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A Fully Self Consistent Implementation of LDA+DMFT in Abinit

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Thanks to F. Jollet, M. Torrent

Outline of the presentation

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- Strong correlation and LDA+DMFT/+U: Reminder/Introduction
- LDA+DMFT and Abinit: Current status
- LDA+DMFT implementation in Abinit
  - Formalism
  - Practical implementation: subroutines, files, modules, link with the code.
  - Input variables
  - Automatic tests
- Test/Check of the implementation
- Some results
- Conclusion: Current status and projects.

Outline of the presentation

- **Strong correlation and LDA+DMFT/+U: Reminder/Introduction**

Strong local correlations: Introduction (I)

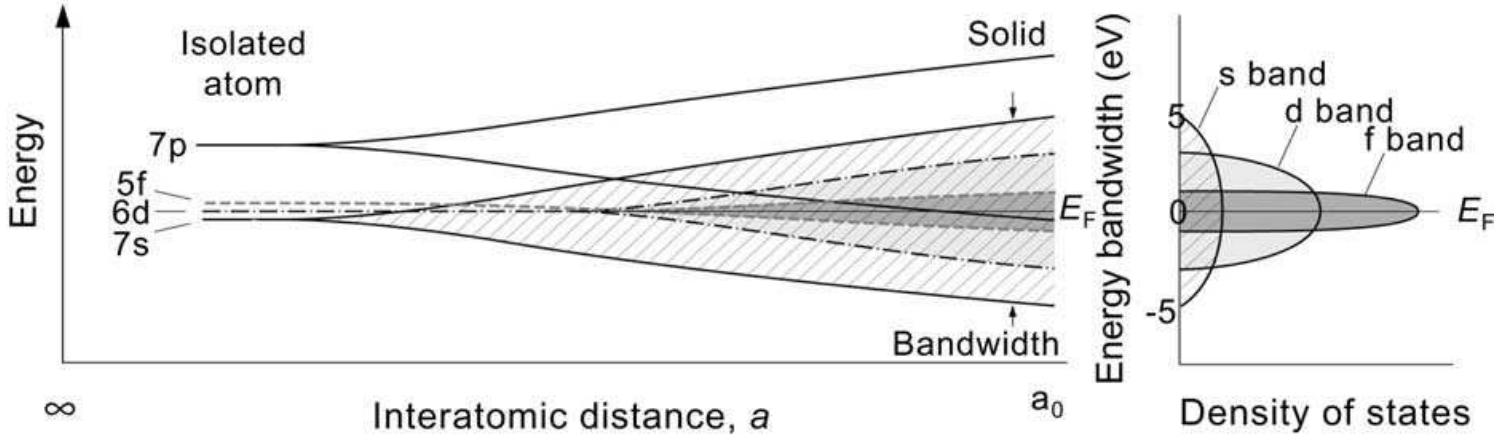

Fig. 5. Schematic of overlapping energy bands and density of states.

- $3d$, $4f$ and $5f$ elements: localized atomic wavefunctions $\Rightarrow$ strong correlations.
  - Small overlap: bands are narrow (width: W).
  - Strong interactions "U" between electrons.

  $\Rightarrow$ From the ratio of U and W, depends the impact of correlations.

U/W$\gg$ 1 : Localization

U/W$\ll$ 1 : Delocalization

Strong local correlations: Introduction (II)

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- Hamiltonian to solve (i: électrons):

$$H = \sum_{i=1}^{N}[-\frac{1}{2}\nabla^2_{\mathbf{r}_i} + V_{\rm ext}(\mathbf{r}_i)] + \frac{1}{2}\sum_{i\neq j}\frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

- Strong local correlation local: For localized orbitals (f, d).
- Other orbitals: DFT(LDA/GGA)

$$\begin{aligned} H_{\rm LDA} &= \text{1 electron LDA term} \\ H_{\rm Correlations} &= \text{Corrected LDA one electron term} + \underbrace{\text{N body interaction term}}_{\frac{U}{2}\sum_{i\neq j}\hat{n}_{\rm i}\hat{n}_{\rm j}} \end{aligned}$$

$$E_{\rm LDA+U} = E_{\rm LDA} - U\frac{N(N-1)}{2} + \frac{U}{2}\sum_{i\neq j} n_i n_j$$

- Limitations of LDA+U: no delocalization, no Kondo effects $\Rightarrow$ DMFT.

The Dynamical Mean Field Theory (DMFT)

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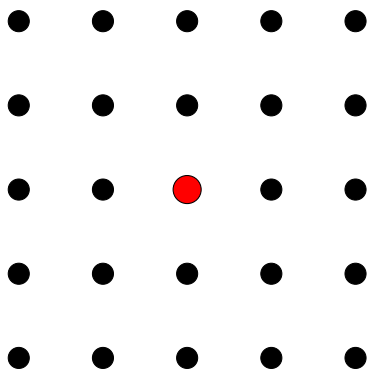
$\Rightarrow$ To describe correlation in solids beyond static mean field theory.

Main idea: An atom is isolated (in red), local correlations are described exactly, effect of the other atoms are gathered in an effective field.



