Weekly Report

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June 23, 2020 - July 1, 2020

1 Introduction

This week I try to use PythTB to generate some band structures and moderate some of the hopping parameters to see how band structures change accordingly. Besides, I also try to reproduce one of the papers in MSE593 courseware for practising. In both work I make some progress and meet a few questions.

2 Progress

2.1 PythTB model

First, I start with a simple and virtual bcc lattice with s,p_x,p_y,p_z spinless bands and take nearest neighbor atoms hopping into account(like NaCl crystal). So a 4*4 Hamiltonian matrix has to be specified. For simplicity, I only consider four types of hoppings, which are:

hop1: hopping between s and three p bands in the same primitive cell.
hop2: hopping between neighboring s bands.
hop3: hopping between neighboring p bands.
hop4: hopping between neighboring s and p bands.
The following picture is the band structure along Γ, H, P, N points.

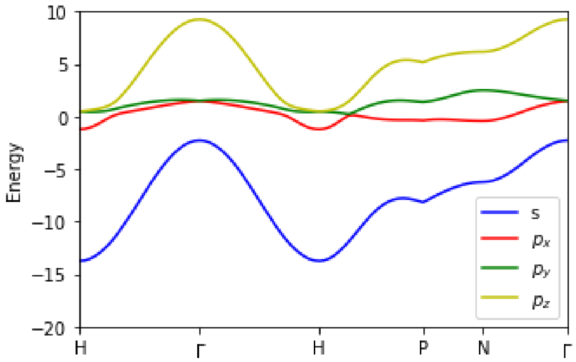
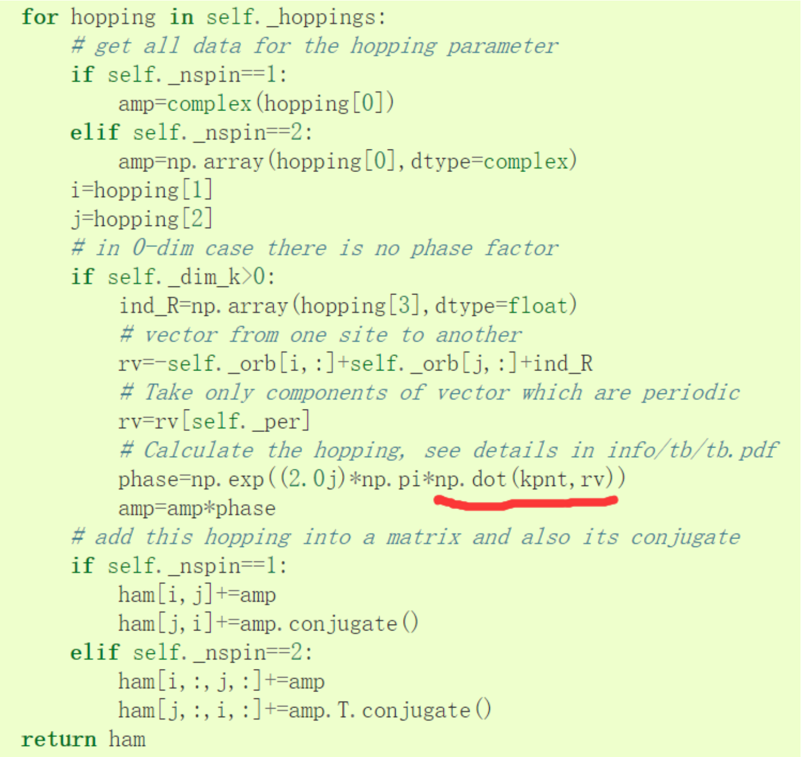


