

Orbital package in WIEN

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1 Description of SRC_orb package

Orbital package calculates the orbitally dependent potentials V_{orb} i.e. potentials which are nonzero in the atomic spheres only and they depend on the orbital state numbers l, m . In the present version the potential is assumed to be independent of the radius vector. V_{orb} acts on specified atoms and specified orbital numbers l only, these are given in the input. Three different potentials are implemented in this package:

- LDA+U. There are three variants of this method, two of them are discussed in [1]:
 1. LDA+U(SIC) - introduced by Anisimov et al. [2]. As discussed in this reference the method approximately corrects the calculation for the self-interaction. The center of gravity of $|l, m_l, m_s \rangle$ states is changed.
 2. LDA+U(AMF) - introduced by Czyzyk and Sawatzky [3] as 'Around the Mean Field' method. In [1] it is denoted as LDA+U(DFT). This version has an advantage that it reduces to LSDA if the occupation of the $|l, m_l, m_s \rangle$ states are independent of m_l . Center of gravity of $|l, m_l, +1/2 \rangle$ is preserved as well as center of gravity of $|l, m_l, -1/2 \rangle$ states, i.e. average exchange splitting is the same as in LSDA.
 3. LDA+U(HMF) - in addition the mean field approximation to the Hubbard model, as introduced by Anisimov et al. [4] is also implemented. Note, however, that it is to be used with LDA (not LSDA) exchange-correlation potential in the spin polarized calculations. Only the center of gravity of all $|l, m_l, 1/2 \rangle, |l, m_l, -1/2 \rangle$ states is preserved. The exchange splitting arises solely due to the presence of LDA+U(HMF) potential.

All variants are implemented in the rotationally invariant way [6].

- Orbital polarization. The additional potential has the form [7]:

$$V_{OP} = c_{OP} \langle L_z \rangle l_z \quad (1)$$

where c_{OP} is the orbital polarization parameter, $\langle L_z \rangle$ is projection of the orbital momentum on the magnetization direction and l_z is single electron orbital momentum component z parallel to \vec{M} .

- Interaction with the external magnetic field. In this case the additional potential has a simple form:

$$V_{B_{ext}} = \mu_B \vec{B}_{ext} (\vec{l} + 2\vec{s}). \quad (2)$$

The interaction with the electronic spin is taken into account by shifting in LAPW0 the spin up and spin down exchange correlation potentials by the energy $+\mu_B B_{ext}, -\mu_B B_{ext}$, respectively. The interaction of B_{ext} with spin could be as well calculated using the 'Fixed spin moment' method. For interaction with the orbital momentum it is necessary to specify atoms and angular momentum numbers for which this interaction will be considered.

In all cases the resulting potential for given atom and orbital number l is the Hermitian, $(2l+1) \times (2l+1)$ matrix. In general this matrix is complex, but in special cases it may be real.

1.1 Contribution to the total energy

For the LDA+U and orbital polarization methods a correction to the total energy appears

$$E_{corr} = E_{orb} - Tr(V_{orb}, \hat{n}) \quad (3)$$

where E_{corr} is the energy correction, E_{orb} is LDA+U or OP energy (e.g. [6]), V_{orb} represents LDA+U or OP potential and \hat{n} is the density matrix. If V_{orb} is calculated separately for spin up and spin down, the above expression holds for both direction of spin:

$$E_{corr}^{\sigma} = E_{orb}^{\sigma} - Tr(V_{orb}^{\sigma}, \hat{n}^{\sigma}) \quad (4)$$

In above expressions the fact that in WIEN instead of calculating the contribution of the kinetic energy to E_{tot} this contribution is expressed through the sum over the eigenvalues of occupied states was taken into account. Providing that E_{orb} is quadratic form in the density matrix (OP, LDA+ U^{AMF} , LDA+ U^{HMF} , but not LDA+ U^{SIC}) the above expressions may be simplified:

$$E_{corr} = -\frac{1}{2}Tr(V_{orb}, \hat{n}) \quad (5)$$

Note that analogous correction applies also for the Hartree term in energy [9]:

$$E_{Hartree} = -\frac{1}{2} \int d\vec{r} \rho(\vec{r}) V_{Hartree} \quad (6)$$

2 Structure of input data

2.1 Input for all potentials

With the exception of 2nd line input uses the free format.

1st line

nmod,natorb,ipr

nmod defines the type of potential 1...LDA+U, 2...OP, 3... B_{ext}

natorb is number of atoms for which orbital potential V_{orb} is calculated

ipr is printing option, the larger is ipr, the longer is output

2nd line

chmix, amix, format(A5,F8.2)

chmix equals either to PRATT or to BROYPD and it determines whether the mixing of V_{orb} follows Pratt or Broyden scheme (analog of MIXER)

amix is coefficient for the mixing.