$\Rightarrow$ Anderson impurity model ....+ Self-consistency

[see review A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg Rev. Mod. Phys. 68, 13 (1996)]

The Dynamical Mean Field Theory (DMFT)

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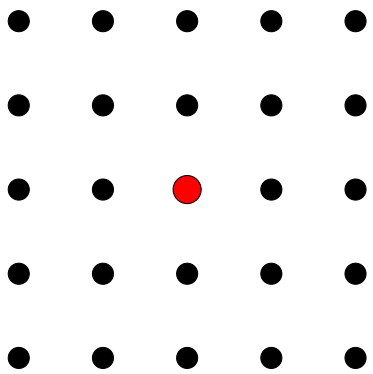
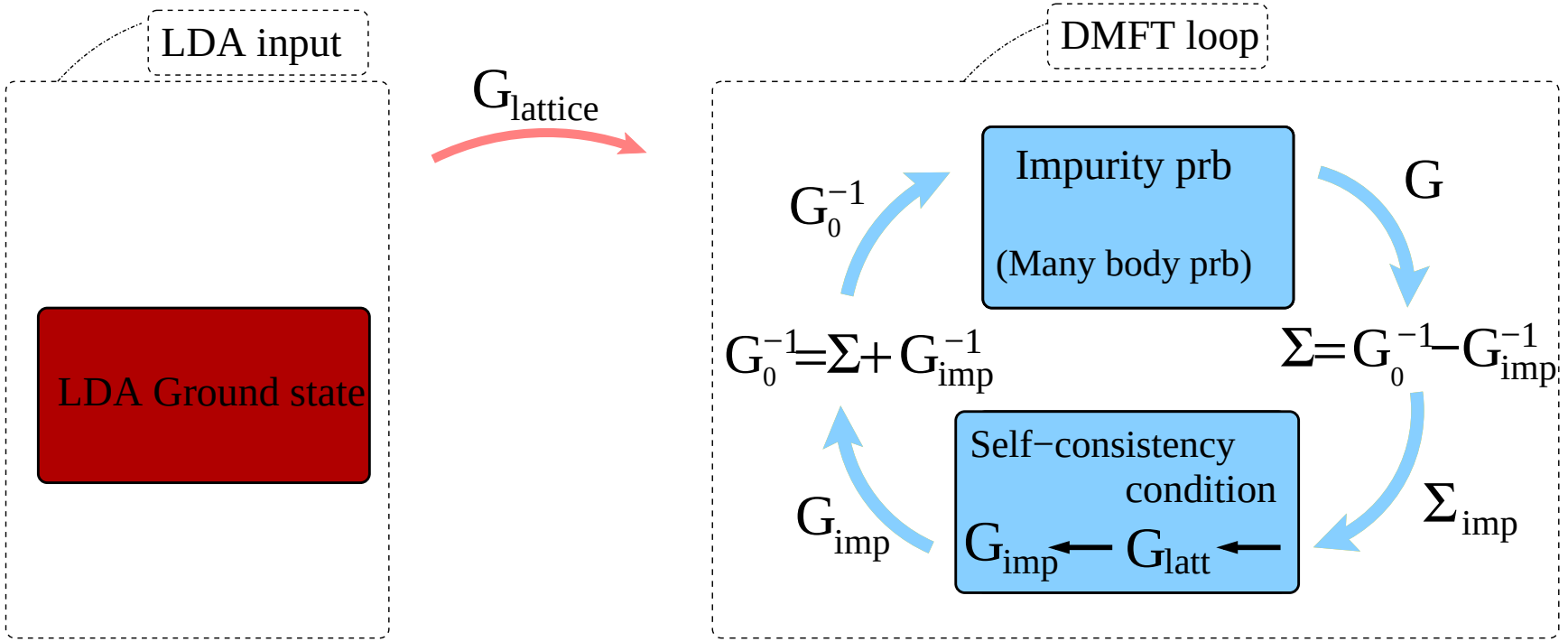


⇒ Anderson impurity model ....+ Self-consistency



Outline of the presentation

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- Strong correlation and LDA+DMFT/+U: Reminder/Introduction
- LDA+DMFT and Abinit: Current status

LDA+DMFT and Abinit: What is available ?

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- **Abinit 6.6.2**
  - DMFT Loop.
  - Anderson model solved with Hubbard I Solver.
  - Self-consistency over electronic Density.
  - Calculation of Total Energy .
- **Currently tested and not yet committed**
  - Anderson model solved with Hirsch Fye Quantum Monte Carlo
  - Calculation of Total Energy in QMC.
- **In progress**
  - Anderson model solved with Continuous Time Quantum Monte Carlo: See the poster of Jordan Bieder !!
- **In project**
  - Forces (and phonons)
  - Improvement of the DMFT self-consistency cycle
  - Spin Orbit (done for NSCF calculations)

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  - **Formalism**

Formalism: LDA+DMFT implementation, Define U and χ

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- Correlated d/f orbitals χ.
  - First implementations: The local correlated subspace is a subset of the basis
    - LMTO Lichtenstein and Katsnelson 1998
  - Maximally Localized Wannier Functions (ML-Wannier)
    - Wanniers or NMTO (Pavarini *et al* 04, Anisimov *et al* 05)
    - Maximally localized Wanniers and NMTO (Lechermann *et al* 06)
  - A simple formalism: Projected local orbitals (Anisimov *et al* 05, Amadon *et al* 08)
    - Independant of the basis
    - Does not require the construction of ML-Wannier
    - Easy to implement in plane waves codes
- Screened Interaction U ? see presentation of Donat Adams

