URSS Summer Research Project

Mathys Potel

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1 Introduction

I'm a second year physicist doing a summer URSS project with the Warwick Centre for Predictive Modelling (WCPM).

2 How did I apply for the URSS project?

The Undergraduate Research Support Scheme (URSS) at Warwick is an opportunity available for all undergraduate students across any department. The idea is that it allows you to experience 6-10 weeks of interdisciplinary research in a field that interests you. The first step in applying for a summer project is to approach an academic who works in a field that you enjoy and drop them an email showing your interest. In my case, I spoke to Dr Albert Bartok-Partay who is affiliated with both the WCPM and the Heterogeneous Systems (HetSys) group, who I'd heard about through one of my lecturers. Once you have spoken to an academic who is willing to do a URSS project with you, you can begin the process of applying through the Warwick URSS website.

It's a fairly straightforward process that requires you to propose an idea for your project which you will have discussed with your academic. The application also allows you to apply for funding to put towards accommodation and general living costs, as well as any costs incurred by the project itself. It is important to note that there is high demand for URSS projects and generally a limited number of projects the URSS body can fund on any given year. However, in some cases, departments may fund projects directly if applications are unsuccessful, although this is not guaranteed.

3 What is my project about?

This project centres around the computational study of Niobium-doped Strontium Titanate Perovskites. Importantly, dopants allow the properties of perovskites to be tuned to better suit certain applications, such as in thermoelectric devices or in photovoltaics. Atomistic simulations of Nb-doped $SrTiO_3$ allow us to study how Sr^{2+} vacancies and Nb clustering affect its thermal conductivity. Simulating the effect of temperature and defect concentration on thermal conductivity is also important and feeds directly into current theoretical and experimental studies at Warwick.

As part of the application process you have to submit a description of how you plan to carry out your project. The following extract from my submission gives some insight into this.

> "The project will use Molecular Dynamics (MD) as a tool for making accurate, good quality descriptions of atomic interactions. I will use a machine-learning based DFT surrogate model, trained on ab initio data developed by the HetSys group to achieve fast and accurate calculations on the structural properties of doped Strontium Titanate. The ab initio data, involving Gaussian Approximation Potentials, will be used in conjunction with a process of active learning to train the model incrementally. I

will then use this model to understand the effect of defect migration and study the structural dynamics of the octahedral lattice including tilting, where LAMMPS will be used for modelling larger systems over longer timescales. To complete a full analysis of doped Strontium Titanate I will also explore the effects of varying conditions, such as temperature and defect concentration."

URSS

UNDERGRADUATE RESEARCH SUPPORT SCHEME

4 How is the project going? Week 1 & 2

Having reached the end of the first two weeks, I cannot stress enough how much I am enjoying my project so far. I have quickly found that there is a great support network in the WCPM group whether that be through the HetSys cohort, the academics or my supervisor, who has been especially good at explaining new concepts in a way that I understand.

In terms of starting the project, the first step was to start learning the theory. Most of the first week was spent trying to understand some of the basic concepts involved in electronic structure theory (see 'Key Theory' below). In general, I spoke with my supervisor a couple of times a week to discuss progress and concepts that I needed to be able to understand. Outside of these meetings I did a combination of online research and reading through textbooks in addition to attending several in-person seminars on related fields of interest.

Aside from the theory, I familiarised myself with the UNIX command line and learnt how to access and run job scripts on Warwick's High Performance Computers (HPCs). I also spent some time getting to grips with the .cell and .param files as part of CASTEP, which is one of the main programs I will be using to model perovskites from a DFT approach. After downloading several useful pieces of software I began the CASTEP tutorial, found on their webpage, to gain a better understanding of how it works. An example of some of the plots I produced from the tutorial are shown below for graphite.