3rd line repeated natorb-times

iatom(i),nlorb(i),(lorb(li,i),li=1,nlorb(i))

iatom - index of atom in struct file

nlorb number of orbital moments for which V_{orb} will be applied

lorb .. orbital numbers repeated nlorb-times

2.2 Input for LDA+U (nmod=1)

4th line

nsic defines 'double counting correction'

- nsic=0 'AMF method' [3]
- nsic=1 'SIC method' [2, 6]
- nsic=2 'HMF method' [4]

5th line repeated natorb-times, for each natorb repeated nlorb-times

U(li,i), J(li,i)

U(li,i), J(li,i) are Coulomb and exchange parameters for LDA+U in Ry for atom type i and for orbital number li

Example of the input file for NiO (LDA+U included for two inequivalent Ni atoms that have indexes 1 and 2 in the structure file):

```

1 2 0          nmod, natorb, ipr
BROYD 0.3      Broyden mixing used with mixing parameter 0.3
1 1 2          iatom nlorb, lorb
2 1 2          iatom nlorb, lorb
1              nsic (LDA+U(SIC) used)
0.59 0.07     U J
0.59 0.07     U J

```

2.3 Input for Orbital Polarization (nmod=2)

4th line

nmodop defines mode of 'OP'

- nmodop=1 (preferred version) - average L_z taken separately for spin up, spin down [8]. The potential is then spin-dependent.
- nmodop=0 - average L_z is the sum for spin up and spin down.

5th line repeated natorb-times

Ncalc(i)

- Ncalc=1 Orb.pol. parameters are calculated ab-initio
- Ncalc=0 Orb.pol. parameters are read from input.

6th line (only if Ncalc=0, then repeated nlorb-times)

pop(li,i) OP parameter in Ry

7th line

xms(1), xms(2), xms(3) direction of magnetization expressed in terms of lattice vectors

Example of the input file for NiO (total $\langle L_z \rangle$ used in (1), OP parameters calculated ab-initio, \vec{M} along [001]):

```

2 2 0          nmod, natorb, ipr, amix
BROYD 0.3      Broyden mixing used with mixing parameter 0.3
1 1 2          iatom nlorb, lorb
2 1 2          iatom nlorb, lorb
0              nmodop
1              Ncalc
1              Ncalc
0. 0. 1.      direction of M in terms of lattice vectors

```

2.4 Input for interaction with B_{ext} (nmod=3)

4th line

B_{ext} external field in Tesla

5th line

xms(1), xms(2), xms(3) direction of \vec{B}_{ext} expressed in terms of lattice vectors

Example of the input file for NiO, ($B_{ext}= 4$ T, along [001]):

```
3 2 0          nmod, natorb, ipr, amix
BROYD 0.3      Broyden mixing used with mixing parameter 0.3
1 1 2          iatom nlorb, lorb
2 1 2          iatom nlorb, lorb
4.            Bext in T
0. 0. 1.      direction of Bext in terms of lattice vectors
```

2.5 Input and output files

In addition to the input data case.inorb, described above, the orbital package needs as input files the density matrices calculated by the LAPWDM program:

case.dmatup, case.dmatdn (eventually case.dmatupdn if $\langle l, m_l, \uparrow | V_{orb} | l, m_l', \downarrow \rangle$ matrix is needed in the LDA+U calculation).

For orbital polarization, and in future also for the LDA+U, the calculation of the Slater parameters F_k is necessary, then Ncalc=1. For this the radial part $u_l(r, E_l)$ of the atomic functions must be known. The energy E_l at which they are calculated is read from corresponding case.scflup or case.scf1dn files. For the same reason the case.vspup, case.vspdn are needed.

The result of the calculation is written in

case.vorbup, case.vorbdn.

3 Calculation and use of the orbital potential

The command to calculate the orbital potential is:

x orb -up (-dn)

or

orb uporb.def (dnorb.def)

In addition the part of V_{orb} which is nondiagonal in spin may be calculated by:

orb dnuorb.def

where dnuorb.def is:

```
5,'case.inorb',      'old',      'formatted',0
6,'case.outdnup',   'unknown', 'formatted',0
11,'case.dmatupdn',  'unknown', 'formatted',0
12,'case.vorbdnup',  'unknown', 'formatted',0
13,'case.vorbdnup_old', 'unknown', 'formatted',0
14,'case.scflup',    'unknown', 'formatted',0
18,'case.vspup',     'unknown', 'formatted',0
20,'case.struct',    'old',      'formatted',0
21,'case.scforbdnup', 'unknown', 'formatted',0
31,'case.br1orbdnup', 'unknown', 'unformatted',0
```

32, 'case.br2orbdnup', 'unknown', 'unformatted', 0

input data case.inorb are unchanged. File case.dmatupd is the $\langle \uparrow || \downarrow \rangle$ block of the occupation matrix as calculated in LAPWDM. Results are in case.vorbdnup.

