Probing Single Crystal Growth Experimentally and through Computational Simulations

Heather Calcaterra
Chemical Engineering, University of Michigan Ann Arbor

REU Program: 2017 Johns Hopkins University Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials Research Experience for Undergraduates (JHU PARADIM REU) Program
JHU PARADIM REU Principal Investigator: Prof. Tyrel McQueen, Department of Chemistry, Department of Materials Science and Engineering, Department of Physics and Astronomy, The Johns Hopkins University
JHU PARADIM REU Mentors: Dr. W. Adam Phelan and Mekhola Sinha, Department of Chemistry, The Johns Hopkins University
Primary Source of PARADIM REU Funding: NSF Materials Innovation Platform Program, Grant # DMR-1539918
JHU PARADIM Tools Used: High pressure optical floating zone furnace, laser diode floating zone furnace, induction furnace
Contact: hcalc@umich.edu, mcqueen@jhu.edu, wphelan2@jhu.edu, msinha4@jhu.edu
Website: http://www.cnf.cornell.edu/cnf_2017reu.html

Abstract:

Advanced electronic, magnetic, and topological materials, such as semimetals, skyrmions, and topological insulators, have attracted significant interest for their predicted applications in quantum computing and information storage. To realize many of the necessary electronic and magnetic properties, these materials need be composed of only a single phase of matter arranged in a continuous, unbroken crystal lattice. This state, known as the single crystalline state, is non-trivial to synthesize in bulk.

One of the biggest challenges in single crystal growth is the low yield of the product, which can result from excessive vaporization and a surrounding dynamic fluid environment. To address this problem, we have created a computational model that simulates actual single crystal growth conditions employed in the laboratory. Furthermore, single crystal samples of proposed topological materials were grown within the laboratory through a variety of synthesis techniques and conditions, with aim to control the composition and yield of the final products. Herein, I present my methods and results in the synthesis, growth, and characterization of novel electronic and topological materials and bulk single crystals, both learned experimentally and through computational simulations.

Summary of Research:

During my REU internship, I have undertaken significant collaborations with various members in academia and industry in creating these novel materials.

In summary, several methods exist to create single crystal samples from bulk. Most apply heat to the polycrystalline sample in order to create a temperature gradient; the method of heating differs between samples. Common techniques include the Bridgman, laser diode floating zone, and high pressure floating zone growths. For the floating zone techniques, the material is melted via irradiation with light; for the Bridgman growth, an induction furnace with heating coils is used to heat the sample. We need to accurately choose the technique carefully to optimize the purity and yield.

When a polycrystalline sample is heated above its melting point, it is inevitable that some vaporization will occur. Depending on materials properties such as vapor pressure, the vaporization can be so significant as to greatly decrease the yield of the final product or even prevent proper melting and crystallization. This can be especially problematic at high pressures of the surrounding 'observer' supercritical fluid, as the solubility of the gas in the dynamic fluid environment will increase. Hence, we would like to find a way to minimize this vaporization.

We utilized COMSOL Multiphysics, a finite element differential equation solver, to investigate this phenomenon and decide what

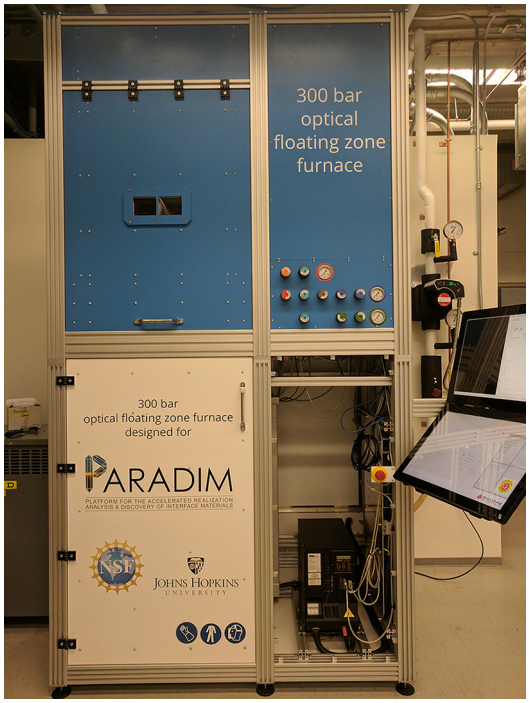

Figure 1: The high pressure floating zone furnace can be used to perform crystal growth under high pressures of inert gases, allowing the formation of materials not accessible under atmospheric pressures.

