Ab initio calculation of effective interaction parameters in Lanthanides

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Why parameters ?

LDA and GGA fail to describe Lanthanides (equilibrium volume, electronic structure...)
Add to DFT Hamiltonian an explicit interaction term

- DFT+U [1], DFT+DMFT [2]
- Introduction of parameters U and J

1. See A. Lichtenstein et al, PRB 52, 5467 (1995) and M. T. Czyzyk and G. A. Sawatzky PRB 49, 14211 (1994)
2. A. Georges et al,Rev. Mod. Phys. 68, 13

Definition [3]

2nd quantization formulation of electronic interaction Hamiltonian (m_i, σ_i indexing correlated orbitals and spin)

$$\hat{H}_{int} = \sum_{m_1,m_2,m_3,m_4,\sigma_1,\sigma_2} U^{\sigma_1,\sigma_2}_{m_1,m_2,m_3,m_4} \hat{c}^{\dagger}_{m_1,\sigma_1} \hat{c}^{\dagger}_{m_2,\sigma_2} \hat{c}_{m_3,\sigma_2} \hat{c}_{m_4,\sigma_1}$$

Direct interaction parameter

$$U = \frac{1}{4} \sum_{\sigma_1,\sigma_2} \frac{1}{(2l+1)^2} \sum_{m_1=1}^{2l+1} \sum_{m_2=1}^{2l+1} U^{\sigma_1,\sigma_2}_{m_1,m_2,m_1,m_2}$$

Exchange interaction parameter

$$J = \frac{1}{4} \sum_{\sigma_1,\sigma_2} \frac{1}{2l(2l+1)} \sum_{m_1=1}^{2l+1} \sum_{m_2=1, m_2 \neq m_1}^{2l+1} U^{\sigma_1,\sigma_2}_{m_1,m_2,m_2,m_1}$$

U and J given as input parameters

3. See also B. Amadon et al, Phys. Rev. B 89, 125110

Calculation of *U* and *J*

Problem : U and J are **parameters** in DFT+U and DFT+DMFT

- Bad for the *ab initio* character of the calculation

But : U and J can be computed with an *ab initio* method : the cRPA [4]

- Full *ab initio* calculation

4. F.Aryasetiawan et al, Phys.Rev.B 70,195104 (2004) and Phys. Rev. B 74,125106 (2006)

Scheme of calculation

Interaction of 2 correlated electrons : screened by surrounding electrons

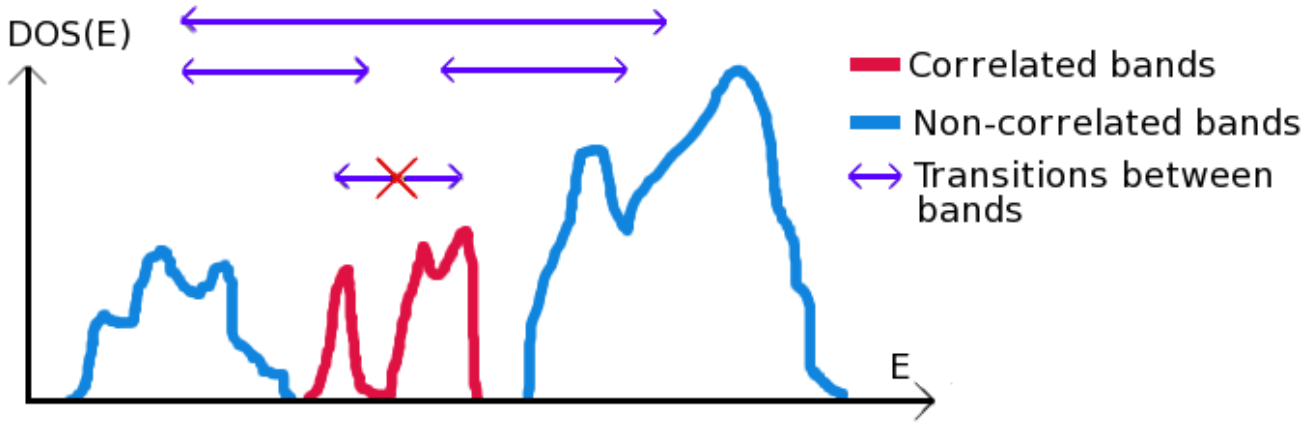
- We must compute a screened interaction W
- To do so, we need to compute a dielectric function ϵ, needing itself a polarization χ (calculated within RPA [5])
- First order perturbation theory : transitions between one-electron bands contribute to χ

$$\chi(r, r', \omega) = \sum_{k_1, n_1, \sigma_1, k_2, n_2, \sigma_2} \frac{\psi^*_{k_1,n_1,\sigma_1}(r)\psi_{k_2,n_2,\sigma_2}(r)\psi^*_{k_2,n_2,\sigma_2}(r')\psi_{k_1,n_1,\sigma_1}(r')}{\omega - \epsilon_{k_1,n_1,\sigma_1} + \epsilon_{k_2,n_2,\sigma_2} \pm i\delta}$$

