Task: LSDA+U

October 23, 2009

Evaluate the LSDA+U fat-bands for the upper and lower Hubbard band in the infinite stacking CaCuO$_2$ model cuprate, time 1:30
This task aims at training the use of LSDA+U and of the fat-band tools.

Hint: Setup the SCF calculations first, before starting to do the band analysis.
Subtasks:

- **The non magnetic LDA calculation**

    1. Setup and run the LDA calculation for the non-magnetic CaCuO$_2$. **calculation time ≈ 2 min**

        symmetry menu
        - space group P4/mmm (123)
            * lattice constants $a_0 = 3.86$ Å $b_0 = a_0$ $c_0 = 3.20$ Å **DO NOT FORGET TO SET THE UNITS TO ÅNGSTRÖM!**
        - Ca at $\frac{1}{2}\frac{1}{2}\frac{1}{2}$
        - Cu at 000
        - O at $\frac{1}{2}00$

        main menu
        - number of k-points = 6 6 6 (This is done to save time in the tutorial. In a real situation one would check k-point convergence.)

        iteration submenu
        - Version: LCIterat

    2. While running, in another window (xterm or similar): open `bravais.ps` in `gv` (ghostview) and get an idea of the unit cell.

    3. Make a list of Wyckoff positions and atomic sites. (search output from top for the string 'UNIT CELL') and make sure that you understand the site numbers and the sort numbers (Wyckoff positions).

    4. When the calculation is converged, calculate the band structure (output file `+band`), bandweights/fatbands (output file `+bweights`)
        To set up the creation of the band-data set in `fedit8.65-32`:

        Bandplot submenu
        - bandstructure plot: ON
        - Weights: ON

        and re-run fplo.

    5. Extract the fat band for the Cu $3d_{x^2-y^2}$ ($m_l = +2$) orbital using `addweights` (use prefactor 1.)

6. Plot fat bands using `fedit8.65-32...  -bandplot`, make sure that you use `+bwsum` instead of `+bweights` as the file to read the weights from (hotkey `WF`), don't forget to set the plot type to bandweights.

7. Visualize the lattice structure. Draw a hand sketch of the CuO_2-plane with the Cu $3d_{x^2-y^2}$ orbital (pointing towards the oxygen).

- **The Anti-ferromagnetic (AFM) LSDA+U calculation**

  1. Evaluate the LSDA+U solution for the anti-ferromagnetic **$CaCuO_2$. calculation time $\approx$ 20 min**

     symmetry menu
     - space group I4/mmm (139)
     - lattice constants $\sqrt{2}a_0$, $\sqrt{2}a_0$, $2c_0$ ... **DO NOT FORGET THE UNITS!**
     - Ca at $0\frac{1}{2}\frac{1}{4}$
     - Cu1 at 000
     - Cu2 at $\frac{1}{2}\frac{1}{2}0$
     - O at $\frac{1}{4}\frac{1}{4}0$

     main menu
     - number of k-points = 6 6 6
     - spin polarized
     - initial spin split switched on!
     - fixed spin moment with zero spin moment (for increased stability)

     iteration submenu
     - Version: LCIterat

     initial spinsplit submenu
     - initial spin split Cu1 +1, Cu2 -1, Ca 0, O 0

     LSDA+U submenu
     - make LSDA+U: ON
     - around mean field functional
     - number of correlated orbitals: 2 ($3d$ on both Wyckoff positions of Cu)
     - def1: Wyckoff position 2, state 3d, $U = 8$ eV, $F_2 = 8.5$ eV, $F_4 = 5.5$ eV, $F_6 = 0$ eV
     - def2: Wyckoff position 3, otherwise same values as def1

        Remark: The fundamental quantities of the LSDA+U implementation are the Slater integrals F_0 to F_6. The first is simply $F_0 = U$. The other three form J, depending on the angular momentum quantum number of the shell.

        s-states: $J = 0$
        p-states: $J = \frac{1}{5}F_2$
        d-states: $J = \frac{1}{14}(F_2 + F_4)$
        f-states: $J = \frac{1}{6435}(286F_2 + 195F_4 + 250F_6)$

        The choice of parameter is quite simple, since one can take $\frac{F_4}{F_2}$ from atomic situations, since it is a pretty local quantity. For $3d$-electrons the ratio is of the order of $0.6\ldots0.75$ and a pratically good choice is $\frac{F_4}{F_2} = \frac{5.5}{8.5}$ since $8.5 + 5.5 = 14$, thus $F_2 = 8.5J, \quad F_4 = 5.5J$.

  2. Make a list of Wyckoff positions and atomic sites. (search output from top for the string 'UNIT CELL') and make sure that you understand the site numbers and the sort numbers (Wyckoff positions). Get an idea of the unit cell. Beware of the body centering.

3. Visualize the lattice structure including site numbers. Draw a hand sketch of the CuO_2-plane with the Cu $3d_{x^2-y^2}$ orbital (pointing towards the oxygen).

