UK High-End Computing Consortium for X-ray Spectroscopy

HPC-CONEXS

Tom Penfold, Sofia Diaz-Moreno, Reinhard Maurer,
Rebecca Ingle, Anna Regoutz, Conor Rankine, Josh Elliott
tom.penfold@newcastle.ac.uk

Newcastle University

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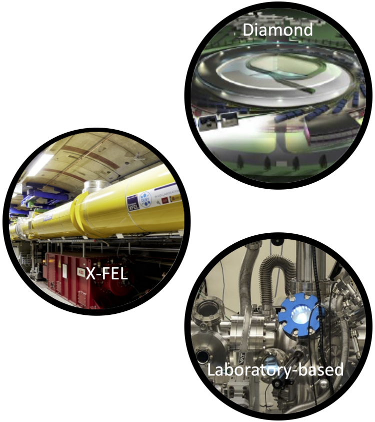
X-ray Spectroscopy



- X-ray spectroscopy probes the nuclear, electronic and spin structure of matters in a full range of sample environments.
- Proliferation of 3^{rd}-generation light sources as well as laboratory-based approaches and X-ray Free Electron Lasers (X-FELs) the capability and accessibility has increased dramatically during the last decade.
- Brings into focus the key challenge: *How can we accurately and efficiently interpret the detailed information contained within each spectra?*
- How can we lead the community ensuring the best practice for the interpretation and analysis of experimental observable?

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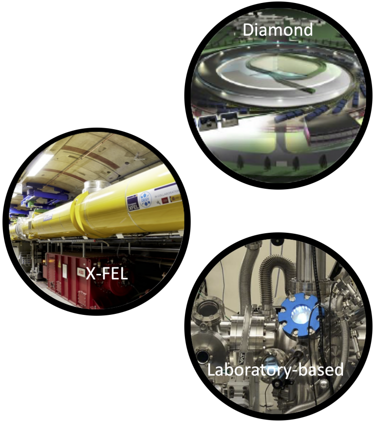
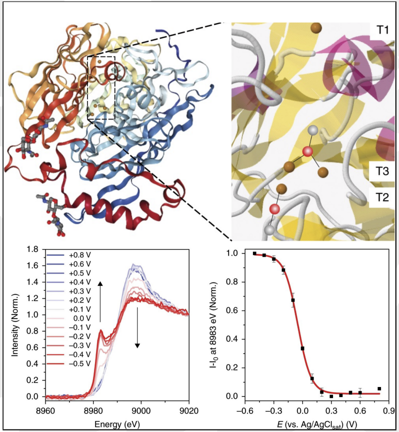
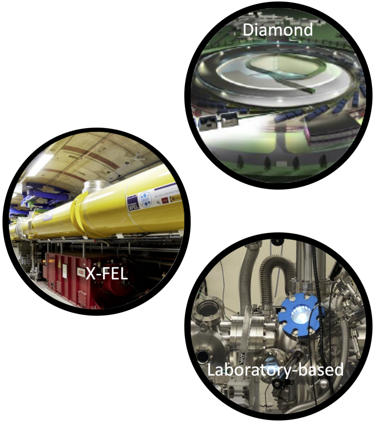
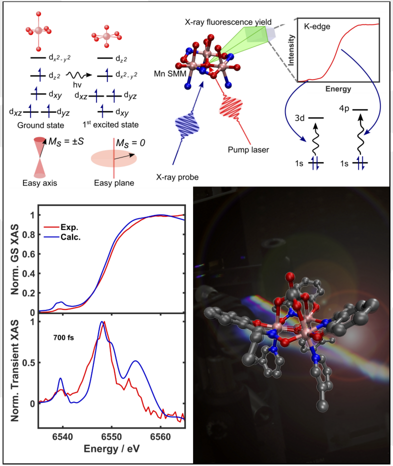