Figure 1: example

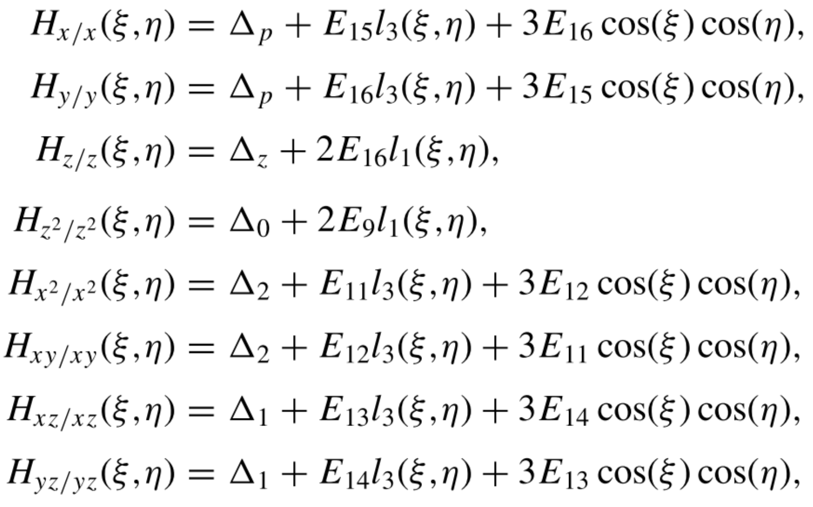
Second, with Pillows package, we can get how band structure change with these four hopping parameters. I don't know how to save the interactive picture, so I will attach the code(both the .ipynb version and .py version) in email.

Then, when it comes to real systems, some difficulties come up in the way. Take MoS2 for example. Setting all the parameters identical to relative papers, PythTB still get the incorrect band structure. So I check the source code of PythTB and finally find the reasons. When generating Hamiltonian matrix, PythTB would simply regard the phase factor to be K values dot relative distance(rv), like figure 2 shows. But in real tight binding model, the relationship between phase factor and K values can be very complex. Here I cut out some elements in figure 3 and 4by



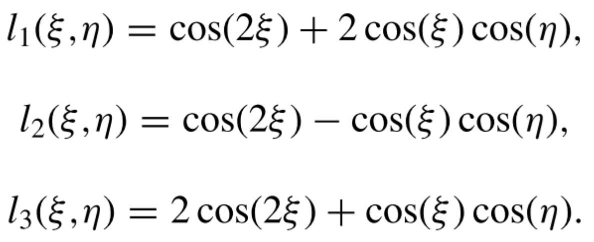
```
for hopping in self._hoppings:
    # get all data for the hopping parameter
    if self._nspin==1:
        amp=complex(hopping[0])
    elif self._nspin==2:
        amp=np.array(hopping[0],dtype=complex)
    i=hopping[1]
    j=hopping[2]
    # in 0-dim case there is no phase factor
    if self._dim_k>0:
        ind_R=np.array(hopping[3],dtype=float)
        # vector from one site to another
        rv=-self._orb[i,:]+self._orb[j,:]+ind_R
        # Take only components of vector which are periodic
        rv=rv[self._per]
        # Calculate the hopping, see details in info/tb/tb.pdf
        phase=np.exp((2.0j)*np.pi*np.dot(kpnt,rv))
        amp=amp*phase
    # add this hopping into a matrix and also its conjugate
    if self._nspin==1:
        ham[i,j]+=amp
        ham[j,i]+=amp.conjugate()
    elif self._nspin==2:
        ham[i,:,j,:]+=amp
        ham[j,:,i,:]+=amp.T.conjugate()
return ham
```

Figure 2: PythTB source code



$$\begin{aligned}
H_{x/x}(\xi,\eta) &= \Delta_p + E_{15} l_3(\xi,\eta) + 3E_{16}\cos(\xi)\cos(\eta),\\
H_{y/y}(\xi,\eta) &= \Delta_p + E_{16} l_3(\xi,\eta) + 3E_{15}\cos(\xi)\cos(\eta),\\
H_{z/z}(\xi,\eta) &= \Delta_z + 2E_{16} l_1(\xi,\eta),\\
H_{z^2/z^2}(\xi,\eta) &= \Delta_0 + 2E_9 l_1(\xi,\eta),\\
H_{x^2/x^2}(\xi,\eta) &= \Delta_2 + E_{11} l_3(\xi,\eta) + 3E_{12}\cos(\xi)\cos(\eta),\\
H_{xy/xy}(\xi,\eta) &= \Delta_2 + E_{12} l_3(\xi,\eta) + 3E_{11}\cos(\xi)\cos(\eta),\\
H_{xz/xz}(\xi,\eta) &= \Delta_1 + E_{13} l_3(\xi,\eta) + 3E_{14}\cos(\xi)\cos(\eta),\\
H_{yz/yz}(\xi,\eta) &= \Delta_1 + E_{14} l_3(\xi,\eta) + 3E_{13}\cos(\xi)\cos(\eta),
\end{aligned}$$