4. When calculation converged, proceed as follows ...

5. Create band structure and fat band data...at the same time...create the local projected DOS (LDOS) for the first Cu site. In fedit:

   main menu
   - switch off 'initial spinsplit'

   bandplot submenu
   - set switches accordingly as before
   - Use rotation of quantization axis: new x-axis=(110).
     Do not forget to switch on 'transform axis'
   - List of special symmetry points:
     * No. of symmetry points: 8

Γ	$0,0,0$
X	$0,\frac{1}{2},0$
M	$\frac{1}{2},\frac{1}{2},0$
Γ	$0,0,0$
Z	$0,0,z$
R	$0,\frac{1}{2},z$
A	$\frac{1}{2},\frac{1}{2},z$
Z	$0,0,z$

     $z = 0.853$
   - set LDOS site list accordingly (note that this list is in general different from the Wyckoff positions)

   after setting all options/flags/data re-run fplo.

6. Draw a picture to understand why we do the rotation (in the rotated frame the $x^2 - y^2$ orbital again points towards the oxygen.)

7. Plot the fat bands for the first Cu site. Use red color for the spin down channel. Identify the local DOS corresponding to the Cu $3d_{x^2-y^2}$ orbitals. (files are named '+ldos....', have a look at the comments in the files)

- **The Ferromagnetic (FM) LSDA+U calculation**

  1. Evaluate the LSDA+U solution for the ferromagnetic $CaCuO_2$ in the same settings as the AFM cell. **calculation time $\approx$ 20 min.**
     Remark: We use the same (larger) unit cell for the FM calculation as for the AFM calculation, which sounds wastefull (and even can invite numerical noice/instable convergence). The main reason is that we want to compare total energies and two different cells differ by their orientation, which leads to small differences in results steming from numerical artifacts like differently oriented finite integration meshes. Although these differences are small in general they still might be of the order of the resulting physical energies and that's why comparing energies is best done in the same setup!

  2. Input: copy the AFM directory to a new (FM) directory. Start from scratch by deleting the files =.dens and =.dmat_init. Remove all old output files: `rm +*`
     - same settings as before except for

     initial spinsplit submenu
     - initial spin split Cu1 +1, Cu2 +1, Ca 0, O 0

     main menu
     - **switch off fixed spin moment!!!**

3

- **make sure the initial spin split is on!**

bandplot menu

- switch off `bandstructure plot` and `weights`

3. Use the mean-field energy of the Heisenberg model $E = \frac{1}{2}J\sum_{ij}\vec{S}_i\vec{S}_j$ to estimate the energy difference between the ferro- and anti-ferro-magnetic state of the CuO_2 plane. Consider nearest neighbor terms only. Compare that to the LSDA+U total energy differences and thus calculate J. (Hint: mean-field means to replace the vector operator by its expectation value. Consider the spin pattern of the plane in the two states and calculate the sum per Cu atom.)

Hints:

- If you forget to set the units to Ångström, the occupation numbers (population analysis) will be messed up. If you do this mistake, stop the calculation and remove the files =.dens and =.dmat_init (if existing) and start over with the corrected settings.

- Use different directories for different input settings.

- Check each calculation for convergence of the iteration and for reasonable population numbers.

- Time estimates suppose that one calculation per processor at a time is running.

- For the cuprates the AMF double counting is not very much different from the Atomic limit double counting, however, the two methods will give somewhat different values for $E_{AMF} - E_{FM}$, if the same U is used. This is a quite general situation; the physical quantities depend on U. In practical applications, one has to study this dependence and make educated choices. There are methods around, which calculate U. The problem with this is that there is no direct connection between these calculated values and their application in an LSDA+U method. There are issues with the projection of the correlated orbitals and even more fundamental ones. (At least that's our heretic opinion.)

Task: LSDA+U
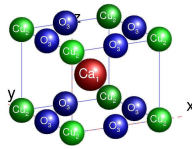# Answers

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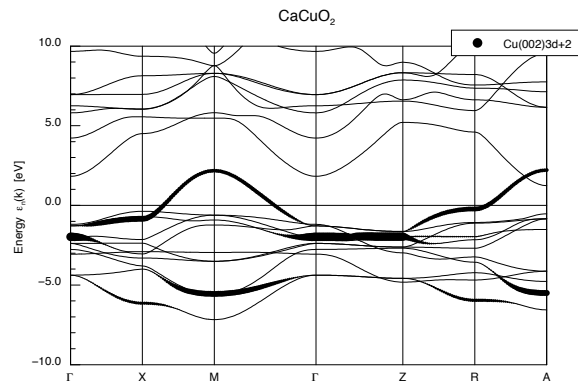
1.

Figure 1: Unit cell for non-magnetic LDA setup

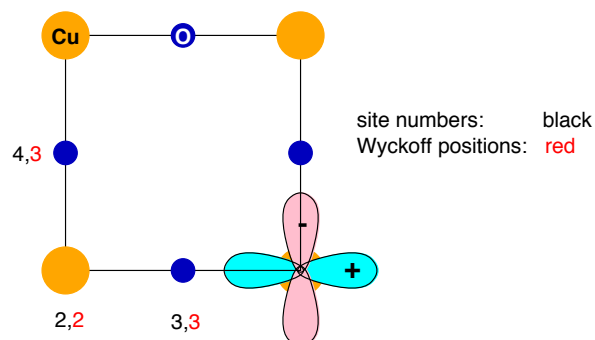2.

