Computational Modeling to Aid in Analysis and Interpretation of Multi-Modal Neutron Experiments

Research Objectives

Significance of

The over-arching goal of our project is create a streamlined workflow for experimental data analysis and interpretation needs of the neutron scattering community using atomistic modeling and simulation. Neutron scattering experiments require users to model and interpret data at the atomic/molecular level. With numerous software applications and a large array of different file formats with each, scientists tend to use a limited (and sometimes dated) subset of software tools to tackle data analysis from neutron experiments. This creates a barrier to use other methods or cutting-edge atomistic modeling softwares in their research that could help in bridging the gap between experiment and theory.

We are currently developing a modeling and analysis workbench called Integrated Computational Environment-Modeling & Analysis for Neutrons, or ICE-MAN. We hope to create a seamless transition from both different types of neutron scattering experimental data and computer modeling and simulation techniques to tackle multi-modal data analysis. An example of a workflow would be the study of a disordered material. First, neutron scattering experiments could yeild the average structure of the material, providing Bragg diffraction data, and the local structure via total scattering, providing the pair distribution function. Molecular dynamics (MD) simulations can provide a trajectory for an atomistic model of the material in time giving an ensemble of possible atomic configurations to compare to experiment. These sampled configurations from the trajectory could be converted and refined with reverse Monte Carlo modeling (RMC) to determine which atomic configurations produce the best fit to both the Bragg diffraction pattern and pair distribution functions produced from the neutron scattering experiments. The MD trajectories could also then feed into software to calculate the inelastic neutron scattering spectrum of the system. This data could feed back into the RMC modeling as a constraint. At present this process would be exceedingly time consuming, involve expertise in at multiple techniques, and most likely present a barrier that would seldom be overcome by general users.

We are requesting XSEDE's HPC resources to carry out this workflow on projects that are already ready to be fed into the pipeline and also for potential projects for the general neutron scattering community, specifically Users of the Spallation Neutron Source instruments.

Proposed Research

Project 1: Investigate structural modifications of irradiated silica for nuclear materials

The first project would be the study of the local structure changes in amorphous silica (SiO2) before and after irradiation of high energy (tens of MeV or more) heavy ion bombardment, or swift heavy ions (SHIs). At these high energies, electronic stopping dominates over nuclear stopping, implying the SHI interacts with the electronic structure of the irradiated target material. The SHI path deposits energy to the electronic structure which then dissipates the energy to the nuclear motion of the material, causing a local heat spike in the material radially perpendicular to the SHI pathway.

Previous studies have looked at the fine structure of SHI tracks (the narrow trails of permanent atomic distortion along penetrating SHI pathways) in thin film amorphous silica using small angle x-ray scattering (SAXS) measurements combine with non-equilibrium MD modeling and simulation techniques. These studies have successfully revealed that SHI tracks form a low density shell around the SHI path of penetration surrounded by an outer shell higher in density. The non-equlibrium MD calculations were carried out using an 20k atom system where the track was produced by using the simple approach of instantaneous heating of the atoms in the simulation cell along the SHI track path. This method has been motivated by the fact that most of the energy transfer from the electronic system to the atomic degrees of freedom occurs on the femtoseconds timescale. The timescale of ionic displacements is much greater, implying the transfer is immediate.

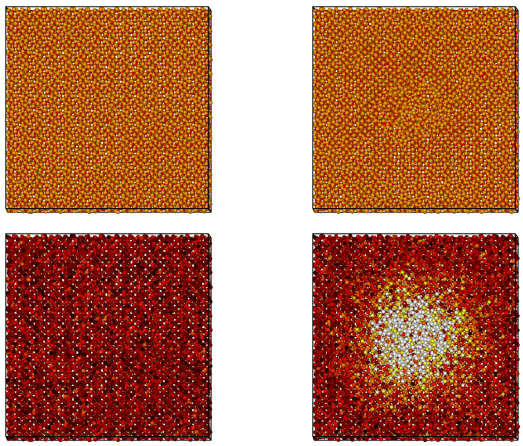

Figure 1: Thermal Spike in SiO2

We intend to carry out similar simulations of comparable size that can be used to elucidate the structural changes observed in neutron scattering experiments of the average and local structure of these polymorphs of silica. Atomistic configurations from the end of this trajectory can then be fed into the RMC modeling to optimize the structure against the experimental data. The results of this study can help understand the fundamental degradation of silica materials exposed radiation damage and also to future work to manipulate nanoclusters within solid silica materials.