Figure 3: Some TB model elements of MoS2



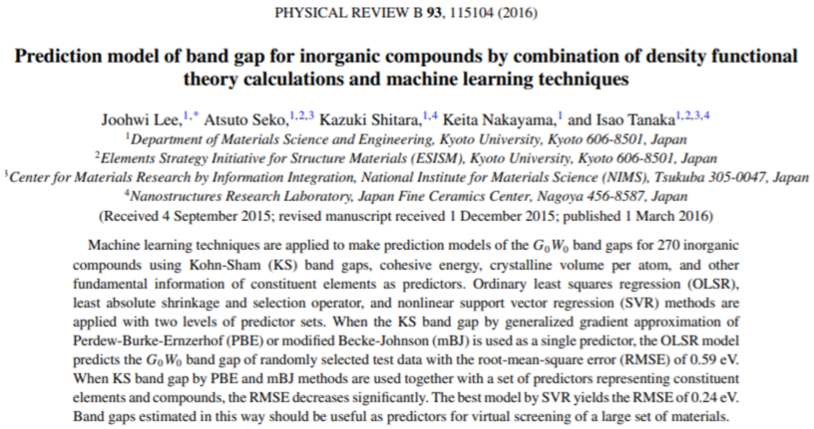
$$\begin{aligned}
l_1(\xi,\eta) &= \cos(2\xi) + 2\cos(\xi)\cos(\eta),\\
l_2(\xi,\eta) &= \cos(2\xi) - \cos(\xi)\cos(\eta),\\
l_3(\xi,\eta) &= 2\cos(2\xi) + \cos(\xi)\cos(\eta).
\end{aligned}$$

Figure 4: Supplment of the figure 3

Therefore, although theoretically we can determine s,p,d orbital by different onsite energy and hoppings, PythTB is not powerful enough to show the band structure of real systems. If we go on this way, maybe for each system we should rewrite PythTB package to get correct Hamiltonian matrix.

2.2 Reproduction of a paper

Here I choose the first paper in the courseware.



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Prediction model of band gap for inorganic compounds by combination of density functional theory calculations and machine learning techniques

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Machine learning techniques are applied to make prediction models of the G_0W_0 band gaps for 270 inorganic compounds using Kohn-Sham (KS) band gaps, cohesive energy, crystalline volume per atom, and other fundamental information of constituent elements as predictors. Ordinary least squares regression (OLSR), least absolute shrinkage and selection operator, and nonlinear support vector regression (SVR) methods are applied with two levels of predictor sets. When the KS band gap by generalized gradient approximation of Perdew-Burke-Ernzerhof (PBE) or modified Becke-Johnson (mBJ) is used as a single predictor, the OLSR model predicts the G_0W_0 band gap of randomly selected test data with the root-mean-square error (RMSE) of 0.59 eV. When KS band gap by PBE and mBJ methods are used together with a set of predictors representing constituent elements and compounds, the RMSE decreases significantly. The best model by SVR yields the RMSE of 0.24 eV. Band gaps estimated in this way should be useful as predictors for virtual screening of a large set of materials.

The paper focuses on the experimental data and three theoretical model of bandgap and tries to use machine learning model to get a modification of theoretical results. The dataset contains 32 compounds which both experimental and theoretical results are aviliable and 238 compounds which only contain theoretical results.

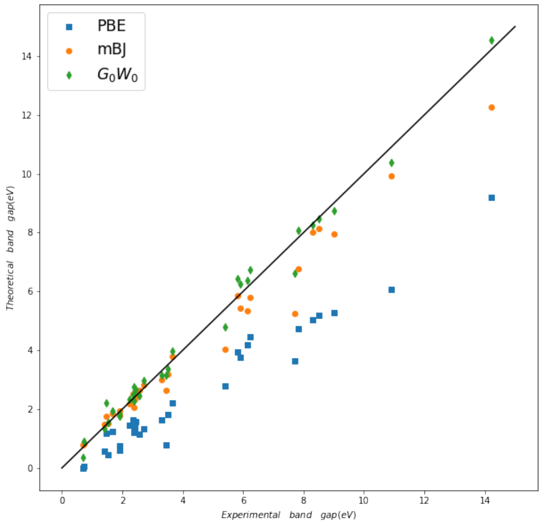

Figure 5: Comparison of theoretical and experimental band gaps

First, I use SVR machine learning model to adjust the experimental data and G_0W_0 theoretical results. Figure 6 shows the result.

