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PNNL-35726

UQ4QM: Uncertainty Quantification for Quantum Materials

Final Report to Chemical Dynamics Initiative

February 2024

W. Steven Rosenthal

U.S. DEPARTMENT OF ENERGY

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Overview

The Uncertainty Quantification for Quantum Materials (UQ4QM) LDRD project focused on developing solutions for strategic two project areas the Chemical Dynamics Initiative (CDi) Use Case 3. This included data-driven approaches toward understanding heterogeneous data, such as multi-fidelity data or data with unknown spatial or temporal perturbations. The applications were varied but the project aims pursued solutions which were amenable to common UQ approaches in order to maximize impact. The two project aims were:

- Materials informatics to predict transition-metal dichalcogenides (TMDs) with potentially useful properties as topological insulators (PI Jenna Pope). An adaptive Gaussian-process-based experimental design tool to was to model the uncertainty in the prediction of energy in these materials using data from a density functional theory (DFT) approach.

- Estimate and reduce uncertainty in time-dependent transmission electron microscopy (TEM) imaging. An algorithm was developed for the identification of common features in a time-series of TEM images with drifting/noisy reference frames. This was combined with a displacement field energy-minimizing iterative alignment tool to facilitate the UQ of chemistry information registered to the aligned pixels in the image.

Summary and Results

The UQ4QM efforts were focused on two collaborations within the existing UC3: a project on predicting structure and energy of transition-metal dichalcogenides (TMDs) (lead by Jenna Pope), and the estimation and UQ of TEM imaging to analyze the structure and composition of materials (lead by Steven Spurgeon). For the TMD task, an adaptive Gaussian-process-based experimental design tool was developed to be integrated with ML codes for the fast relaxation of structures for DFT analysis. This workflow was to be used to build a parameterized surface for the prediction of energy in these materials. A paper on the computation and curation of a dataset used for partial training was submitted in Q4 of FY22. For the TEM imaging task, an algorithm was developed for the identification of common features in a time-series of TEM images with drifting/noisy reference frames. This was combined with an iterative alignment tool to facilitate statistical analysis and reduction in variance for chemistry information registered to the aligned pixels in the image.

To help drive the selection and analysis of TMD candidates for analysis, a multi-fidelity hierarchical model was developed to relate the structure and energetics of TMDs with spin-orbit spillage (SOS) calculations. A multi-scale Gaussian adaptive sampler was implemented to facilitate the design of thermodynamic ensembles of TMD configurations and the acquisition of structure and energetics data from novel DFT calculations. A sequence of ensembles would be selected using the Bayesian optimization framework and generated to guide the selection of TMD compositions toward regions of high SOS with the fewest number of novel SOS computations. (See Figure 1.) The SOS model developed was not sufficiently tractable to drive the outer loop of the multi-scale error model. An alternative dataset was generated from an ML-training optimization application to develop the two-scale Gaussian adaptive sampler. The framework was constrained to match the hierarchical structure of the TMD sampler so that the model remained relevant to the application when the SOS model was optimized.