Formalism: The projected scheme

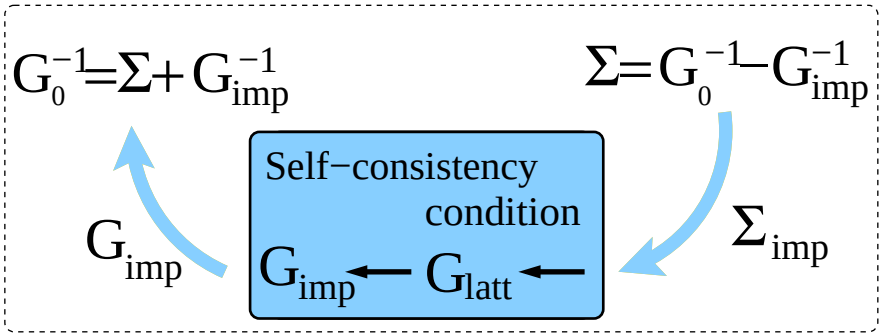
- The number of Bloch states N is an input for the calculation.
- The definition of χ_m depends on N.

Projectors: $P_{m\nu}(\mathbf{k}) \equiv \langle \chi_{\mathbf{k}m} | \Psi_{\mathbf{k}\nu} \rangle$, $P^*_{\nu m}(\mathbf{k}) \equiv \langle \Psi_{\mathbf{k}\nu} | \chi_{\mathbf{k}m} \rangle$.

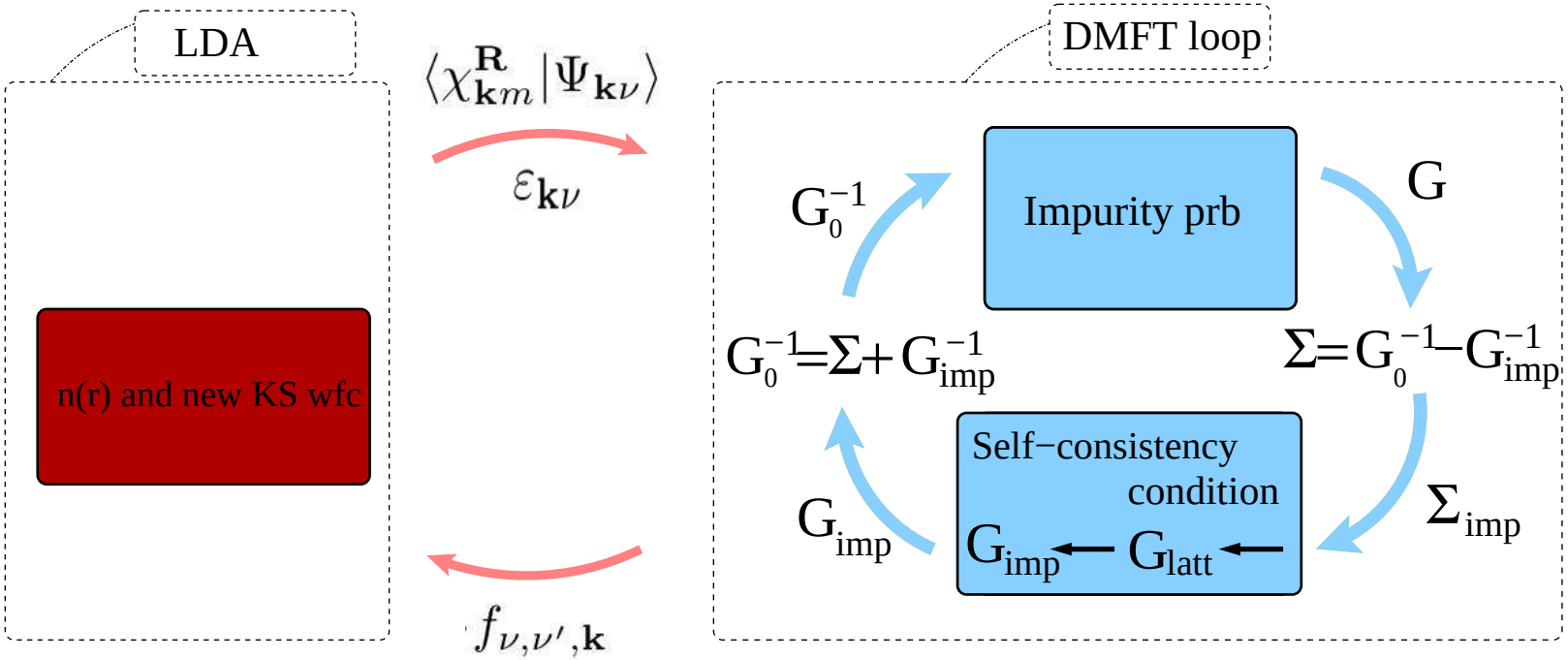
$$\Delta\Sigma^{\rm bloch}_{\nu\nu'}(\mathbf{k}, i\omega_n) = \sum_{mm'} P^*_{\nu m}(\mathbf{k}) \left(\Sigma^{\rm imp}_{mm'}(i\omega_n) - \Sigma^{\rm dc}_{mm'}\right) P_{m\nu}(\mathbf{k})$$

