

Tutorial on DFT+U

The projected density of states of NiO.

This tutorial aims at showing how to perform a DFT+U calculation using Abinit (see also [Amadon2008a] ([../theory/bibliography#amadon2008a](#)))

You will learn what is a DFT+U calculation and what are the main input variables controlling this type of calculation.

It is supposed that you already know how to do PAW calculations using ABINIT. Please follow the two tutorials on PAW in ABINIT (PAW1 ([../paw1](#)), PAW2 ([../paw2](#))), if this is not the case.

Important

All the necessary input files to run the examples can be found in the `~abinit/tests/` directory where `~abinit` is the absolute path of the abinit top-level directory.

To execute the tutorials, you are supposed to create a working directory (`Work*`) and copy there the input files and the *files* file of the lesson.

The *files* file ending with `_x` (e.g. *tbase1_x.files*) **must be edited** every time you start to use a new input file. You will discover more about the *files* file in section 1.1 ([../guide/abinit#intro](#)) of the help file.

To make things easier, we suggest to define some handy environment variables by executing the following lines in the terminal:

```
export ABI_HOME=Replace_with_the_absolute_path_to_the_abinit_top_level_dir

export ABI_TESTS=$ABI_HOME/tests/

export ABI_TUTORIAL=$ABI_TESTS/tutorial/           # Files for base1-2-3-4, GW ...
export ABI_TUTORESPFN=$ABI_TESTS/tutorespfn/       # Files specific to DFPT tutorials.
export ABI_TUTOPARAL=$ABI_TESTS/tutoparal/         # Tutorials about parallel version
export ABI_TUTOPLUGS=$ABI_TESTS/tutoplugs/         # Examples using external libraries.
export ABI_PSPDIR=$ABI_TESTS/Psps_for_tests/       # Pseudos used in examples.

export PATH=$ABI_HOME/src/98_main/:$PATH
```

The examples in this tutorial will use these shell variables so that one can easily copy and paste the code snippets into the terminal (**remember to set `ABI_HOME` first!**)

The last line adds the directory containing the executables to your `PATH`^[7] so that one can invoke the code by simply typing *abinit* in the terminal instead of providing the absolute path.

Finally, to run the examples in parallel with e.g. 2 MPI processes, use *mpirun* (*mpiexec*) and the syntax:

```
mpirun -n 2 abinit < files_file > log 2> err
```

The standard output of the application is redirected to `log` while `err` collects the standard error (runtime error messages, if any, are written here).

This tutorial should take about 1 hour to complete.

0 Short summary of the DFT+U method

The standard Local Density Approximation (LDA), where the exchange and correlation energy is fit to homogeneous electron gas results, is a functional that works well for a vast number of compounds. But, for some crystals, the interactions between electrons are so important that they cannot be represented by the LDA alone. Generally, these highly correlated materials contain rare-earth metals or transition metals, which have partially filled *d* or *f* bands and thus localized electrons.

The LDA tends to delocalize electrons over the crystal, and each electron feels an average of the Coulombic potential. For highly correlated materials, the large Coulombic repulsion between localized electrons might not be well represented by a functional such as the LDA. A way to avoid this problem is to add a Hubbard-like, localised term, to the LDA density functional. This approach is known as LDA+U (actually DFT+U). In the actual implementation, we separate localized *d* or *f* electrons, on which the Hubbard term will act, from the delocalized ones (*s* and *p* electrons). The latter are correctly described by the usual LDA calculation. In order to avoid the double counting of the correlation part for localized electrons (already included in the LDA, although in an average manner), another term - called the double-counting correction - is subtracted from the Hamiltonian.

In Abinit, two double-counting corrections are currently implemented:

-The Full localized limit (FLL) [*Liechtenstein1995*] (*../theory/bibliography#liechtenstein1995*)
(*usepawu* (*../variables/paw#usepawu*)=1)

-The Around Mean Field (AMF) [Czyzyk1994] ([../theory/bibliography#czyzyk1994](#)) (usepawu ([../variables/paw#usepawu](#))=2)

For some systems, the result might depend on the choice of the double-counting method. However, the two methods generally give similar results.

1 Ground state calculation of NiO using LDA

Before continuing, you might consider to work in a different subdirectory as for the other tutorials. Why not Work_dftu? In what follows, the names of files will be mentioned as if you were in this subdirectory.