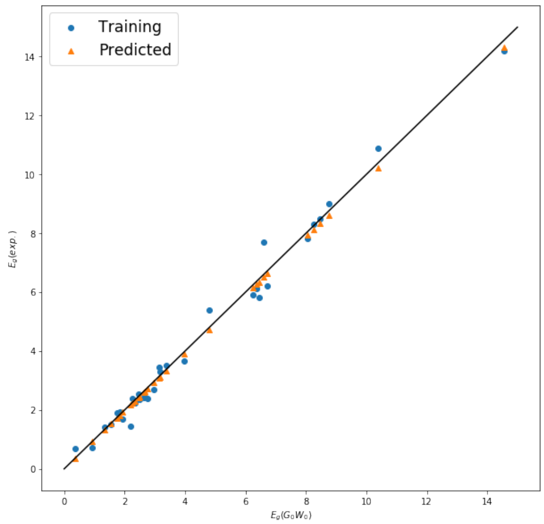

Figure 6: SVR model of experimental data and G_0W_0 theoretical results

Then, for the compounds which experimental datas are absent, I try to use the model to get the modified results.

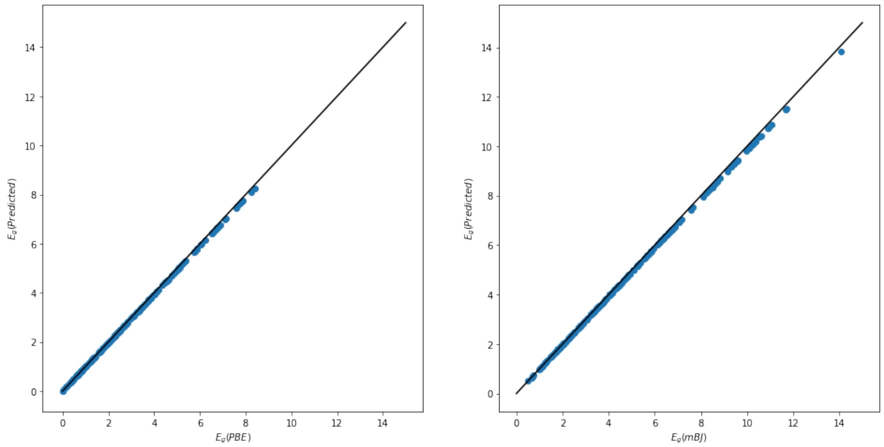

Figure 7: Using SVR model to modify PBE and mBJ computational results

I don't draw the learning curve or get RMSE values because during the work I realise some questions that have confused me for a long time.

1. The training set is too small, which leads to the result not so accurate or reliable. We only have 32 data for training and it is even not enough for one batch, which makes the process more like a data fitting instead of

machine learning. This problem also exists in my own work. In material science, the dataset is usually very small compared to Computer Science field. So collecting enough data and selecting proper machine learning model(perhaps self-supervised learning?) would be a challenge.

2. This kind of model don't use feature function or something and the machine learning parameters are a black box, so it lacks of physical interpretability. We can't get an explanation about why computational results differ from experimental data and what the offset terms stand for.

3 Summary

This week I mainly play with PythTB and some machine learning papers. Both work are not going smoothly. If we want to see how band structure change with specific band hoppings, we have to turn to other tools. What's more, maybe I can try other papers in the courseware that are more meaningful to reproduce next week.

4 Questions

Some of my questions are listed in section 2.2. Besides, I have a question about TB model:

I'm wondering how the papers about TB model like 2 and 3 draw their band structure picture? Are they code for the different systems or there exist some mature tools that can do such jobs?

5 Reference

1. Joohwi Lee, Atsuto Seko, Kazuki Shitara, Keita Nakayama,and Isao Tanaka, Prediction model of band gap for inorganic compounds by combination of density functional theory calculations and machine learning techniques, DOI:10.1103/PhysRevB.93.115104

2. E. Cappelluti, R. Rold´an, J. A. Silva-Guill´en, P. Ordej´on, and F. Guinea1, Tight-binding model and direct-gap/indirect-gap transition in single-layer and multilayer MoS2, DOI:10.1103/PhysRevB.88.075409

3. Yaohua Tan, Michael Povolotskyi, Tillmann Kubis, Timothy B. Boykin, and Gerhard Klimeck, Transferable tight-binding model for strained group IV and III-V materials and heterostructures, DOI:10.1103/PhysRevB.94.045311