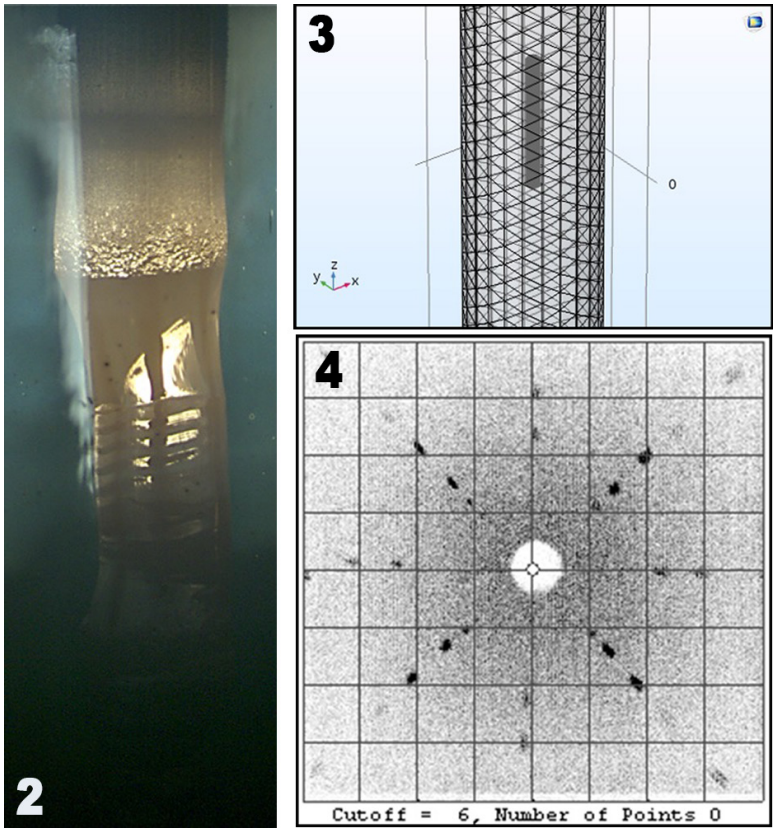

Figure 2, left: The crystal growth SmB_6, a material with predicted topological insulating character, is performed in the high pressure floating zone furnace. Figure 3, top right: Visual representation of the computational model created in COMSOL. The outer cylinder is the protective quartz container and the inner rod is the polycrystalline melt material. Figure 4, bottom right: Laue spots from the grown crystal show good symmetry, indicating the formation of a single crystal.

parameters might be ideal for crystal growth. We sought to investigate the effects of pressure, temperature gradient, and other materials-specific properties on the surrounding fluid environment in accordance with the Navier-Stokes Equations.

For our computational model, we measured dimensions that describe the laser diode floating zone furnace found in PARADIM. We defined the border of the simulation as being made of quartz, which is used as the protective shield in the furnace. The rod of polycrystalline material was created using average dimensions of an actual precursor rod, and we defined the rod properties as several different commonly grown species, such as silicon and titanium oxide. Finally, the simulation space with observer gas of either argon or oxygen, and the pressure exerted by these gases was changed sequentially.

Results and Conclusions:

We were able to model key parameters governing single crystal growth in the high pressure floating zone furnace and adjust them as needed. Difficulties in accurately defining the temperature gradient between the rod and the surrounding quartz tube prevented convergence of our simulation. This suggests to us that the complexity of the surroundings in growing single crystals is non-negligible and further studies must be conducted.

On the experimental end, it is important to ensure that our material is actually a single crystal after performing the melt. Crystallinity is determined by seeing how the material interacts with light, commonly using a technique called Laue diffraction. Symmetric spots show that the material satisfies Bragg's condition and the beam is parallel to a high-symmetry direction of the crystal, hence suggesting single crystallinity of a target material, SmB_6. However, in growing this material we had significant vaporization, solidifying the need for our working computational model.

Future Work:

During my REU, I have collaborated with users to plan, synthesize, and characterize single crystals such as the SmB_6 shown, using the materials by design method. These grown crystals have novel electronic and magnetic properties, such as topologically insulating character, which is helpful for next generation electronic devices and quantum computing.

However, there is still much work to do in minimizing vaporization and creating a more accurate simulation. In real experiments, there are often two rods, a seed and a feed rod, but only one rod was included in our simulation for simplicity. Furthermore, we described the observer gases as stationary. In the future, we would like to be able to simulate conditions for more complex electronic materials grown in PARADIM, such as SmB_6 that was grown experimentally. However, priority should be given to efforts in accurately describing the temperature gradient between the seed and feed rods and on ensuring the convergence of the Navier-Stokes equations used in the model.

Acknowledgements:

I thank NSF, PARADIM, and JHU for their funding. Additionally, I would like to thank Dr. W. Adam Phelan, Mekhola Sinha, Prof. Tyrel McQueen, and all the McQueen Lab members for their help.

References:

[1] W. Adam Phelan, Scientific Reports. 6, 20860 (2016).