Scientific Computing

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A (concise) overview of SCD's current capabilities/activities in *Modelling and Simulation* of Materials and Molecules

on behalf of

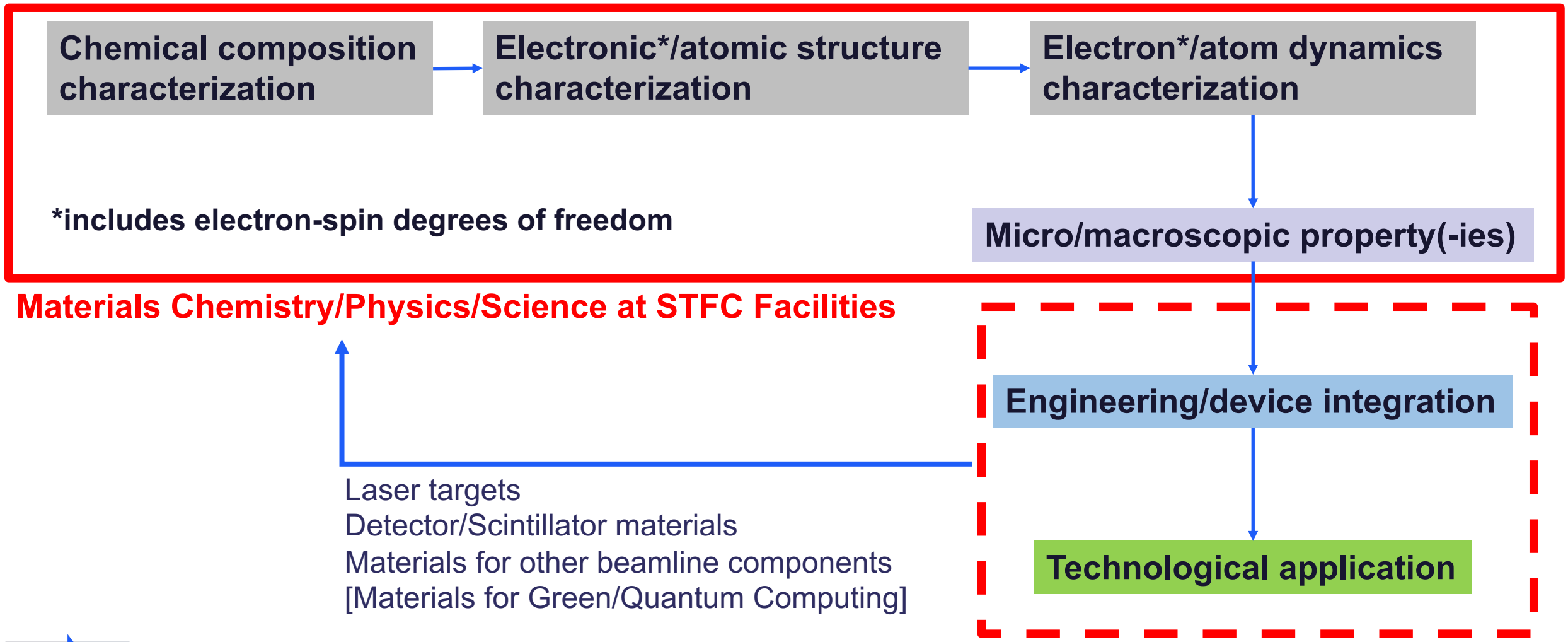
Ilian Todorov, Silvia Chiacchiera, Michael Seaton, Vlad Sokhan, Chin Yong, Benjamin Speake, Noel Vizcaino

Thomas Keal, Andrey Brukhno, Andrew Duff, Thomas Durrant, Rajany KV, You Lu, John Purton, Kakali Sen, Joseph Thacker

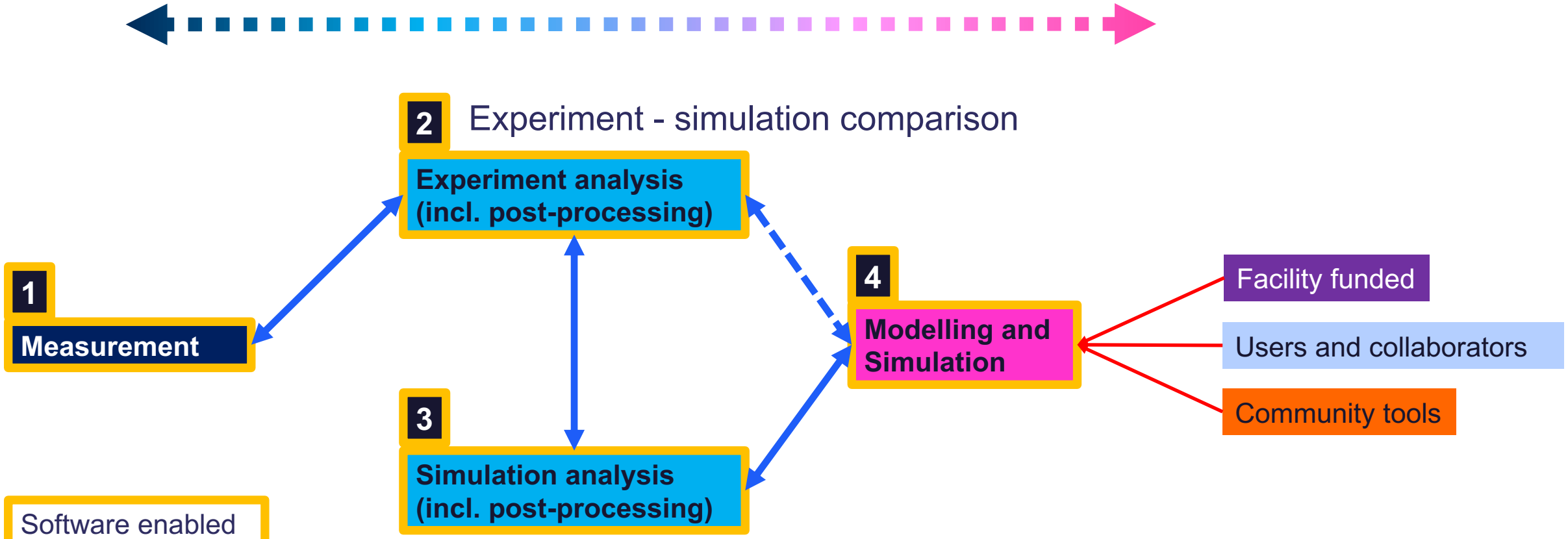
Alin Elena, Federica Zanca, Elliott Kasoar

Leon Petit, Jerome Jackson, Martin Plummer, Manuel Dos Santos Dias, Peter Elliott, Ivan Scivetti, John Trail, Paolo Trevisanutto, Ian Bush, Christopher Marooney, Subindev Devadasan, Adam Jackson, Dominik Jochym, Leandro Liborio, Marcello Puligheddu, Sathya Sai Seetharaman, Barry Searle, Kane Shenton, Jacob Wilkins

Let's start by trying to agree on the scope



A workflow across STFC facilities



- Extraction of an observable or feature (which may not be directly measurable…) using theory
- Simulation-based bridging of different observables

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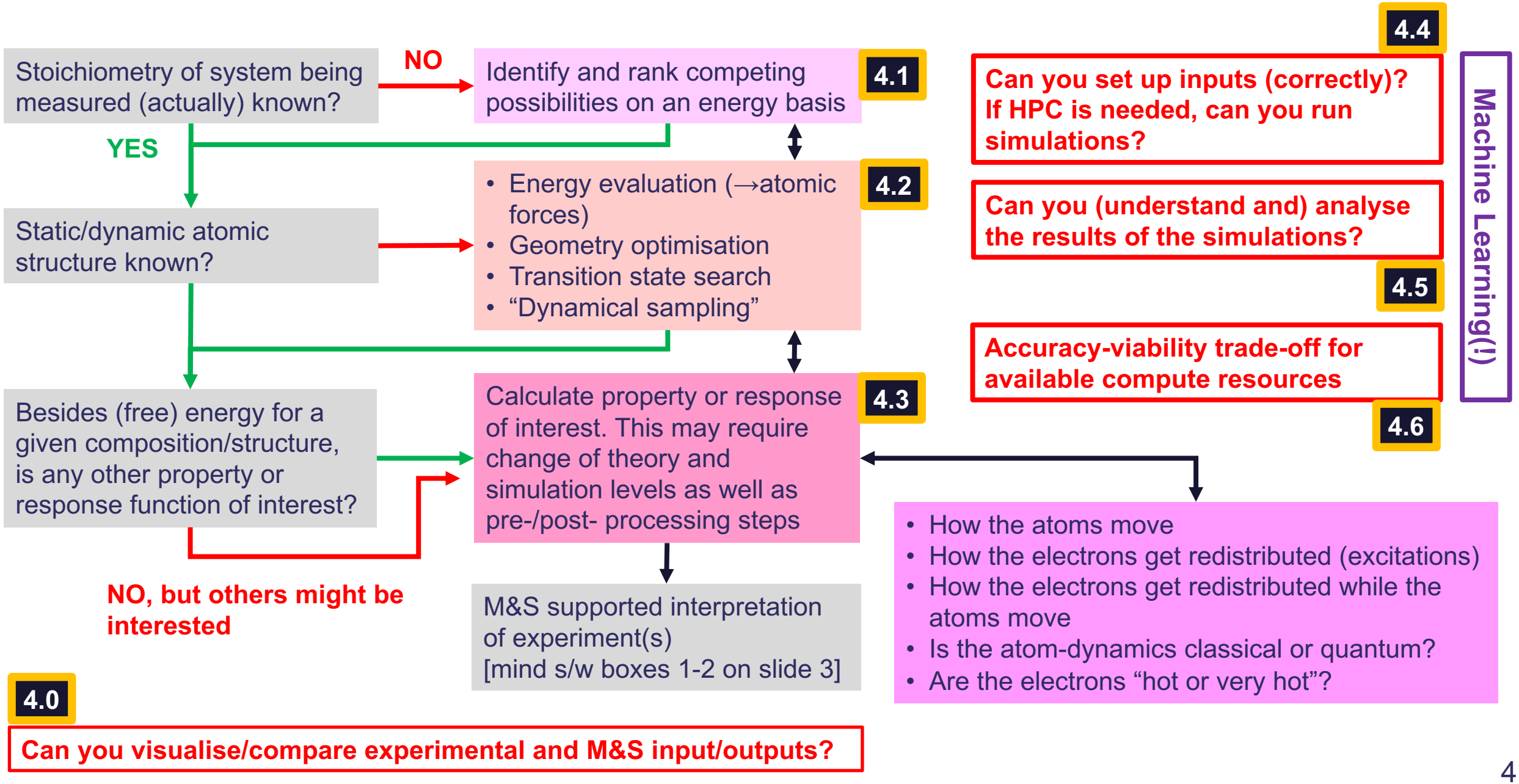
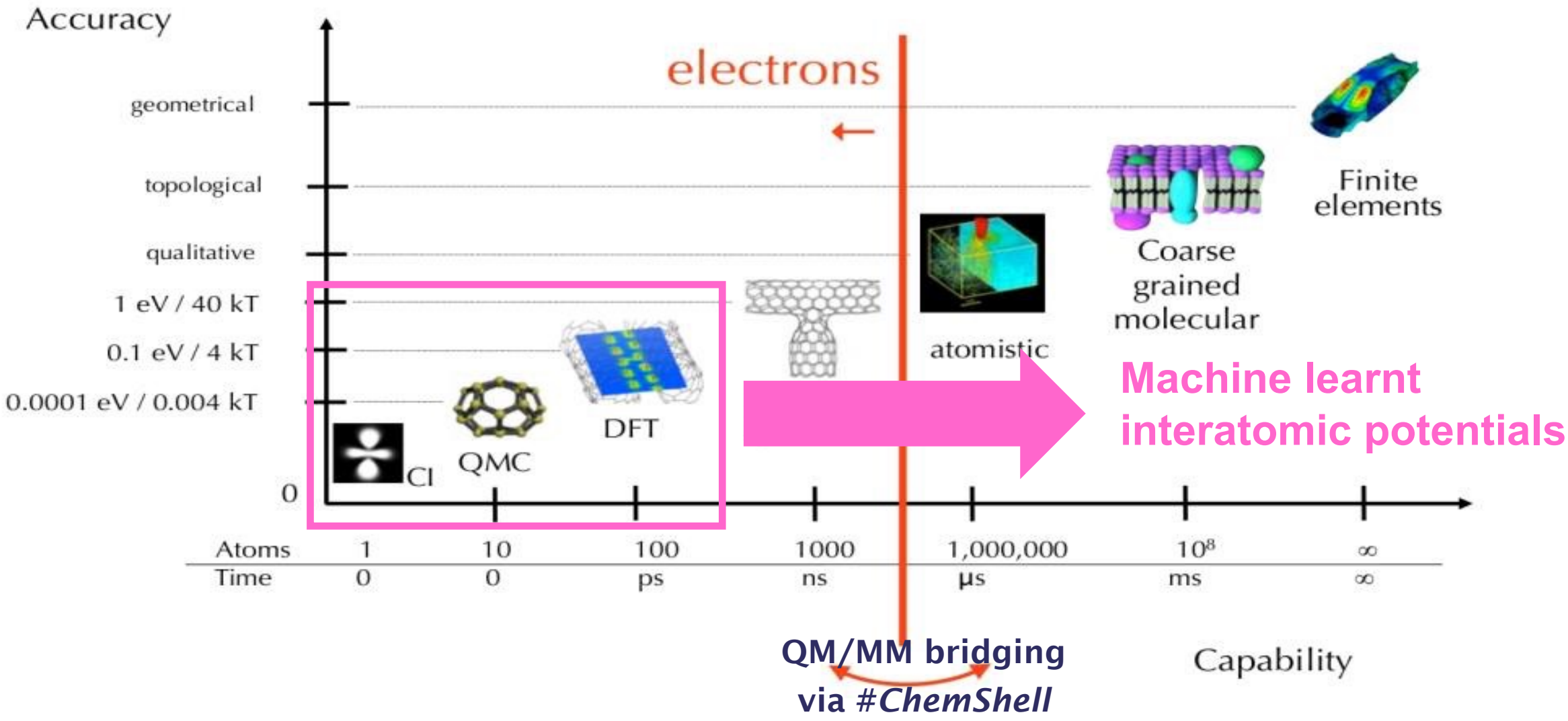
Let's expand (minimally) the "4:M&S"…



A multi-scale Modelling and Simulation platform



Computational Materials and Molecular Science Theme

Currently 4 groups: ~40 staff-members with expertise across 2, 3 and 4

- **Classical Materials and Molecular Modelling** (Ilian Todorov)
- **Multiscale Materials Chemistry** (Thomas Keal)
- **Data-driven Materials and Molecular Science** (Alin-Marin Elena)
- **Theoretical and Computational Materials Physics** (Gilberto Teobaldi, interim)

SCD-CMMS' software menu as of today (with working hyperlinks…)

- **2: experimental data postprocessing (EXAFS)**
  - DL_EXCURV

- **2-3: experiment-simulation comparison (INS)**
  - Abins
  - Euphonic
  - INTONE (not released yet)

- **2-3: experiment-simulation comparison ([QE]NS])**
  - MDMC (not released yet)

- **2-3: experiment-simulation comparison (μSR)**
  - The Muon Spectroscopy Computational Project
  - Muon Galaxy

- **2-3: experiment-simulation comparison (EQCM)**
  - ALC_EQCM

- **4.0: Visualisation**
  - DL_Visualize

- **4.1: Stochiometric resolution**
  - ALC_EQCM

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – first principles**
  - CASTEP
  - CP2K
  - CRYSTAL
  - ELK
  - ONETEP
  - QUESTAAL
  - W90

SCD-CMMS' software menu as of today (with working hyperlinks…)

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – parameterised Hamiltonians**
  - DL_FIELD
  - DL_POLY
  - DL_MONTE
  - ACDC (not released yet)

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – Coarse-grained/mesoscale lattice models**
  - DL_MESO

- **4.2-4.3-4.4: Energy (forces) evaluation + responses/dynamics – multi-scale (QM/MM)**
  - ChemShell
  - DL_FIND

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – machine learned potentials**
  - APD
  - Janus

- **4.4: Input set-up and workflow management**
  - Aiida-MLIP
  - ALC_EQCM
  - ALC_SUTOR
  - DL_FIELD
  - DLPOLY-PY
  - Muon Galaxy
  - Shapespyer

- **4.5: output analysis**
  - ALC_TRAJECTORY
  - DL_ANALYSER
  - Shapespyer

- **4.6: viability-accuracy trade-offs**
  - Extended pool of highly experienced people
  - Continuous algorithmic and parallelism improvements

ps about software for 4.2-4.3:

The previous software lists have been limited to the software we develop/co-develop. However, quite a few of us know how to skilfully use (and are licensed to use) also other very popular simulation codes NOT included in the previous slides.

MORE EXPLICITLY:
If you or your users really need to use LAMMPS, GROMACS, Quantum Espresso, VASP, Exciting, Yambo, Ocean, Molpro, NWChem, etc, bar licensing caveats and ALC funding strings, we would be open to it. As a matter of fact, we may have likely been using it already for benchmarking and validation…

Some considerations about balancing M&S demands

- Level of M&S accuracy vs **compute resources available** (SCARF or beyond SCARF? How to access it?)
- Setting discussion and possible work on *generative AI for training* aside, software documentation and continuous training as well as beamscientist(s) engagement is essential for **software uptake** to happen. This requires (£-)support beyond the code development, validation and release…
- Software improvements around software types #1 and #2 (slide-3) tend to show "**value at the beamline**" earlier than #3 and #4. However, interpretation and **cross-correlation** with other observables require #3 and #4. #4 may require additional external collaboration (*Theory development* has been left loose on purpose).
- Work at #3 and #4 can facilitate contact also with **laboratory or EPSRC-funded facility measurements**, which may add to the "users' experience".
- Interfaces with **ALC *Data*** and ***Machine Learning*** themes have been purposefully left loose/undefined.
- Balance between SCD's "**software expertise provision**", "**in silico added insights**", and **"scientific collaboration/partnership**" is and will be Facility and user-dependent**. One size fits all is most likely not possible.** We are flexible and can respond to different inclinations/requests.