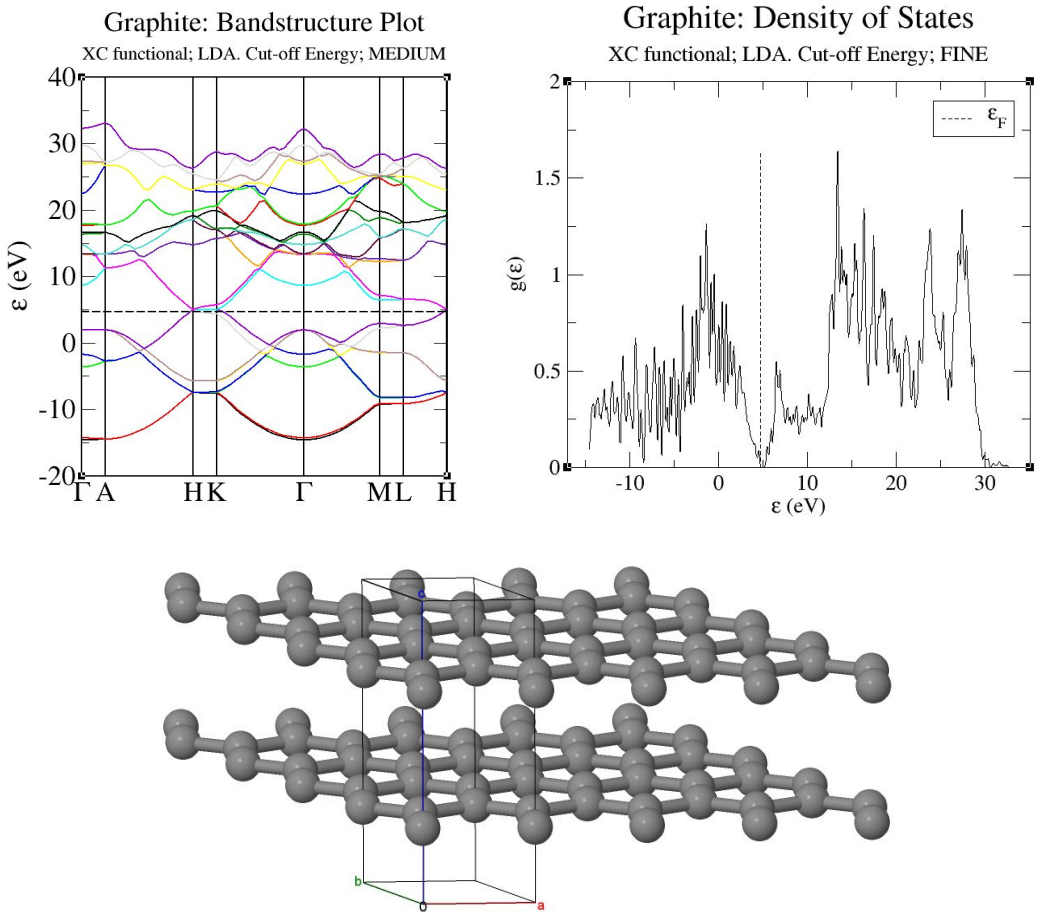

Figure 1: Graphite Structure; Periodic unit cell repeated in 5 x 5 grid

4.1 Key theory from Week 1 and 2:

- Crystal Structure (including Basis and Bravais Lattice)
- Space Groups (including Translation and Point Groups)
- Wigner-Seitz cells, Brillouin Zones and Reciprocal Space
- Excitations and Bloch's Theorem in relation to Periodic Systems
- Born-Von Karmen Periodic Boundary Conditions
- The basics of Density Functional Theory (DFT) and importance of functionals
- Plane waves basis sets and the Schrodinger Equation
- Kohn-Sham equations
- Born Oppenheimer Molecular Dynamics (MD) and the B.O. approximation
- Relation of the B.O. approximation to constructing wavefunctions
- Potential Energy Surfaces (PES) and relation to adiabatic theorem
- Hellman-Feynman theorem; in particular, the forces involved
- Pseudopotentials; these are of particular importance to running MD calculations
- K-point sampling (including an understanding of reducible and irreducible k-points)

5 How is the project going? Week 3 & 4

Over the last two weeks I've spent most of my time working my way through the CASTEP tutorial which introduces the basics of this computational modelling software. The tutorial covers most of the important aspects required to be able to run the program including areas such as bandstructure, density of states, convergence testing, geometry optimization, exchange-correlation functionals and molecular dynamics. Having now finished the tutorial I have a more coherent understanding of how the software works and the general approach you need to take to be able to run your own simulations on other materials.

At the end of the 4th week I attended a two-day conference about CASTEP. This was an amazing opportunity that allowed me to attend a total of 13 different talks on research involving the use of CASTEP. This experience gave me the chance to interact with other researchers as well as the developers themselves and also gave me ideas about posters which will be useful when I create my own poster at the end of this project.

In addition to the technical aspects of learning CASTEP and attending the CASTEP conference, I have also been keeping up to date with the theory-side of the project. Below I have listed an update on some of the content I have covered in weeks 3 and 4.

