dir (TASK=2 and INSTR=3)
```

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

# How simulations worked together with INS experiments: examples





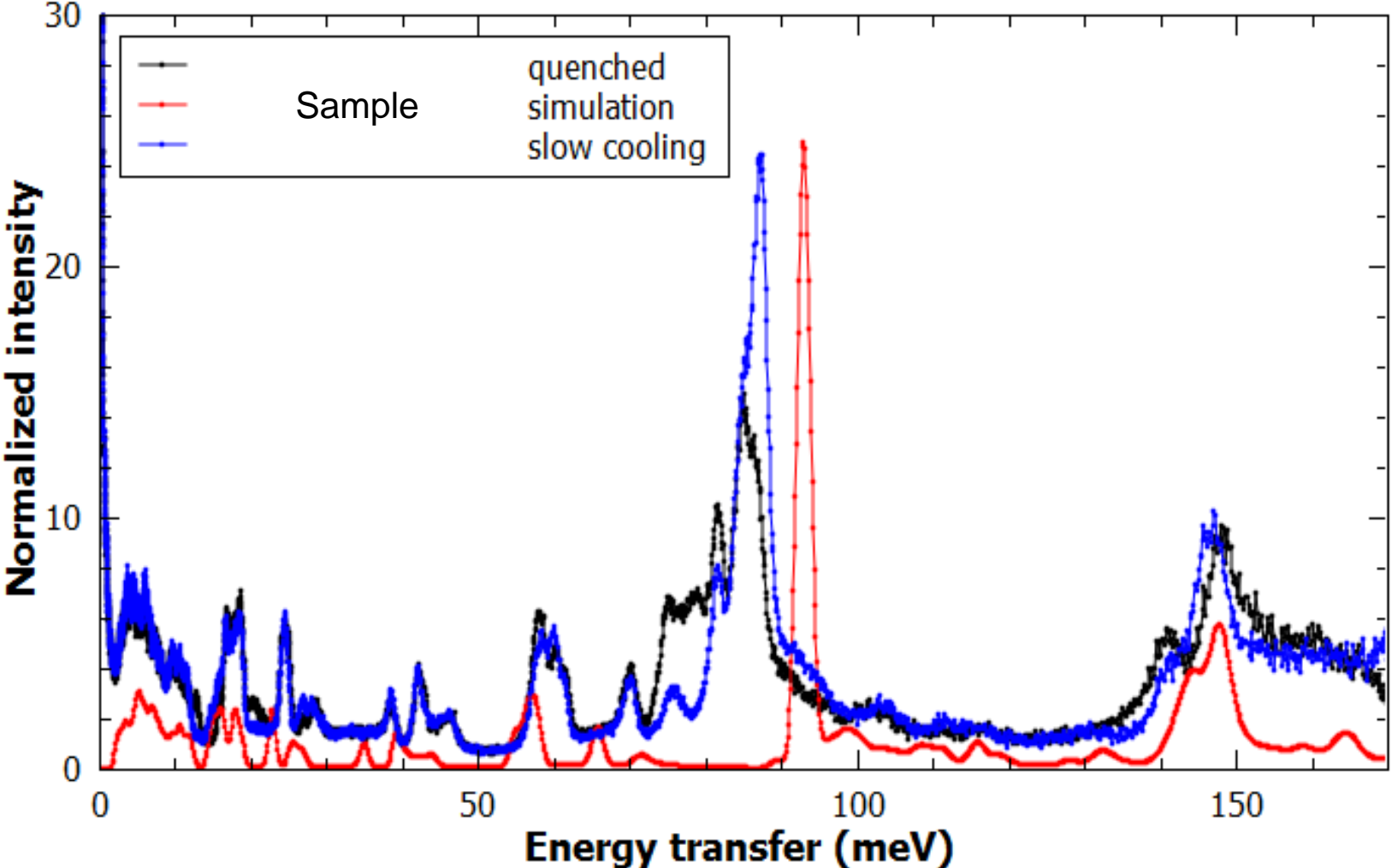
# Simulation helped users to make decisions on-the-fly

```
[yyc@or-condo-login02 ~]$ ls -lhr  
-rw-r--r-- 1 yyc users 3.6K Nov  4 15:50 cell  
-rw-r--r-- 1 yyc users 1.1K Nov  4 15:50 param  
-rw-r--r-- 1 yyc users 3.9K Nov  4 15:51 _PhonDOS.cell  
-rw-r--r-- 1 yyc users  735 Nov  4 15:52 _PhonDOS.param  
-rw-r----- 1 yyc users 1.1M Nov  4 16:46 .castep  
-rw-r----- 1 yyc users 7.3M Nov  5 06:15 _PhonDOS.phonon  
-rw-r----- 1 yyc users 232K Nov  5 06:15 _PhonDOS.castep  