Copy the files *tdftu_1.in* and *tdftu_x.files* from *\$ABI_TUTORIAL/Input* to your *Work_dftu* directory with:

```
cd $ABI_TUTORIAL/Input
mkdir Work_dftu
cd Work_dftu
cp ../tdftu_x.files . # You will need to edit this file.
cp ../tdftu_1.in .
```



View tests/tutorial/Input/tdftu_x.files



View tests/tutorial/Input/tdftu_1.in

Now run the code as usual. The job should take less than 30 seconds on a PC 3 GHz. It calculates the LDA ground state of the NiO crystal. A low cutoff and a small number of k-points are used in order to speed up the calculation. During this time you can take a look at the input file.

The NiO crystallizes in the rocksalt structure, with one Ni and one O atom in the primitive cell (the crystallographic primitive cell). However, NiO is known to exhibit an antiferromagnetic ordering at low temperature (along the $\langle 111 \rangle$ direction). From the electronic point of view, the true unit cell has two Ni and two O atoms: the local magnetic moment around the first Ni atom will have a sign opposite to the one of the other Ni atom.

You should take some time to examine the values used for the input variables *xred* ([../variables/basic#xred](#)), *rprim* ([../variables/basic#rprim](#)) (note the last line!), *typat* ([../variables/basic#typat](#)), *spinat* ([../variables/gstate#spinat](#)), *nsppol*

(../../../../variables/basic#nspol), and nspden (../../../../variables/gstate#nspden), that define this antiferromagnetic ordering along the $\langle 111 \rangle$ direction (of a conventional cubic cell).

If you take a look at the output file (tdftu_1.out), you can see the integrated total density in the PAW spheres (see the PAW1 (../paw1) and PAW2 (../paw2) tutorials on PAW formalism). This value roughly estimate the magnetic moment of NiO:

Integrated total density in atomic spheres:

| Atom | Sphere radius | Integrated_up_density | Integrated_dn_density | Total(up+dn) |
|------|---------------|-----------------------|-----------------------|--------------|
| 1 | 2.30000 | 9.05536980 | 7.85243738 | 16.9078 |
| 0718 | 1.20293241 | | | |
| 2 | 2.30000 | 7.85243738 | 9.05536980 | 16.9078 |
| 0718 | -1.20293241 | | | |
| 3 | 1.21105 | 1.82716080 | 1.82716080 | 3.6543 |
| 2159 | -0.00000000 | | | |
| 4 | 1.21105 | 1.82716080 | 1.82716080 | 3.6543 |
| 2159 | 0.00000000 | | | |

Note: Diff(up-dn) can be considered as a rough approximation of a local magnetic moment.

The atoms in the output file, are listed as in the `typat` (../../../../variables/basic#typat) variable (the first two are nickel atoms and the last two are oxygen atoms). The results indicate that spins are located in each nickel atom of the doubled primitive cell. Fortunately, the LDA succeeds to give an antiferromagnetic ground state for the NiO. But the result does not agree with the experimental data.

The magnetic moment (the difference between up and down spin on the nickel atom) range around 1.6-1.9 according to experiments ([Cheetham1983] (../../../../theory/bibliography#cheetham1983), [Neubeck1999] (../../../../theory/bibliography#neubeck1999), [Sawatzky1984] (../../../../theory/bibliography#sawatzky1984), [Hufner1984] (../../../../theory/bibliography#hufner1984)) Also, as the Fermi level is at 0.22347 Ha, one can see that the band gap obtained between the last occupied (0.20672 Ha, at k point 2) and the first unoccupied band (0.23642 Ha, at kpoint 3) is approximately 0.8 eV which is lower than the measured value of 4.0-4.3 eV (This value could be modified using well-converged parameters but would still be much lower than what is expected).

Making abstraction of the effect of insufficiently convergence parameters, the reason for the discrepancy between the DFT-LDA data and the experiments is first the fact the DFT is a theory for the ground state and second, the lack of correlation of the LDA. Alone, the homogeneous electron gas cannot correctly represent the interactions among d electrons of the Ni atom. That is why we want to improve our functional, and be able to manage the strong correlation in NiO.

2 DFT+U with the FLL double-counting

As seen previously, the LDA does not gives good results for the magnetization and band gap compared to experiments. At this stage, we will try to improve the correspondence between calculation and experimental data. First, we will use the DFT(LDA)+U with the Full localized limit (FLL) double-counting method.

FLL and AMF double-counting expressions are given in the papers listed above, and use the adequate number of electrons for each spin. For the Hubbard term, the rotationally invariant interaction is used.

Note

It is important to notice that in order to use LDA+U in Abinit, you must employ PAW pseudopotentials.