An (incomplete) overview of previous/ongoing projects and/or untapped capabilities across 2-4.X (see slides 3&4)

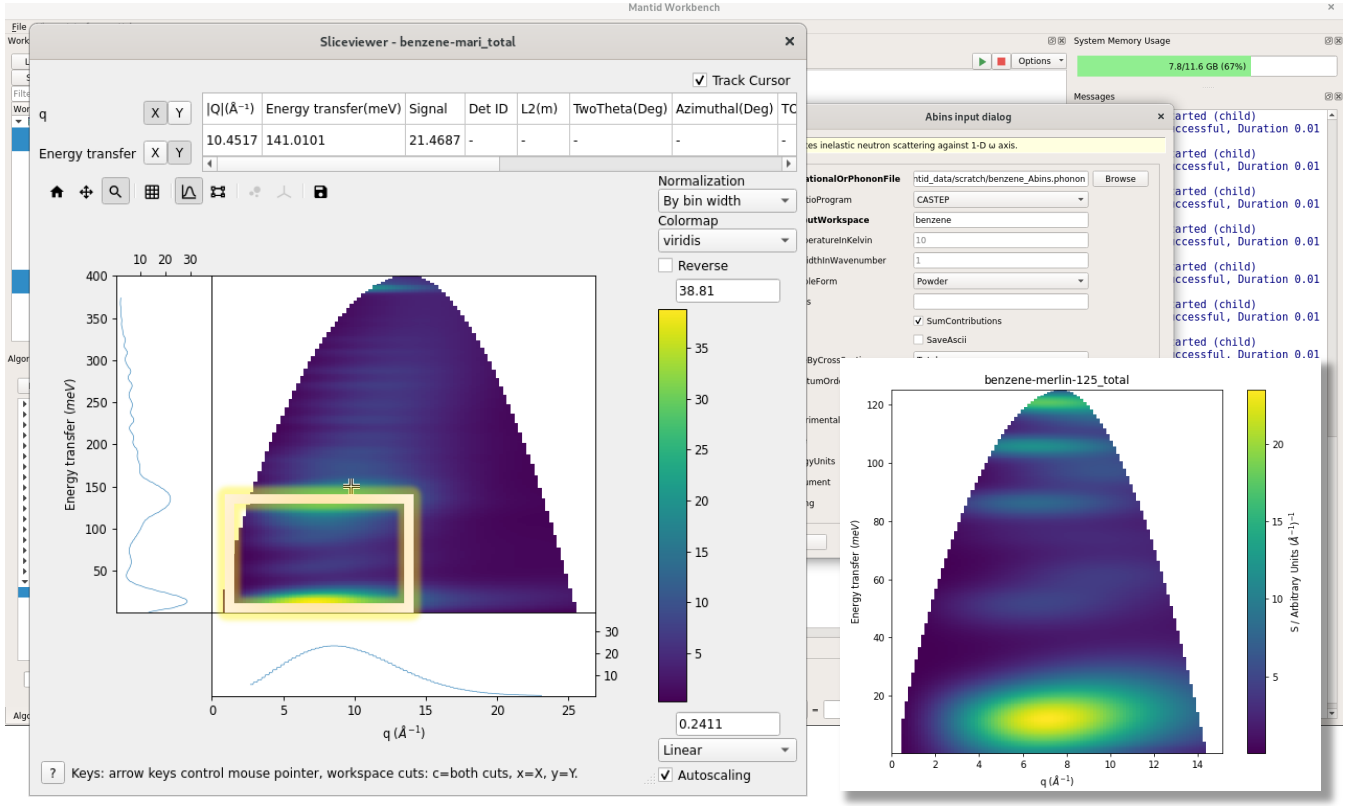
2-3: AbINS – *Ab initio inelastic neutron scattering*

- Simulated INS experiments with data from popular atomistic codes *(e.g. CASTEP, VASP, GAUSSIAN…)*

- Successor to CLIMAX, aCLIMAX codes used for ISIS TOSCA beamline (and predecessor TXFA…)
  - Model intended for molecular spectroscopy (incoherent scattering)

- Python code in Mantid: available on beamlines, VMs (IDAaaS), Win/Mac/Linux

- Has grown to support more beamlines at ISIS (MAPS/MARI/MERLIN) and elsewhere (ILL Lagrange, PANTHER)



- Beamlines can be compared when planning experiments... this is not yet standard practice!

[Abins v1 (mantidproject.org)]

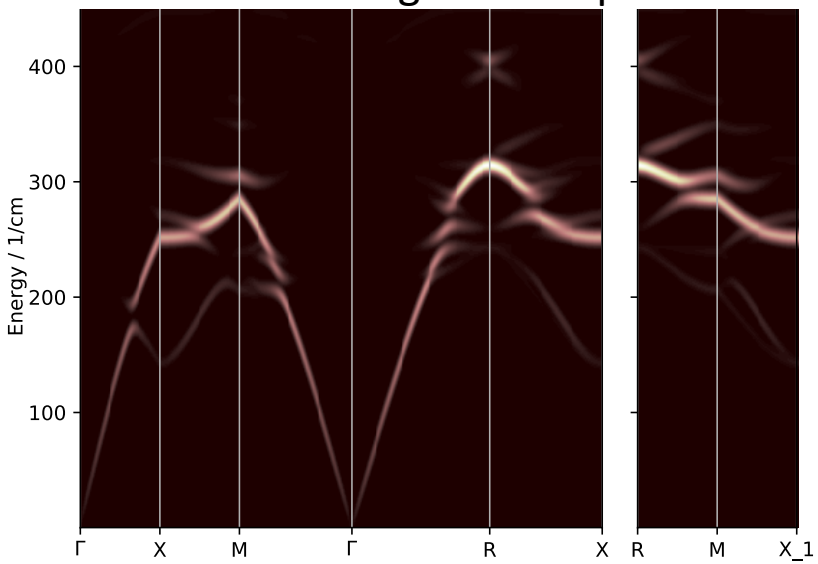
Credit: Adam Jackson, Dominik Jochym et al.

2(-3): Euphonic – *q*-point sampling backend and more

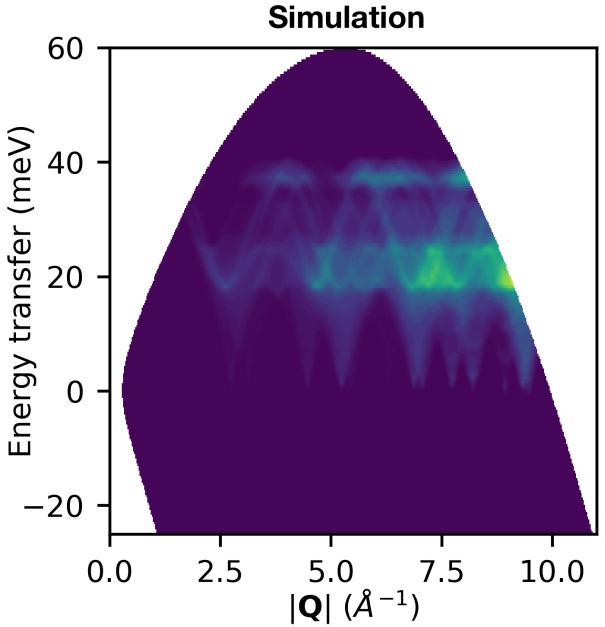
- Created as part of “Proper analysis of Coherent Excitations” (PACE) project; modernisation of the Horace package
- Horace: 4-D analysis from time-of-flight data for ISIS MAPS/MARI/MERLIN/LET
- Euphonic provides simulation for Horace, decoupled into a “friendly” Python+C package
- Coherent scattering on fine ***q***-mesh: generally suitable for inorganic crystals

https://github.com/pace-neutrons/Euphonic

“Coherent-weighted” dispersion



Numerical powder-averaging



- OpenMP parallelism performs well on IDAaaS
- ***q***-point interpolation also used by AbINS for incoherent scattering
- *Mantid manages Euphonic as a Conda dependency*
- Command-line tools: intended as a demo of library, but have proven useful for quick simulation work

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Credit: Becky Fair, Adam Jackson, Dominik Jochym et al.

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2-3: INTONE – *Ab initio dynamical structure factor for quantum materials*

Manuel dos Santos Dias
Kiran Jonathan

First-principles materials properties

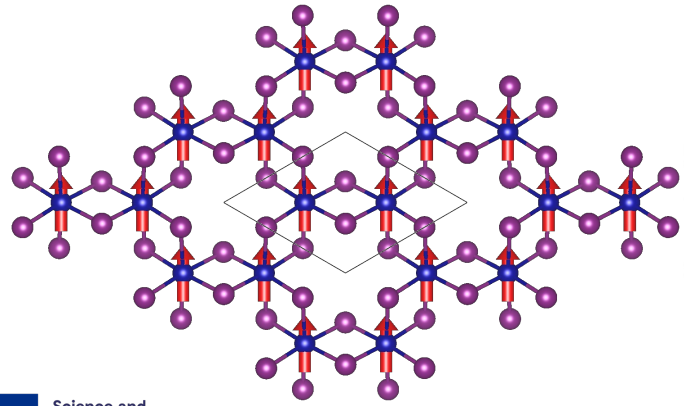
Model parameters
Spin wave theory

Parameter-free susceptibilities

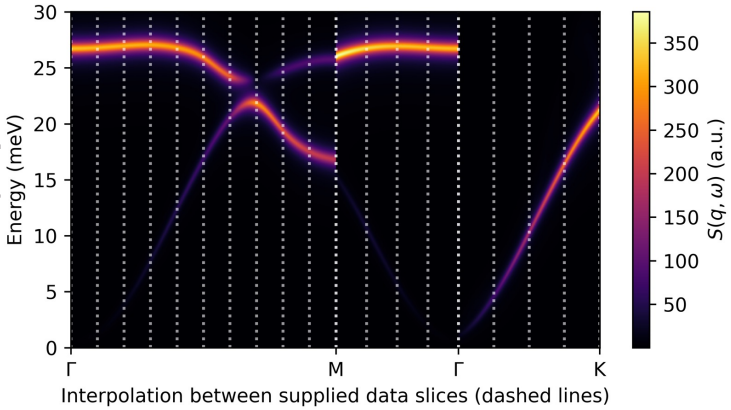
Theoretical spectroscopy
- $S(\mathbf{q},\omega)$ from magnons, plasmons, …
- Cuts from 4D S(q,w) datasets
- Powder average, co-alignment effects
- Complementary to MD approaches
- Interpret and design experiments

QUANTUMESPRESSO
JuKKR
QUESTAAL

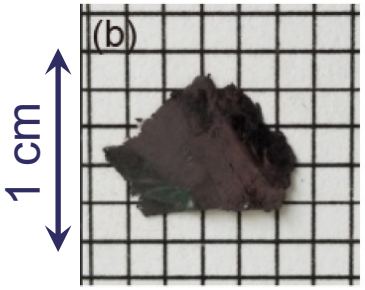
Case study: 2D CrI_3



Susceptibility data from turboMagnon (QE)



Next: effects from co-aligned crystals



20-30 crystals co-aligned for INS

PRX **8**, 041028 (2018)

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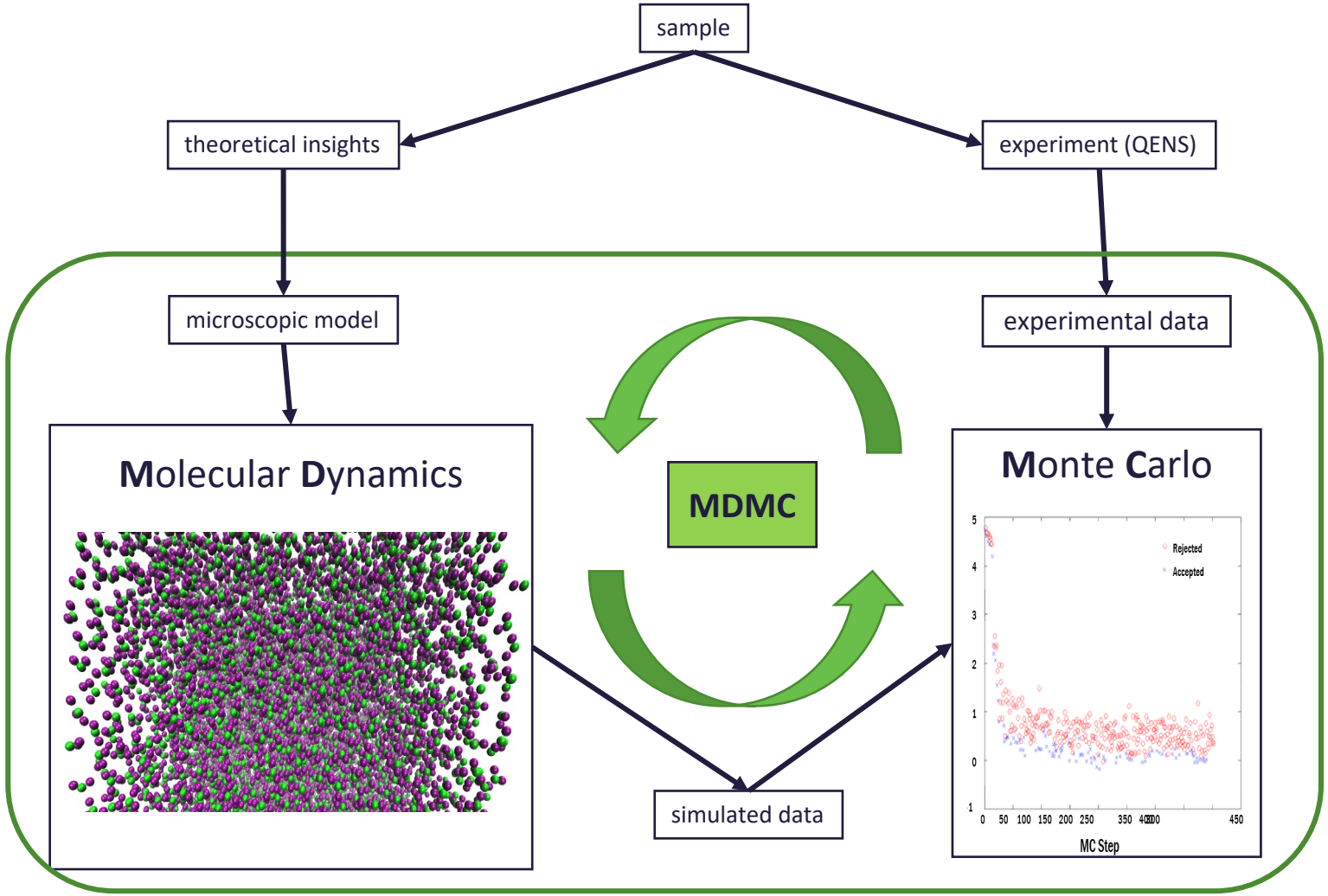
2-3: MDMC – optimising molecular dynamics potentials to dynamical structure factor data



- Simulations Engines:
  - Lammps
  - DL_POLY
  - Others
- Restructuring of the project to support more MD Engines and other simulators, MC, hMC
- Refining against other observables than structure factor, e.g. diffusion coefficients
- UX friendly for both simulators and experimentalists, MDANSE?

proof of concept: AJ Markvardsen, RL McGreevy, RAL-TR-2017-014

Credit: Alin Elena (SCD), Jacob Wilkins (SCD), Franz Lang (ISIS), Maciej Bartkowiak (ISIS)

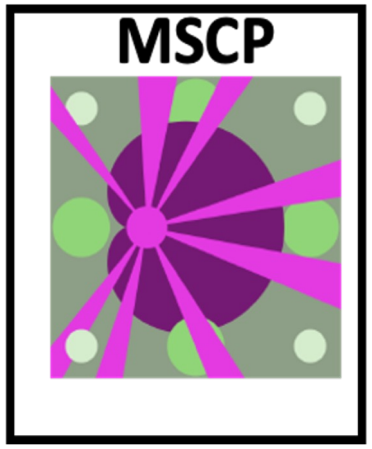
2(-3): Muon Spectroscopy Computational Project

Sustainable, User-friendly and Reproducible Software Tools for Interpreting Muon Experiments

https://muon-spectroscopy-computational-project.github.io/

`MuDirac`
treat muonic atoms

`pymuon-suite`
muon stopping sites



`pm-nq`
muon quantum effects

`Muspinsim`
spin dynamics of systems of a muon plus other spins

Muon

Galaxy PROJECT

`Muon Galaxy` *Open source, web-based GUI for MSCP's tools*

https://muongalaxy.stfc.ac.uk/

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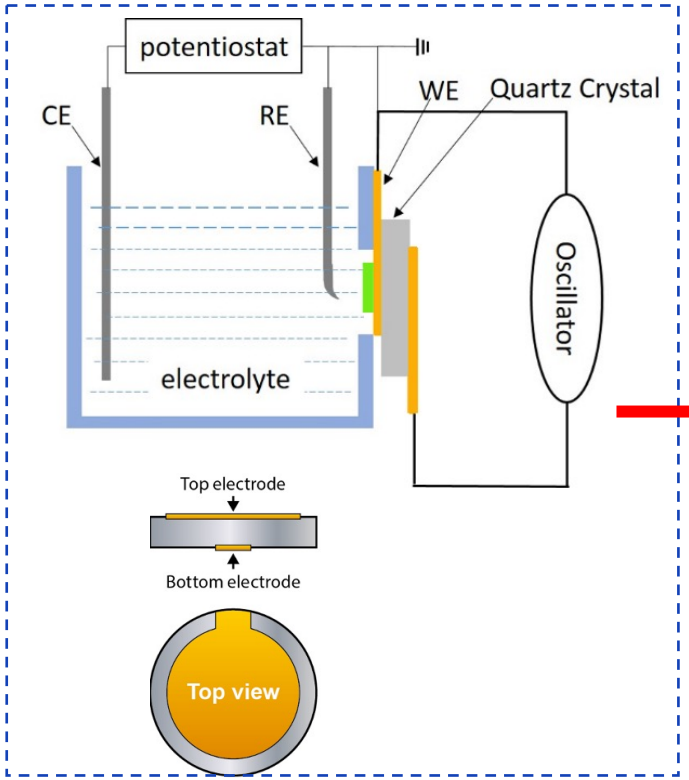
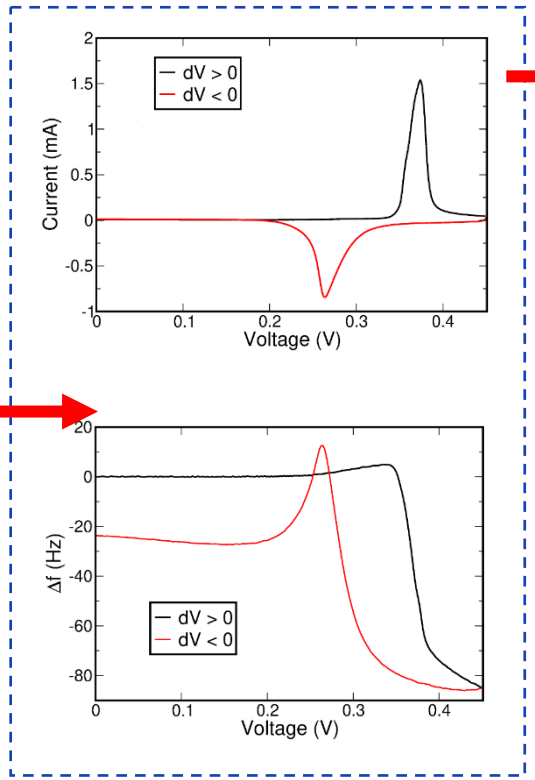
4.1: ALC_EQCM: software for electrochemistry