F. Cresphilho and co-workers Nat. Comm. **11**:316 (2020)

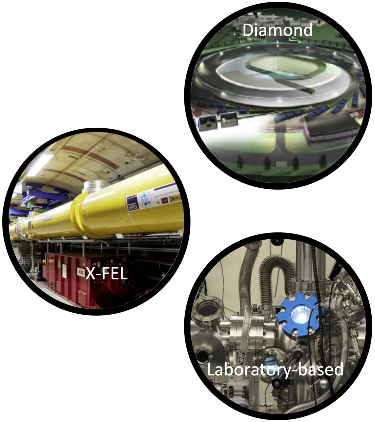
Modern X-ray Spectroscopy

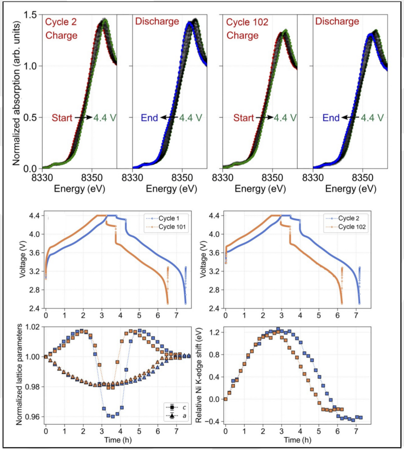




Liedy *et al.* Nature Chem. **12** (2020) 452.

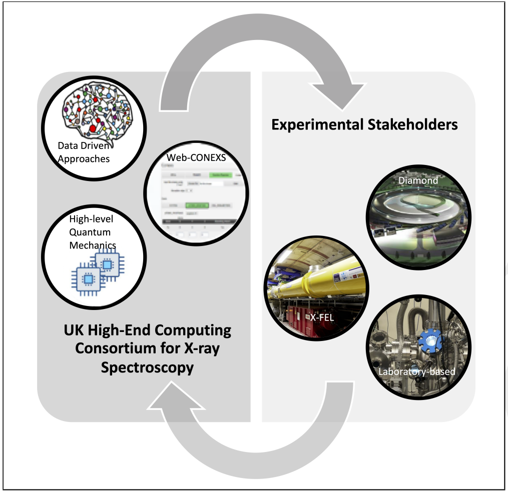
Modern X-ray Spectroscopy





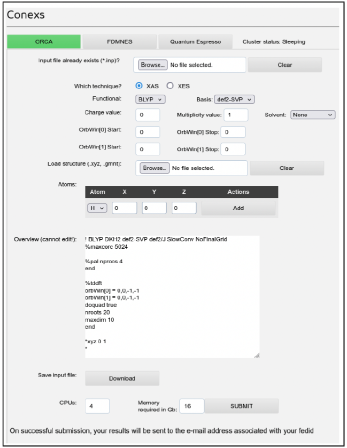
L. Piper and co-workers PRX Energy **3**:013004 (2024)

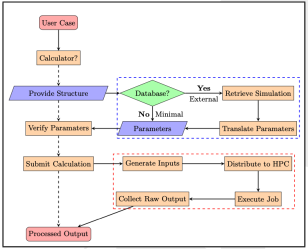
HPC-CONEXS Objectives



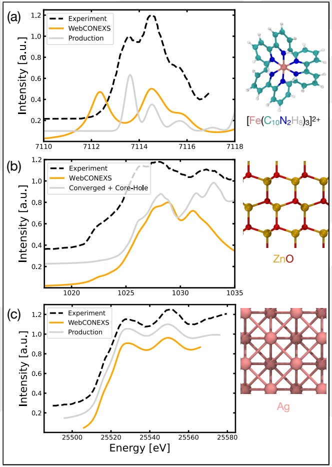
- **Development of First Principles Methods**
  - Development of new scalable first-principles quantum-mechanical methodologies.
  - Adaptation of existing codes for modern computer architectures to enhance performance
  - Implementation of automation and analysis techniques to streamline workflows
- **Development of Machine Learning Models**
  - Development of new machine learning approaches to enhance data interpretation.
  - Analysis of machine learning methodologies to enhance interpretability and to quantify uncertainties.
  - Development and analysis of advanced training sets for machine learning techniques.
- **Experiment-Theory Collaboration**
  - Application of state-of-the-art techniques to collaborative theory/experiment investigations..
  - Application of machine learning models to interpret complex data.