An extension of this project would be to incorporate recently models that have been developed to address more realistic modeling of the electronic heat conduction to the atomic degrees of freedom. In these models, the solution of the electronic heat conduction is embedded into the MD simulations via including an electronic continuum used to solve a heat equation for modeling the energy transport between the atomic and electronic subsystems. The two-temperature model formalism of including the electron-phonon cou-

pling has been previously used in modeling and simulating SHI bombardment of alpha-quartz, laser ablation, and shock simulations. Interestingly to our ICE-MAN project, this extension would allow us to bridge a quantum-to-molecular multiscale-modeling barrier. An important input parameter into the two-temperature model is the electronic specific heat. Recently, it has been shown that the electronic temperature depedence of the electronic specific heat can have an effect on the morphology of the SHI track that is formed. To produce this relation, one calculates the difference in internal energy with change in the electronic temperature using finite-temperature density functional theory. Previous work of others have used the popular opEn Source Package for Research in Electronic Structure, Simulation, and Optimization (Quantum ESPRESSO) and the Vienna ab initio simulation package (VASP) softwares with proven scalability. Thus, we hope to use this project as a proof-of-concept workflow for ICE-MAN to: begin with quantum calculations, use outputs of these calculations as inputs into non-equilibrium MD simulations, and then use the atomic configurations of outputs from these simulations as inputs into RMC modeling to optimize the atomic structures against neutron scattering data.

Project 2: Modeling of amorphous germanium and silicon to determine high-pressure phase transformations

The second project we would like to undertake is trying to determine different structural changes of high-pressure amorphous germanium (a-Ge) and silicon (a-Si) from different forms of compression or indentation loads that result in new materials for industry applications. Similar to our first project, we will be modeling systems based on silicon that have been probed via neutron scattering studies to determine structure changes while under high-pressure, being compressed in diamond anvil cells or being indented using diamond tips. Under compression, these samples become metallic and do not return to their initial state upon releasing the pressure. Based on either indentation or compression, different structures are observed with improved electronic and photovoltaic properties with cheaper manufacturing methods.

Our modeling and simulation effort will attempt to find atomistic models that best fit neutron scattering experiments probing the average and local atomic structure via diffraction and pair distribution functions along with vibrational density of states from vibrational spectroscopy. To create models of a-Ge and a-Si in agreement with all experimental data is still a current research challenge, and to date no such model exists. We have developed a structural relaxation program based on the Wooten-Winer-Weaire method to produce realistic random-network models of a-Ge and a-Si via the Atomic Simulation Environment (ASE). From this program, we have initial amorphous structures that retain their four-fold coordination and bond-angle deviations in qualitative agreement with experiment. We use these structures to then perform a range of equilibrium or non-equilibrium MD simulations to best replicate different pressure loading and thermal annealing cycles. We feed atomic configurations from the MD trajectories into reverse RMC modeling to optimize the structure against the experimental data taken throughout thermal and pressure load cycles and the send this back to the MD simulations for further refinement and progess in the cycle. Experimentally constrained methods have been pursued in recent studies with success using very similar Monte Carlo relaxation methods. These include

pure Monte Carlo using just the experimental data and hybrid approaches that add an emperical potential as an additional constraint. Our hope that using ICE-MAN's unique capability of lowering the barrier for data cross-over from the MD simulations to the RMC optimization to fit to experimental data will allow a larger search in the phase space and accelerate the convergence of a best fit to a variety of experimental data with strong emphasis on neutron data for these materials.

Project 3: Modeling oxygen insertion in one-dimensional channels of shafarzikite-like structures

Recent study of schafarzikite-like (FeSb2O4) materials have been shown to store relatively large amounts of oxygen (3.5% mass) within the structure at low temperatures (350 C) without decomposition, leading to new possibilities as potential oxygen storage materials.