$$G^{\rm bloch}_{\nu\nu'}(\mathbf{k}, i\omega_n) = \left\{ \frac{1}{(i\omega_n + \mu - \varepsilon_{\mathbf{k}\nu})\delta_{\nu\nu'} - \Delta\Sigma^{\rm bloch}_{\nu\nu'}(\mathbf{k}, i\omega_n)} \right\}_{\nu\nu'}$$

$$G^{\rm imp}_{mm'}(i\omega_n) = \sum_{\mathbf{k},\nu\nu'} P_{m\nu}(\mathbf{k})\, G^{\rm bloch}_{\nu\nu'}(\mathbf{k}, i\omega_n)\, P^*_{\nu m}(\mathbf{k})$$



Formalism: Self Consistency over electronic density



- The introduction of correlations changes the electronic density
- To do the DMFT loop for a given LDA iteration, one needs:
  - The LDA eigenvalues $\epsilon(\mathbf{k}, n)$ computed for a given density $n(\mathbf{r})$.
  - The projections $\langle \chi_m | \Phi(\mathbf{k}, n) \rangle$ computed for a given density $n(\mathbf{r})$.
- From DMFT,non-diagonal occupations $f_{n,n'}(\mathbf{k})$ are used to compute the density

Formalism: Self Consistency over electronic density

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$$n(\mathbf{r}) = \widetilde{n}'(\mathbf{r}) + n^{1'}(\mathbf{r}) - \widetilde{n}^{1'}(\mathbf{r})$$

with

$$\widetilde{n}'(\mathbf{r}) = \sum_{n,n',\mathbf{k}} f_{n,n',\mathbf{k}} \widetilde{\Psi}^*_{n,\mathbf{k}}(\mathbf{r}) \widetilde{\Psi}_{n',\mathbf{k}}(\mathbf{r})$$

and,

$$\begin{aligned} n^{1'}(\mathbf{r}) &= \textstyle\sum_{ij} \rho'_{ij} \phi_i(\mathbf{r}) \phi_j(\mathbf{r}) \\ \widetilde{n}^{1'}(\mathbf{r}) &= \textstyle\sum_{ij} \rho'_{ij} \widetilde{\varphi}_i(\mathbf{r}) \widetilde{\varphi}_j(\mathbf{r}) \end{aligned}$$

and

$$\rho'_{ij} = \sum_{n,n',\mathbf{k}} f_{n,n',\mathbf{k}} \langle \widetilde{\Psi}_{n,\mathbf{k}} | \widetilde{p}_j \rangle \langle \widetilde{p}_i | \widetilde{\Psi}_{n',\mathbf{k}} \rangle$$

$$E_{\mathrm{LDA+DMFT}} = E_{\mathrm{LDA}} - \sum f^{\mathrm{lda}}_{\mathbf{k},n} \epsilon_{\mathbf{k},n} + \sum f^{\mathrm{dmft}}_{\mathbf{k},n} \epsilon_{\mathbf{k},n} + E_U - E_{\mathrm{DC}}$$

(two ways..)

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  - **Practical implementation: subroutines, files, modules, link with the code.**

Subroutines and modules (mainly) in 68_dmft


```
../66_paw/m_paw_dmft.F90   # Contains Data for DMFT modules and SCF
../66_paw/*pawu*.F90       # input for DMFT

datafordmft.F90            # Prepare Projections for DMFT
psichi_renormalization.F90 # Renormalize projections

m_matlu.F90     #  Operator in the local density MATrix for LpawU
m_oper.F90      #  General OPERator expressed in KS or local basis
m_green.F90     #  Green function (frequency or time dependant)
m_self.F90      #  Self energy
m_hu.F90        #  Interaction Hamiltonian U and J between electrons
m_energy.F90    #  Total energy

dmft_solve.F90  # Main routines for the DMFT Loop  (called in vtorho.F90)
dyson.F90       # Dyson Equation
fermi_green.F90 # Compute Fermi level
newton.F90      #  Search for Fermi Level with newton method

impurity_solve.F90  # Solve impurity model
hubbard_one.F90     # Hubbard One solver
ldau_self.F90       # Compute LDA+U Self Energy
qmc_prep.F90        # Preparations for QMC

local_ks_green.F90     # Print Green function
spectral_function.F90  # Print Spectral Function (For Hubbard I only)
interfaces_68_dmft.F90

../*/mkrho.F90...  # Compute electronic density from DMFT occupations.
```