-rw-r--r-- 1 yyc users 3.3M Nov  5 08:56 aclimax
```

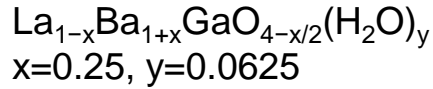
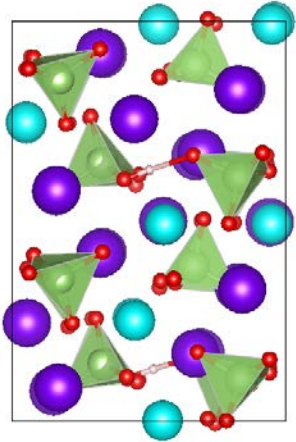
```
[yyc@analysis-node02 manualreduce]$ ls -lhr  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 12:34 VIS_20557_5K_for_0.9hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 13:28 VIS_20559_50K_for_0.9hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 14:23 VIS_20561_75K_for_0.9hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 15:56 VIS_20563_100K_for_0.9hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 17:21 VIS_20565_125K_for_0.9hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 18:44 VIS_20567_150K_for_0.9hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 20:23 VIS_20570_175K_for_1.2hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 21:58 VIS_20572_200K_for_1.2hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  5 23:29 VIS_20574_225K_for_1.2hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  6 01:00 VIS_20576_250K_for_1.2hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  6 02:28 VIS_20578_275K_for_1.2hr.nxs  
-rw-rwx---+ 1 yyc users 2.2M Nov  6 03:57 VIS_20580_300K_for_1.2hr.nxs
```

Simulation was started at the beginning of the experiment. By the time when experimental data were collected, the calculation was already finished with theoretical