You should run abinit with the *tdftu_2.in* input file. This calculation takes less than 30 seconds on a PC 3.0 GHz During the calculation, you can take a look at the input file.



[View tests/tutorial/Input/tdftu_2.in](#)



Some variable describing the LDA+U parameters have been added to the previous file. All other parameters were kept constant from the preceding calculation. First, you must set the variable `usepawu` (`../../variables/paw#usepawu`) to one (for the FLL method) and two (for the AMT method) in order to enable the LDA+U calculation. Then, with `lpawu` (`../../variables/paw#lpawu`) you give for each atomic species (`znuc1` (`../../variables/basic#znuc1`)) the values of angular momentum (*l*) for which the LDA+U correction will be applied. The choices are 2 for d-orbitals and 3 for *f*-orbitals. You cannot treat

s and p orbitals with LDA+U in the present version of ABINIT. Also, if you do not want to apply LDA+U correction on a species, you can set the variable to -1. For the case of NiO, we put `lpawu` (`../..../variables/paw#lpawu`) to 2 for Ni and -1 for O.

Finally, as described in the article cited above for FLL and AMF, we must define the screened Coulomb interaction between electrons that are treated in LDA+U, with the help of the variable `upawu` (`../..../variables/paw#upawu`) and the screened exchange interaction, with `jpawu` (`../..../variables/paw#jpawu`). Note that you can choose the energy unit by indicating at the end of the line the unit abbreviation (e.g. eV or Ha). For NiO, we will use variables that are generally accepted for this type of compound:

```
upawu 8.0 eV
jpawu 0.8 eV
```

You can take a look at the result of the calculation. The magnetic moment is now:

```
Integrated total density in atomic spheres:
-----
Atom  Sphere radius  Integrated_up_density  Integrated_dn_density  Total(up
+dn)  Diff(up-dn)
  1      2.30000      9.28514439           7.53721910      16.8223
6349   1.74792528
  2      2.30000      7.53721910           9.28514439      16.8223
6349  -1.74792528
  3      1.21105      1.84896670           1.84896670       3.6979
3339   0.00000000
  4      1.21105      1.84896670           1.84896670       3.6979
3339   0.00000000
Note: Diff(up-dn) can be considered as a rough approximation of a local mag
netic moment.
```

NiO is found antiferromagnetic, with a moment that is in reasonable agreement with experimental results. Moreover, the system is a large gap insulator with about 5.0 eV band gap (the 24th band at k point 3 has an eigenenergy of 0.15896 Ha, much lower than the eigenenergy of the 25th band at k point 1, namely 0.24296 Ha). This number is very approximative, since the very rough sampling of k points is not really appropriate to evaluate a band gap, still one obtains the right physics.

A word of caution is in order here. It is NOT the case that one obtain systematically a good result with the LDA+U method at the first trial. Indeed, due to the nature of the modification of the energy functional, the landscape of this energy functional might present numerous local minima.

Unlike LDA+U, for the simple LDA (without U), in the non-spin-polarized case, there is usually only one minimum, that is the global minimum. So, if it converges, the self-consistency algorithm always find the same solution, namely, the global minimum. This is already not true in the case of spin- polarized calculations (where there might be several stable solutions of the SCF cycles, like ferromagnetic and antiferromagnetic), but usually, there are not many local minima, and the use of the `spinat` (`../../../../variables/gstate#spinat`) input variables allows one to adequately select the global physical characteristics of the sought solution.

By contrast, with the U, the `spinat` (`../../../../variables/gstate#spinat`) input variable is too primitive, and one needs to be able to initialize a spin-density matrix on each atomic site where a U is present, in order to guide the SCF algorithm.

The fact that `spinat` (`../../../../variables/gstate#spinat`) works for NiO comes from the relative simplicity of this system.

3 Initialization of the density matrix

You should begin by running the `tdftu_3.in` file before continuing.

In order to help the LDA+U find the ground state, you can define the initial density matrix for correlated orbitals with `dmatpawu` (`../../../../variables/paw#dmatpawu`) To enable this feature, `usedmatpu` (`../../../../variables/paw#usedmatpu`) must be set to a non-zero value (default is 0). When positive, the density matrix is kept to the `dmatpawu` (`../../../../variables/paw#dmatpawu`) value for the `usedmatpu` (`../../../../variables/paw#usedmatpu`) value steps. For our calculation(`tdftu_3.in`), `usedmatpu` (`../../../../variables/paw#usedmatpu`) is 5, thus the spin-density matrix is kept constant for 5 SCF steps.