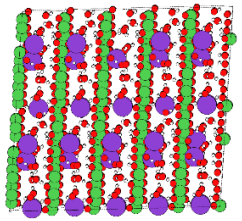
EQCM device



EQCM data



ALC_EQCM



Atomistic models

HPC

Motivation

Complex electrochemical reactions involve many species.
Lack of quantitative stoichiometric resolution during cycling

Strategy

- Use EQCM values of mass and charge changes to obtain the set of multiple stoichiometric solutions
- Generate atomistic models compatible with EQCM data
- Compute structures to screen relative energies thanks to **automatically prepared, sanity checked input files for standard Density Functional Theory codes** (CASTEP, ONETEP, CP2K, VASP, QE…)

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Ivan Scivetti et al.,

1) ACS Appl. Energy Mater. 3, 3347 (2020)
2) Comput. Mater. Sci. 218, 111968 (2023)
3) https://github.com/stfc/alc_eqcm

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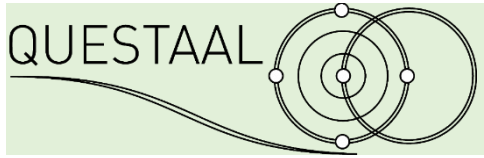
4.2-4.3 DFT and beyond-DFT methods and codes

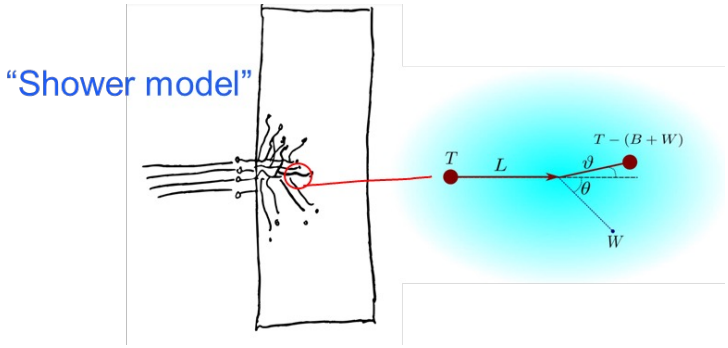
- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – first principles**
  - CASTEP
  - CP2K
  - CRYSTAL
  - ELK
  - ONETEP
  - QUESTAAL
  - W90



- **Different** scaling (→ compute cost, speed) and (numerical) accuracy
- **Different** functionalities available
- **Different** stage of maturity (→ documentation and ease of use)
- "One size fits all does not exist" (→"***what is the best code [for a given application]?***")

4.4: ALC_SUTOR

Python Suite for Monte-Carlo scattering calculations starting from the Energy Loss Function (ELF) generated by Linear Response TD-DFT calculations



Inelastic scattering

LR –TDDFT calculations of the ELF up to 60-100 eV (QE, Turboeels Lanczos algo, ELK, Questaal, etc) (k->0)

Sutor ELF for energies from 60,100 eV to 300 keV are merged from the NIST scattering tables in (k->0) https://physics.nist.gov/PhysRefData/FFast/html/form.html

Sutor: TDDFT and NIST ELF are merged to provide the ELF up to 300 keV and extended at finite k

Sutor: Extension of the ELF at finite k: (TD-DFT) or Drude model

Sutor: Integration in k and ω. Determination of the Inelastic mean free path and the Cumulative Probabilities at different Kinetic energies.

Elastic scattering

ELSEPA code: Determination of the cumulative elastic probabilities for different kinetic energies

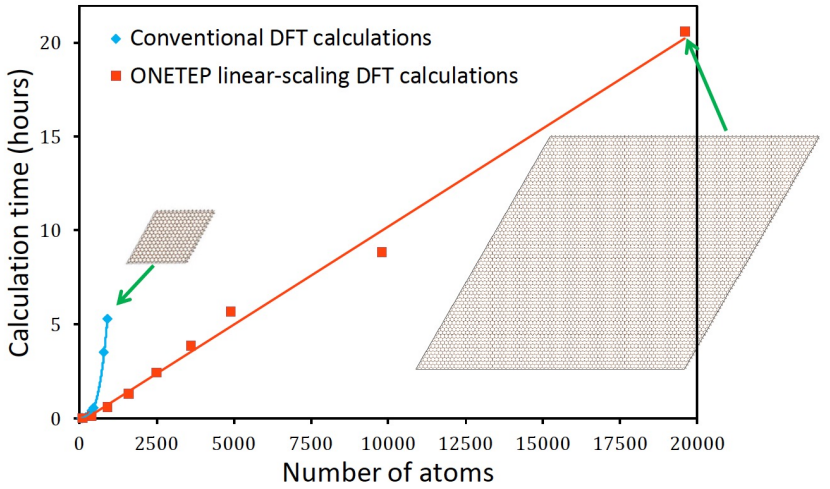
Seed Montecarlo code: **determination of reflective electron energy loss and Transmission, dose deposition, and secondary electrons**

stfc/ALC_SUTOR (github.com)
(Paolo Trevisanutto)

4.2-4.3: beyond linear-response – Better-scaling RT-TD-DFT for electron (-ion) dynamics

$$i\frac{\partial}{\partial t}\psi_j(\mathbf{r},t) = \left[\frac{1}{2}\left(-i\boldsymbol{\nabla} - \frac{1}{c}\mathbf{A}_{\mathrm{ext}}(t)\right)^2 + v_{\mathrm{ext}}(\mathbf{r},t) + v_{\mathrm{HXC}}[n](\mathbf{r},t)\right]\psi_j(\mathbf{r},t) \qquad n(\mathbf{r},t) = \sum_j |\psi_j(\mathbf{r},t)|^2$$

- **Predictive** first principles method, parameter free, unbiased
- **Quantum Mechanical** → capture coherences
- Includes electronic structure
- **Non-linear effects** automatically captured. Pump-probe experiments and not so low laser power ones [$A_{ext}(t)$ term…]
- Good for first **~100fs** (with electron-ion coupling can go longer)



www.onetep.org

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Credit: Peter Elliott

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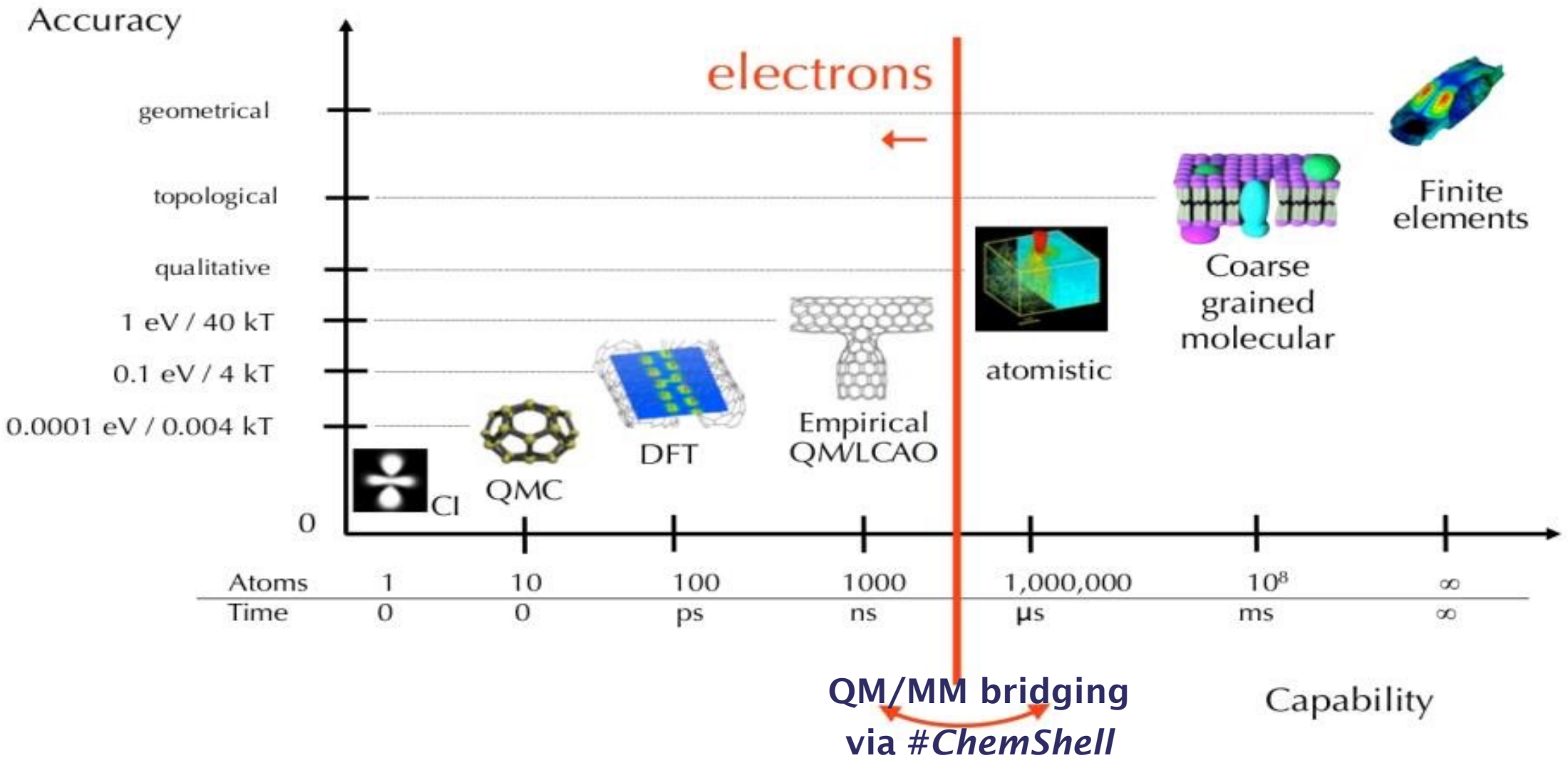
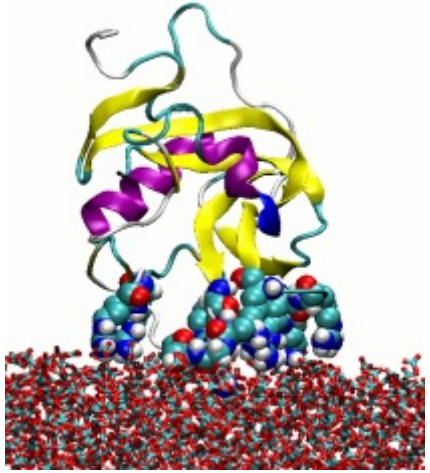
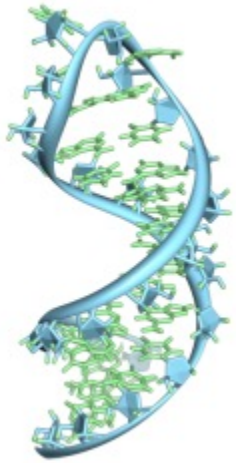
4.2-4.3: a multi-scale Modelling and Simulation platform



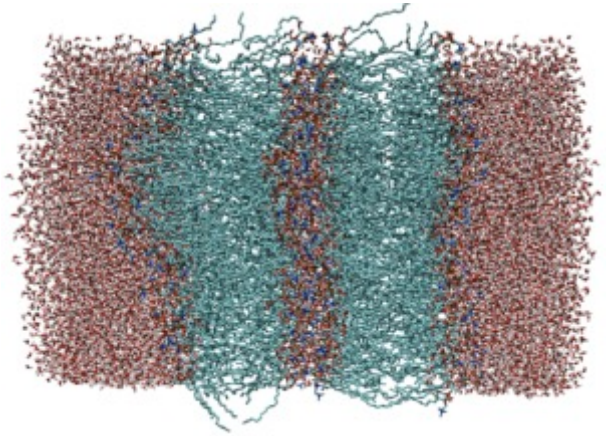
4.2-4.3: Molecular Dynamics with parameterised Hamiltonians – DL_POLY



Proteins
solvation & binding

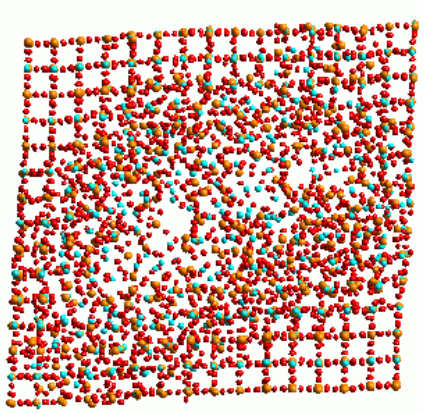


DNA strands
dynamics

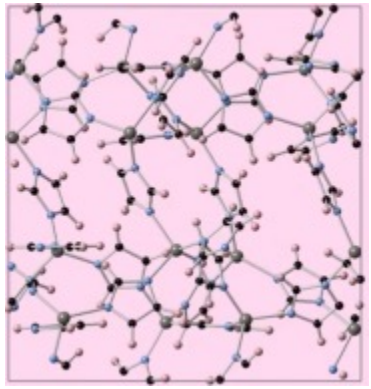


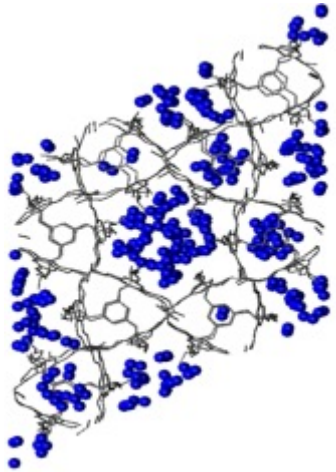
Membranes' processes

Drug polymorphs
& discovery

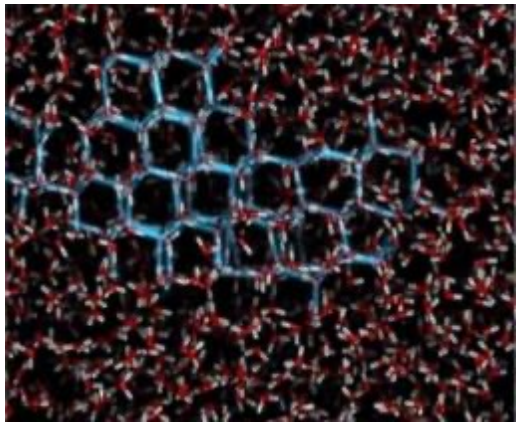


Crystalline & Amorphous
Solids – damage and recovery





Dynamic processes in
Metal-Organic & Organic Frameworks



Dynamics at Interfaces &
of Phase Transformations

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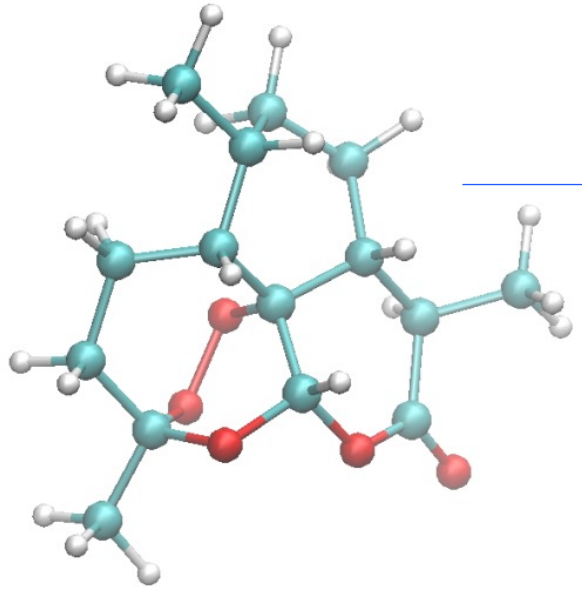
4.4: DL_FIELD

Capability to identify the exact chemical nature of every atom in the system and assign universal notation for a range of different force field schemes.