predication available to be compared with experiment. This eventually led to a critical decision made by the user (see next slide).

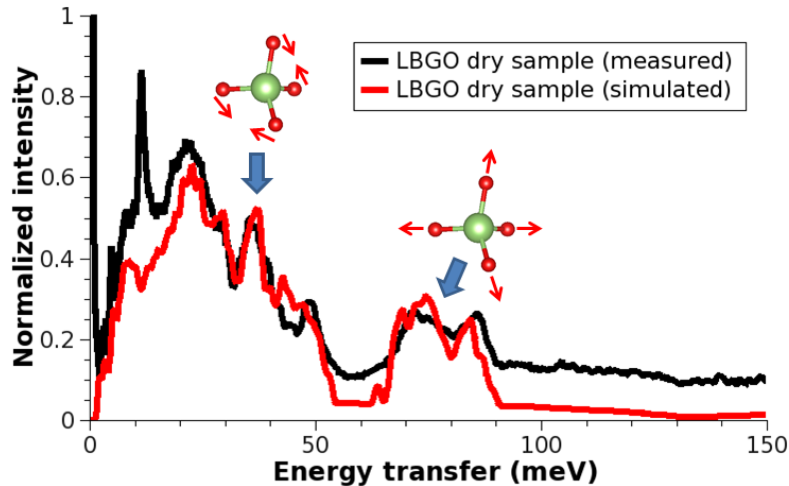
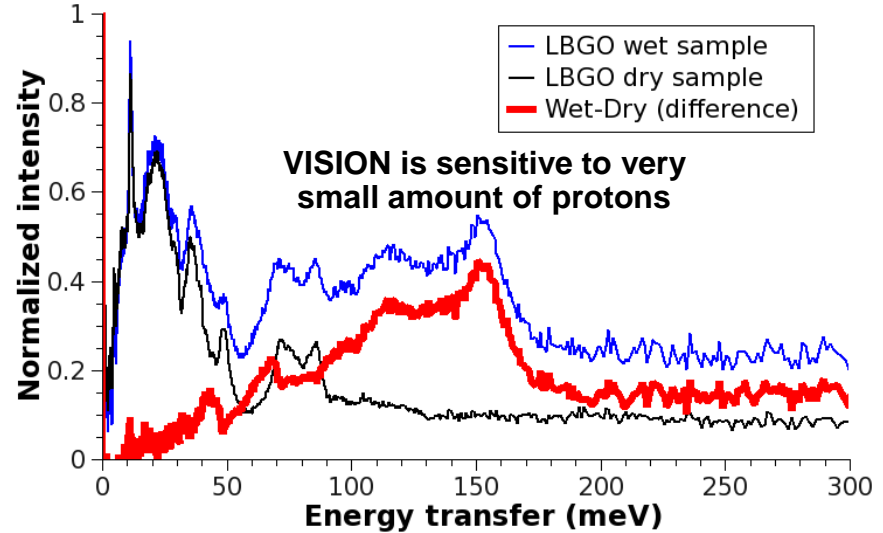
# Simulation helped users to make decisions on-the-fly



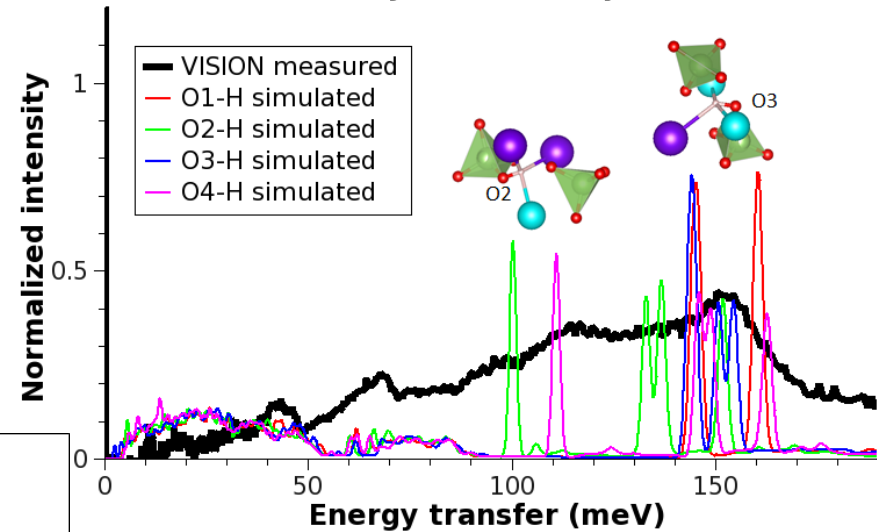
# Simulation led to key findings based on INS data



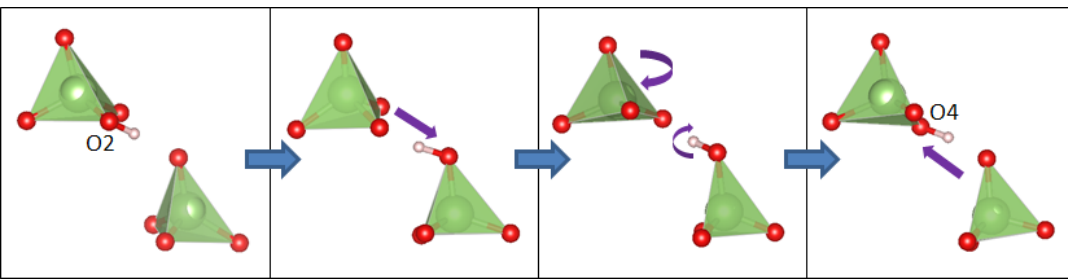
La: cyan  
Ba: purple  
Ga: green  
O: red



**Wet-Dry difference spectra**

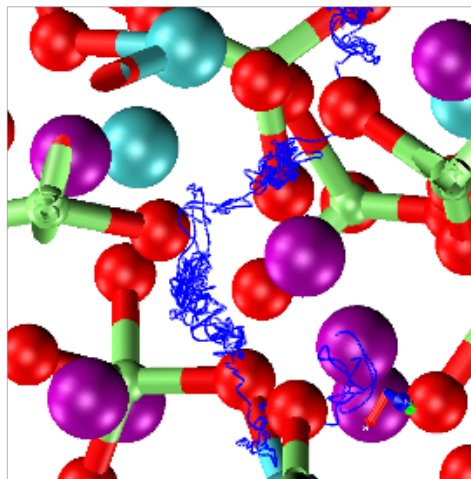


VISION+modeling identifies various local chemical environment for the protons, and reveals the proton transport mechanism



