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Modelling of Muon Experiments in Battery Materials with Muon Galaxy

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Muons and the Muon Spectroscopy Computational Project

Muons are sub-atomic particles that are generated at target station 1 in ISIS, at STFC. Muons are 100% spin polarised particles with spin ½, which can be thought of as either light protons or very heavy electrons.

Contrary to what happens in a neutron or x-ray experiment, muons are not diffracted by the sample: they are implanted into the sample, and knowing their implantation site is crucial for the interpretation of muon experiments.

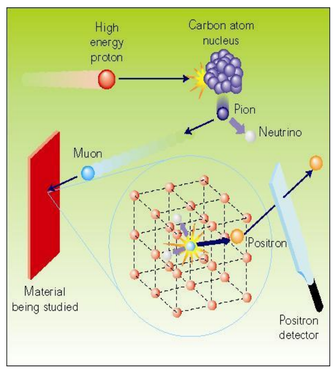


Figure 1: A schematic description of Muon experiment.

The MSCP[2] develops software tools for tackling computational challenges in muon spectroscopy. These tools can be used for:

- Identify the **muon stopping site(s)** in a crystalline system
- Simulate the **spin dynamics** of a system containing a muon, electrons, and atomic nuclei, with various experimental setups and couplings
- Fit a spin dynamics simulation to experimental data

These tools can be chained together to form an analysis pipelines known as workflows. We release the tools as Python packages: **pymuon_suite** and **muspinsim,** which can be installed using pip or conda, or run online using **Muon Galaxy**.

Muon Galaxy

Galaxy is an open-source web platform for data intensive research. It allows users to **run complex workflows** and visualise results without any programming experience.

Analysis in Galaxy is **easily reproducible**, as a consistent computational environment is used for every job, and all data files are stored in a 'history'. Data and workflows can be **easily shared** with other researchers or made public; publishing a workflow alongside a paper is a great way to ensure reproducibility.

Galaxy is well established in the biology community, but we have launched an instance specifically for muon science – known as **Muon Galaxy**[3]. We are making all our tools available in Muon Galaxy, along with tutorials, example workflows, and visualisations.

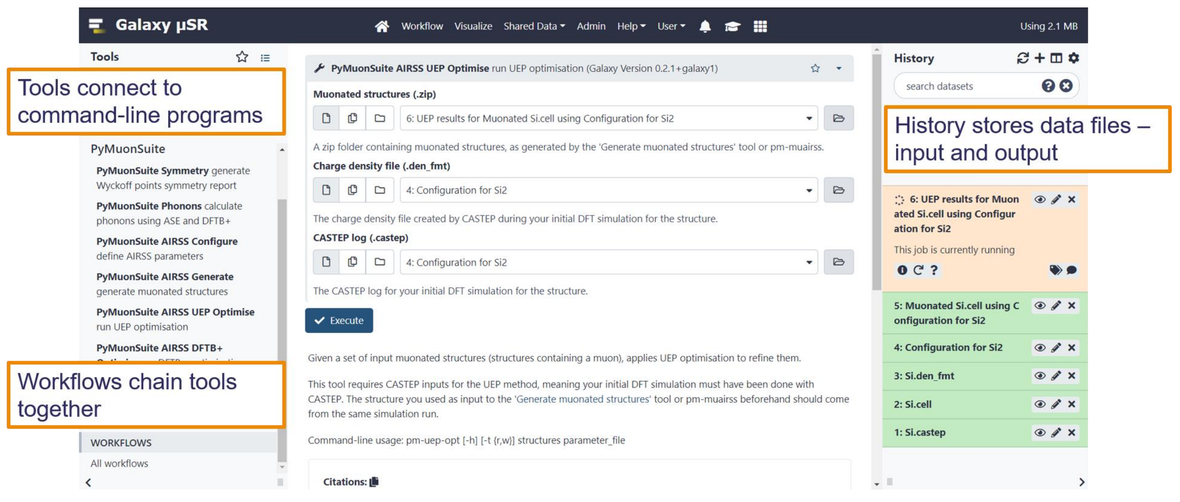


Figure 2: The muon galaxy interface with a description of the features provided.

Lithium-ion Battery Materials

Lithium-ion batteries consist of negative and positive electrodes composed by two lithium insertion materials.

During the charging process, lithium ions are inserted into the solid matrix that forms the positive and negative electrodes without destructing their core structures.

During the discharge process, lithium ions are extracted from the electrodes. As this happens, electrons are simultaneously extracted from one electrode and injected into another electrode, storing and delivering electrical energy[1].

Figure 3 exemplifies how a Li-battery operates. In spite of a long research history on Li-ion batteries, the value of the Li diffusion coefficient, D_{Li}, is still difficult to determine with any reliability.