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  - **Input variables**

Input variables


```
# == LDA+U
usepawu    1       # Use U Hamiltonian
dmatpuopt  3       # Choose density matrix (only for print out)
lpawu  2           # Use Hubbard U for l=lpawu
upawu 4.0 0.0 eV   # Value of U for the two species
f4of2_sla 0.0 0.0 # Value of F4/F2 to compute <m1m2|1/r|m3m4>

# == LDA+DMFT
usedmft   1        # Enable DMFT
dmftbandi 6        # value of the Initial KS BAND to use
dmftbandf 12       # value of the Final KS BAND to use
dmft_nwlo 100      # Nb of freq W in the LOg Grid (sums are parallelized)
dmft_nwli 100000   # Nb of matsubara freq W (LInear Mesh)
dmft_iter 10       # nb of ITER for the dmft loop
dmftcheck 1        # Enable CHECKs (symetry, projection, fourier)
dmft_solv 2        # choice of the SOLVer
dmft_rslf 1        # Read SeLF energy from file (for restart)
dmft_mxsf 0.7      # MiXing coefficient for SelF energy
dmft_dc 1          # Double Counting for DMFT
```

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  - **Automatic tests**

Automatic tests


```
v6/t07  LDA  calculation through the DMFT Loop (check)
v6/t45  DMFT calculation with U=0, and U/=0 for several solvers
v6/t46  DMFT calculation with two Ni atoms.
v6/t47  DMFT calculation for f-orbitals (Gd)
```

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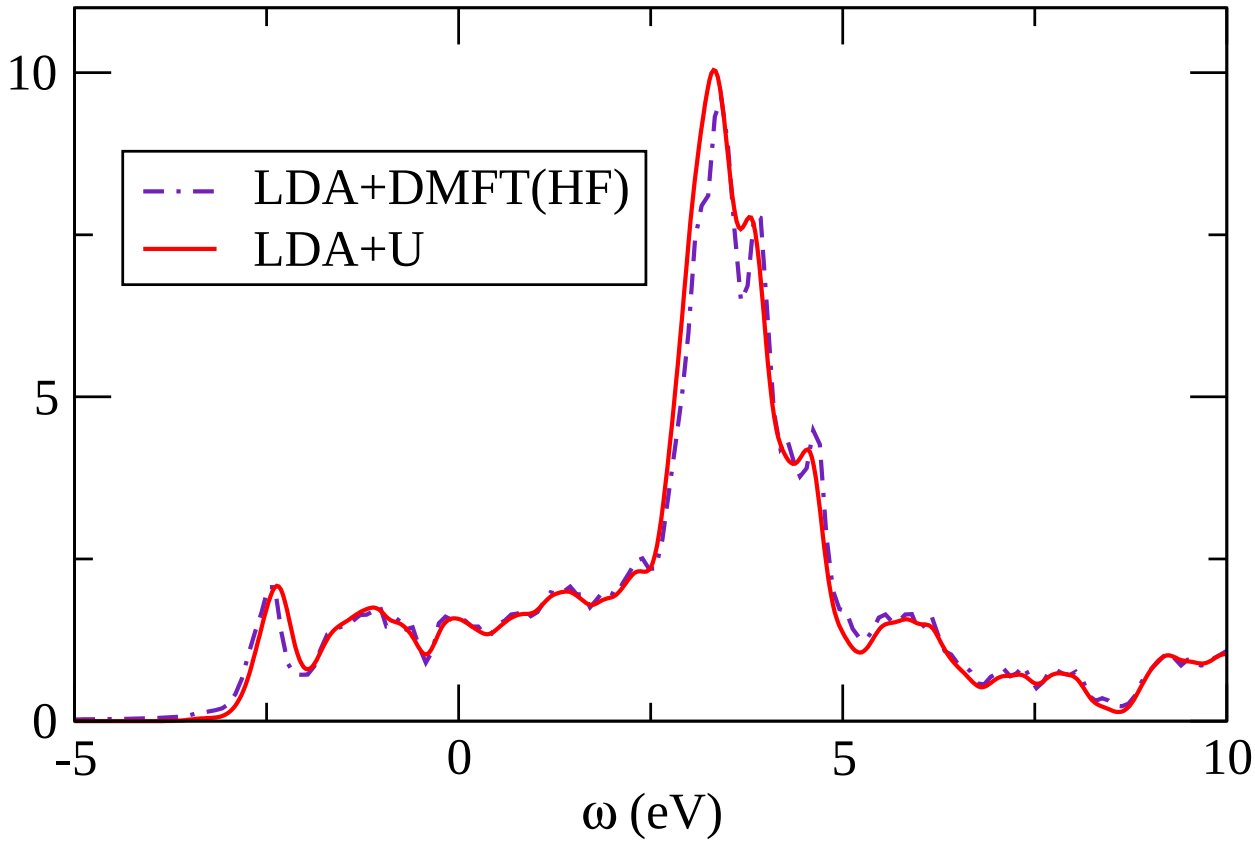
- **Test/Check of the implementation**