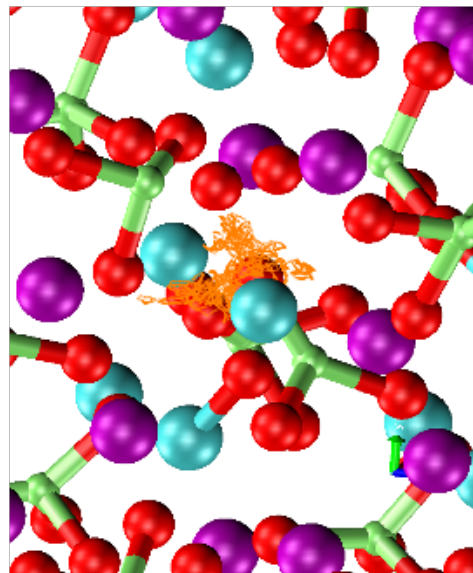
# Simulation led to key findings based on INS data

(a) **Ba-rich (H1)**

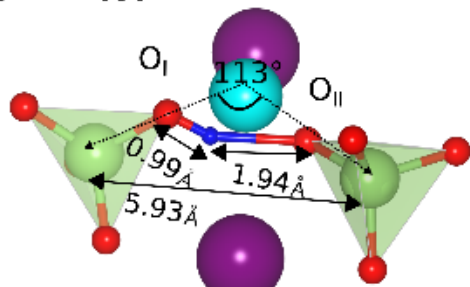


(c)

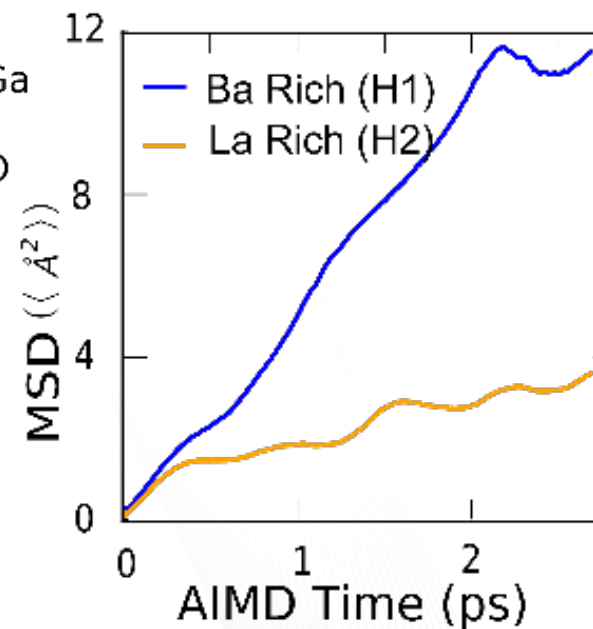
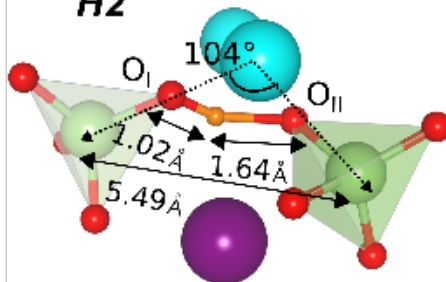
(b) **La-rich (H2)**



(c) **H1**



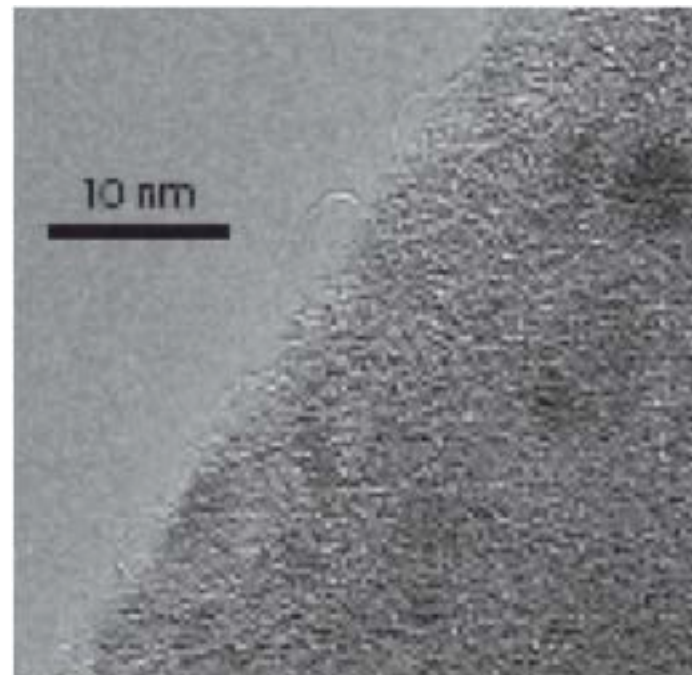
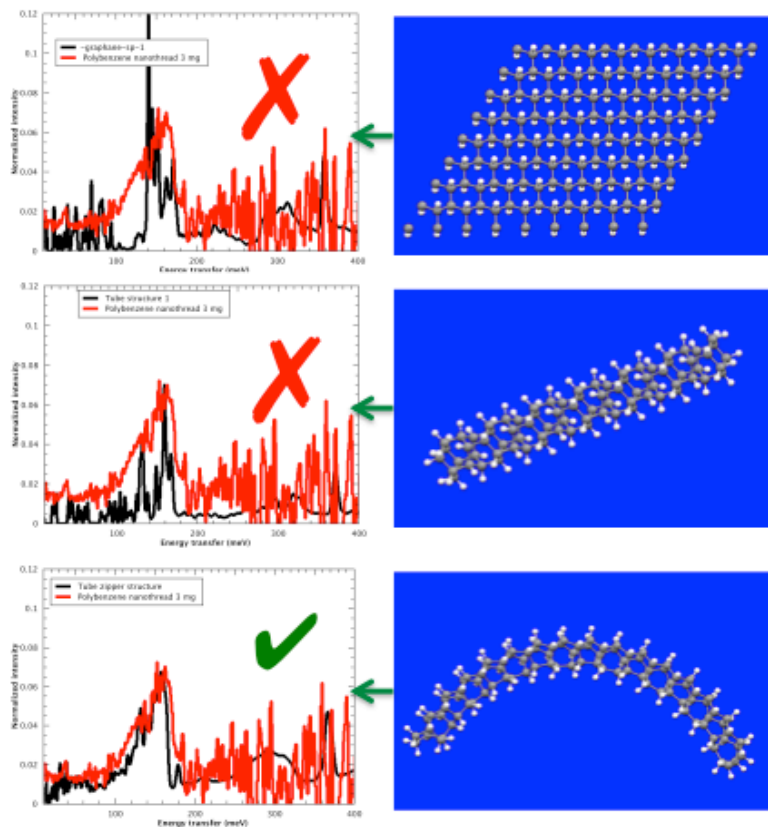
(d) **H2**



Cheng Y.Q., Balachandran J., Bi Z., Bridges C.A., Paranthaman M.P., Daemen L.L., Ganesh P., Jalarvo N.,  
The influence of the local structure on proton transport in a solid oxide proton conductor  $\text{La}_{0.8}\text{Ba}_{1.2}\text{GaO}_{3.9}$   
Journal of Materials Chemistry A, 5, 15507–15511 (2017).