5. Cf. **A Collective Description of Electron Interactions**, D. Bohm and D. Pines (1951-1953)

The cRPA approximation

- Define **correlated orbitals** (as Wannier functions)
- Suppress transitions between selected bands (**correlated bands**) from the calculation of χ, giving a "constrained" χ_r



- Compute the corresponding screened interaction

$$W_r(\omega) = [I - v\chi_r(\omega)]^{-1} v$$

- Compute dynamical U coefficients

$$U^{\sigma,\sigma'}_{m_1,m_3,m_2,m_4}(\omega) = < m_1^{\sigma} m_3^{\sigma'} | W_r(\omega) | m_2^{\sigma} m_4^{\sigma'} >$$

- Deduce static U and J

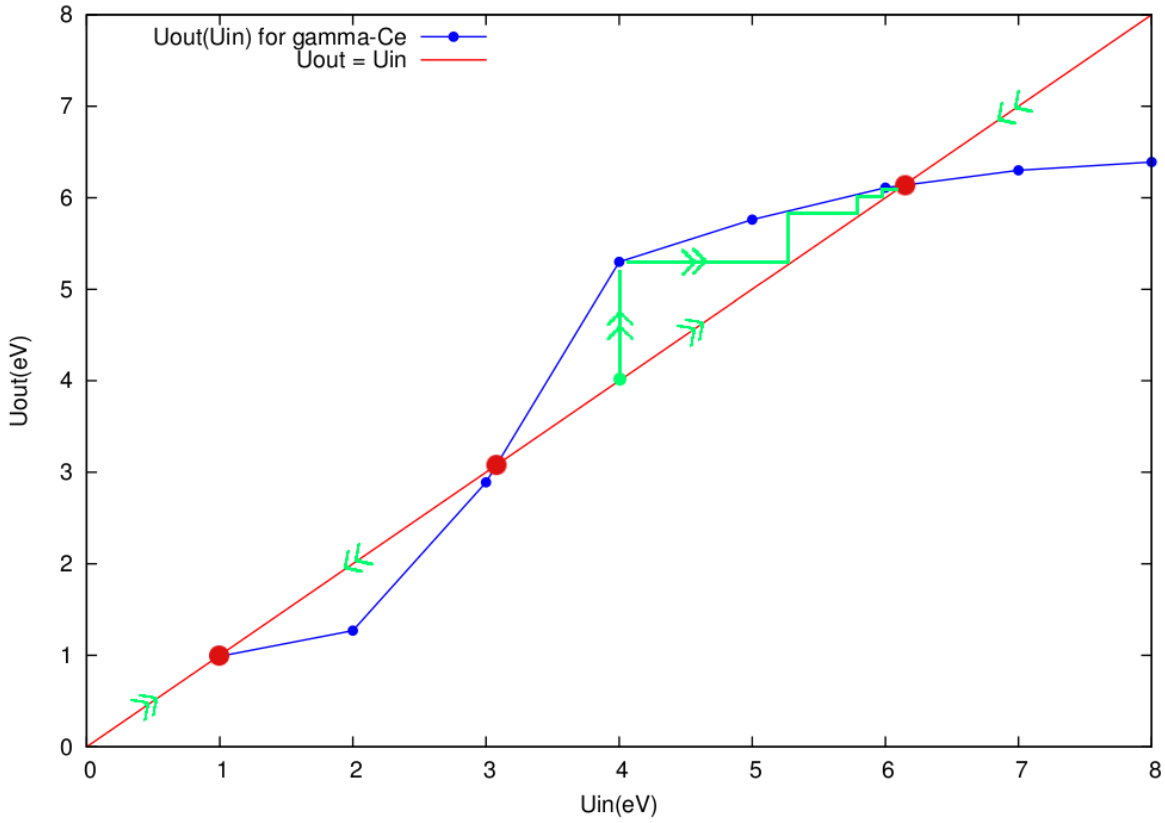
Self-consistent scheme

U and J can be calculated self-consistently

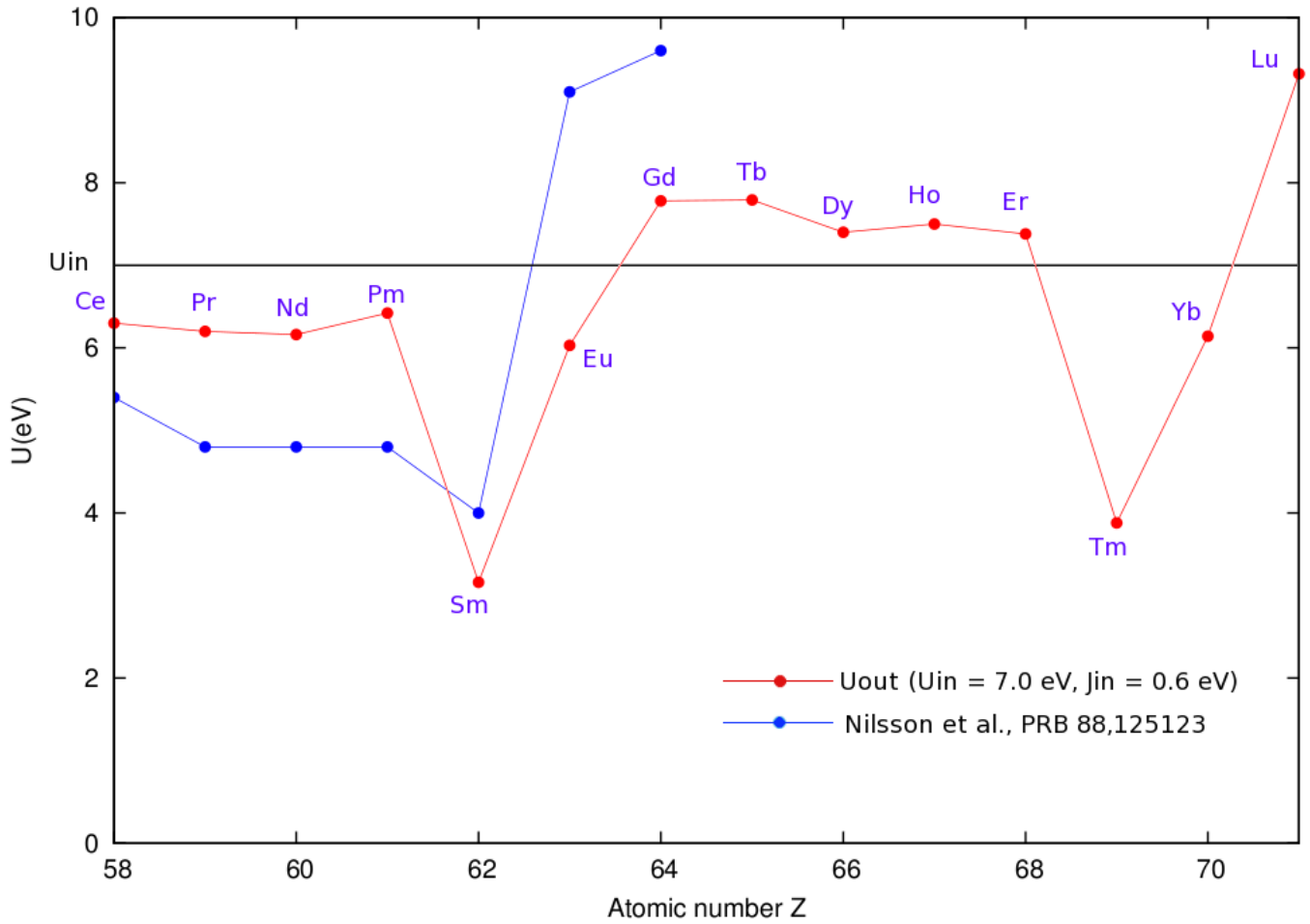
- Choose starting values U_{in}, J_{in}
- Compute new values U_{out}, J_{out} using cRPA
- Check self-consistency

Self-consistent values of U (γ-Ce)

$U_{out}(U_{in})$ for γ-Ce, with J fixed to 0.6 eV.



Non-self-consistent values of U



Conclusion

Limitations of our method

- DFT+U limitations : the chosen value of U must correctly reproduce the physics of the system
- Only $f - f$ interactions are regarded as "correlated". Considering $d - d$ and $d - f$ interactions would be desirable [6]

Perspectives

- Different choices for the construction of correlated orbitals and bands to substract in χ
- Towards a full self-consistent framework : GW+DMFT [7]

6. P. Seth et al., arXiv :1508.07466
7. Biermann, S. et al, Phys.Rev.Lett., American Physical Society, 2003, 90, 086402

Thank you for your attention !