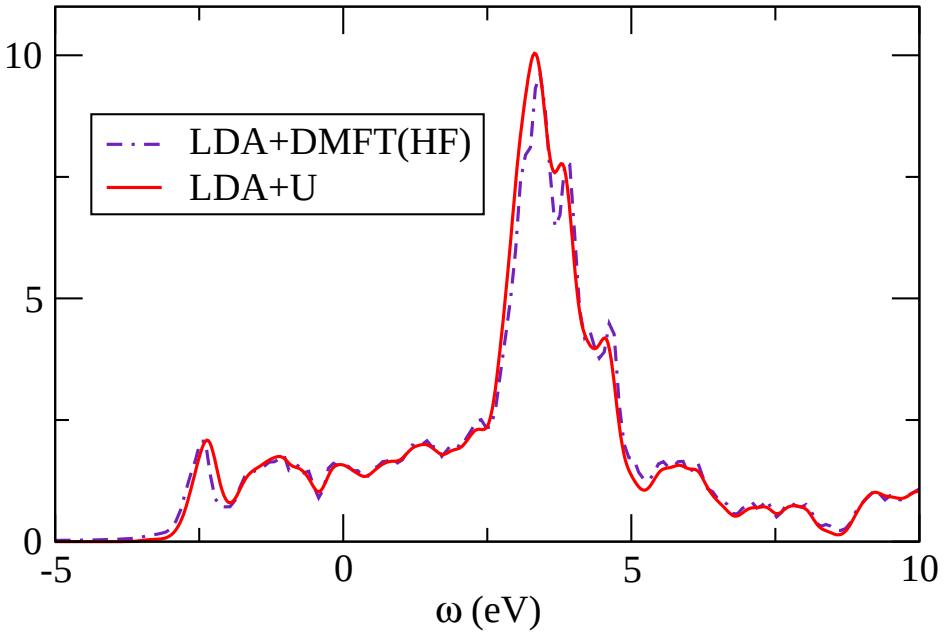
Implementation: Checks (I)



Spectral function of γ-cerium, LDA+U and LDA+DMFT (Static)

With the LDA+U Self-energy, the DMFT recover the LDA+U results at convergence of the KS basis.

Implementation: Checks (II)



	a (a.u.)	B_0 (GPa)
PAW/LDA+U	9.58	32
PAW/LDA+DMFT(HF)	9.59	31

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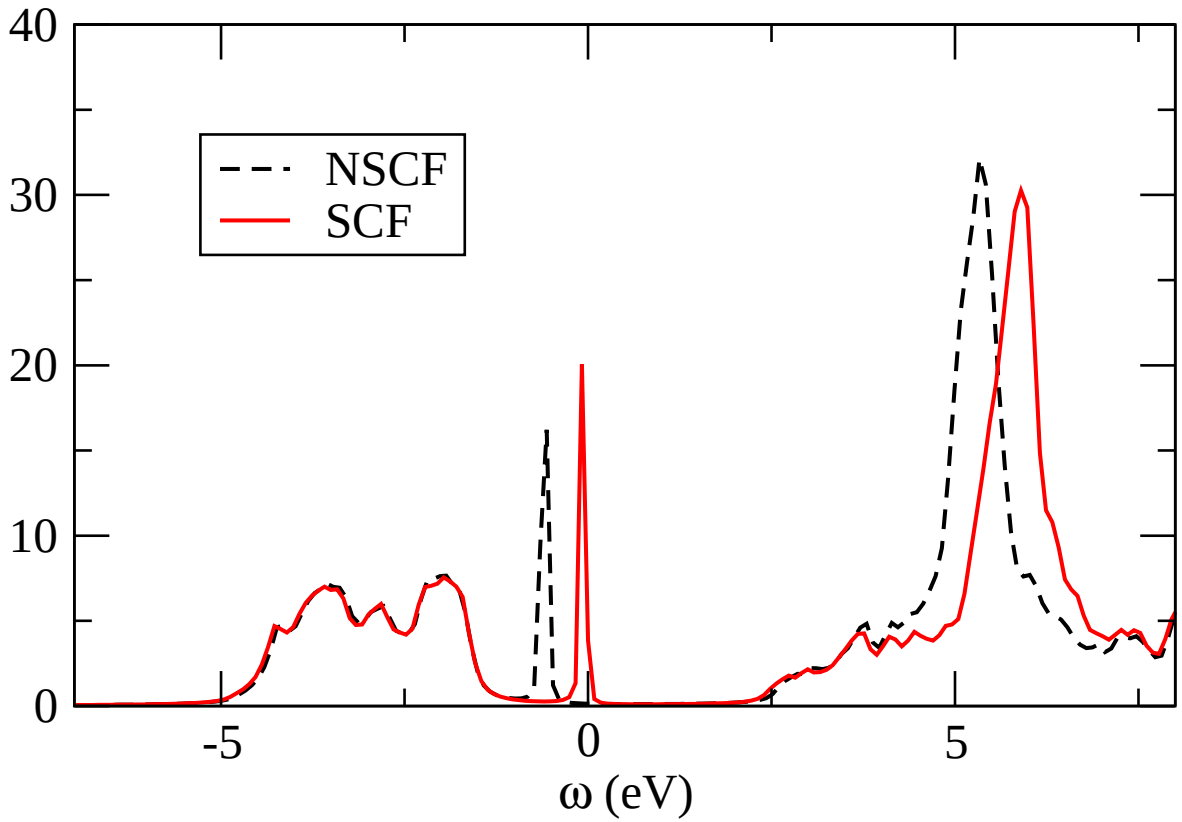
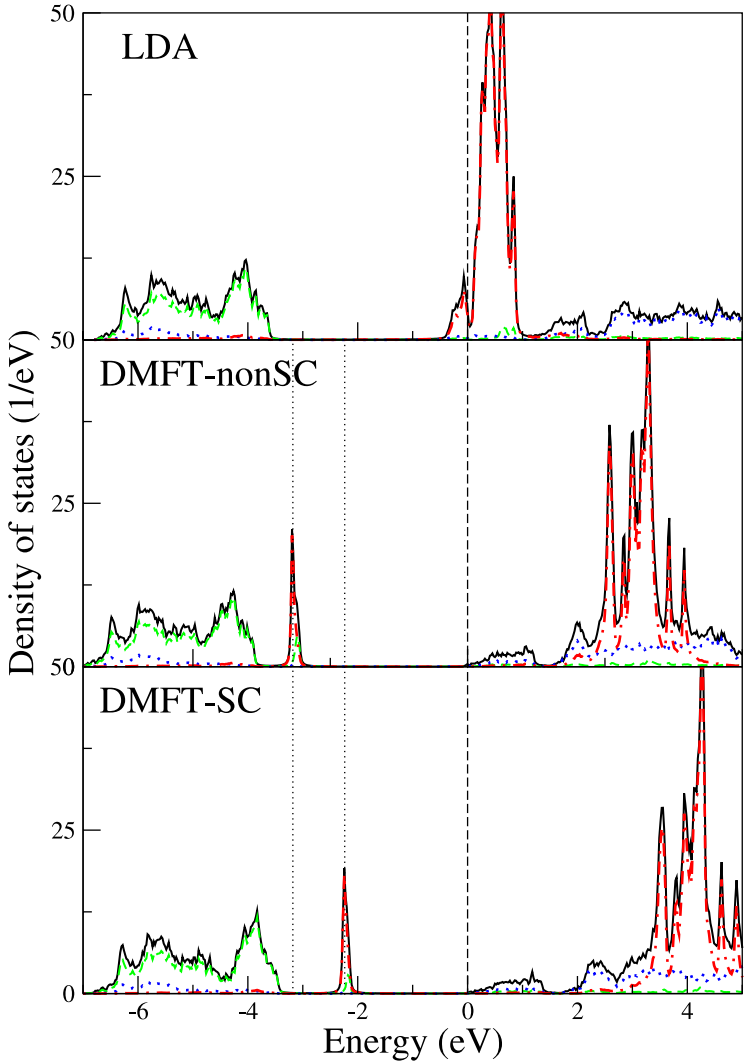
- **Some results**