# Simulation enabled structural determination from INS spectra

3 mg of nanothread sample

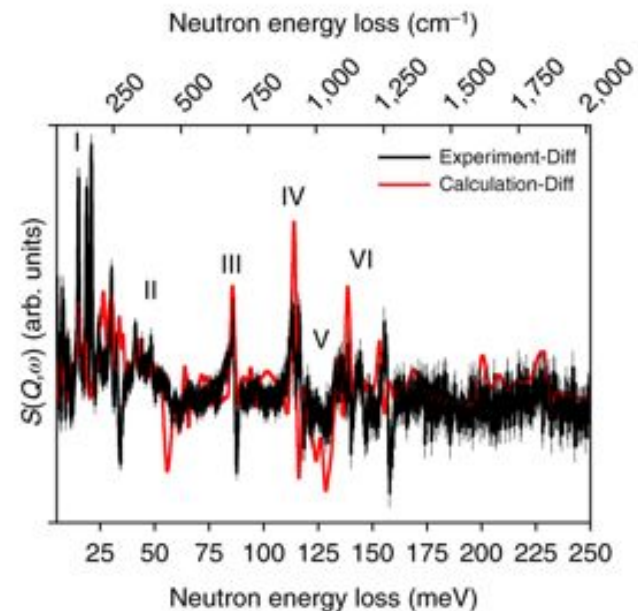
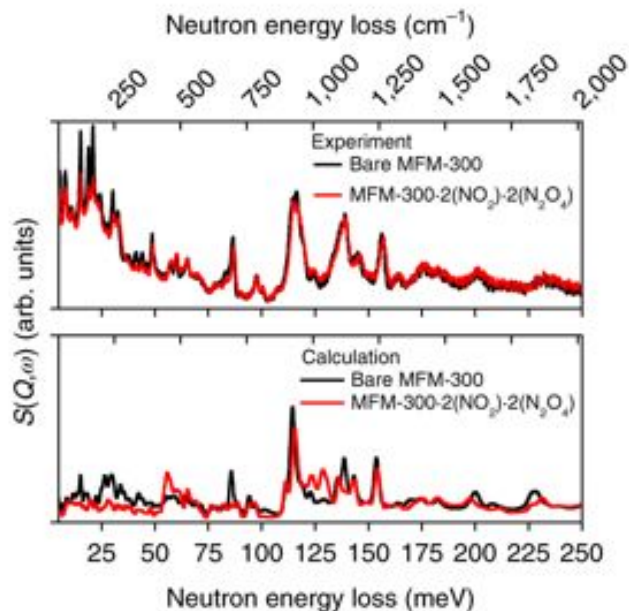
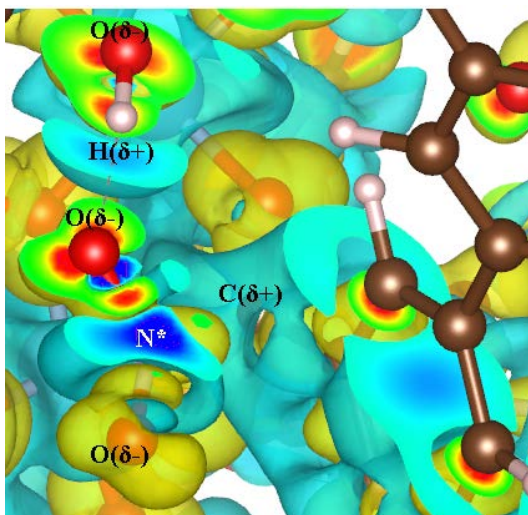
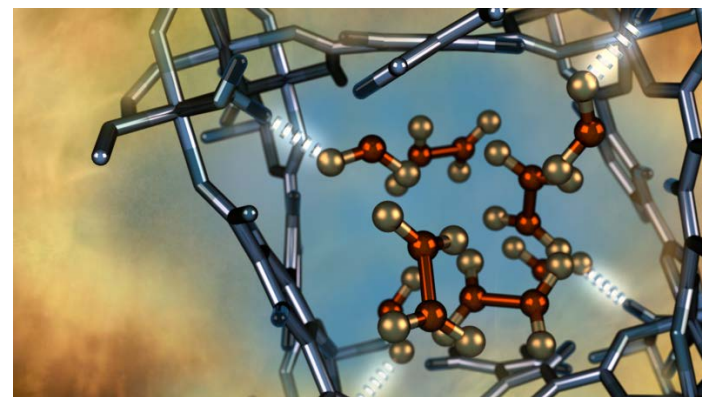
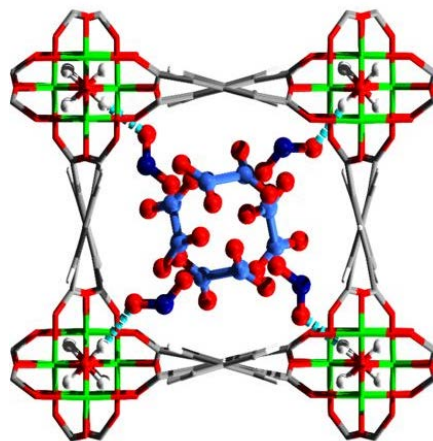
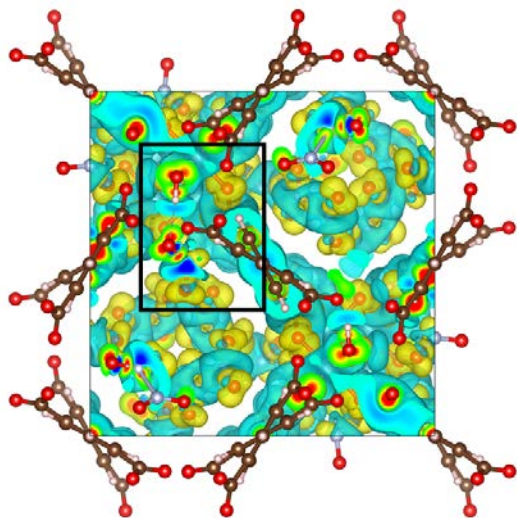


Collaboration with Malcolm Guthrie, John Badding, Vin Crespi. Original publication on carbon nanothreads: *Nature Materials*, **14**, 43 (2014)

Comparison of the experimental data from VISION and a series of DFT calculations of hypothetical structures that contain  $sp^3$  carbon and the correct stoichiometry (C:H ratio 1:1) allows us to determine which structure corresponds to the measured spectra.



# Simulation revealed fundamental mechanism behind small differences

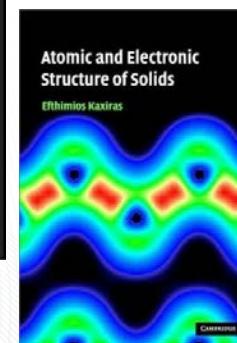