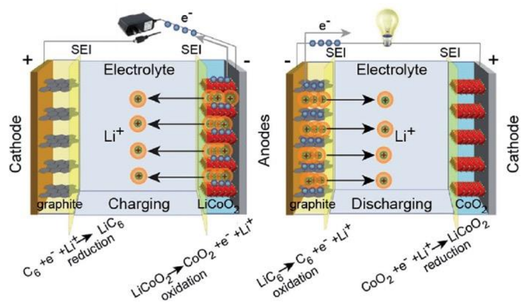


Figure 3: A schematic illustration on a lithium-ion battery consisting of two lithium insertion electrodes.

Why muons?

Knowing the muon stopping site helps to determine whether the muon is either likely to diffuse with Li when T is increased or to remain at its stopping site. If the muon site is stable with respect to Li diffusion, the Li diffusion coefficient, D_{Li}, is given by:

$$D_{Li} = \sum_{i=1}^{n} \frac{1}{N_i} Z_{\nu i} \, s_i^2 \, \boldsymbol{\nu}$$

N_i is the number of Li sites in a given diffusion path; $Z_{\nu i}$ is the Li-vacancy fraction; s_i the jump distance; and $\boldsymbol{\nu}$ is the magnetic field fluctuation rate at the muon site, which is measured in a muon experiment.

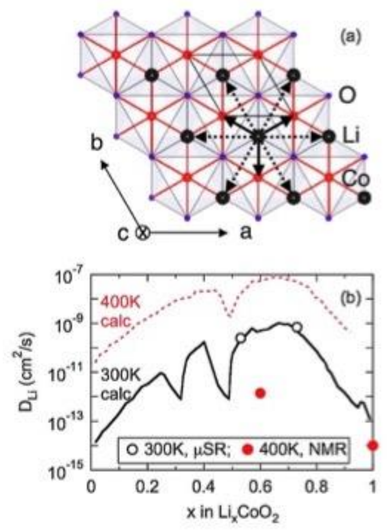
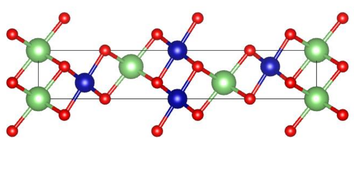


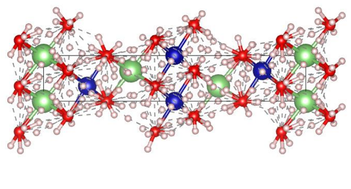
Figure 4: (a) Li diffusion paths. (b) Comparison of D_{Li} values measured with NMR and muons experiments and estimated with computer simulations[4].

The Workflow to obtain Muon stopping sites

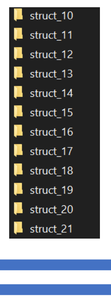
DFT pure host



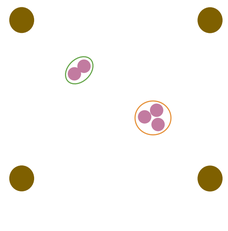
Create randomly populated muonated structures



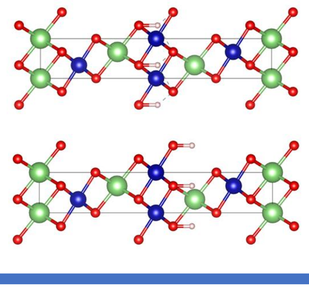
Relax electrostatic forces on muons



Cluster relaxed structures

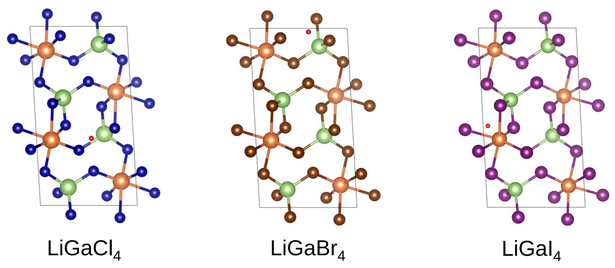


μ^+ stopping sites



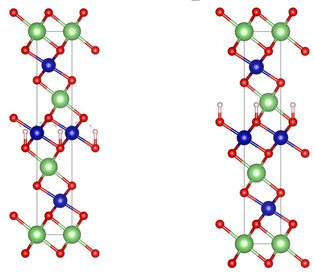
Using the UEP method[5] with the above workflow in Muon Galaxy, the most stable stopping sites for $LiGaX_4$ and $LiCoO_2$ are shown in the figures below:

$LiGaX_4$ (X=Br, Cl, I)



$LiGaCl_4$ $LiGaBr_4$ $LiGaI_4$

$LiCoO_2$



The sites obtained correlate to sites that remain unchanged when the Lithium ions diffuse through the material during the battery's charge and discharge process, allowing for accurate determination of the diffusion coefficient value.

References

[1] Journal of Power Sources 174 (2007) 449–456

[2] https://muon-spectroscopy-computational-project.github.io/

[3] https://muongalaxy.stfc.ac.uk/

[4] PRL 103, 147601 (2009)

[5] J. Chem. Phys. 28 July 2020; 153 (4): 044111

MSCP

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ISIS Neutron and Muon Source