Artemisinin - drug against malaria



Molecular structure
(starting structures)

1 - O_acetal
2 - Cq_alkane
3 - HC_alkane
4 - Cp_alkane
5 - Cs_alkane
6 - HC_alkane
7 - Cs_alkane
8 - HC_alkane
9 - Ct_alkane
10 - HC_alkane
11 - HC_alkane
12 - Ct_alkane
13 - Cq_alkane
14 - HC_alkane
15 - Ct_alkane
16 - O_peroxide
17 - C_acetal
18 - OL_ester
19 - HC_alkane
20 - HC_acetal
21 - HC_alkane
22 - O_peroxide
23 - C_ester
24 - OE_ester
25 - Ct_alkane
26 - Cp_alkane
27 - HC_alkane
…
…

Atomistic chemical information
(useful for analysis)

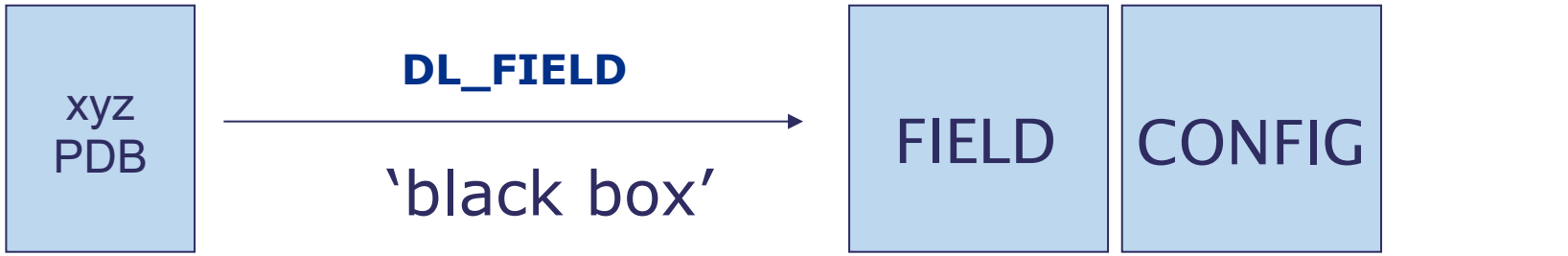
```
Generated by DL_FIELD v3.40
Units kcal/mol
Molecular types 1
Molecule name not_define
nummols 1
atoms 42
OS      15.99940   -0.40000   1   0
CT      12.01150    0.40000   1   0
HC       1.00797    0.06000   1   0
CT      12.01150   -0.18000   1   0
CT      12.01150   -0.12000   1   0
HC       1.00797    0.06000   1   0
CT      12.01150   -0.12000   1   0
HC       1.00797    0.06000   1   0
...
```

Force field model
(MD simulations)

One-step conversion process

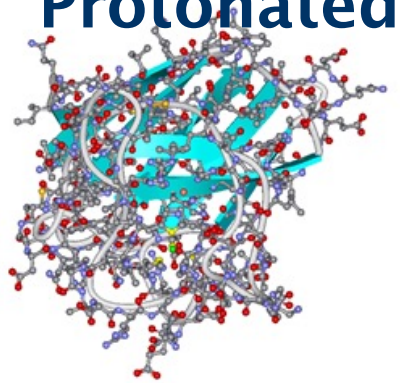
4.4: DL_FIELD

- **Flesh on bone** – chemical typing and identification, via DL_F notation, forming complete chemistry perspective
- **Organic Fields** – AMBER+Glycam, CHARM, OPLS-AA, PCFF, Drieding, CHARM19 (united atom) + many others
- **Inorganic Fields** – some access for organic/inorganic interfaces including core-shell polarisation, etc.
- **Solvation Features, Auto-CONNECT feature** for mapping complex random structures such as gels and random polymers
- Input units freedom and molecular rigidification



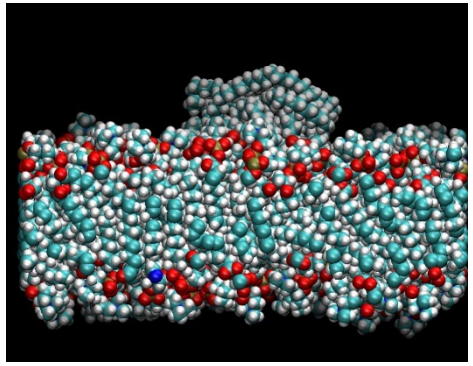
SOD

Protonated

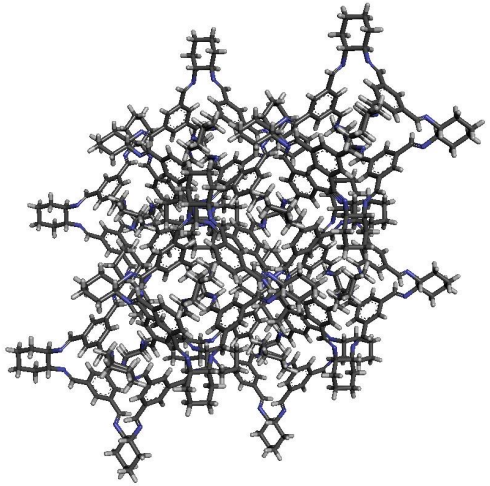


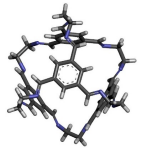
4382 atoms (excluding water)
19400 bond interactions
7993 angles interactions
13000 dihedral interactions
730 VDW interactions

4.2-4.4: DL_FIELD to DL_POLY Studies

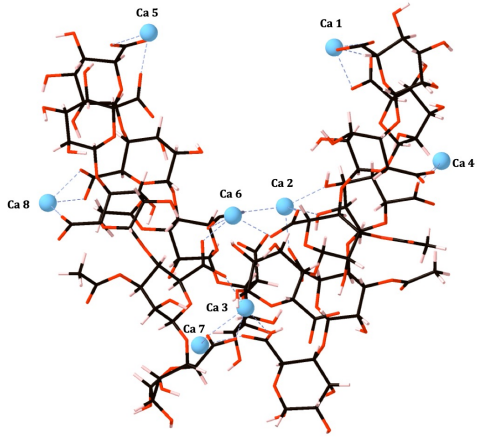


Polymer on lipid membrane

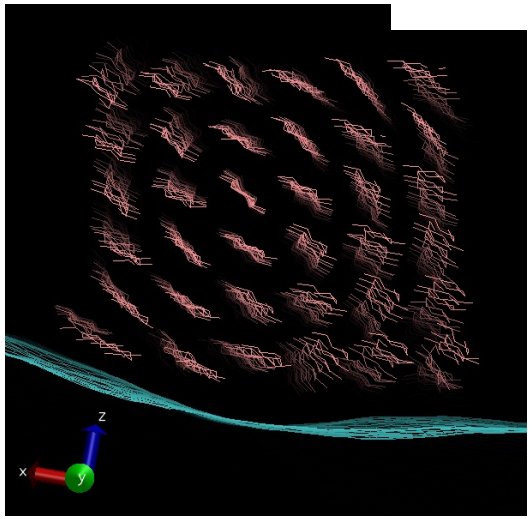




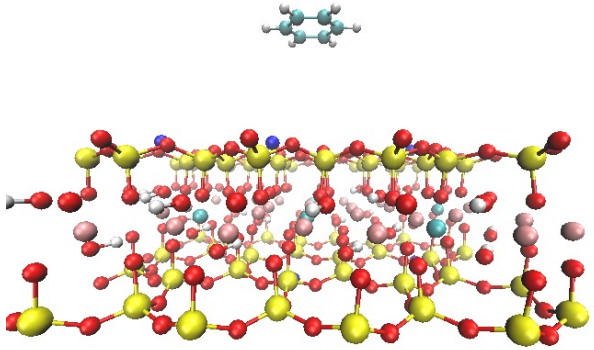
Molecular cages



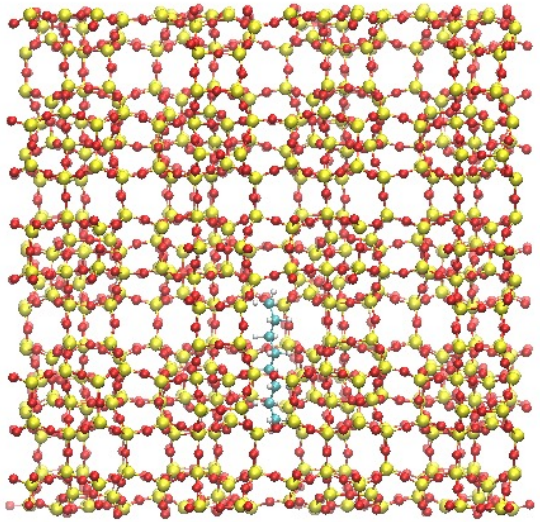
Biomolecules



Graphene-cellulose contact adhesion



Bio-inorganic systems
(montemorillonite+organic)



Zeolite+ adsorbates (hydrocarbons, organic molecules, gas, etc)

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Central Laser Facility

Science and Technology Facilities Council
ISIS Neutron and Muon Source

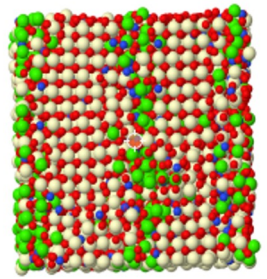
diamond

Science and Technology Facilities Council
Scientific Computing

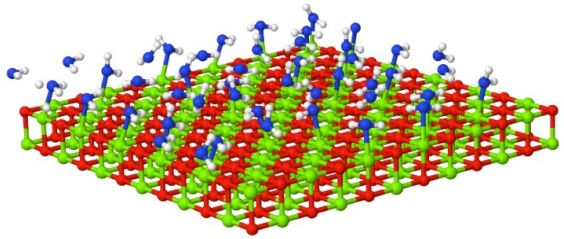
4.2-4.3: Monte Carlo with parameterised Hamiltonians – DL_MONTE

- **A stochastic method to study phase equilibria, (meta-) stability and coexistence**
  – *getting cool science out of "dice throwing"*

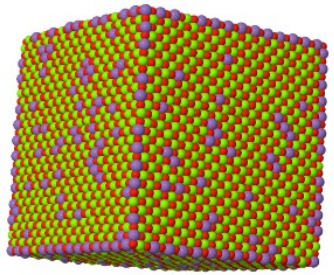
- **Advantages of MC beyond MD**
  – atom/molecule insertions/deletions/mutations → **chemical equilibrium**
  – "artistic" moves and pathways to skip energy barriers → **metastability & coexistence**
  – efficient biased exploration of configuration space → **thermodynamic equilibrium**
  – powerful at riding on free-energy landscapes → **transition states & rare events**



- **Lack of time & forces**
  – no mapping onto real time scale -> **kinetic/transport properties inaccessible**
  – no momenta / real forces -> **slow motion in dense systems**



- **Broad application scope**
  –useful for calculation of **phase diagrams** and associated **thermodynamic properties** in bulk and at interfaces especially for materials and condensed matter systems
  – opportunities to develop enhancements and explore applications in biological systems



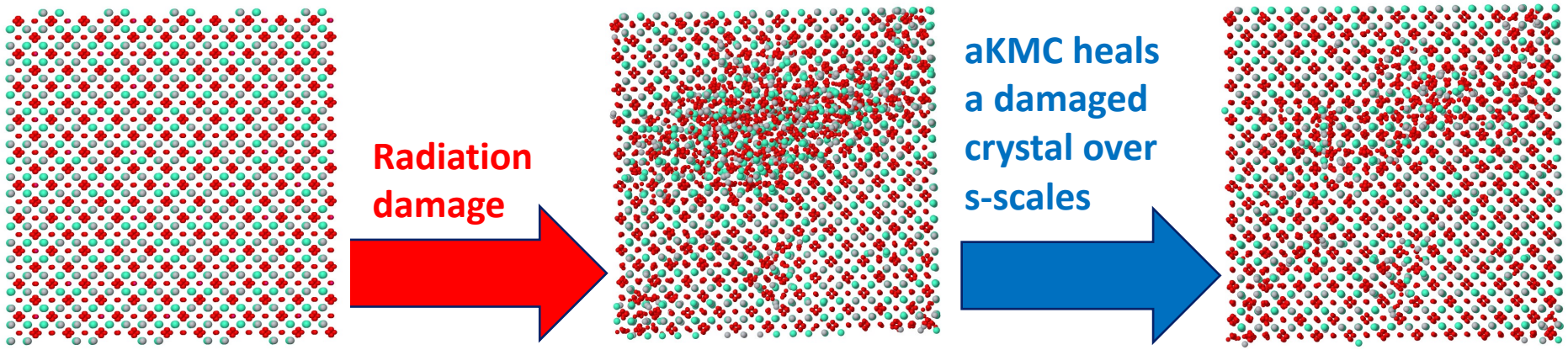
4.2-4.3: adaptive kinetic Monte Carlo (aKMC) with parameterised or first principles (or ML-derived) Hamiltonians – ACDC

- Kinetic Monte Carlo (**KMC**) - simulates **state-to-state kinetics of a rare event system**. Rare events correspond to the thermal activation of atoms from one energy basin to another on the potential energy surface. If the rates of these transitions are known, KMC can be used to simulate kinetics over long time scales.

- Adaptive Kinetic Monte Carlo (**aKMC**) is a method for determining all of the transitions from each state on the fly, **eliminating the need to use a pre-defined rate-list**.

- Molecular Dynamics timescales are inaccessible and insufficient for capturing the dynamics of rare events

- Many technological important processes and materials rely on **solid state diffusion**!
  - Growth of thin films: magnetic recording media, electronic semiconductor devices, LEDs, optical coatings**.**
  - Defect mobility and clustering: batteries and solid oxide fuel cells, nuclear materials



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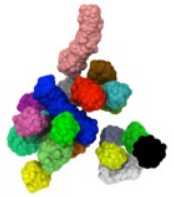
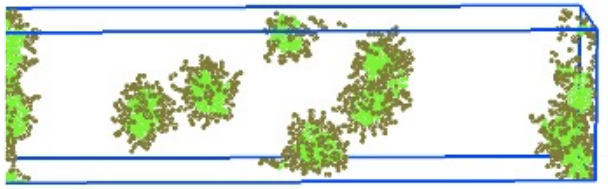
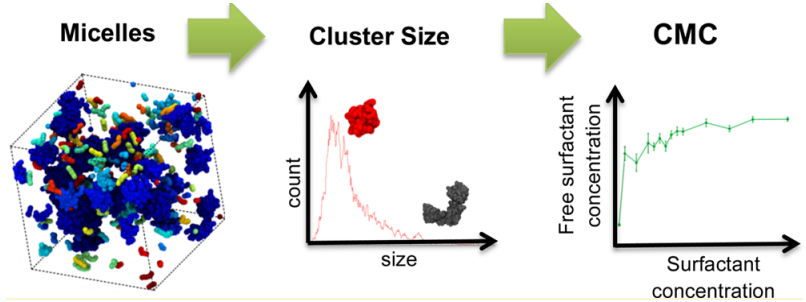
4.2-4.3 DPD and LBE – DL_MESO





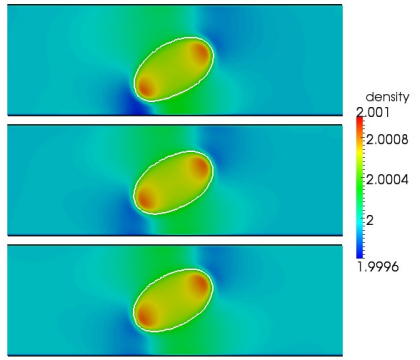
Dissipative Particle Dynamics (DPD)

- Particle based method akin to Molecular Dynamics
- Bottom-up approach to describe **fluidic systems** at the engineering scale
- Used for simulation of **mesophases of colloids** (strong industrial application – Unilever, DOW, Syngenta, Infineum) and their **rheological behaviour** (mixing and dynamics of phases, including CMC, micellization, vesicle formation, condensation, shear flows LEBC)
- Novel DPD interaction (nDPD), capable to model true interfaces with **access to solid and vapour**

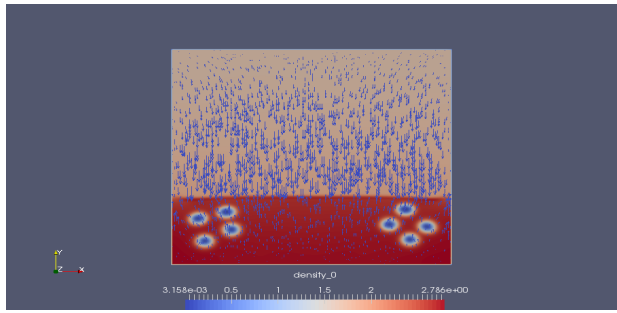


Lattice Boltzmann Equation (LBE)

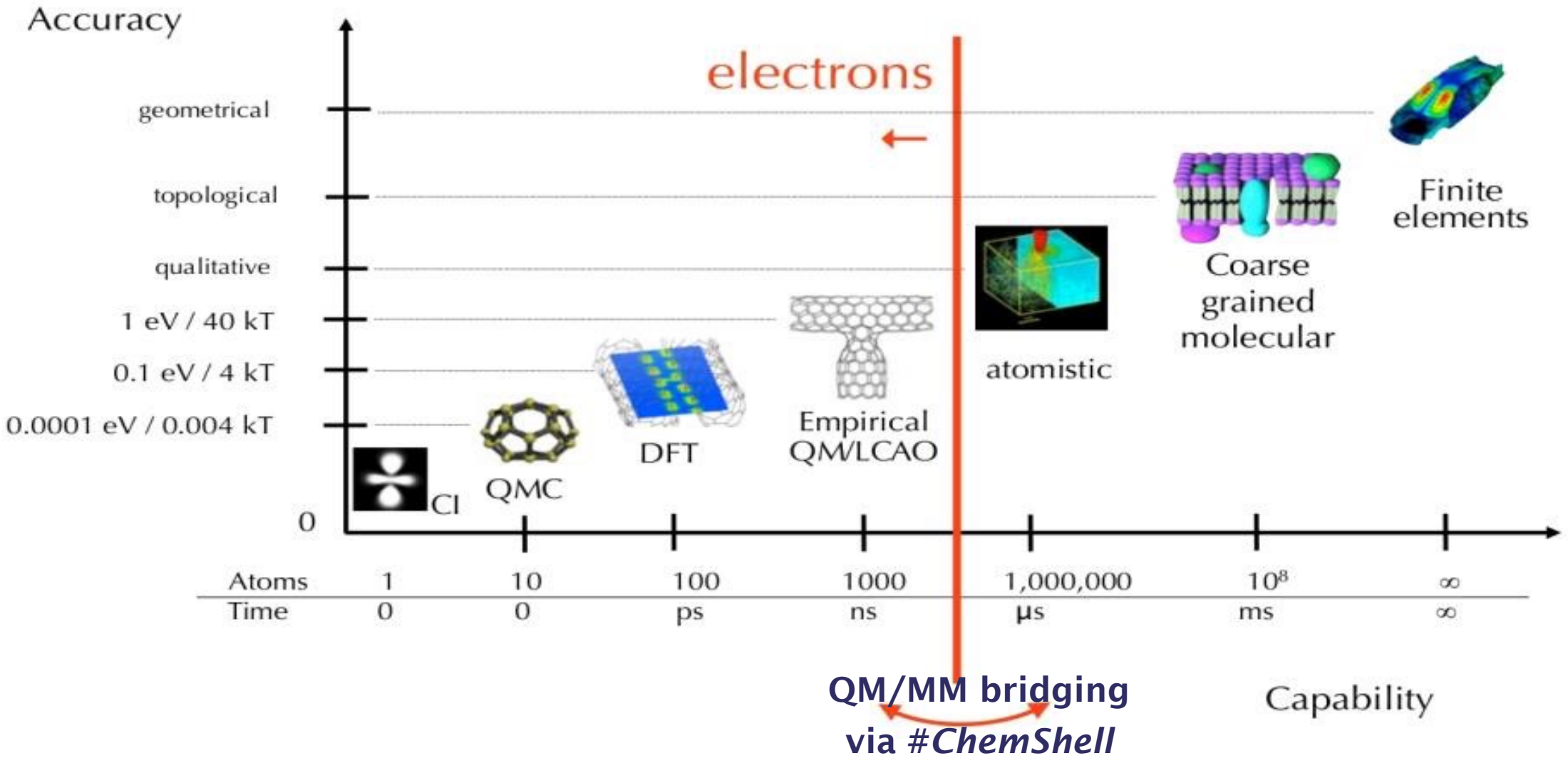
- Mesh based method akin to cellular automata
- Top-down, statistical mechanics approach to particle motion, governed by collisions and propagation
- Target engineering scale behaviour of **fluidic flows** and **interface dynamics**
- Examples: von Karman instability, convections, resonant acoustic mixing of suspensions