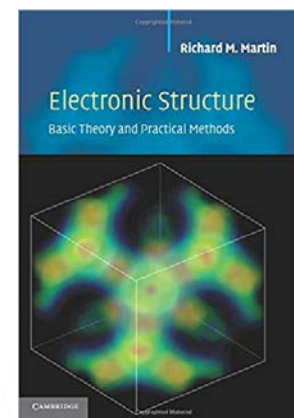
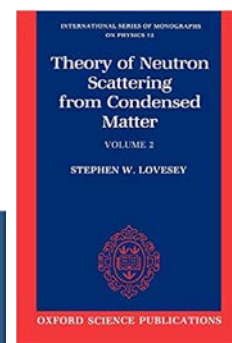
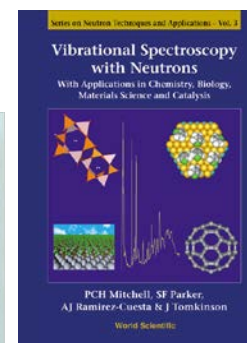
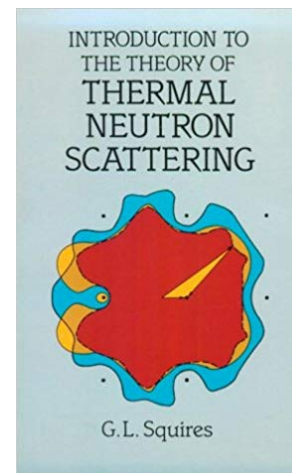


X. Han, Nature Materials (2018)  
<https://doi.org/10.1038/s41563-018-0104-7>



# References and recommended reading

- Neutron scattering theory
  - G. L. Squires, Introduction to the Theory of Thermal Neutron Scattering
  - S. W. Lovesey, The Theory of Neutron Scattering from Condensed Matter
  - P. C. H. Mitchell, S. F. Parker, A. J. Ramirez-Cuesta, J. Tomkinson, Vibrational Spectroscopy with Neutrons
- Density functional theory
  - R. M. Martin, Electronic Structure: Basic Theory and Practical Methods
  - E. Kaxiras, Atomic and Electronic Structure of Solids



# Questions?



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