In most cases the orbital potential is used in spin-polarized calculations with spin-orbit coupling switched on, so that the orbital momentum is nonzero. To use orbital potential in non spin-polarized calculations or in spin-polarized ones, but without the s-o coupling, makes sense for LDA+U method only. In the latter case ($l_z=0$) the symmetry of system is preserved in presence of the orbital potential (in particular spin and direct space remain decoupled in the spin-polarized systems). The most effective approach is then to add V_{orb} to the Hamiltonian in LAPW1.

3.1 Spin-polarized calculations with spin-orbit coupling

The situation is more complicated in spin-polarized calculations with spin-orbit coupling, as the simultaneous presence of the s-o coupling and magnetization leads to the reduction of the symmetry of system. Only those symmetry operations remain which leave the magnetization unchanged. These form a subgroup of the original symmetry group and may be divided in two subsets:

Type A-operations that do not invert the magnetization (e.g. identity, inversion, rotations with the rotation axis parallel to magnetization).

Type B-operations invert the magnetization and must be multiplied by the time inversion in order to leave the system unchanged (e.g. mirror planes, magnetization in the mirror plane).

Problems arise because of the type B-operations. The struct file contains symmetry operations that are not ordered with respect to their type. It may therefore happen that when in LAPW1 the program searches for symmetry operation transforming the equivalent atom into the reference atom the B-type operation is found. The calculation is then incorrect as incorrect form of V_{orb} is added to the hamiltonian. The error may be avoided if

- Only A-type operations are retained in struct file.
- Symmetry operations in struct file are ordered so that A-type operations come first and check is made that all equivalent atoms are transformed into the reference atoms by A-type operations.
- V_{orb} is added to hamiltonian not in LAPW1, but in LAPWSO.

Note that in first and second case complex LAPW1 must be used, while in the third case real LAPW1 is retained (for systems with the inversion center). From above analysis follows that in the spin-polarized calculations with s-o coupling the orbital potential should be preferably included in LAPWSO and not in LAPW1. Additional advantage is that then the spin-up, spin-down cross-term $\langle l, m_l, \uparrow | V_{orb} | l, m_l', \downarrow \rangle$ may also be considered.

The struct file for spin-polarized calculation with s-o coupling and/or V_{orb} may be easily created using newly developed package SRC_symmetso which should be released soon as a part of WIEN2k.

4 WIEN2k packages modified for presence of V_{orb}

4.1 LAPW0 package

For the interaction of the electronic spin with the external magnetic field ($nmod=3$) the spin up and spin down exchange correlation potentials are shifted by the energy $+\mu_B B_{ext}$, $-\mu_B B_{ext}$, respectively. To this end small modifications were made of:

lapw0.f
xcpot1.f
xcpot3.f

The program searches for file case.vorbup, if it finds it, imfield is put equal to 1 (imfield=0 otherwise). If imfield=1, B_{ext} energy is added to V_{xc} in e atomic spheres (xcpot1.f) and in the interstitial region (xcpot3.f).

No changes of the input data are needed.

4.2 LAPW1 package

The matrix elements of the additional orbital potential are added to the matrix elements of the usual LAPW hamiltonian in the new subroutine horb.frc. Other subroutines modified are:

```
inilpw.f
calkpt.frc
```

No changes of the input data are needed.

The command calling LAPW1 is
x lapw1 -orb (-up) (-dn)

4.3 LAPWSO package

LAPWSO was recently modified substantially (inclusion of relativistic $p_{1/2}$ orbitals, speeding up the calculation), detailed description will be given elsewhere [10].

The command calling LAPWSO for spin-polarized calculation with orbital potential is
x lapwso -orb -up [-c]

4.4 LAPWDM package

With respect to the WIEN2k release there is one more output file:

```
case.dmatupdn
containing ↑, ↓ cross term of the occupation matrix.
```

No change of the input data, no change of the calling command.

4.5 MIXER package

Small modification of scfana.f in MIXER package was made in order to add the extra orbital energy EORB, calculated in SRC_orb, to the total energy.

