Bulk-Derived Local Pseudopotentials Generator

Based on Modified ABINIT 7.0.5 Package

This manual that accompanies the generator of bulk-derived pseudopotentials was written by Mohan Chen on May 2016.

1. Bulk-Derived Local Pseudopotential (BLPS)

The Wu-Yang method [1] implemented in ABINIT yields an effective potential $V_{eff}(\vec{r})$, which is also named 'bulk-derived local pseudopotential (BLPS)' [2] due to the fact that the charge densities of bulk materials are used as references during the potential optimization. The Wu-Yang functional is defined as

$$W\left[v_{eff}(\vec{r}),\rho_0(\vec{r})\right]=T_s\left[v_{eff}(\vec{r})\right]+\int dr\left[\rho(\vec{r})-\rho_0(\vec{r})\right]V_{eff}(\vec{r}),$$

where the target electron density $\rho_0(\vec{r})$ is typically generated via density functional theory (DFT) computations using a non-local pseudopotential (NLPS). In our implementations, once a BLPS is generated by the ABINIT-BLPS code, the structure factor $S(\vec{q})$ in $V_{eff}(\vec{q})$ (the Fourier counterpart of $V_{eff}(\vec{r})$) should be removed to yield a BLPS of an atom as:

$$V_{atom}(q)=\frac{V_{eff}(\vec{q})}{S(\vec{q})},$$

where $q=|\vec{q}|$. We only work with the non-Coulombic part of $V_{atom}(q)$ to avoid the singularity at $q=0$. The non-Coulombic part is written as

$$V_{nc}(q)=V_{atom}(q)+\frac{4\pi z}{q^2}.$$

In principle, $V_{nc}(q=0)$ can be any given value. However, a spline fit is applied to obtain $V_{nc}(q)$ at small q. Thus, $V_{nc}(q=0)$ should be carefully chosen.

2. Install the package

Here we introduce how to download and install the BLPS patch.

1) Download and compile the ABINIT 7.0.5 package from the ABINIT website.

2) Enter the directory of the ABINIT-BLPS package. Make sure both *'inv_KS_code'* and

'tools' directories exist.

3) Enter the *'inv_KS_code'* directory and run the script *'install.sh'* to install the BLPS patch for ABINIT. You will see:

Path-file path_to_abinit-7.0.5 not found
Please enter directory of compiled original abinit-7.0.5

Please enter the directory of the compiled original abinit-7.0.7 package.

Enter the directory where you installed the ABINIT 7.0.5 package.

Please enter target directory for install of inv_KS
This directory should not exist yet!

Enter a new directory path where you want to install the ABINIT-BLPS generator (follow the instructions and confirm with 'yes' to complete the installation). Next, please wait until the BLPS patch is fully installed. You will find the package in the new directory and the executable 'abinit' is in the 'src/98_main' directory.

3. Procedures to Generate a BLPS

Here we discuss the detailed procedures to generate a BLPS:

1) Perform Kohn-Sham DFT computations to obtain bulk properties of target systems. In general, these properties include lattice constants, bulk moduli, and equilibrium volumes of various crystal structures such as body-centered-cubic (bcc), face-centered-cubic (fcc), simple cubic (sc) and cubic diamond (dia) structures. These results will be used to benchmark the results obtained from computations based on a BLPS.

2) Perform ABINIT computations (with a NLPS) with the geometries of target systems. There are two detailed steps:

   a. Prepare the input files of ABINIT. Use *"prtden 1"* in the input file (probably this is a default setting) of ABINIT to print out the electron density of the system.

   b. Use *cut3d* (a small program provided by the ABINIT package) to transform the charge density file (a file typically ends with *"_DEN"*) from unformatted to formatted. The new formatted electron density file should be named *"refden.in"*. When the *cut3d* program is used, please follow the procedures as indicated here:

Question 1: What is the name of the 3D function (density, potential or wavef) file?
Answer: the output name of the electron density file (a file typically ends with *"_DEN"*).
Question 2: Does this file contain formatted 3D ASCII data (=0)
or unformatted binary header + 3D data (=1) ?
or ETSF binary (=2) ?
Answer: 1
Question 3: What is your choice ?

Answer: 5 (3D formmated data)
Question 4: Enter the name of an output file:
Answer: refden.in
Question 5: More analysis of the 3D file?
Answer: 0 (No).

A script is provided to setup a system as described above. The script is in

Example/1_ABINIT_BLPS/bcc/step1.sh

3) ABINIT-BLPS computations in four steps:

   a. Create a new directory (suggested).

   b. Prepare the *param.in* file that is an extra input file of ABINIT-BLPS in the current working directory as:

```
1       loadDen
0       LoadVr
100     Maxcount
0       bKEDFcc
0       bAtomV
1       bAllowsym
1.0e-3  nontmpTol
BCC     str
1.0e-3  stoptol
1.0e-5  penLambda
1       bspin
```

      Explanation for each parameter is provided in Appendix A.

   c. Use the same input files of ABINIT from step (2) and **a new trial local pseudopotential file named *tmp.lps*. The trial local pseudopotential should be generated by the user (see Appendix B).**

   d. Run the ABINIT-BLPS computations and obtain "*res_vion.dat",* which is the target BLPS with structure factors. Meanwhile, a file named "*invKS_log"* will be generated recording the optimization information.