Results: Spectral function of Ce_2O_3



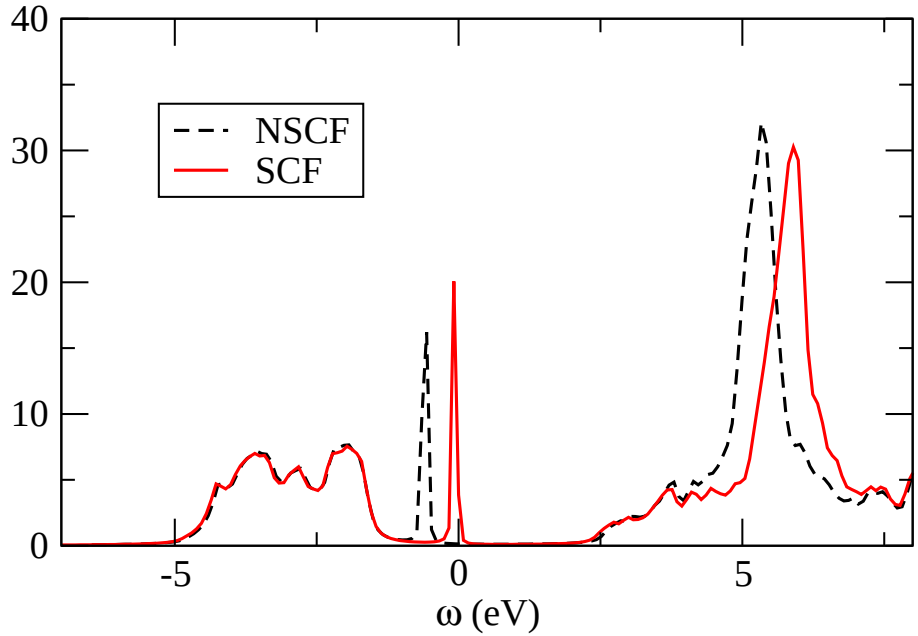
Spectral function of Ce_2O_3, in LDA+DMFT (Hubbard I) with and without self-consistency. Self-consistent calculation is in better agreement with experiment (2.4 eV).

Results: Spectral function of Ce_2O_3



LMTO-ASA +DMFT

L. Pourovskii, B. A., S. Biermann and A. Georges PRB (2007)



PAW + DMFT

B. Amadon Arxiv 2011

Results: Structural parameters of Ce_2O_3

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	a (A)	B_0 (Mbar)
Exp(Barnighausen 1985)	3.89	1.11
PAW/LDA+DMFT (H-I) NSCF	3.76	1.7
PAW/LDA+DMFT (H-I) SCF	3.83	1.6
ASA/LDA+DMFT(H-I) NSCF (Pourovskii 2007)	3.79	1.6
ASA/LDA+DMFT(H-I) SCF (Pourovskii 2007)	3.81	1.6

Lattice parameter a and Bulk modulus B_0 of Ce_2O_3.