[View tests/tutorial/Input/tdftu_3.in](#)

In the log file (not the usual output file), you will find for each step, the calculated density matrix, followed by the imposed density matrix. After the first 5 SCF steps, the initial density matrix is no longer imposed. Here is a section of the log file, in which the imposed occupation

matrices are echoed:

```
-----
Occupation matrix for correlated orbitals is kept constant
and equal to initial one !
-----
```

```
== Atom    1 == Imposed occupation matrix for spin 1 ==
  0.90036    0.00000   -0.00003    0.00000    0.00000
  0.00000    0.90036   -0.00001    0.00000    0.00002
 -0.00003   -0.00001    0.91309   -0.00001    0.00000
  0.00000    0.00000   -0.00001    0.90036   -0.00002
  0.00000    0.00002    0.00000   -0.00002    0.91309
```

```
== Atom    1 == Imposed occupation matrix for spin 2 ==
  0.89677   -0.00001    0.00011   -0.00001    0.00000
 -0.00001    0.89677    0.00006    0.00001   -0.00010
  0.00011    0.00006    0.11580    0.00006    0.00000
 -0.00001    0.00001    0.00006    0.89677    0.00010
  0.00000   -0.00010    0.00000    0.00010    0.11580
```

```
== Atom    2 == Imposed occupation matrix for spin 1 ==
  0.89677   -0.00001    0.00011   -0.00001    0.00000
 -0.00001    0.89677    0.00006    0.00001   -0.00010
  0.00011    0.00006    0.11580    0.00006    0.00000
 -0.00001    0.00001    0.00006    0.89677    0.00010
  0.00000   -0.00010    0.00000    0.00010    0.11580
```

```
== Atom    2 == Imposed occupation matrix for spin 2 ==
  0.90036    0.00000   -0.00003    0.00000    0.00000
  0.00000    0.90036   -0.00001    0.00000    0.00002
 -0.00003   -0.00001    0.91309   -0.00001    0.00000
  0.00000    0.00000   -0.00001    0.90036   -0.00002
  0.00000    0.00002    0.00000   -0.00002    0.91309
```

Generally, the LDA+U functional meets the problem of multiple local minima, much more than the usual LDA or GGA functionals. One often gets trapped in a local minimum. Trying different starting points might be important...

4 AMF double-counting method

Now we will use the other implementation for the double-counting term in LDA+U (in Abinit), known as AMF. As the FLL method, this method uses the number of electrons for each spin independently and the complete interactions $U(m_1, m_2, m_3, m_4)$ and $J(m_1, m_2, m_3, m_4)$.

As in the preceding run, we will start with a fixed density matrix for d orbitals. You might now start your calculation, with the *tdftu_4.in* and *tdftu_4.files*, or skip the calculation, and rely on the reference file provided in the *\$ABI_TUTORIAL/Refs* directory. Examine the *tdftu_4.in* file.



View tests/tutorial/Input/tdftu_4.in

The only difference in the input file compared to *tdftu_3.in* is the value of *usepawu* (*../../variables/paw#usepawu*) = 2. We obtain a band gap of 4.3 eV. The value of the band gap with AMF and FLL is different. However, we have to remember that these results are not well converged. By contrast, the magnetization,

| Atom | Sphere radius | Integrated_up_density | Integrated_dn_density | Total(up+dn) |
|------|---------------|-----------------------|-----------------------|--------------|
| 1 | 2.30000 | 9.24026835 | 7.56013140 | 16.8003 |
| 9975 | 1.68013694 | | | |
| 2 | 2.30000 | 7.56013140 | 9.24026835 | 16.8003 |
| 9975 | -1.68013694 | | | |
| 3 | 1.21105 | 1.84683993 | 1.84683993 | 3.6936 |
| 7986 | -0.00000000 | | | |
| 4 | 1.21105 | 1.84683993 | 1.84683993 | 3.6936 |
| 7986 | 0.00000000 | | | |

Note: Diff(up-dn) can be considered as a rough approximation of a local magnetic moment.

is very similar to the LDA+U FLL. In fact, this system is not very complex. But for other systems, the difference can be more important. FLL is designed to work well for crystal with diagonal occupation matrix with 0 or 1 for each spin. The AMF should be used when orbital occupations are near the average occupancies.

5 Projected density of states in LDA+U

Using *prtdos* (*../../variables/files#prtdos*) 3, you can now compute the projected d and f density of states. For more information about projected density of states, for more details see the PAW1 (*../paw1*) tutorial.