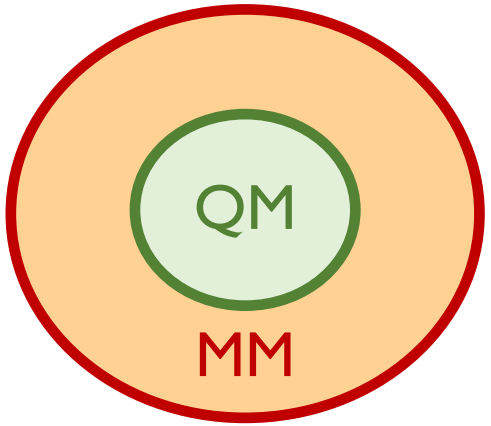
Both methods operate at mesoscopic (phenomenological) length and time scales between those of atomistic and continuum methods



4.2-4.3: a multi-scale simulation and modelling platform



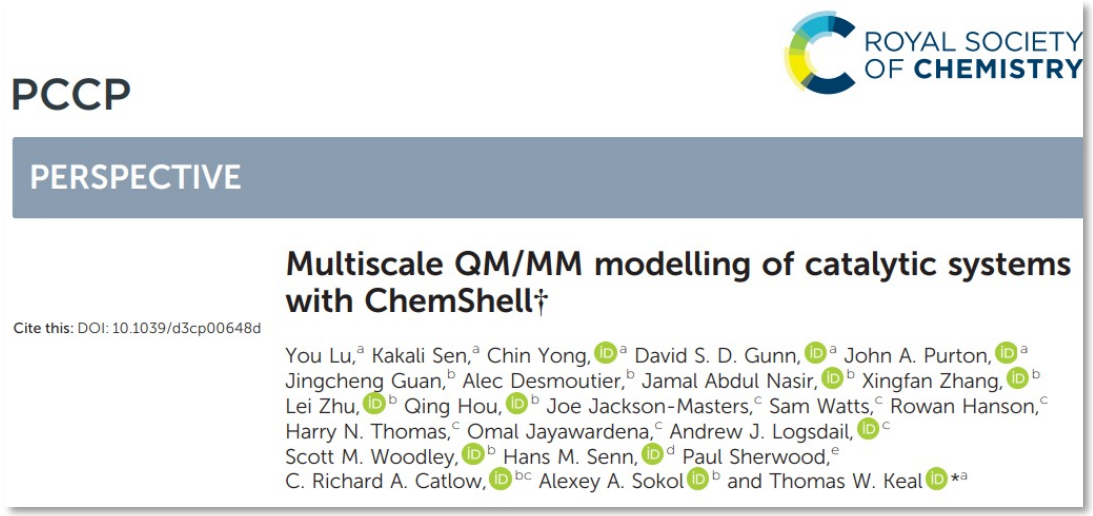
4.2-4.3-4.4: Multiscale QM/MM modelling



- **Well established approach to overcoming size limit on electronic structure calculations**
  - Combine quantum and classical methods:
    - QM where high accuracy is required (e.g. active site)
    - Efficient MM calculation for the environment

ChemShell

- Scriptable computational chemistry environment
- Particular focus on QM/MM calculations for complex chemical systems
- Flexible, modular design, integrating with a wide range of QM and MM packages


PCCP

ROYAL SOCIETY OF CHEMISTRY

PERSPECTIVE

Multiscale QM/MM modelling of catalytic systems with ChemShell†

Cite this: DOI: 10.1039/d3cp00648d

You Lu,[a] Kakali Sen,[a] Chin Yong,[a] David S. D. Gunn,[a] John A. Purton,[a] Jingcheng Guan,[b] Alec Desmoutier,[b] Jamal Abdul Nasir,[b] Xingfan Zhang,[b] Lei Zhu,[b] Qing Hou,[b] Joe Jackson-Masters,[c] Sam Watts,[c] Rowan Hanson,[c] Harry N. Thomas,[c] Omal Jayawardena,[c] Andrew J. Logsdail,[c] Scott M. Woodley,[b] Hans M. Senn,[d] Paul Sherwood,[e] C. Richard A. Catlow,[bc] Alexey A. Sokol[b] and Thomas W. Keal[*a]

Phys. Chem. Chem. Phys., 2023, **25**, 21816-21835

UKRI Science and Technology Facilities Council

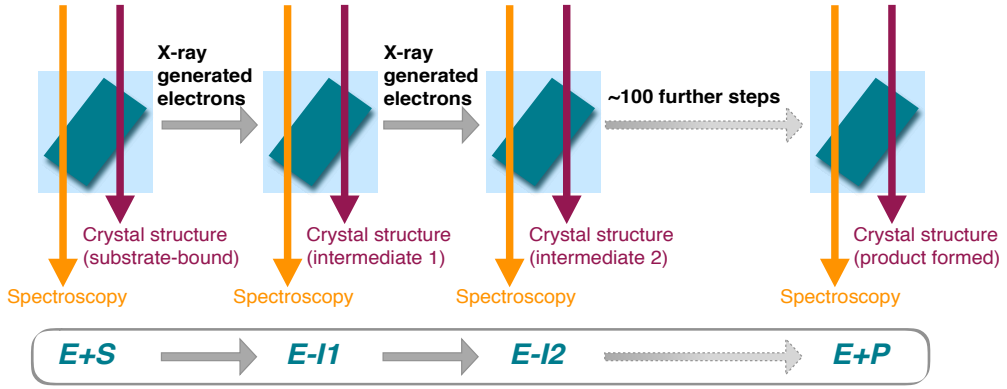
Credit: Thomas Keal

chemshell.org

@chemshproject

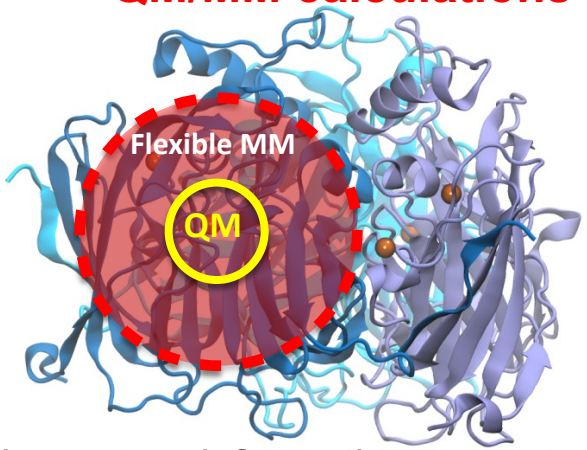
QM/MM energy profiles

Serial crystallography



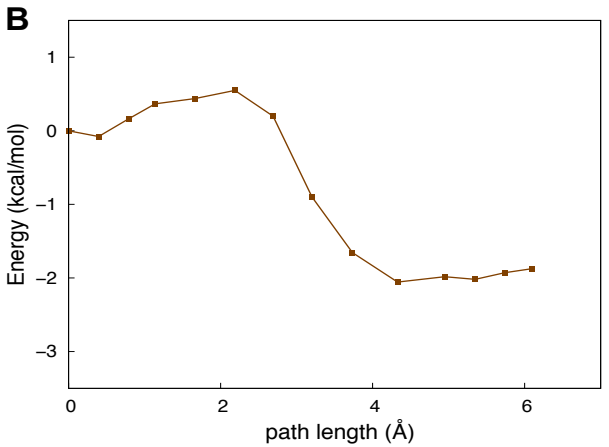
Catalytic "movie" of redox reaction in the crystal

QM/MM calculations



Complementary information on protonation and oxidation states and reaction mechanisms

- X-ray crystal structures are the starting point for setting up multiscale models
- Example is a BBSRC-funded collaboration with Prof. Mike Hough (Diamond) and researchers at Essex and Bristol
- ALC funding could be used to improve accuracy, accessibility, automation/throughput, and handling of ensembles of structures from time resolved experiments



QM/MM calculated reaction path following reduction of a nitrite-bound copper nitrite reductase

K. Sen, M.A. Hough, R.W. Strange, C. Yong, T.W. Keal, *J. Phys. Chem. B,* 2021, **125**, 9102

QM/MM Vibrational spectroscopy

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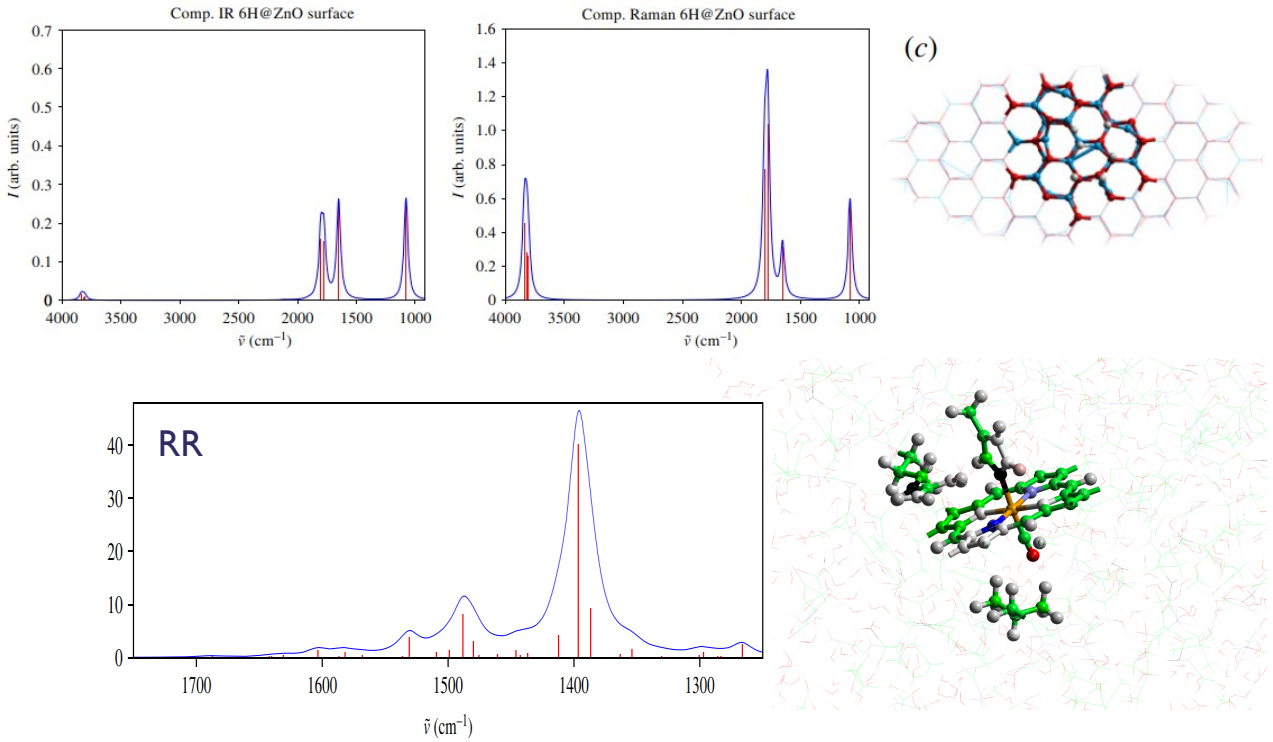
Engineering and Physical Sciences Research Council



PHILOSOPHICAL TRANSACTIONS A

royalsocietypublishing.org/journal/rsta

Research

Cite this article: Guan J *et al*. 2023 Computational infrared and Raman spectra by hybrid QM/MM techniques: a study on molecular and catalytic material systems. *Phil. Trans. R. Soc. A* 20220234. https://doi.org/10.1098/rsta.2022.0234

Received: 23 December 2022
Accepted: 4 April 2023

Computational infrared and Raman spectra by hybrid QM/MM techniques: a study on molecular and catalytic material systems

Jingcheng Guan[1], You Lu[2], Kakali Sen[2], Jamal Abdul Nasir[1], Alec W. Desmoutier[1], Qing Hou[1,9], Xingfan Zhang[1], Andrew J. Logsdail[3], Gargi Dutta[1,5], Andrew M. Beale[1,4], Richard W. Strange[6], Chin Yong[2], Paul Sherwood[7], Hans M. Senn[8], C. Richard A. Catlow[1,3,4], Thomas W. Keal[2] and Alexey A. Sokol[1]

- QM/MM simulations of IR, Raman and Resonance Raman spectra can help interpret experimental data, e.g. by identifying transient species in catalytic cycles with a realistic model of the chemical environment
- ALC PhD studentship (with Prof. Richard Catlow, UCL, and project partners Prof. Stewart Parker, ISIS and Dr Paul Donaldson, CLF) started Oct 2023, to apply these EPSRC-funded developments to investigate high temperature superconductivity in cuprates

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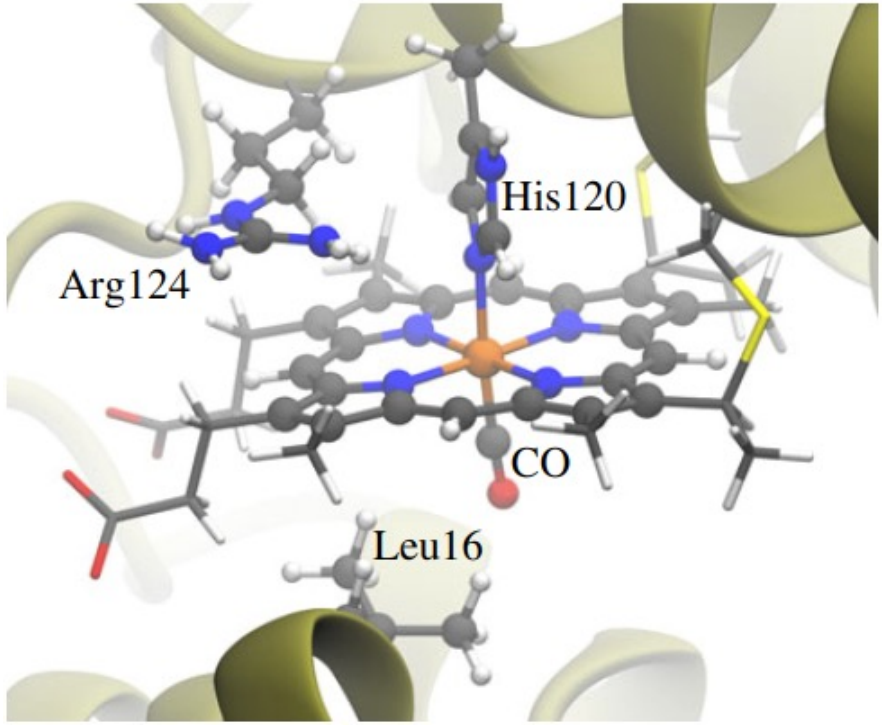
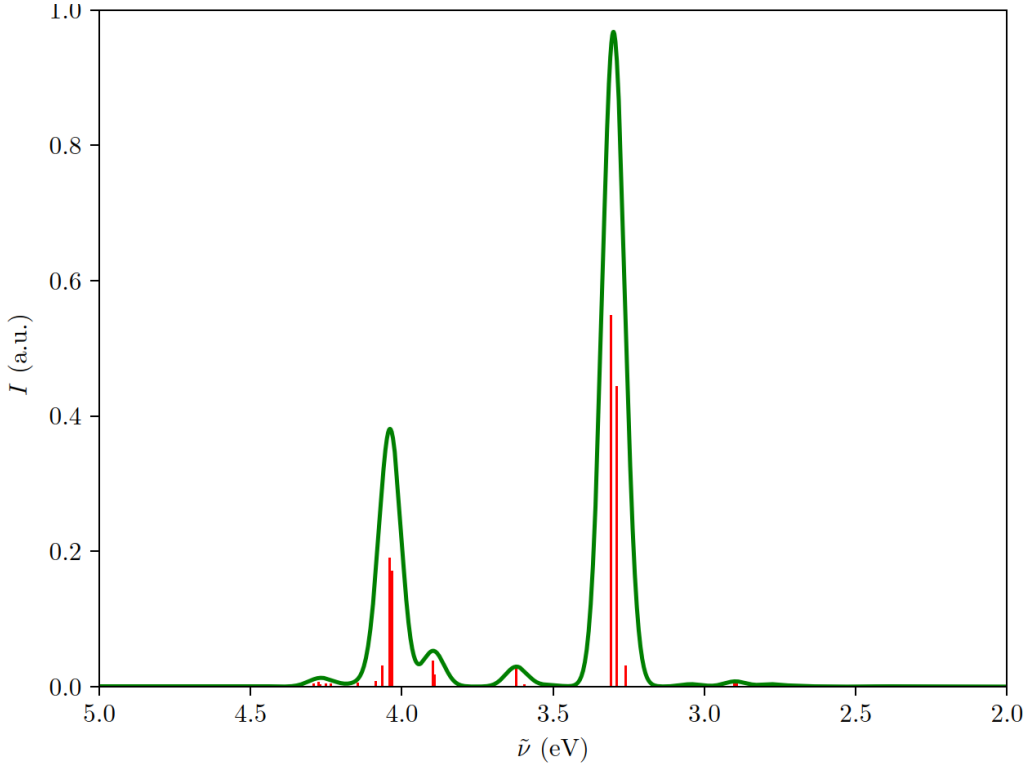
- Additional ALC funding would enable better integration with facilities software (e.g. AbINS for inelastic neutron scattering), accelerate progress on anharmonic corrections and address coupling of electronic and vibrational degrees of freedom