Beginning the journey to Archer2: web-CONEXS



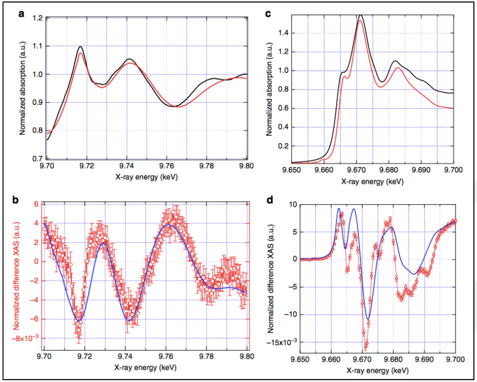


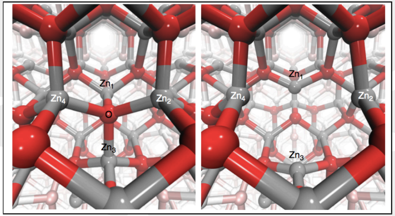
Establish easy to use tool, which can be begin analysis and training in computational spectroscopy.

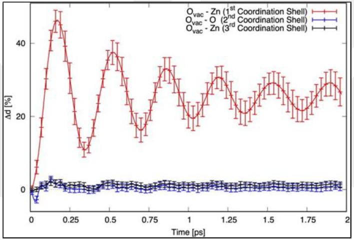


Probing hole trapping in ZnO nanoparticles

Time-resolved XANES probed structural dynamics after above band-gap excitation.

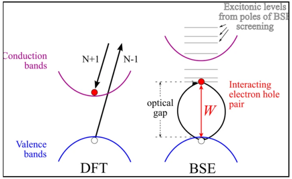




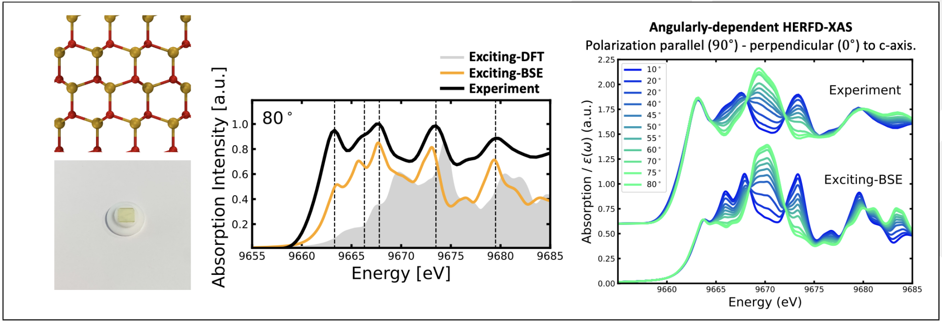


Penfold *et al.* Nat. Comm. **9**:478 (2018).
Milne *et al.* Struc. Dyn. **10**:064501 (2023)

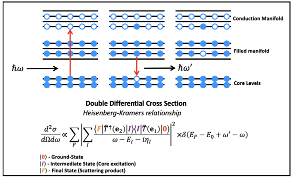
How can we improve simulations? (Joshua Elliott; Diamond Light Source)



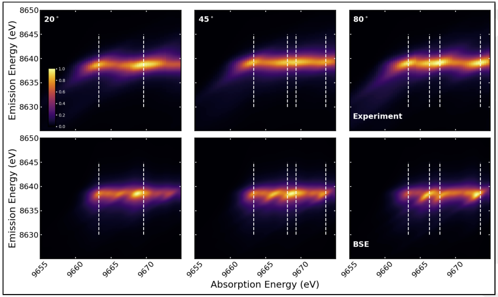
- Simulations Carried out using Exciting Code.
- Bethe-Salpeter Equation formalism provide significantly better agreement with the experiment.
- Fully account for changes associated with polarization of incident beam

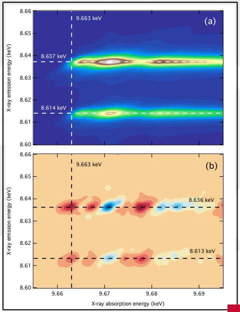


Beyond First-order Spectroscopy (Joshua Elliott; Diamond Light Source)

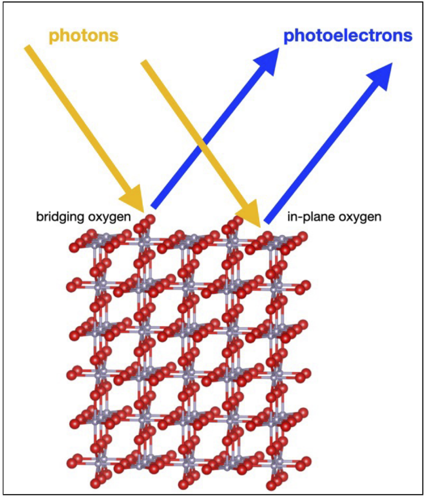


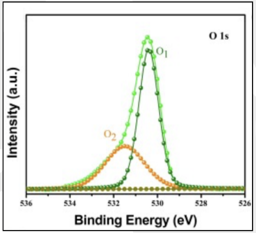
- Simulated RIXS maps correlate DFT ground state, BSE K-edge excited states and BSE L-edge excited states.
- Fully converged simulations require 58k BSE transitions at the K-edge and 121k BSE transitions at the L-edge.