Specifically, oxygen insertion into cobalt and lead doped derivatives of shafarzikite show promise in applications of electrocatalysis due to their unique 1-D cation channels with high peroxide anion mobility and potential for high, directed electronic conductivity. We plan to analysis the neutron scattering data with RMC modeling to clarify the structures that occur before and after oxidization and also variants of the material with different doping agents. Structures generated from the RMC modeling can then be used to construct nudge elastic band calculations using density functional theory to determine transition state energy barriers during the oxidation process. Results from this study would help in clarifying the proposed defect cluster produced by the low temperature oxidation reaction.

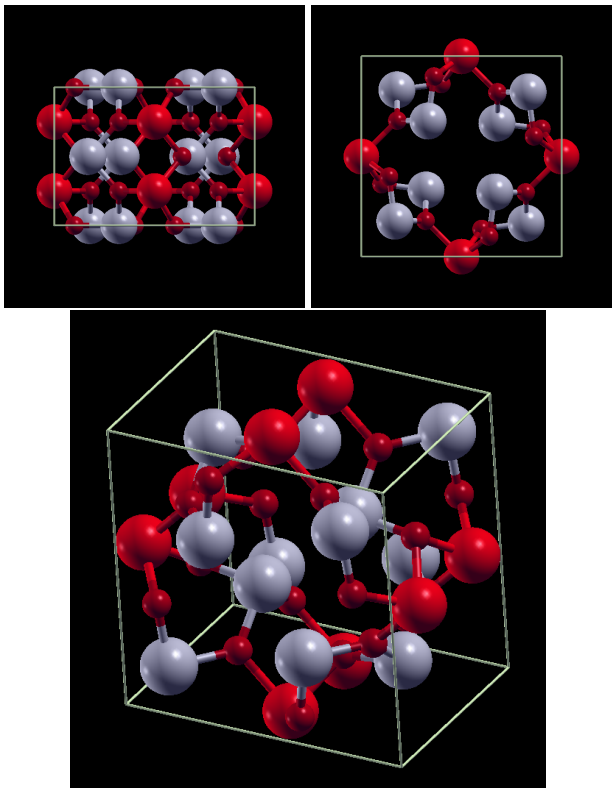

Figure 2: Schafarzikite

Propose the research projects that we can answer.

3) Sankar's project

Computational Methodology (applications/codes)

LAMMPS

The main engine for the MD simulations will be the Large-Scale Atomistic/Molecular Massively Parallel Simulator (LAMMPS) software. This code is highly-scalable for a variety of machine architectures: IBM BG/L, Cray XT3, Cray XT5, and Intel clusters with GPUs (Comet). It can take advantage of GPU and Intel MIC accelerators with good performance scaling. For documentation on general benchmarking of LAMMPS, the following website contains several benchmark problems for a variety of machines: http://lammps.sandia.gov/bench.html.

DFT Codes

Currently, the VISION neutron vibrationl spectrometer instrument at the SNS has its own dedicated computer cluster, VirtuES, for carrying out computer modeling as integral part of the neutron data analysis and interpretation of the spectra (discussed more in the Additional Comments). This cluster has a variety of DFT codes installed for carrying out eletronic struturce calculations to determine vibrational density of states spectra. Vision Users have a suite of codes to choose from: VASP, Quantum ESPRESSO, CASTEP, ABINIT, CP2K, RMG-DFT, and GULP. Also, the O'Climax software is currently being developed by the VISION team as software that can generate calculated incoherent INS spectra from the output of these listed DFT codes.

A major thrust of the ICE-MAN project is to have an interface to the O'Climax code within the first year of the project. This implies that ICE-MAN will also interface with these DFT codes to create the workflow between these codes and eventually feed in to the structural modeling. A subset of these codes will be used for our quantum calculation work so explicit scaling and performance measurements are not show. We will only present a small test case for the Quantum ESPRESSO code only for the schafarzikite project.