Results: Converged Electronic Density.

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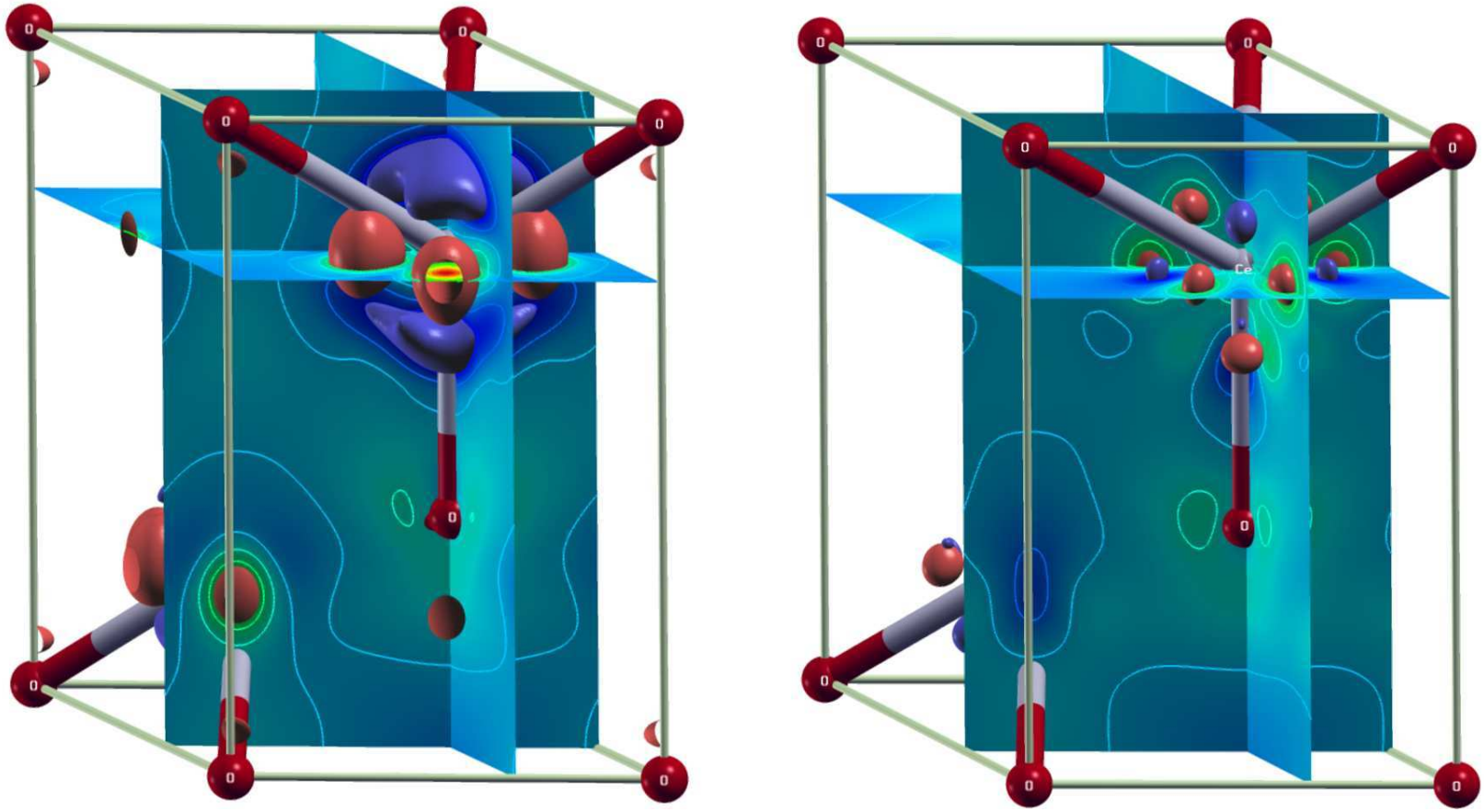

Difference between electronic densities computed in the LDA+U (left)/LDA+DMFT (right) and in LDA for Ce_2O_3.

Blue (resp. green-red) area corresponds to positive (resp negative) value of the difference.

Implementation of the LDA+DMFT (self-consistency)

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	a (a.u.)	B_0 (GPa)
Exp[Jeong 2004]	9.76	19/21
PAW/LDA+DMFT NSCF (H-I)	9.41	38
PAW/LDA+DMFT SCF (H-I)	9.58	36
ASA/LDA+DMFT NSCF (H-I)	9.28	50
ASA/LDA+DMFT SCF (H-I)	9.31	48

Lattice parameter a and Bulk modulus B_0 of γ Cerium according to experimental data and calculations

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Conclusion

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- Fully self consistent implementation of LDA+DMFT in PAW.
- Spectra and total energy in Hubbard I.
- Implementation with modules: code is easier to read.
- Quantum Monte Carlo solvers are in progress (Poster of Jordan Bieder)
- Forces and Spin orbit are in project for 2011.

See details on **arxiv:cond-mat/1101.0539**.