QM/MM excited electronic states

- Straightforward to use e.g. TD-DFT (or higher theory level) in the context of hybrid QM/MM calculations to generate a UV-vis spectrum of a complex chemical system

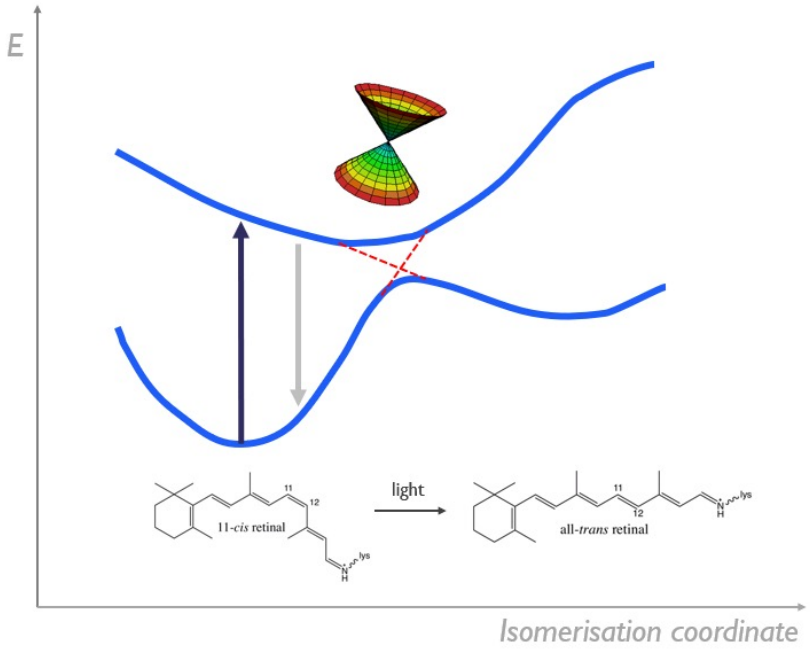




Science and Technology Facilities Council

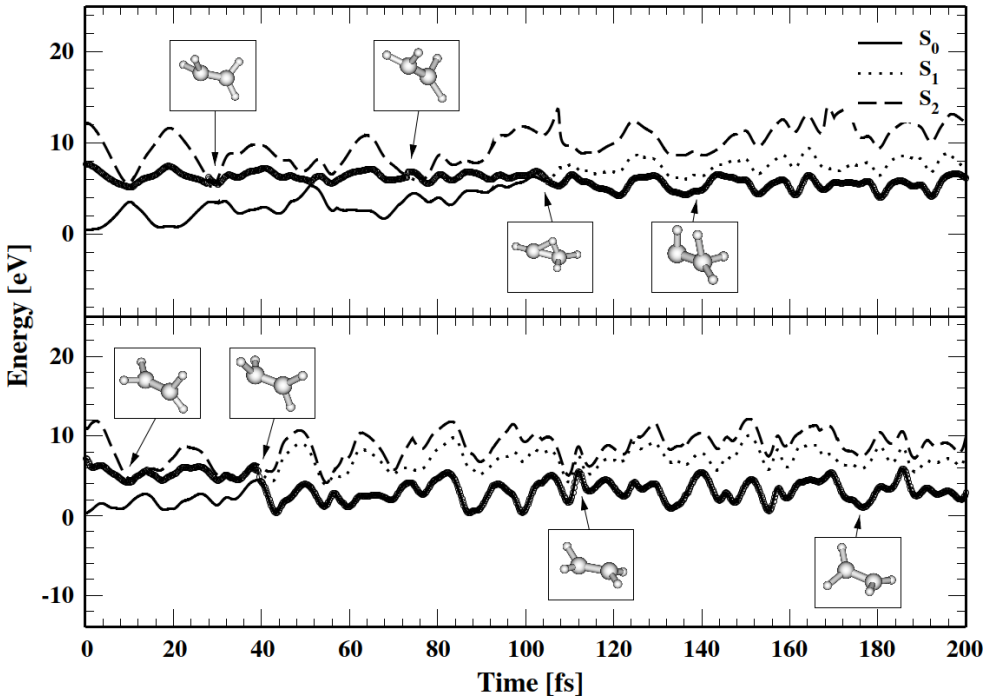
J. Guan, Y. Lu, K. Sen, T.W. Keal et al, *Phil. Trans. R. Soc. A*, 2023, **381**, 20220234

Surface crossing optimization methods



- Explore photochemical reaction mechanisms

Ultrafast electron dynamics



- Enables comparisons with time-resolved experiments

ALC funding will enable integration of these methods into the latest version of ChemShell and further development to support the work of e.g. CLF-ARTEMIS

Science and Technology Facilities Council

J. Kästner, J.M. Carr, T.W. Keal, W. Thiel, A. Wander, P. Sherwood, *J. Phys. Chem. A,* 2009, **113**, 11856
E. Fabiano, T.W. Keal, W. Thiel, *Chem. Phys.,* 2008, ***349***, 334
Z. Lan, Y. Lu, E. Fabiano, W. Thiel, *ChemPhysChem*, 2011, **12**, 1989

SCD-CMMS' software menu as of today (with working hyperlinks…)

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – parameterised Hamiltonians**
  - DL_FIELD
  - DL_POLY
  - DL_MONTE
  - ACDC (not released yet)

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – Coarse-grained/mesoscale lattice models**
  - DL_MESO

- **4.2-4.3-4.4: Energy (forces) evaluation + responses/dynamics – multi-scale (QM/MM)**
  - ChemShell
  - DL_FIND

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – machine learned potentials**
  - APD
  - Janus

- **4.4 Input set-up and workflow management:**
  - Aiida-MLIP
  - ALC_EQCM
  - ALC_SUTOR
  - DLPOLY-PY
  - Muon Galaxy
  - Shapespyer

- **4.5 output analysis:**
  - ALC_TRAJECTORY
  - DL_ANALYSER
  - Shapespyer
  - DLPOLY-PY

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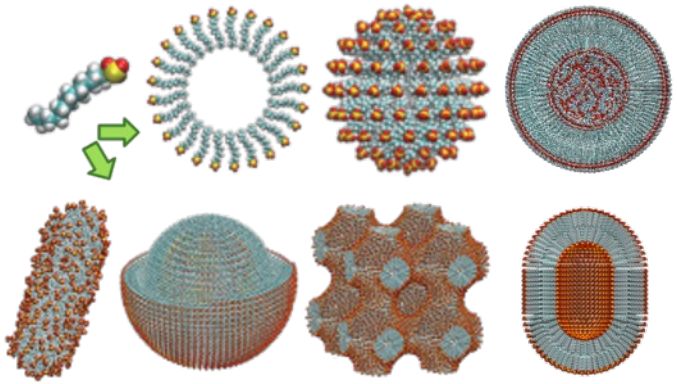
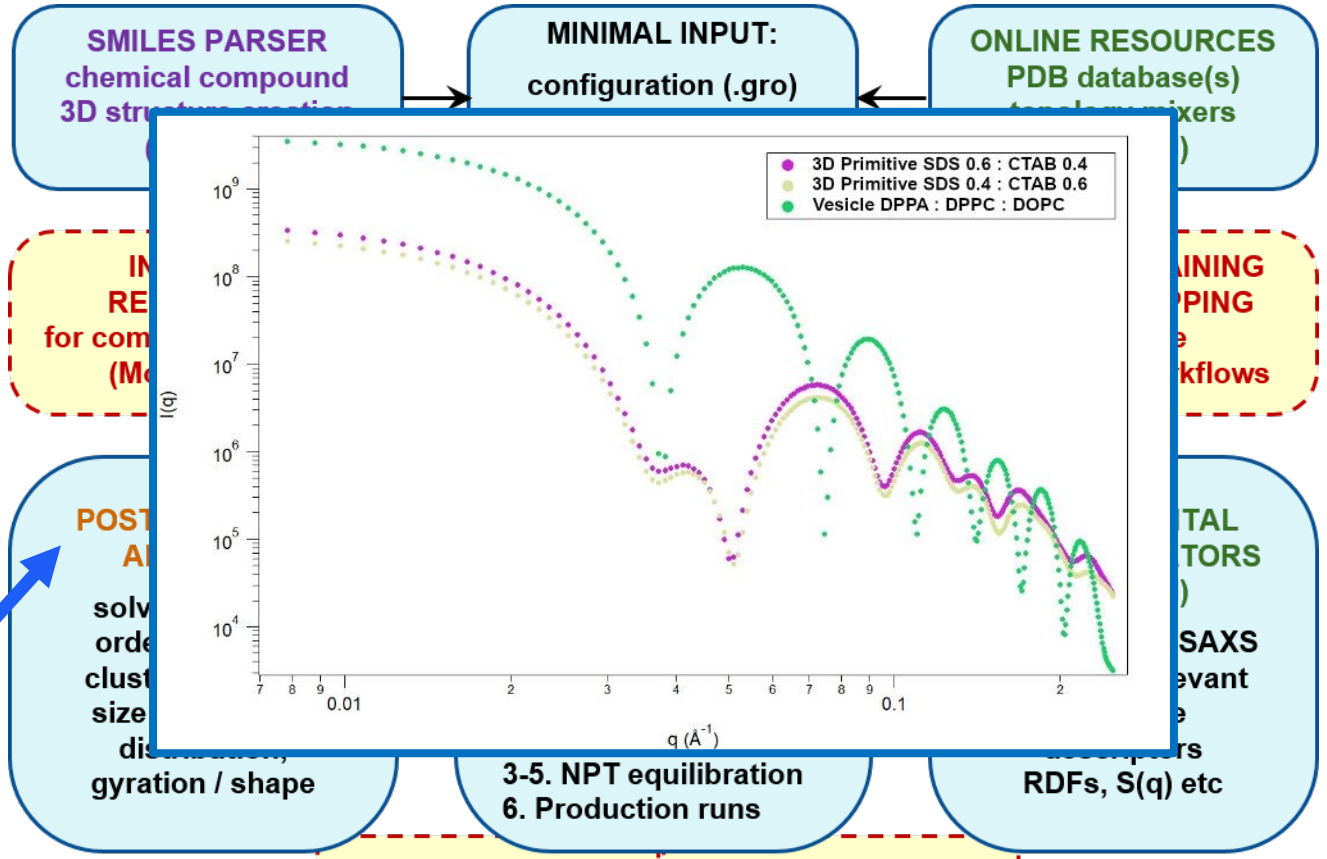
4.4-4.5: Shapespyer – a python framework for soft matter simulations

Project overview

- **Generation, simulation & analyses**
  of pre-equilibrated molecular structures
  for functional soft matter research
- **Target structures / trajectories**
  for academic and industrial R&D;
  Linking with SANS, SAXS experiments
- **Provide automated workflows**
  Python library, APIs, Bash scripts
- **Partners / collaborators:**
  Maggie Holme, *Chalmers, Sweden*
  Hanna Barriga, *Karolinska Institutet, Sweden*
  Tom Headen, *ISIS, UK*
  SasView Team, *International*





www.scd.stfc.ac.uk/Pages/Shapespyer.aspx
gitlab.com/simnavi/shapespyer

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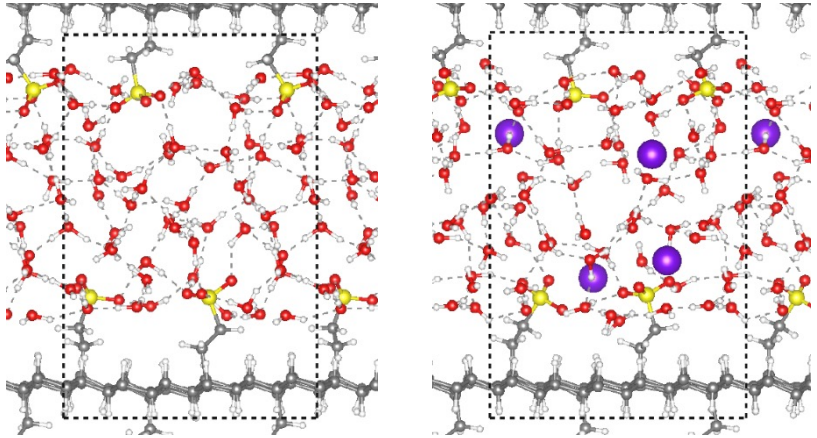
Andrey Brukhno (Co-I), *Computational Chemistry, SCD*

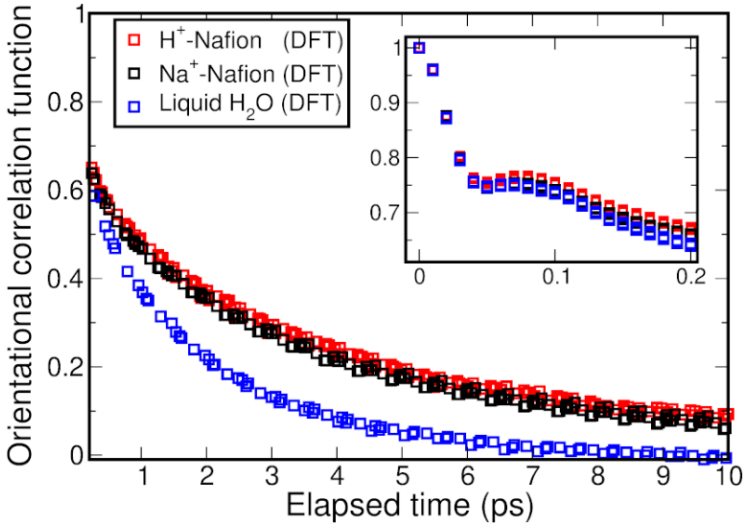
PI: James Doutch, *ISIS,* **Co-I's: Tim Snow,** *Diamond,* **Michael Seaton, John Purton,** *SCD*

4.5: ALC_TRAJECTORY

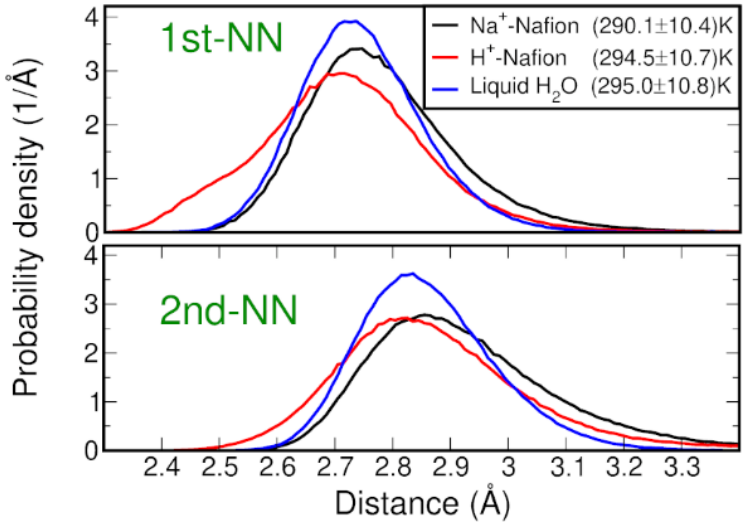
Fortran suite to compute **orientational** and **transfer correlation functions** from **reactive** Molecular Dynamics trajectories (generated at any level of theory)

Originally developed to interpret transient laser-spectroscopy experiments on proton-conducting Nafion membranes at CLF-ULTRA (Paul Donaldson)





The dynamical orientation of nano-confined H_2O in H^+- and Na^+-Nafion is similar…

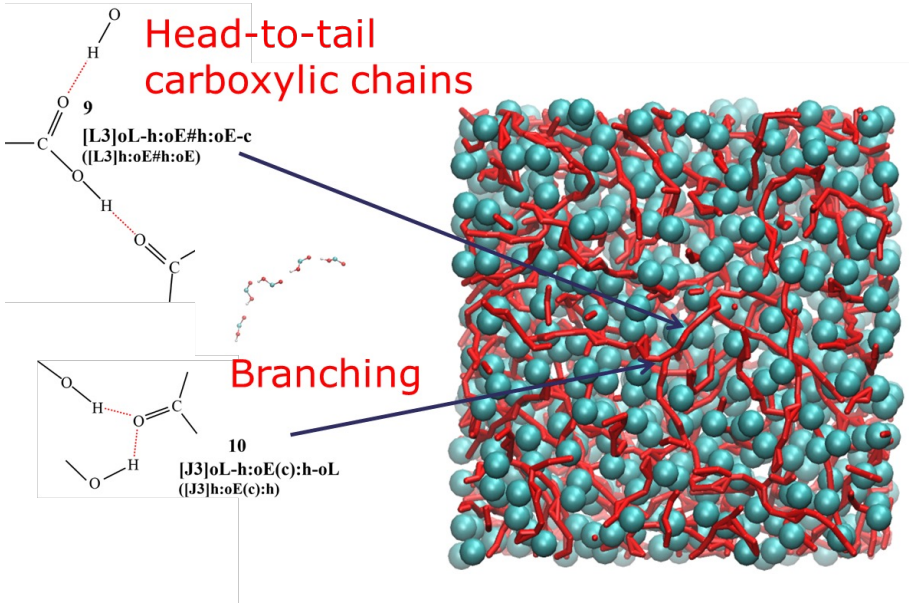


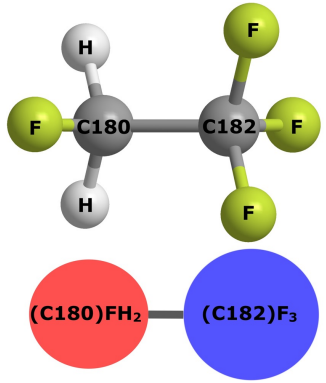
…but the intermolecular-interactions are stronger in H^+-Nafion, in agreement with 2D-IR experiments

