Creation of an XAS and EELS Spectroscopy Resource within the Materials Project using FEFF9

Alan Dozier[1], Kristin Persson[2], Shyue Ping Ong[3], Kiran Mathew[2], Chen Zheng[3], Chi Chen[3], Joshua Kas[4], Fernando Vila[4], and John Rehr[4]

[1.] NIOSH/CDC, Cincinnati, OH, USA
[2.] Material Science and Engineering/UC, Berkeley, CA, USA
[3.] NanoEngineering/UC, San Diego, CA, USA
[4.] Physics/University of Washington, Seattle, WA, USA

We describe the development of an X-ray Absorption Spectroscopy (XAS) and Electron Energy Loss Spectroscopy (EELS) resource in the Materials Project (MP) [1] database as part of the Local Spectroscopy Data Infrastructure Project [2]. This integration within the MP allows material properties and the associated spectroscopies to be calculated using NERSC (National Energy Research Scientific Computing Center) computing facilities and retrieved through either the user portal or the Representational State Transfer (REST) interface [1]. This ability to search the MP database through its REST interface and carry out a comparative analysis with experimental spectra introduces an efficiency in characterization that will aid in understanding material functionality in various technologically important areas such as semi-conductors, batteries, reactions, etc.

The Materials Project uses high-throughput computing to calculate selected properties of all materials in the Inorganic Crystal Structure Database (ICSD). These properties are all amenable to high-throughput computing, including phase diagrams, density of states (DOS), band structure, elasticity, dielectric functions, XRD spectra, etc. Aperiodic structures are generally not amenable to high-throughput computing using conventional Density Functional Theory (DFT) codes, as the computation time becomes excessive. The FEFF9 [3] code gets around this limitation by using an efficient quantum mechanical approach based on real-space Green's function multiple scattering theory. This greatly increases its computational efficiency and makes it an excellent choice for high-throughput computing applications. Quality control is provided by comparison with available experimental spectra, and in the near future, users will be able to upload their own spectra, which can become part of the database. Through this feedback, more refined sets of FEFF9 "intelligent default" input parameters for differing atomic environments can be derived and the quality of results improved. In the first few months of initial runs, K-shell XANES for about 8,000 materials have been computed and we expect over 30,000 materials by the end of the first year. However, this is just the starting point, and these spectra will be evaluated and refined. Once we are confident of these results, we will make them available on the MP web portal and expand the computations to include additional edges, more materials, and also EELS. When the spectra are published on the MP web portal, the FEFF9 input file will be provided so that users can refine their own calculations.

The open-source software that enables the MP was primarily developed by the MP team, but many other collaborators worldwide have made significant contributions. The middleware, Pymatgen [4], Fireworks [7] and Atomate [8], handle high throughput workflow generation as well as processing of the generated data. Pymatgen [4] handles the creation of inputs and outputs for quantum mechanical codes and contains applications for finding materials in the database based on user defined properties, modifying structures, several battery material search functions, the REST interface, and other functionality. Users can download this software, use the built in REST interface to the database, and write their own applications to retrieve

data to characterize structures and processes. Some examples are *in situ* reactions [5], spectroscopic mapping [6], and aperiodic structures. Fireworks [7] provides the infrastructure to manage the execution of massive amounts of workflows in a consistent and fool-proof manner. The Atomate [8] package aids in the composition of the workflows used to compute various types of spectra as well as much more complicated workflows such as fitting of the experimental EXFAS data to the ones computed for the selected list of scattering paths.

The big news for the electron microscopy community is that in the future, atomic resolution experimental EELS spectra from aperiodic structures such as grain boundaries, defects, interfaces, etc. can be compared with spectra computed from hypothesized structures. This will be made possible by the future ability for users to upload their structures and have the MP compute the spectra from a particular location in the structure. Users will also be able to use the software infrastructure developed as part of this initiative to write their own applications to aid in this analysis.