5.1 Key Theory from Week 3 & 4:

- Bandstructure Plots and Density of States
- Conduction and Valence Bands
- High symmetry points/lines in the Brillouin Zone
- Fast Fourier Transforms (FFTs)
- Exchange-Correlation effects and use of XC functionals to model them
- XC potentials used in Kohn-Sham DFT
- Nearly Free Electron Model (NFEM) and the Tight Binding Model
- The many-body Hamiltonian
- Calculation of electron density using Fourier transforms and grids

6 How is the project going? Week 5 & 6

Over weeks 5 and 6 the focus of the project has shifted towards modelling Strontium Titanate in more detail. Week 5 involved lots of convergence testing for the geometry optimisation of $SrTiO_3$ in its single cell form (i.e. 1×1×1) as well as the plotting of electronic bandstructure. Building on the knowledge gained from the tutorials covered in previous weeks, this allowed a good starting point for exploring Strontium Titanate in more depth.

In week 6 the use of machine-learning was introduced in order to speed up DFT calculations on larger systems. Constructing $SrTiO_3$ systems on a 2×2×2 scale allowed for much more flexibility when it came to observing effects such as octahedral tilting and will also be important when trying to introduce dopants to the system. However, 2×2×2 systems are much more computationally intense which is where the machine-learning algorithm comes in.

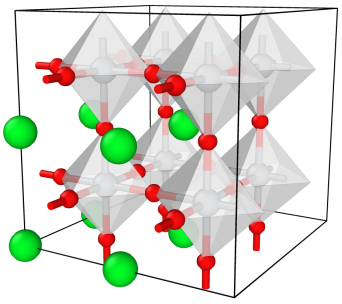

Figure 2: A pure $SrTiO_3$ 2×2×2 supercell. The green spheres are Stronitum, red are Oxygen and the grey cores of the polyhedra are Titanium.

The approach works by switching back and forth between the DFT CASTEP calculations, which are accurate but computationally draining, and GAP (Gaussian Approximation Potentials) which is the machine-learning algorithm that interpolates the interatomic PES (Potential Energy Surface). Once I had practiced using the machine-learning tools to run some simple Ab Initio Molecular Dynamics (AIMD) on Silicon Carbide (SiC) I ran some similar tests but with $SrTiO_3$. As before, at this stage a lot of convergence testing was required to determine suitable input values for this 2×2×2 system.

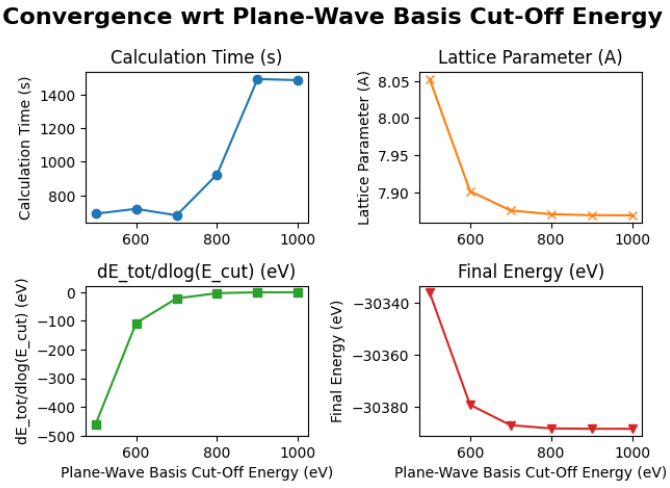

Figure 3: An example of some of the convergence testing done on the $SrTiO_3$ 2×2×2 supercell.

In addition to this work on the project, I was also invited to attend the HetSys conference in the Scarman Conference Centre on campus. Over the course of 3 days I got the chance to listen to numerous talks about the current progress being made in this field of research. This gave me a useful insight into the type of work that a PhD might entail if I were to pursue this branch of science further.

7 How is the project going? Week 7 & 8

These last two weeks have highlighted 3 main areas to explore in relation to building a database for Strontium Titanate. These fall loosely under the following categories; "Pure $SrTiO_3$", "Pure $SrTiO_3$ with Noise" and "Niobium-doped $SrTiO_3$". In week 7 much of the focus was on building the suitable models for these three categories and performing geometry relaxations on them. As part of this I had to explore different ways of doping the 2×2×2 system with Niobium, whilst exploiting symmetries, so that eventually the model could be trained to recognise different doped configurations. Given the computational cost of relaxing 2×2×2 systems, I initially used coarse k-point grids to check the code ran as expected before switching over to a finer k-point grid.

Much of week 7 was spent analysing the data from the output files of these relaxations. To do this I familiarised myself with the "grep" commands and wrote several python scripts to extract, manipulate and plot the required data. Below is an example showing how three different tolerances are converged over 36 LBFGS iterations (with magnified plots on the right) for a system of Strontium Titanate with added noise.