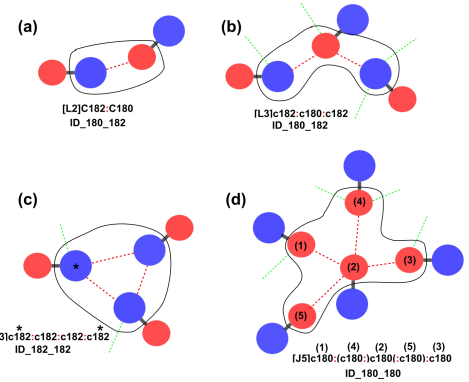
https://github.com/STFC/ALC_TRAJECTORY (Ivan Scivetti)

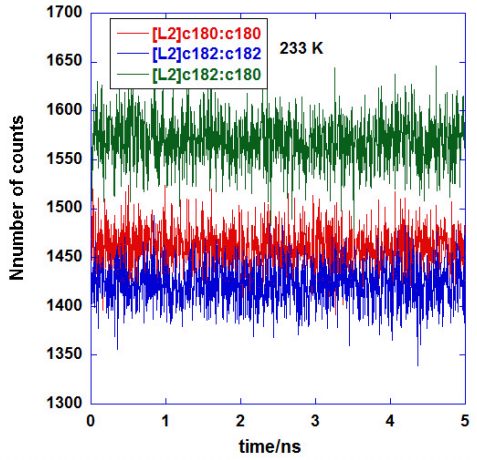
4.5: DL_ANALYSER

Unique feature: DANAI - a standard expression system to annotate specific atomic interactions.
Useful for *automated* rationalisation of structured interactions in complex condensed phase systems









At low temperatures, the predominant interactions in liquid is the 'head-to-tail' interactions, [L2]c182:c180

Journal of Molecular Liquids 383(1):121993
DOI: 10.1016/j.molliq.2023.121993

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Scientific Computing

Credit: C. W. Yong and I. T. Todorov
Molecules (2018), **23**, 36

diamond

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SCD-CMMS' software menu as of today (with working hyperlinks…)

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – parameterised Hamiltonians**
  - DL_FIELD
  - DL_POLY
  - DL_MONTE
  - ACDC (not released yet)

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – Coarse-grained/mesoscale lattice models**
  - DL_MESO

- **4.2-4.3-4.4: Energy (forces) evaluation + responses/dynamics – multi-scale (QM/MM)**
  - ChemShell
  - DL_FIND

- **4.2-4.3: Energy (forces) evaluation + responses/dynamics – machine learned potentials**
  - APD
  - Janus

- **4.4 Input set-up and workflow management:**
  - Aiida-MLIP
  - ALC_EQCM
  - ALC_SUTOR
  - DLPOLY-PY
  - Muon Galaxy
  - Shapespyer

- **4.5 output analysis:**
  - ALC_TRAJECTORY
  - DL_ANALYSER
  - Shapespyer
  - DLPOLY-PY

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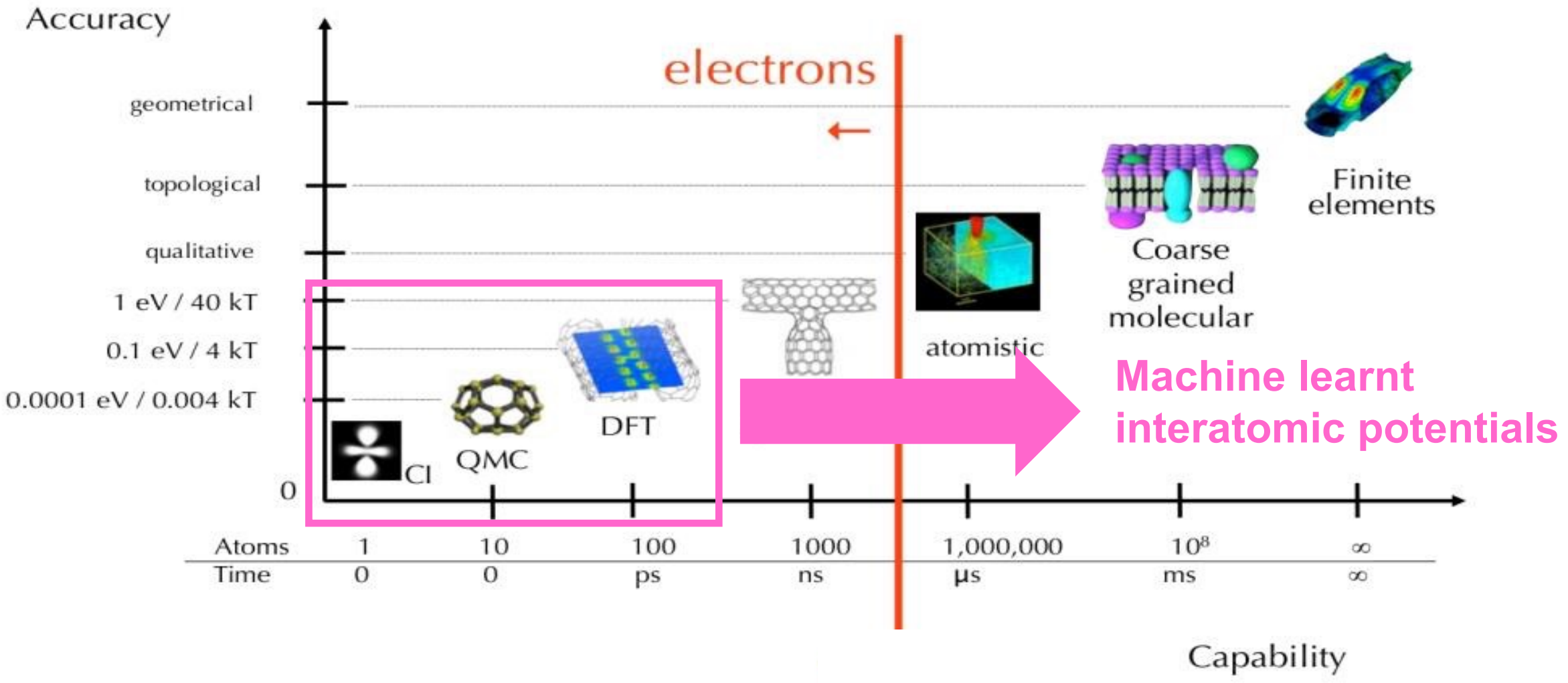
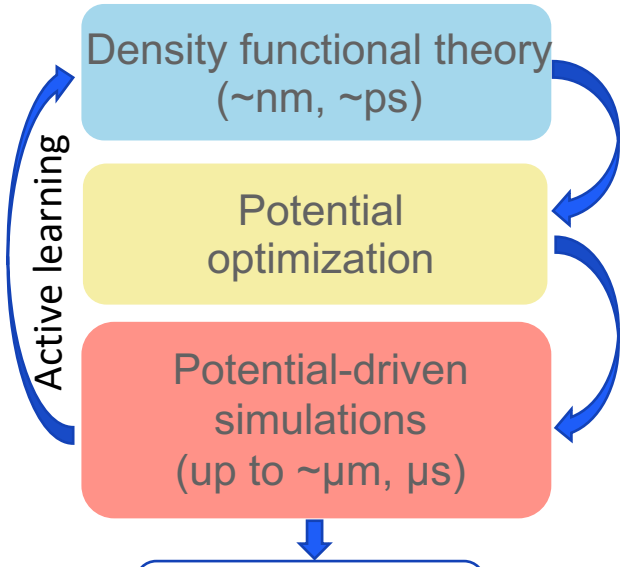
4.2-4.3: a multi-scale Modelling and Simulation platform



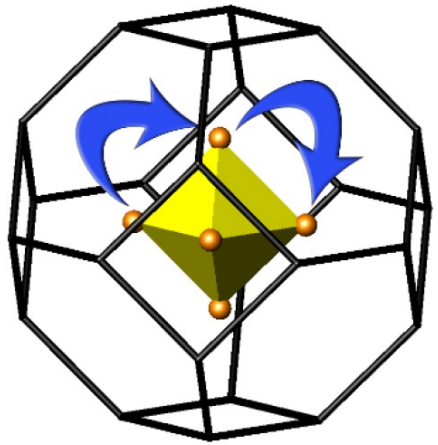
4.2-4.4: Automated ML-potential development (APD) workflow

- Black box solution to automate interatomic potential development, including: density functional theory, potential optimization, molecular dynamics simulations, active learning and property calculations
- https://gitlab.com/AndyDuff123/automated-potential-development
- Funded by Ada Lovelace Centre, in collaboration with ISIS (David Voneshen, Russell Ewings, Helen Walker)
- Supports reference-free MEAM potentials, being extended to MTP ML potentials
- Methodology paper applied to $BaZrO_3$: Comp. Phys. Comm. **293**, 108896 (2023)



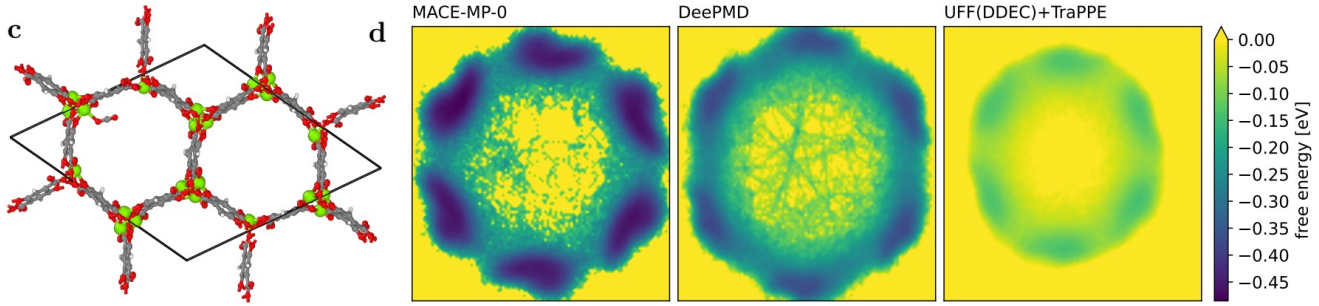
Beyond Rattling: Tetrahedrites as Incipient Ionic Conductors

- First case study involving the APD workflow, in collaboration with University of Reading and ISIS
- Thermoelectrics allow waste heat conversion to useful energy
- New material class, 'incipient ionic conductors' discovered that can achieve this while mitigating material decomposition, often inherent with such materials
- High impact publication: Adv. Mat. **35**, 2306088 (2023)
- ISIS Science Highlight: https://www.isis.stfc.ac.uk/Pages/Science-Highlights.aspx



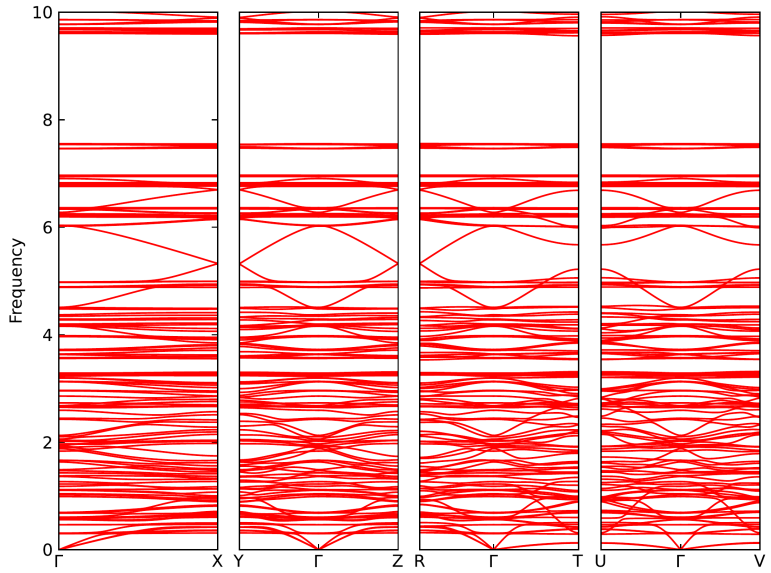
Credit: Andrew Duff

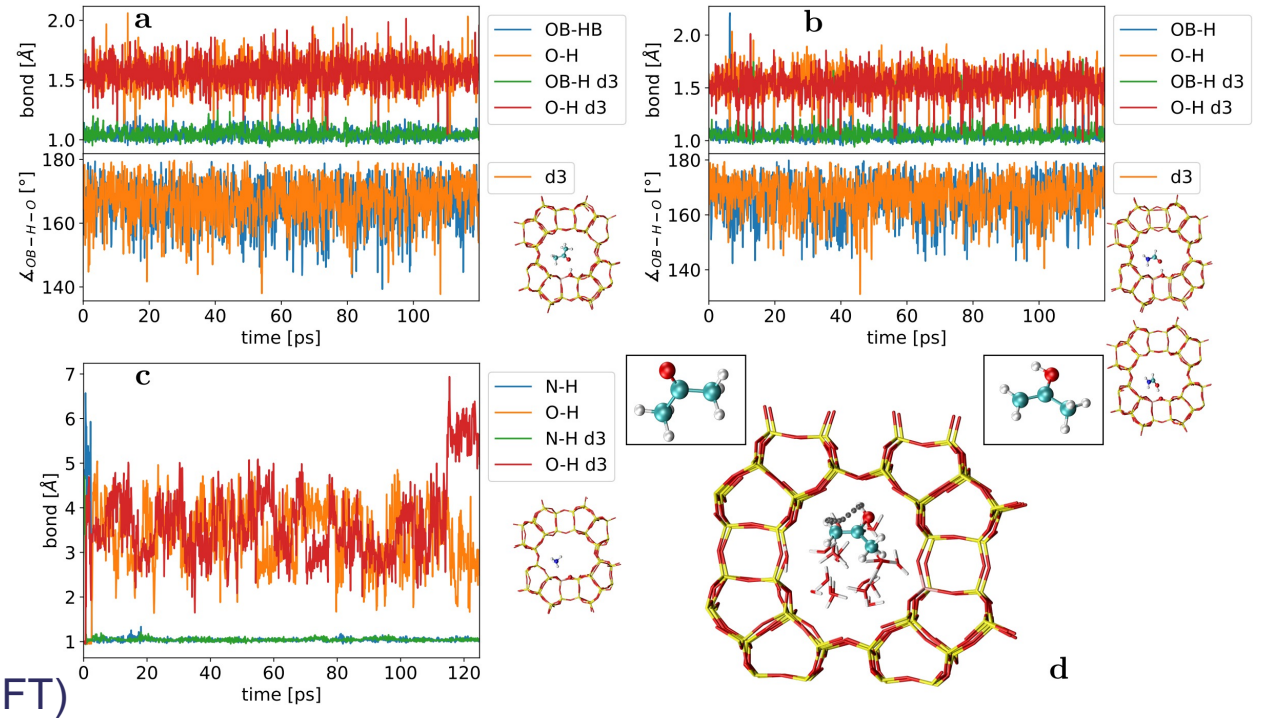
4.2-4.3: MACE machine learnt interatomic potentials



- condensed phases: cresol, water
- SrTiO3 – pimd
- Confined systems, endofuleranes
- Catalysis in MOFs

Metal Organic Frameworks – CO2 **adsorption** in Mg-MOF-74*





Phonon calculations – IRMOF 10, 1h(MACE) vs weeks (DFT)
https://arxiv.org/abs/2401.00096

Deprotonation of small molecules(acetone a, formamide b, NH3 c) in zeolites at Bronstead acid sites*, d) acetone enolization

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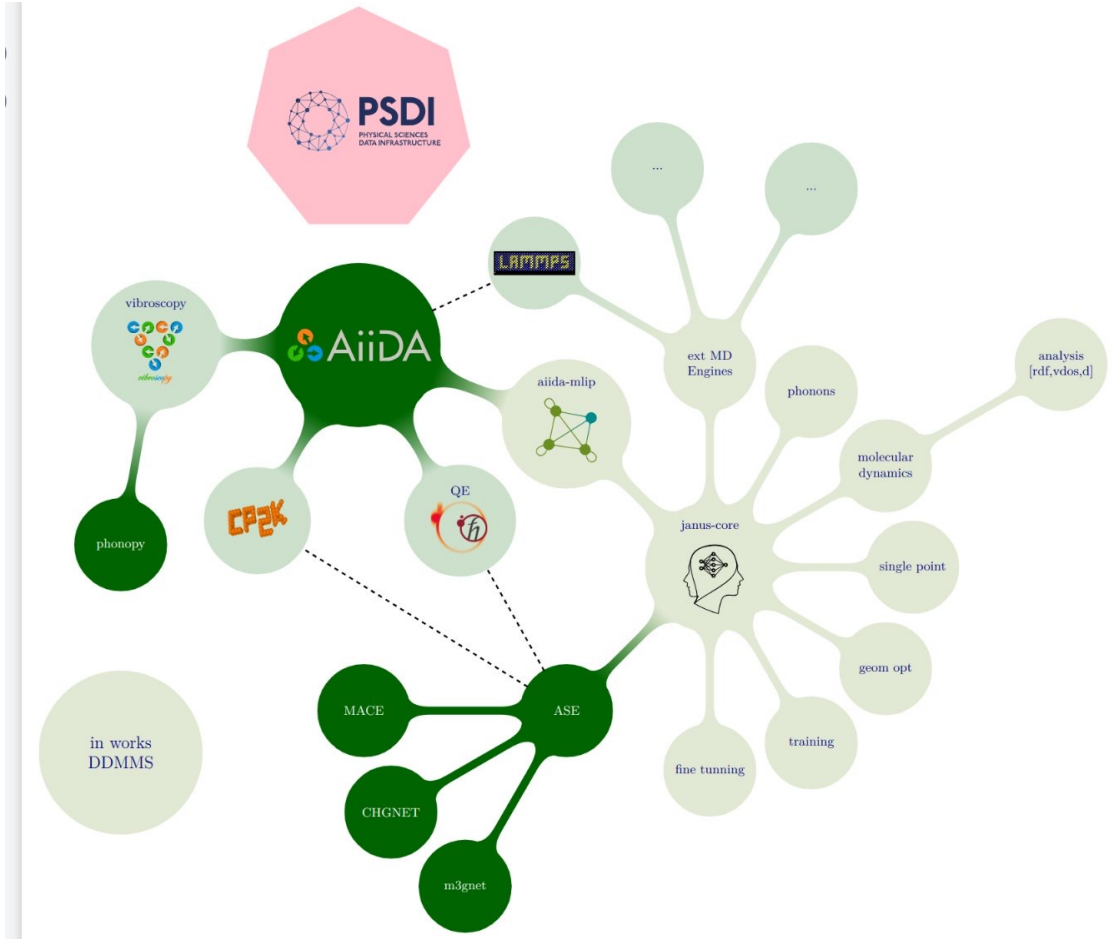
Credit: Alin Elena