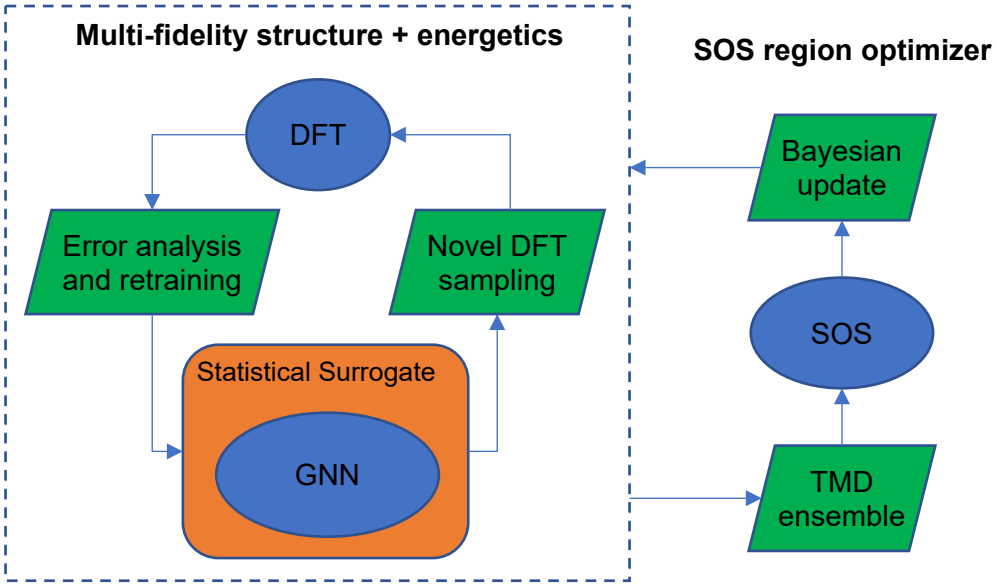

Figure 1: Schematic of multi-fidelity error surface for adaptive sampling of TMD configurations.

To reduce uncertainty in TEM spectra due to time-dependent sample movement, distortion, and degradation, an algorithm for optimal flow mapping was developed to align

the underlying spatial distributions of each frame indicated by the dynamics of observed features. (See Figure 2.) Key regions of interest were identified in coordination with the SMEs as the outermost “edges” of non-overlapping particles. Dynamical drift vectors were estimated by optimizing the local offset between key regions, and the drift field was regularized by a displacement strain penalty function. This automated the process of aligning key regions of interest in subsequent TEM images and determined a corresponding sequence of optimal flow maps to realign the underlying spatial support for the entire image stack. The result enabled more accurate quantification and reduction of variance in TEM spectral data without assuming the rigidity or integrity of target sample, only that the timescale of deformation and degradation was sufficiently greater than the imaging timescale.

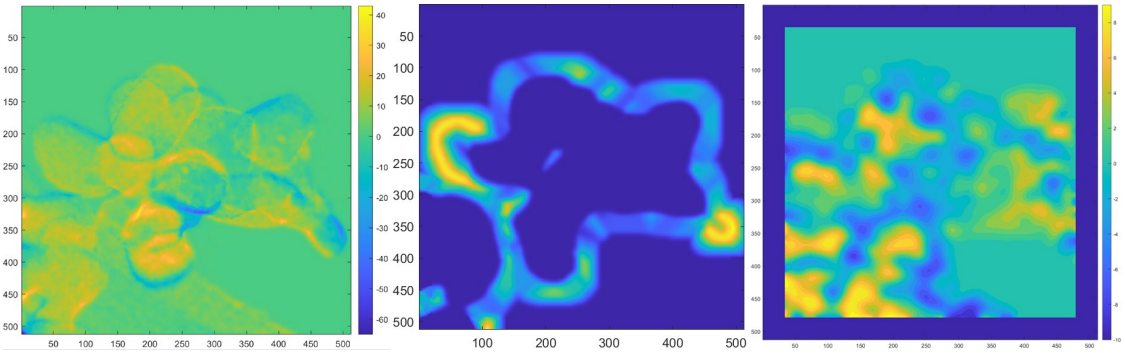

Figure 2: Quantification and minimization of displacement fields of TEM spectra from imaging data. (Left) Intensity differential of TEM images; (center) regions of interest; (right) optimal flow map displacement (x-coordinate).

Under UQ4QM, work was also concluded on a paper for the Adsorbed Iodine Chemistry project (PI T. Blake). Two software stacks for kinetics parameter estimation and spectral uncertainty quantification were developed. The first enabled the calculation of kinetics parameter uncertainties for concentration profiles measured by integrated 1H NMR spectra. It adapted to poor or missing data and unknown initial conditions and reaction rate parameters. The second was a spectral analysis package and user interface (UI) that used a partial time series of 1H NMR spectra to deconvolute overlapping spectra and estimate kinetics and spectral uncertainties. This information was integrated to automate similar 1H NMR analysis for the remaining time series to reduce analysis time from technicians. These tools were applied to DABCO+Iodine reactions in liquid phase for several iodine sources, solvents, and temperatures, and provided the data analytics for an iodine chemistry paper with T. Blake and team.

Acknowledgments

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