Figure 2: Resulting non-magnetic LDA fat bands

3.

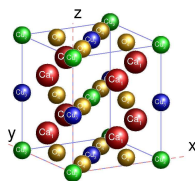Figure 3: non-magnetic CuO$_2$-plane

4.

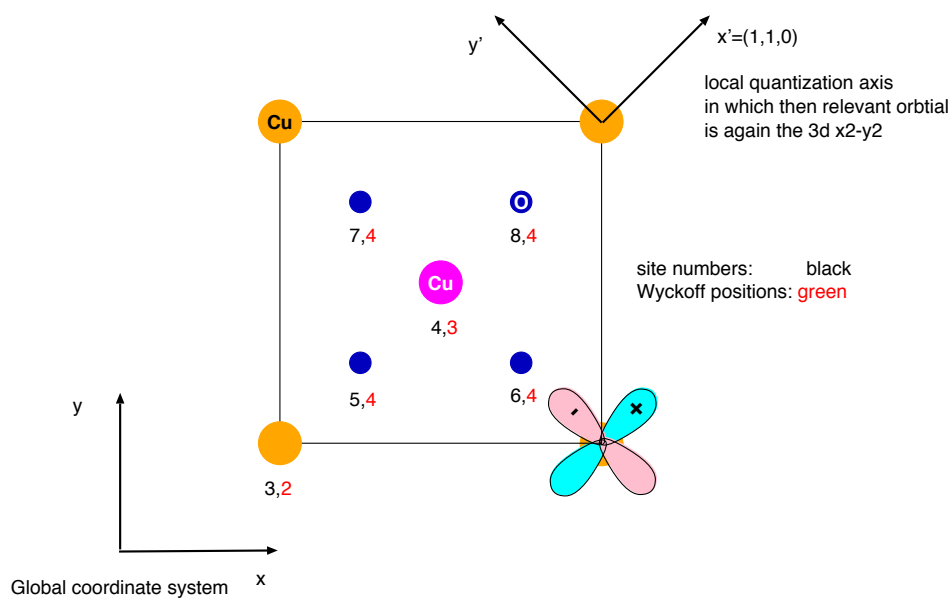Figure 4: Unit cell for AFM LDA+U setup
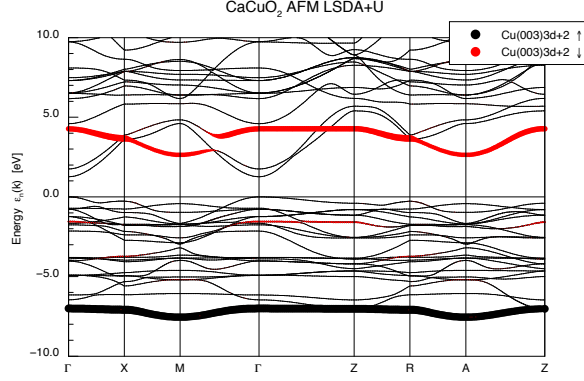
5.

Figure 5: AFM CuO$_2$-plane
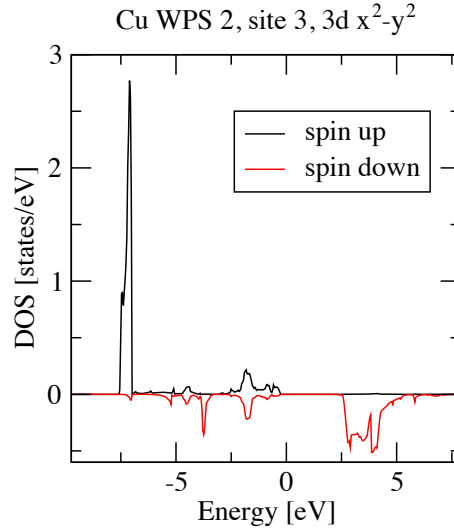
6.

Figure 6: Resulting AFM LDA+U fat bands

7.

Figure 7: LDOS Cu $3d\ x_2 - y_2$, AFM LDA+U

8. $E = \frac{1}{2}J\sum_{ij}\vec{S}_i\vec{S}_j$, each copper has four copper neighbours with spin $\frac{1}{2}$ alined along the z-axis. For the anti-ferromagnetic solution the spins are antiparallel, $E^{AFM} = \frac{1}{2}J\cdot4\cdot\left(-\frac{1}{2}\cdot\frac{1}{2}\right) = -\frac{J}{2}$, and for the ferromagnetic solution the spins are parallel, $E^{FM} = \frac{1}{2}J\cdot4\cdot\left(\frac{1}{2}\cdot\frac{1}{2}\right) = \frac{J}{2}$. Thus,

$$E^{FM} - E^{AFM} = J,$$

Numerically we get: $E^{AFM} = -4962.0984$ Ha, $E^{FM} = -4962.0897$ Ha. These are the energies for 2 copper sites. The model energy is for one site. So we devide the energy difference by two! $J = 4.3$ mHa $= 117$ meV.