References:
[1] Shyue Ping Ong, Shreyas, Anubhav Jain, *et al*, Computational Materials Science, **97** (2015), p. 209., www.materialsproject.org
[2] NSF Award #1640899.
[3] J.J. Rehr, J.J. Kas, F.D. Vila, *et al,* Phys. Chem. Phys. Chem., **12** (2010), p. 5503.
[4] S.P. Ong, W.D. Richards, A. Jain, *et al*, Computational Materials Science, **68** (2012), p. 314.
[5] Gary Jacobs, Yaying Ji, Burtron H. Davis, *et al*, Applied Catalysis A, **333** (2007), p. 177.
[6] P. Rulis, A.R. Lupini, S.J. Pennycook, *et al*, Ultramicroscopy, **12** (2009), p. 1472.
[7] Jain, A., Ong, S. ets al, FireWorks: a dynamic workflow system designed for high-throughput applications. Concurrency Computat.: Pract. Exper., 27: 5037–5059.
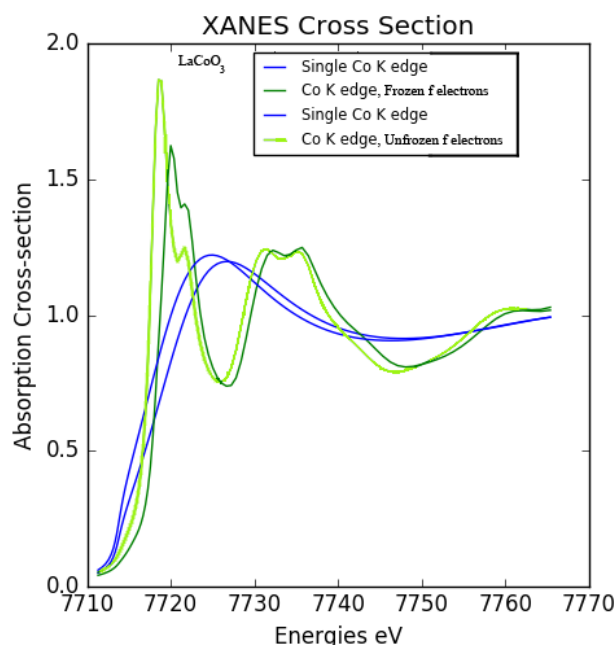[8] https://github.com/hackingmaterials/atomate

Figure 1. Example result of user created software using Pymatgen to plot two different FEFF9 calculations of the Co K edge in LaCoO$_3$, one with frozen *f*-electrons and the other without.
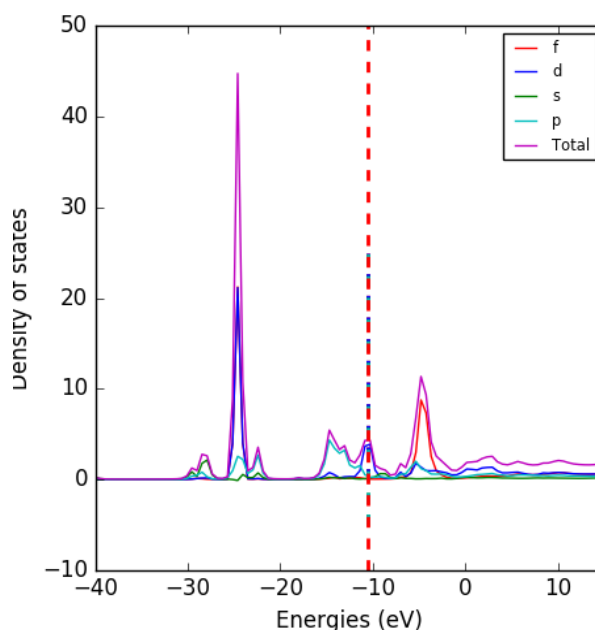

Figure 2. Example of user created software using Pymatgen to plot the DOS for LaCoO$_3$ computed using FEFF9.