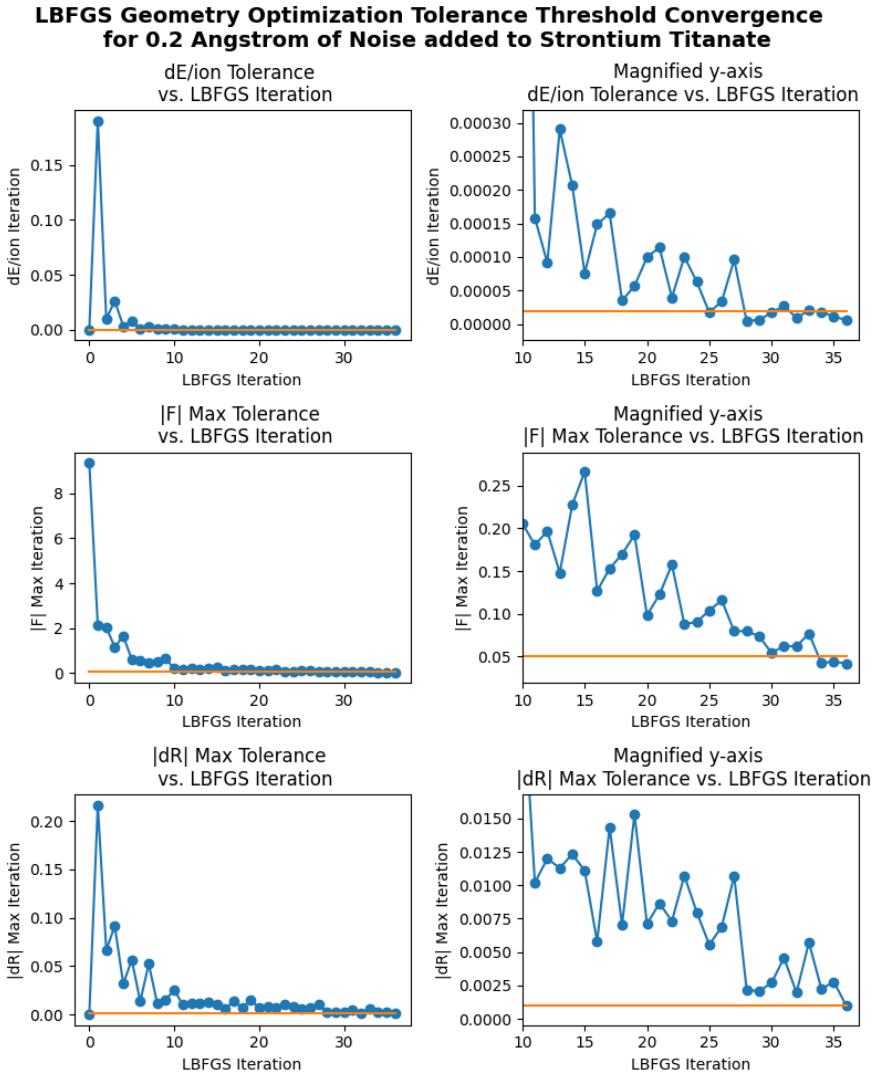

Figure 4: Geometry optimization of pure Strontium Titanate with 0.2 Angstrom of added noise.

In the last week of the project I ran some AIMD simulations using the relaxed Strontium Titanate structures. In order to analyse these simulations I wrote several more python scripts to extract and plot the relevant data which allowed me to keep track of how well the machine-learning model was coping. In conjunction with this I produced some more visualisations of the Strontium Titanate structures in OVITO, a software I have used throughout this project. This meant I was able to fine-tune some of the input parameters including the 2-body descriptor and some of the refitting tolerances to ensure the model was as accurate as possible.

Alongside the work I did on my project, my supervisor also invited me to attend the 'GAP and (M)ACE Conference' held here at Warwick. I got the opportunity to listen to a whole range of talks including presentations on current research in this field, talks on the latest software developments and even some tutorial sessions. At the end of the second day of the conference there was also a "Poster Session" where about two dozen researchers presented their work in a more informal setting. This allowed for more in depth discussions as well as giving me inspiration for designing my poster - the final output of my URSS project.

8 Final Remarks

Completing the URSS project over the summer between my 2^{nd} and 3^{rd} year has been an incredibly valuable experience. Thanks to my supervisor, Dr Albert Bartok-Partay, I have had the opportunity, over the last few months, to delve into the world of condensed matter physics and get an insight into what computational research entails. I have been lucky enough to attend numerous seminars, conferences and presentations in related fields and have had the chance to interact with some of the students and academics involved in this area of research. One of the main challenges I faced whilst doing the project was the steep learning curve involved with learning the theory in conjunction with the coding. However, thanks to the amazing support and help from my supervisor this was one of the things I found most enjoyable about the project and is one of the reasons I would thoroughly recommend doing a URSS summer project to any 2^{nd} or 3^{rd} year students considering it.