Accurate Calculation of Core Binding Energies (Johannes Lischner; Imperial)





- SnO_2 is an important material for gas sensor applications. However, the microscopic mechanism by which adsorbed gas molecules modify the conductivity remains ill-understood.
- *Ab initio* calculations of core hole binding energies show that different oxygen species, such as bridging oxygen atoms or surface in-plane oxygen atoms, exhibit very different binding energies.

Characterizing MoleculeMetal Surface Chemistry (Reinhard Maurer; Warwick)

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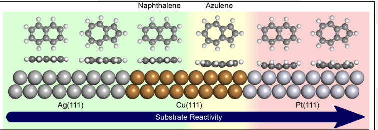
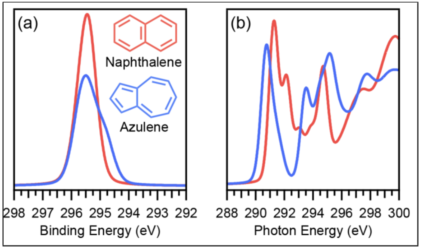
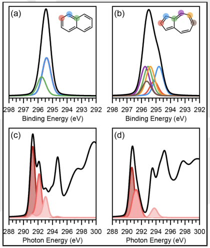


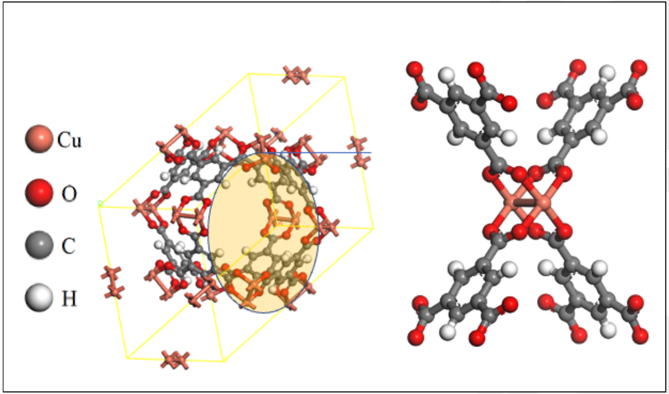
Adsorption Energy (kJ/mol)	118	128	135	173	298	335
Adsorption Height (Å)	3.10	2.94	2.96	2.33	2.09	2.10
Net Charge Transfer (e)	0.06	0.01	0.06	-0.49	0.03	0.02
Structural Deformation	None	None	None	Weak	Strong	Strong
Molecule-Metal Hybridisation	Weak	Weak	Weak	Intermediate	Strong	Strong