4.2-4.5: machine learnt interatomic potentials software infrastructure

- Reproducible and reusable workflows for MLIPs **aiida-mlip**:
  - ▷ Geometry optimization
  - ▷ Single point calculations
  - ▷ Molecular Dynamics (MD)
- Different MLIPs: MACE, M3GNET, CHGNET
- Code based on **janus-core**
  - ▷ Additional features: training functionalities?
  - ▷ Integration with **ABCD** database
    - Unstructured data, stored via JSON/dictionary objects
    - Atomic positions, forces, energies, stresses, charges, dipoles...
    - (PSDI) Metadata?
      - ▷ User-defined properties
      - ▷ Integration with OpenSearch
      - ▷ Highly scalable
      - ▷ Complex and efficient queries



https://github.com/stfc/janus-core

https://github.com/stfc/aiida-mlip

Credit: Alin Elena

Opportunities [1/2]

- Exploit existing tools to support user communities or experiments
- Connect users to experts to help their science
- Tools to simplify the use of simulation for users
  - Workflows and Web UI's
  - Generation of input files and optimization
  - Prepare a model to compare to data
- Support experiment design
- New Theory and Approaches

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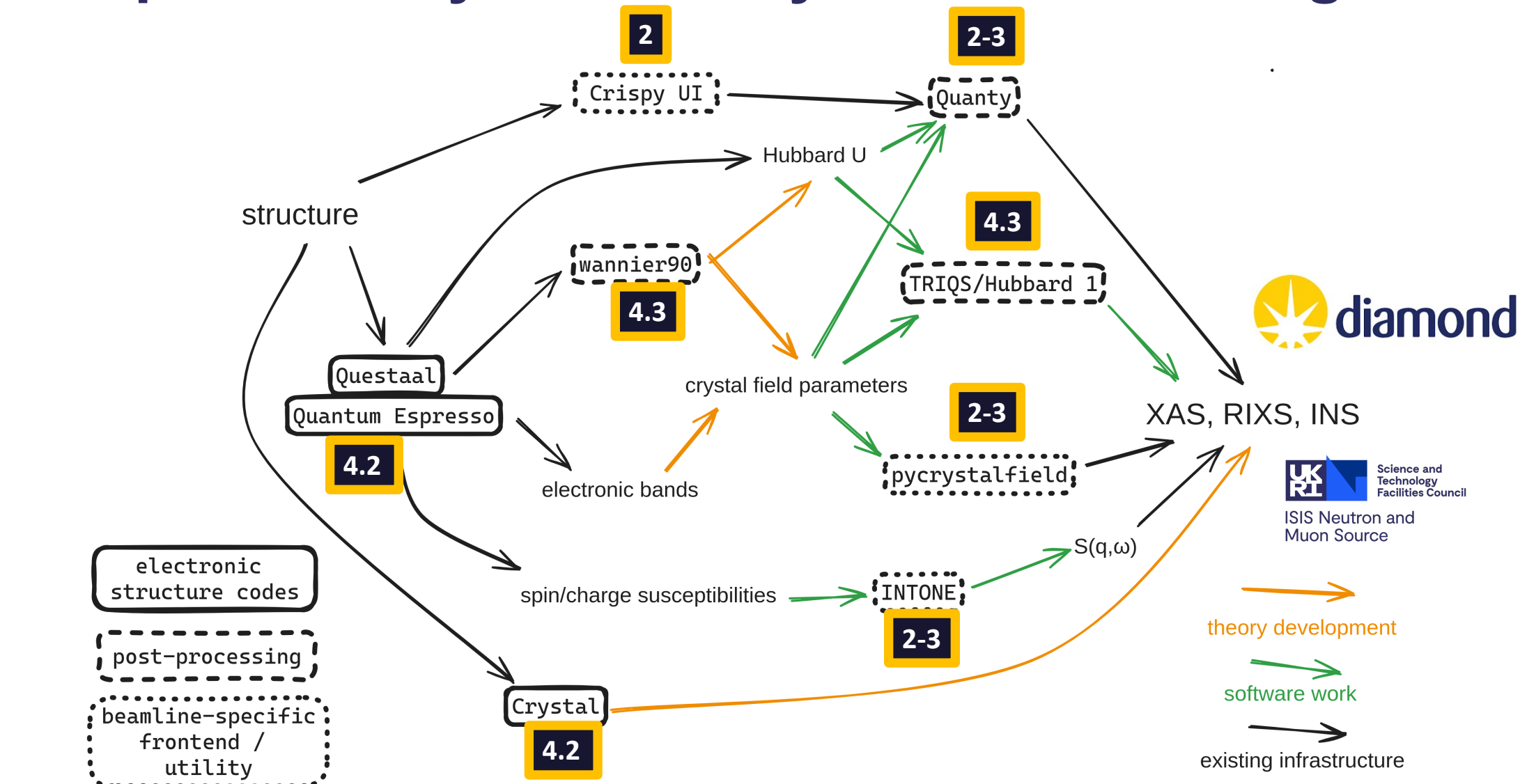
Opportunities [2/2]

- How the atoms move
- How the electrons get redistributed (excitations)
- How the electrons get redistributed while the atoms move
- Is the atom-dynamics classical or quantum?
- Are the electrons “hot or very hot”?

We can develop tools to:

- Bridge length and time scales
- Better treatment of solid materials vs isolated atom approaches
- Expand theory to match experimental observables

Example: DFT/beyond-DFT crystal-field modelling workflow



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Credit: Leon Petit, Jerome Jackson, Barry Searle & Manuel dos Santos Dias

Example: from static/dynamic atomic structures (stoichiometry) to electronic properties and response functions (across facilities)

Chemical composition characterization → Electronic*/atomic structure characterization → Electron*/atom dynamics characterization → Micro/macroscopic property(-ies)

*includes electron-spin degrees of freedom

Materials Chemistry/Physics/Science at STFC Facilities

2-4.6

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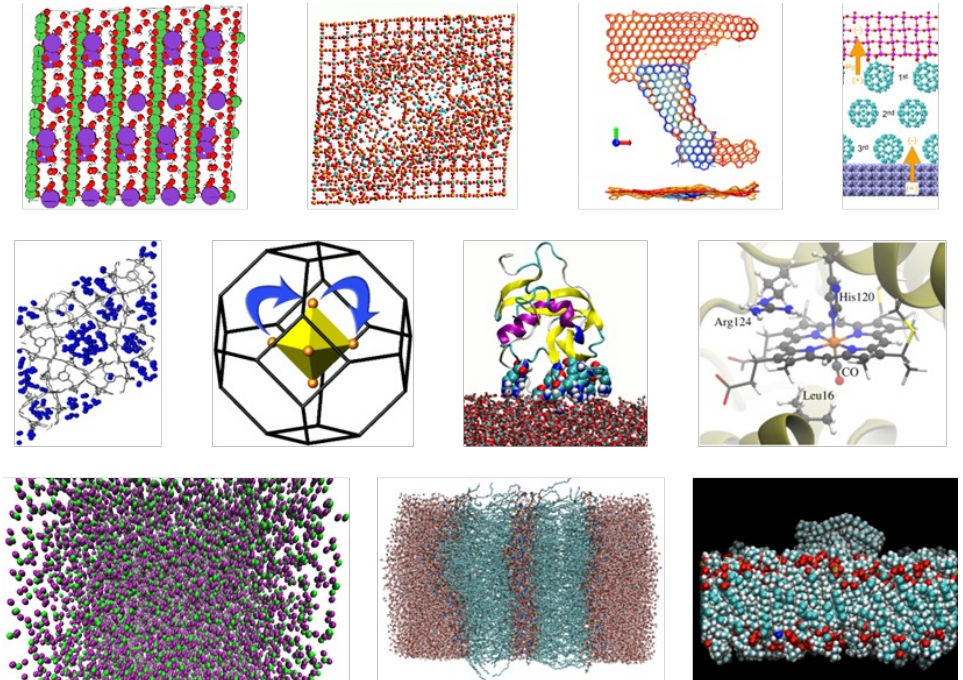


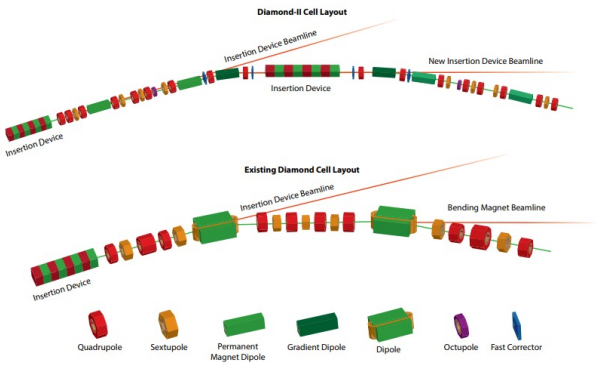
*Disordered materials, "surface science", magnetism and bio-molecules **may** be presenting the "lowest" hanging fruits (neglects human factors)*

Example: matter out of equilibrium and/or at the extreme…

CLF UK XFEL

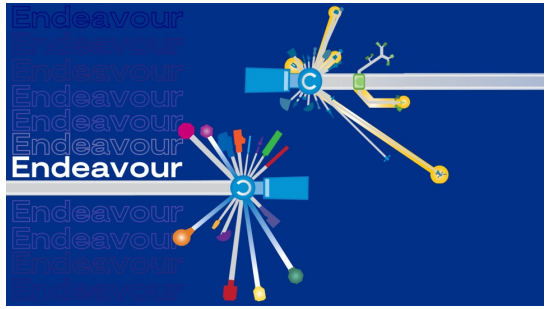
CLF HiLUX upgrades to Ultra and Artemis

CLF-EPAC

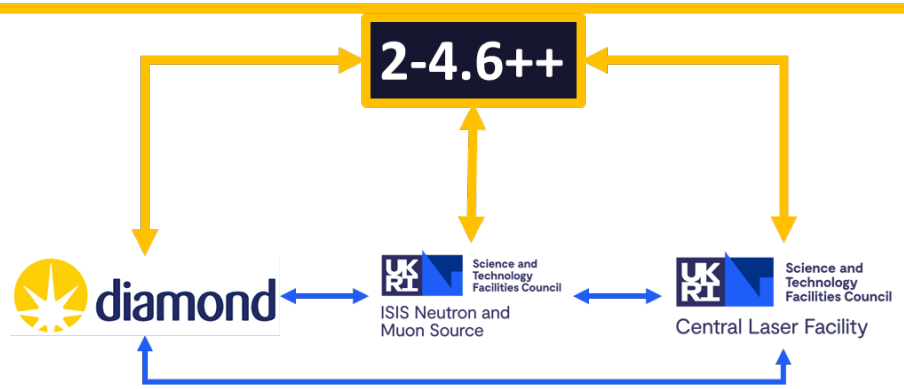


Diamond-II - - Diamond Light Source

- (operando) Electrochemistry and Photo-Electrochemistry
  - Chemistry/Physics in open-boundary conditions
- Role of static/dynamic nano-structuring for
  - Bound and unbound electronic excitations
  - “beyond-linear/perturbative” responses
  - Warm Dense Matter (pre-plasma) regimes
  - Energy dissipation/damage pathways
  - […]



ISIS The Endeavour Programme (stfc.ac.uk)



Ada Lovelace Centre

A scientific computing centre with the primary objective of enhancing the scientific impact of the STFC large-scale National Facilities

From isolated projects to a wider strategy

- Objectives and vision aligned to facilities needs and support science impact
- Long-term and sustained delivery in strategic areas
- Coordinated delivery of cross-cutting activities and complex goals

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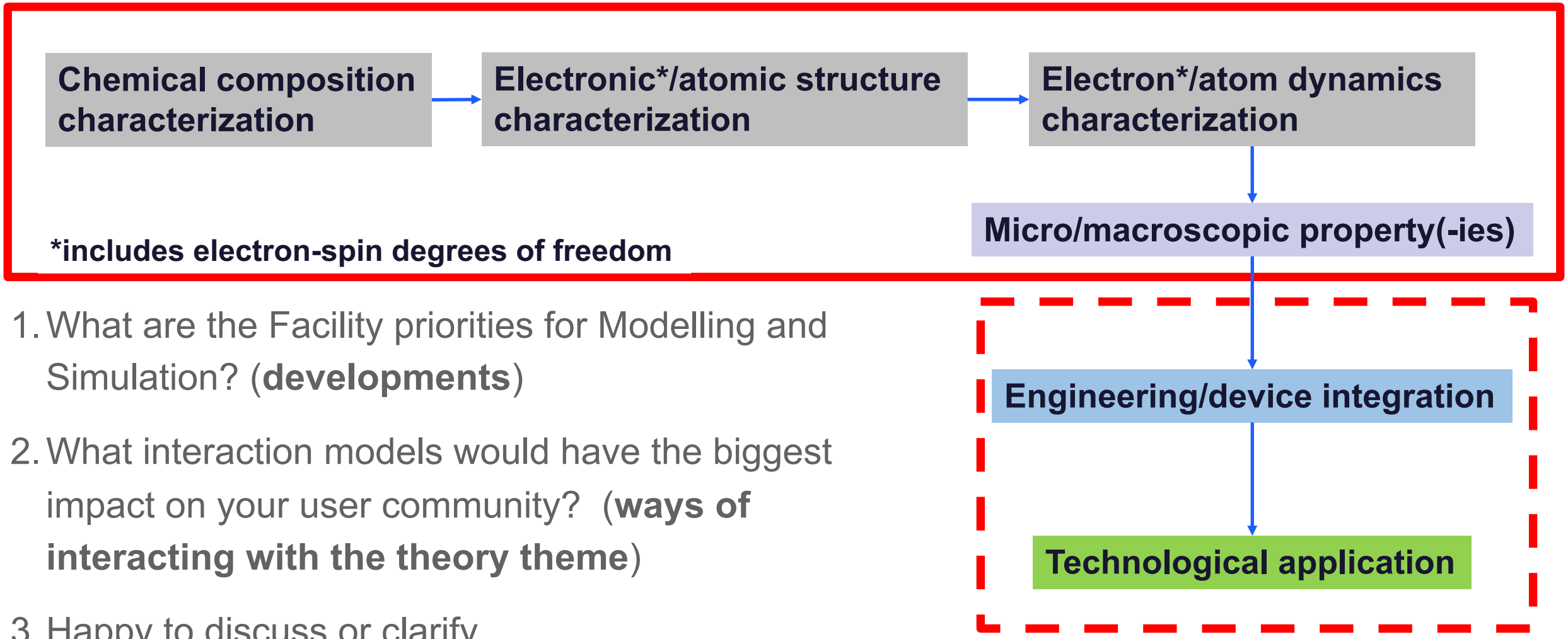
To close it how we started it + questions for you



1. What are the Facility priorities for Modelling and Simulation? (**developments**)
2. What interaction models would have the biggest impact on your user community? (**ways of interacting with the theory theme**)
3. Happy to discuss or clarify

Appendix 1 - Levels of theory and description of materials and molecules

- **First principles - materials and molecules**
  - Density Functional Theory (DFT)
  - Time-dependent DFT (TD-DFT)
  - Variational Multiple Scattering Theory (KKR)
  - Dynamic Mean Field Theory (DMFT)
  - (quasiparticle self-consistent) GW (QSGW)
  - DMFT-QSGW
  - Bethe Salpeter Equation (BSE)

  - Quantum Monte Carlo (QMC)
  - Post Hartree-Fock wavefunction methods

- **First principles - atoms**
  - R-matrix theory
  - Dirac equation

- **Parameterised atomistic Hamiltonians**
  - Density Functional Based Tight Binding (DFTB+)
  - Modified embedded atom method (MEAM)
  - Classical force fields (FFs)
  - Reactive FFs

- **Coarse-grained/mesoscale lattice models**
  - Dissipative Particle Dynamics (DPD)
  - Lattice Boltzmann Equation (LBE)

- **Multi-scale models**
  - QM/MM
  - [QC/QM/MM]

- **Machine Learning**
  - First principles derived interatomic potentials (MTP, MACE, CHGNET, M3GNET, NEQUIP,…)
  - Physics-Informed Neural Networks (PINNs) for Partial Differential Equations and iterative optimization problems

Appendix 2 - algorithmic modelling expertise (a selection of)

- Iterative self-consistent field optimisations
- Geometry optimisation
- Vibrational analysis (analytical, finite-difference, perturbative, real-time)
- Molecular Dynamics
- Monte Carlo
- (adaptive) kinetic Monte Carlo
- Hybrid rare event sampling methods (hMC, metadynamics, temperature-accelerated MD, etc)
- Excited-state real-time and surface hopping dynamics
- [Path integrals molecular dynamics]
- Perturbative/real-time evaluation of response functions
- Integration of hybrid first principles/analytical approaches to approximate complex response functions
- Integration of hybrid first principles/experimental input to approximate complex response functions