RMCProfile

The majority of fitting to neutron scattering data will be carried out using the RMCProfile software to perform reverse Monte Carlo optimization modeling. This software is able to fit many data types simultaneously (Neutron and X-ray total scattering, Bragg diffraction profiles, EXAFS, and single crystal diffuse scattering) and use a range of constraints to produce atomic models that are consistent with all the available data. The fitting will be carried out in a "perfectly parallel" (or "embarrassingly parallel") manner where an array of fits will be carried out simultaneously on serial processors with no communication between the processes. This makes the Open Science Grid XSEDE resource optimial to carry out the fitting optimization. The ICE-MAN project is an extension of the ICE project,

which is already able to handle the launching and monitoring of local and remote jobs, visualizing and analyzing data, and managing data transfers. Thus, we will work towards automating a seamless transition from taking data located on an HPC resource generated by atomistic modeling and simulation and transferring it to the High Throughput Computing (HTC) resource where it will automatically launched for the fitting, perserving HPC SUs for jobs that can make use of the resource. This project would provide an impetus to push parallelization of the RMCProfile software to handle the large atomistic configurations generated and make use of the HPC resources, reducing optimization timescales.

Comet

Comet is the ideal resource for our research for the following reasons:

1. It provides a heterogenous research platform that is capabable of providing domain-specific, ideal architectures. The compute nodes are ideal for our large-scale atomistic simulations and the large memory nodes are ideal for our quatum calculation work. The GPU nodes are available to tackle very large problem sizes that make use of massive level of parallelization efficiently

2. We are targeting the Oak Ridge National Laboratory Leadership Computing Facility resources (i.e. Titan) to launch ICE-MAN. However, due to all nodes containing a GPU on Titan, we can only use the machine to its full potential for a subset of our project. Using Comet as the first target machine allows us access to the resources we will eventually use, broaden the scope of machines that ICE-MAN can utilize, and be accessible to a larger part of the research community.

3. Members of the team already have experience using Comet and have had publications as a direct result of the allocations awarded on the machine.

4. The Data Oasis Lustre parallel file system ensures plenty of scalable storage available for the jobs run on the machine.

Open Science Grid

The Open Science Grid would be an optimal resource to use for launching multple arrays of reverse Monte Carlo jobs in a high throughput manner for large spanning of the phase space. The ICE-MAN project could take advantage on its already-underlying remote job launching capabilities to transfer work to the appropriate computational resource.

Performance and Scaling

Computational Research Plan

We propose to complete the following work on Comet

Justification for Service Units (SUs) Requested

Table 1 summarizes the jusitification for the requested resources to begin our project. The resources are

Table	Project	Machine	Program	Service-Units	Storage (GB)
1	Irradiated SiO2	Comet/Oasis	LAMMPS	300,000	500
2	High-pressure a-Ge/a-Si	Comet/Oasis	LAMMPS	300,000	500
3	Oxygen in FeSb2O4	Comet/Oasis	QE	100,000	500
4	RMC modeling	OSG	RMCProfile	25,000	100

Table 1: Summary of requested service units for projects

Like it?

Additional Comments

Currently, there is a shared comuter cluster available for computations for the group, the Virtual Experiments in Spectroscopy with neutrons (VirtuES) cluster. This machine was made available in 2015 as a funded Laboratory Directed Research and Development project for the VISION neurton vibraional spectrometer at the SNS. The machine consists of 2500+ cores with nodes consisting of two 16-core Intel Xeon E5-2698 v3 running at 2.30GHz. VirtuES is dedicated to the VISION beamline, the first SNS instrument that has computer modeling as integral part of the neutron data analysis and interpretation of the spectra. This cluster mainly used for running DFT calculations to help in data analysis and interpretation of User's neutron data. Our current proposed projects and ICE-MAN development comes secondary to Users needs.

There is also an open-research cluster, called Analysis, available at the SNS for Users to analyze and visualize their data from neutron experiments. This cluster is mainly for accessing and reducing User's data and not suited for HPC applications. MTM has currently just finished using a initial startup allocation for 100,000 total SUs from a previous project on the two XSEDE HPC resources at the San Diegoe Supercomputer Center: Comet and Gordon (50,000 SUs each)