A script to copy data from step 1 and then setup this calculation is provided:

Example/1_ABINIT_BLPS/bcc/step2.sh

4) Because the effective potential contains $S(q)$ and is stored in "*res_vion.dat"*. Follow the described steps to remove the structure factor $S(q)$ from the effective potential:

   a. Prepare the general ABINIT input files.

   b. Change the name of the "*res_vion.dat"* file to "*refpot.in"*.

c. Change the parameter *loadDen* in *param.in* from *1* to *0*.

d. Change the parameter *LoadVr* in *param.in* from *0* to *1*.

e. Change the parameter *bAtomV* in *param.in* to *1*.

f. Run the ABINIT-BLPS code and obtain the BLPS file "*c_vag.out*" without $S(q)$.

A script to copy data from step 2 and setup this calculation is provided:

Example/1_ABINIT_BLPS/bcc/step3.sh

5) Repeat the above steps (a) to (f) for all of the bulk structures until one gets all of the "*c_vag.out*" files.

6) Run the *step4.sh* script (in the Example/1_ABINIT_BLPS directory) to collect and sort all the data from *c_vag.out* files into a new file named '*BLPS.txt*'. Specifically, the *step4.sh* script will call the *sort_Vq.py* script to generate a '*raw_data.txt*' file. Note that at this step the $\frac{4\pi z}{q^2}$ term is added to the effective potential to cancel the $-\frac{4\pi z}{q^2}$ term in the effective potential.

7) Fitting procedures: Use the script '*Example/2_codeBLPS/fitting_BLPS.py*' to obtain the final BLPS (make sure the *"BLPS.txt"* file is in the same directory). The command is:

python fitting_BLPS.py Atom_Number N_e V(q=0) r_{cut}

where four parameters need to be input:

a. *"Atom_Number"* is the atomic number.

b. N_e is the valence electron number of the element.

c. $V(q=0)$ is a manually tuned value which affects BLPS at small q.

d. r_{cut} is a real-space radius cutoff beyond which $V_{eff}(r)$ becomes $-Z/r$ with Z being the number of valence electrons of the atom.

8) An effective potential (BLPS) is generated after step (7): the real space potential '*psp.rlpot*' is for ABINIT while the reciprocal potential '*psp.recpot*' is for PROFESS. Note that the $V_{nc}(q=0)$ and r_{cut} are chosen empirically. An extra step is needed to obtain the final *'psp.rlpot'* if non-linear core corrections are used (see Appendix B). We suggest the user first tune $V_{nc}(q=0)$ by setting the r_{cut} as a reasonable value between 4.0 and 7.0 Bohr (a.u.).

9) The final step is to validate the accuracy of the BLPS.

APPENDIX A. The 'param.in' File

Here are the detailed explanations for each parameter in the *'param.in'* file.

loadDen	Load density or not, should be set 1 when doing inverting-KS calculations.
LoadVr	Load potential or not, should be set to 1 when removing the structure factor $S(q)$ from the bulk-derived local potential.
Maxcount	Maximal bfgs iterations allowed
bKEDFcc	Usually set to 0; If set to 1, the code does nonlocal inverting on the density.
bAtomV	1: the code will remove the structure factors; 0: the code inverts the Kohn-Sham equation.
bAllowsym	1: the code allows symmetry (for bVatom=1 case), 0: the code does not allow symmetry.
nontmpTol	Tolerance of convergence of optimization when program decides to stop.
str	Which structure it is (This parameter is in the input file but performs no function at all actually, will be removed in future).
stoptol	Stopping tolerance for energy changes.
penLambda	Coefficients for the penalty function.
bspin	1: spin non-polarized case; 2: spin polarized case, where you need to provide two density files for spin up and down, respectively.

APPENDIX B. Trial Pseudopotentials and Non-Linear Core Corrections

A local pseudopotential file (LPS) is needed as a trial potential in order to perform Wu-Yang optimizations. One suggestion is to obtain this trial potential from a NLPS. In general, the format 6 (a special format described on ABINIT website) for ABINIT pseudopotentials generated by FHI98PP code is used for a NLPS and trail potential. However, ABINIT-BLPS generates a BLPS stored in format 8 (another special format described on ABINIT website) which utilizes an uniform real-space grid starting from zero. Detailed information for both formats of pseudopotentials can be found on the ABINIT website. Specifically, a NLPS can be simply transformed into a trial LPS by setting the parameters 'lmax' and 'lloc' (on the third line of a NLPS) to zero, then only a local potential and its associated wave function for a particular angular momentum channel are kept in the trial potential file. If necessary, non-linear core corrections (NLCC) can be included in the trial potential file; a positive 'fchrg' parameter indicates that NLCC is used.

After a BLPS is generated by ABINIT-BLPS, the real-space BLPS will be written into a new file with a uniform grid. Note that the NLCC will not be automatically written into the pseudopotential file even it is used in the trial potential. Thus, one needs to first cut the NLCC part from a NLPS file (NLCC.txt) and use a script to transform the NLCC to a uniform real-space grid (the script is provided in Example/3_codeNLCC/uniform_grid_NLCC.py). Note that this uniform grid should be exactly the same for both BLPS and NLCC in order to

be readable by ABINIT. The file that stores the NLCC for use in ABINIT is named 'ABACUS_NLCC.txt' while the other file named 'PROFESS_NLCC.txt' can be used in PROFESS. One also needs to paste the 'ABACUS_NLCC.txt' file into the BLPS file (for ABINIT), and update the information for the three parameters: 'rchrg, fchrg, qchrg' before testing a BLPS.

REFERENCE

[1] Q. Wu and W. T. Yang, J. Chem. Phys. **118**, 2498 (2003).

[2] B. J. Zhou, Y. A. Wang, and E. A. Carter, Physical Review B **69**, 125109 (2004).

[3] C. Huang and E. A. Carter, Phys Chem Chem Phys **10**, 7109 (2008).