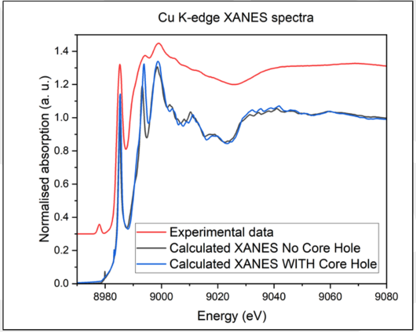




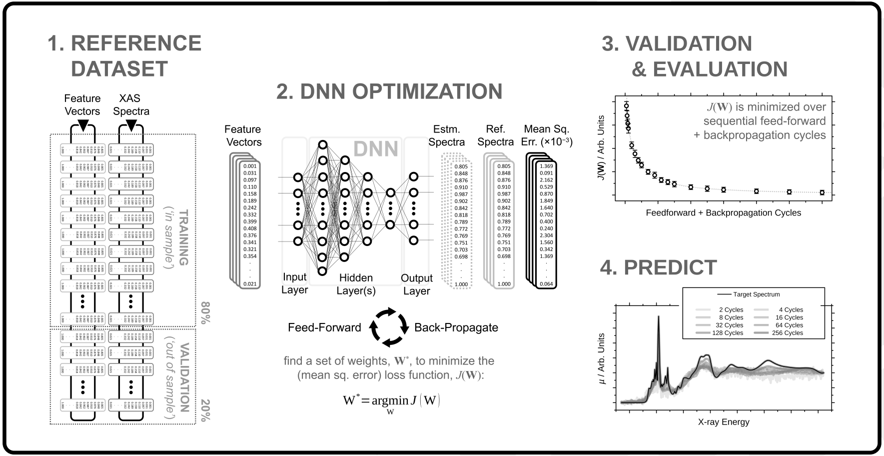
Hall *et al.* J Phys. Chem. C **127**:1870-1880 (2023)

Understanding XANES of Cu-BTC (Iuliia Mikulska; Diamond)



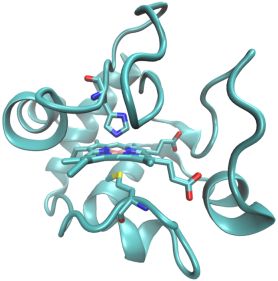


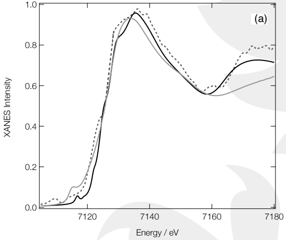
XANESNET DNN

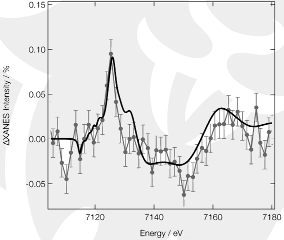


C Rankine *et al.* J. Phys. Chem. A, **124** (2020) 4263

Augmenting Interpretation using ML Models



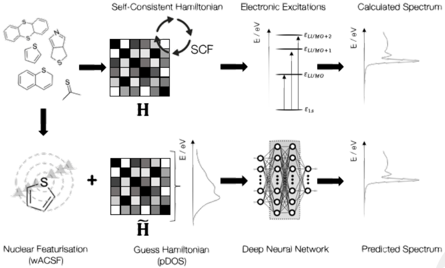


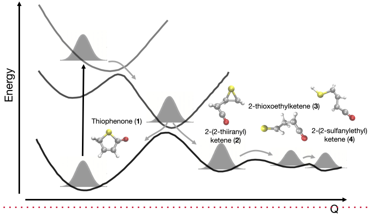


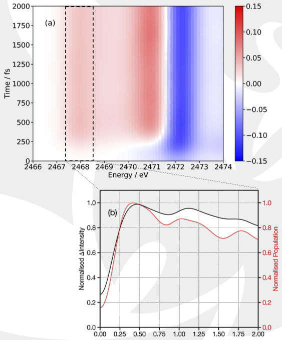
- Ground state, with the exception agrees well with experiment and DNN predictions.
- Small elongation of Fe-N_P (0.05 Å) , larger expansion (0.1 Å) of Fe-N_{Im} and Fe-S shows little or no change , but sensitivity is weak.
- Refinement using DNN agrees with expected structure.

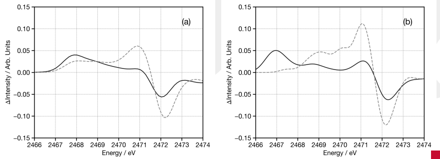
C. Bacellar *et al.* PNAS **117** (2020): 21914.

p-DOS descriptor









Summary

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Development and Application of First Principles Method

- MD and spectral simulations describes hole dynamics within ZnO nanoparticles.
- Improvements achieved using BSE, well suited for Archer2.
- Accurate computation of core-binding energies for surfaces.

Development and Application of Machine Learning Models

- New p-DOS descriptor provides new perspective of ML models in XS.
- Archer2 infrastructure facilitates generation of large training sets.

Training and Dissemination

- Web-CONEXS provides a user friendly framework for computing and analysing spectra.
- Developed for new users seeking to gain experience with HPC and simulation.

Acknowledgements

Management Committee:
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Web-CONEXS:
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Christopher Reynolds, Jacob Filik

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Core Binding Energies:
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Molecule-Metal Surfaces:
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Cu-BTC MOF:
Iuliia Mikulska

Machine Learning XAS:
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https://xfel.ac.uk