5 Running the calculation with the orbital potential

The density matrix calculation must be inserted in the usual WIEN iteration loop (after running LAPW2), followed by the calculation of the orbital potential.

Once case.vorbup, case.vorbdn were calculated, they are copied to case.vorbup_old, case.vorbdn_old, which are used in the next iteration loop for mixing of the old and new potential. A UNIX script (joborb) for running the spin-polarized calculation in WIEN2k November 2001 release, that includes orbital potential, and the spin-orbit coupling looks as follows:

```
file=$1
num=$2
count=0
while [ $count -lt $num ]
do
    count='expr $count + 1'
```

```

x lapw0
cat $file.scf0>>$file.scf
cp $file.clmsum $file.clmsum_old
cp $file.clmup $file.clmup_old
cp $file.clmdn $file.clmdn_old
cp $file.vorbup $file.vorbup_old
cp $file.vorbdn $file.vorbdn_old
x lapw1 -c -orb -up
cat $file.scf1up>>$file.scf
x lapw1 -c -orb -dn
cat $file.scf1dn>>$file.scf
x lapwso -c -up
cat $file.scfso>>$file.scf
cp $file.energydum $file.energydumdn
cp $file.energydumdn $file.energydumup
x lapw2 -c -up -so
cat $file.scf2up>>$file.scf
x lapw2 -c -dn -so
cat $file.scf2dn>>$file.scf
x lapwdm -c -up -so
cat $file.scfdmup>>$file.scf
x orb -up
cat $file.scforbup>>$file.scf
x orb -dn
cat $file.scforbdn>>$file.scf
x lcore -up
cat ni.scfcup>>ni.scf
x lcore -dn
cat ni.scfcdn>>ni.scf
x mixer
mv ni.scfm ni.scf
done

```

For the above case the ordering of the symmetry operations in struct file is needed (as discussed above). Remember that the symmetry must be reduced (simultaneous presence of the magnetization and the spin-orbit coupling). This script does not include the spin-up, spin-down cross-term of V_{orb} and it cannot be used for LDA+U(HMF), this special case is run as nonmagnetic, with the exception of lapwso (simple way to do it is to copy nonmagnetic files: case.vector, case.vsp, case.vns on magnetic case.vectorup, case.vectordn etc.)

Analogous script, but without the orbital field (to be used for spin-polarized LDA+U calculation, no interest in orbital effects)

```

file=$1
num=$2
count=0
while [ $count -lt $num ]
do
    count='expr $count + 1'
x lapw0
cat $file.scf0>>$file.scf
cp $file.clmsum $file.clmsum_old
cp $file.clmup $file.clmup_old
cp $file.clmdn $file.clmdn_old

```

```

cp $file.vorbup $file.vorbup_old
cp $file.vorbdn $file.vorbdn_old
x lapw1 -orb -up
cat $file.scf1up>>$file.scf
x lapw1 -orb -dn
cat $file.scf1dn>>$file.scf
x lapw2 -up
cat $file.scf2up>>$file.scf
x lapw2 -dn
cat $file.scf2dn>>$file.scf
x lapwdm -up
cat $file.scfdmup>>$file.scf
x lapwdm -dn
cat $file.scfdmdn>>$file.scf
x orb -up
cat $file.scforbup>>$file.scf
x orb -dn
cat $file.scforbdn>>$file.scf
x lcore -up
cat ni.scfcup>>ni.scf
x lcore -dn
cat ni.scfcdn>>ni.scf
x mixer
mv ni.scfm ni.scf
done

```

Finally the LDA+U non-spin polarized calculation may be run using the script:

```

file=$1
num=$2
count=0
while [ $count -lt $num ]
do
    count='expr $count + 1'
    x lapw0
    cat $file.scf0>>$file.scf
    cp $file.clmsum $file.clmsum_old
    cp $file.vorbup $file.vorbup_old
    x lapw1 -orb
    cat $file.scf1>>$file.scf
    x lapw2
    cat $file.scf2>>$file.scf
    x lapwdm
    cat $file.scfdm>>$file.scf
    cp $file.dmat $file.dmatup
    cp $file.dmat $file.dmatdn
    cp $file.scf1 $file.scf1up
    x orb -up
    cat $file.scforbup>>$file.scf
    cp $file.vorbup $file.vorb
    x lcore
    cat ni.scfc>>ni.scf
    x mixer

```

```
mv ni.scfm ni.scf
done
```

These scripts are called by a command:

```
joborb case n
```

where n is the number of iterations required.

References

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