Using Density Functional Theory to Predict the Formation Energies of AI/ML Generated Superconducting Structures

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Abstract

The Applied Physics Laboratory (APL) at Johns Hopkins University (JHU) is programming an AI/ML algorithm that can generate potential superconducting structures (PSS). The project is to utilize Density Functional Theory (DFT) to calculate the formation energies of these PSSs and feed the data gathered back into the algorithm for training purposes. What was found is that the calculations for the cuprates (copper-oxygen) compounds $BaSrLiCu_3O_5$ and $BaSr_2LiCu_4O_7$ were completed but were not energetically favorable relative to the carbonate (carbon-oxygen) oxide reagents. In addition, many other cuprates were not completed in a DFT calculation because of the exotic crystal layout of the PSSs. $Rb_2NaSnBr_6$ shows to be a promising candidate; however, more investigation is necessary. What these calculations show is that DFT is able to model different reactions and investigate the energetic favorability.

1 Introduction

Superconductivity is a massive scientific frontier in science and is heavily investigated by numerous lab groups across the globe due to their lack of resistivity and perfect diamagnetic properties. It is a state of matter that is only achieved through low temperatures approaching subzero (0K) predominately around 0K-10K; however, some superconductors are recorded to ascend higher than 100K. Potential applications of superconductors are the power distribution industry as electricity is more conserved because there is no energy lost in resistance, Maglev trains that utilize superconductors that oppose all magnetic fields allowing the trains to float on frictionless tracks, magnetic resonance imaging (MRI) devices which are a huge application of superconductors, etc. The problem with superconductors is that the state of matter only exhibits at critically low temperatures, therefore, it is difficult to apply without a way to bring the temperature of the said material to their respected critical temperatures T_c. Essentially, this can lead to an immense cost of operations using an expensive substance such as liquid helium to bring the superconductor to T_c.

With the emerging field of artificial intelligence (AI) and machine learning (ML), the Applied Physics Laboratory (APL) at Johns Hopkins University (JHU) is attempting to push further discovery of superconductors by programming their own AI/ML algorithm to see if it can potentially discover new superconducting structures, which may assist in the discovery of a superconductor with a T_c of relative room temperature.

The algorithm generates numerous potential superconducting structures (PSS), which is then narrowed down with scientific crystal structure intuition and it is also synthesized to test

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for superconductivity. This report will cover a Density Functional Theory (DFT) approach to the process of training this APL AI/ML algorithm, as the results gained in this process are then fed back into the algorithm which will then generate more plausible superconducting structures. It will also show the capabilities that DFT has in modeling different reactions.

2 Methodology

The formation energies E_f of PSSs were the focus of this project. DFT calculations were performed on all structures including the PSSs and relevant reagents, potentially used in the synthesis process. A free open-source package, Quantum Espresso (QE) was used for the calculations, in which it contains a number of pseudo-potentials, coded by independent and inter-operable groups. The primary functional type used was the Perdew-Burke-Ernzerhof Generalized Gradient Approximation (PBE-GGA) for all structures, the self-consistent field (SCF) calculation was used to find enthalpy of the PSSs and reagents, and the calculations are all done at OK. The structures for the reagents were all downloaded from Materials Project. To accept the accuracy of the DFT calculation, it must complete as that proves the calculation process was properly executed. Once completed, a stoichiometry equation is set up with a PSS being one of the products, along with the relevant reagents and extra products as equation (1) specifies. The formation energy of the PSS is then calculated using equation (2) where the difference of the sum of E_f of products ($\Sigma E_f Prod$) and the sum of E_f of reagents ($\Sigma E_f Reag$), divide it by the sum of atoms of the current PSS (ΣPSS). If E_f equals a positive, DFT predicts the PSS will not form, and if negative, DFT predicts the PSS might form. Data is then compared to experimental data, and fed back into the algorithm.

$$Reagents \rightarrow PSS + Products \quad (1)$$

$$\frac{\Sigma E_f Prod - \Sigma E_f Reag}{\Sigma PSS} \quad (2)$$

3 Results and Discussion

Numerous calculations were done on multiple PSSs with most not converging under a SCF calculation (mostly the cuprates did not converge), however, for the sake of this report, we will condense the findings down to $Rb_2NaSnBr_6$ shown in figure 1, $BaSrLiCu_3O_5$ shown in figure 2, and $BaSr_2LiCu_4O_7$ shown in figure 3. The table provided displays different stoichiometry equations and their E_f relative to the reaction. When dealing with all three of the PSSs compounds, they are predicted to be energetically favorable with the elements as their reagents. With $BaSrLiCu_3O_5$ and $BaSr_2LiCu_4O_7$ as the respected carbonates (carbon-oxygen) and oxides for reagents, they are predicted to not be energetically favorable. For $Rb_2NaSnBr_6$ more equations with different reagents were tested with the compound showing energetically favorable reactions until all reagents were compounds of the respected element needed for $Rb_2NaSnBr_6$.

Given the predictions of the calculations, this could indicate that the PSSs the algorithm generates are likely not possible with current synthesis because compound-based reagents that are typically used in synthesis, especially considering the cuprates (copper-oxygen), and the equations are not coming out as energetically favorable. A potential explanation is due to the PSSs not existing, therefore, the DFT calculations are predicting a positive energetic favorably as so. Further investigation is necessary as the calculations could be implemented differently in QE as well as the data being compared to more experimental data. $Rb_2NaSnBr_6$ still needs to be synthesized. What is shown is that DFT models different reagents to better guide the experimentalist on their synthesis.

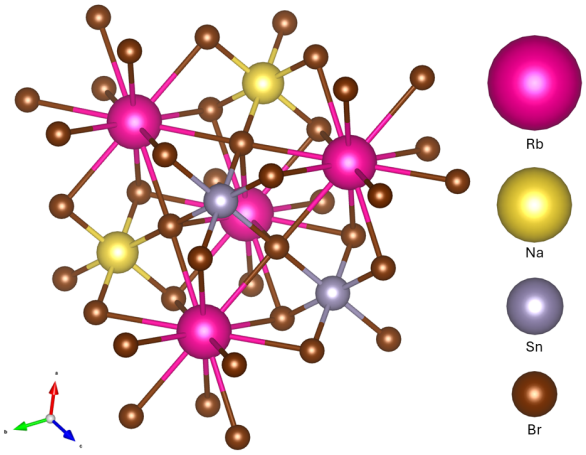

Figure 1: $Rb_2NaSnBr_6$

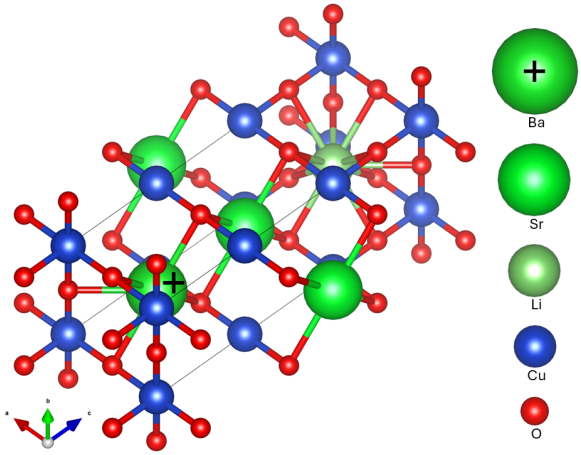

Figure 2: $BaSrLiCu_3O_5$

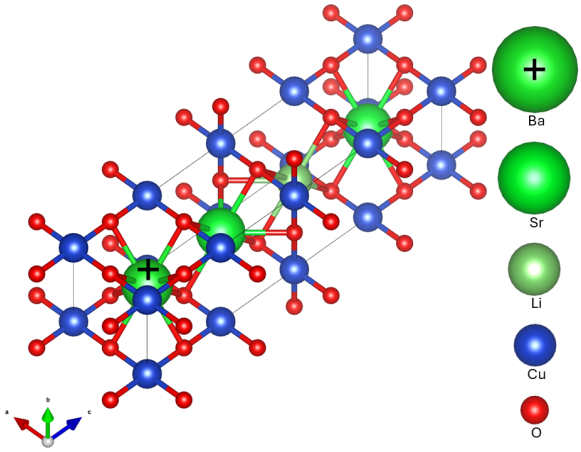

Figure 3: $BaSr_2LiCu_4O_7$

PSS	Reagent	PSS + Product	E_f
$BaSrLiCu_3O_5$	$Ba + Sr + Li + Cu_3 + O_5$	$BaSrLiCu_3O_5$	-
$BaSrLiCu_3O_5$	$2BaCO_3 + 2SrOC_3 + Li_2CO_3 + 6CuO$	$2BaSrLiCu_3O_5 + 5CO_2 + \frac{1}{2}O_2$	+
$BaSr_2LiCu_4O_7$	$Ba + Sr_2 + Li + Cu_4 + O_7$	$BaSr_2LiCu_4O_7$	-
$BaSr_2LiCu_4O_7$	$2BaCO_3 + 4SrCO_3 + Li_2CO_3 + 8CuO$	$2BaSr_2LiCu_4O_7 + 7CO_2 + \frac{1}{2}O_2$	+
$Rb_2NaSnBr_6$	$Rb_2 + Na + Sn + Br_6$	$Rb_2NaSnBr_6$	-
$Rb_2NaSnBr_6$	$2RbBr + NaBr + SnBr2 + Br$	$Rb_2NaSnBr_6$	-
$Rb_2NaSnBr_6$	$2RbBr + Na + SnBr_4$	$Rb_2NaSnBr_6$	-
$Rb_2NaSnBr_6$	$2RbBr_3 + Na + Sn$	$Rb_2NaSnBr_6$	-
$Rb_2NaSnBr_6$	$4RbBr + 2NaBr + SnBr_2 + SnBr_4$	$2Rb_2NaSnBr_6$	+

4 Conclusion

Given the many structures generated by the AI/ML algorithm rejected, many PSSs calculations not completing (mostly being the cuprates), and the lack of energetically favorable reactions with the PSSs that do complete in the DFT calculation, the APL AI/ML algorithm has much development to undergo. Ultimately, there are numerous amounts of PSS that are worth looking into, given how $Rb_2NaSnBr_6$ looks promising. The data found during this project will be fed back into the algorithm which will then improve the accuracy.