DE LA RECHERCHE À L'INDUSTRIE

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Implementation of "charge-only-DFT"+U and k-resolved spectral function in DFT+DMFT.

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abidev 2019

1. ”charge-only-DFT”+*U*

"charge-only-DFT"+U

- Usual "spin DFT"+U : sDFT+U

$$\begin{aligned} E &= -\sum_{\sigma,\nu}\int \psi_\nu^{\sigma *}(\mathbf{r})\frac{\nabla^2}{2}\psi_\nu^{\sigma}(\mathbf{r})d\mathbf{r} + \int d\mathbf{r} v_{\rm ext}(\mathbf{r})n(\mathbf{r}) + E_{\rm Hartree}[n(\mathbf{r})] \\ &+ E_{\rm xc}[n^{\uparrow}(\mathbf{r}), n^{\downarrow}(\mathbf{r})] + E_{\rm ee}^{U,J}[n^{\uparrow}_{m,m'}, n^{\downarrow}_{m,m'}] - E_{\rm DC}^{U,J}[N^{\uparrow}, N^{\downarrow}] \end{aligned}$$

- "charge-only DFT"+U : DFT+U (or nsDFT+U) (Park *et al* PRB 2016)

$$\begin{aligned} E &= -\sum_{\sigma,\nu,\mathbf{k}}\int \psi_\nu^{\mathbf{k}\sigma *}(\mathbf{r})\frac{\nabla^2}{2}\psi_\nu^{\sigma}(\mathbf{r})d\mathbf{r} + \int d\mathbf{r} v_{\rm ext}(\mathbf{r})n(\mathbf{r}) + E_{\rm Hartree}[n(\mathbf{r})] \\ &+ E_{\rm xc}[n^{\uparrow}(\mathbf{r}) + n^{\downarrow}(\mathbf{r})] + E_{\rm ee}^{U,J}[n^{\uparrow}_{m,m'}, n^{\downarrow}_{m,m'}] - E_{\rm DC}^{U,J}[N^{\uparrow} + N^{\downarrow}] \end{aligned}$$

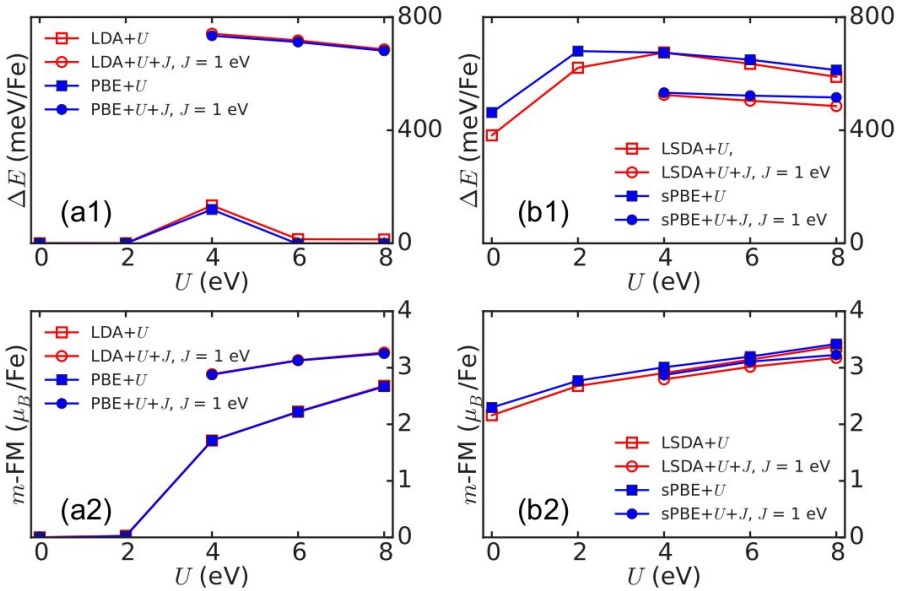
where $n^{\sigma}_{m,m'}$ is the density matrix for correlated electron ($m, m' = -l, \ldots + l$)

$$n^{\sigma}_{m,m'} = \sum_{\mathbf{k},\nu} f_\nu^{\mathbf{k},\sigma} \langle \psi_\nu^{\mathbf{k},\sigma} | \chi_{m'} \rangle \langle \chi_m | \psi_\nu^{\mathbf{k},\sigma} \rangle \quad \text{and} \quad N^{\sigma} = \sum_m n^{\sigma}_{mm}$$

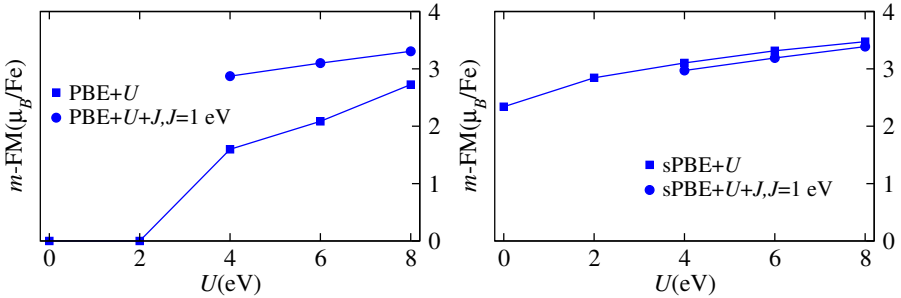
Input variables

`usepawu=1` (usual) sDFT+U (FLL double counting)
`usepawu=4` (new) nsDFT+U (FLL double counting)

Comparison to previous calculations on Iron



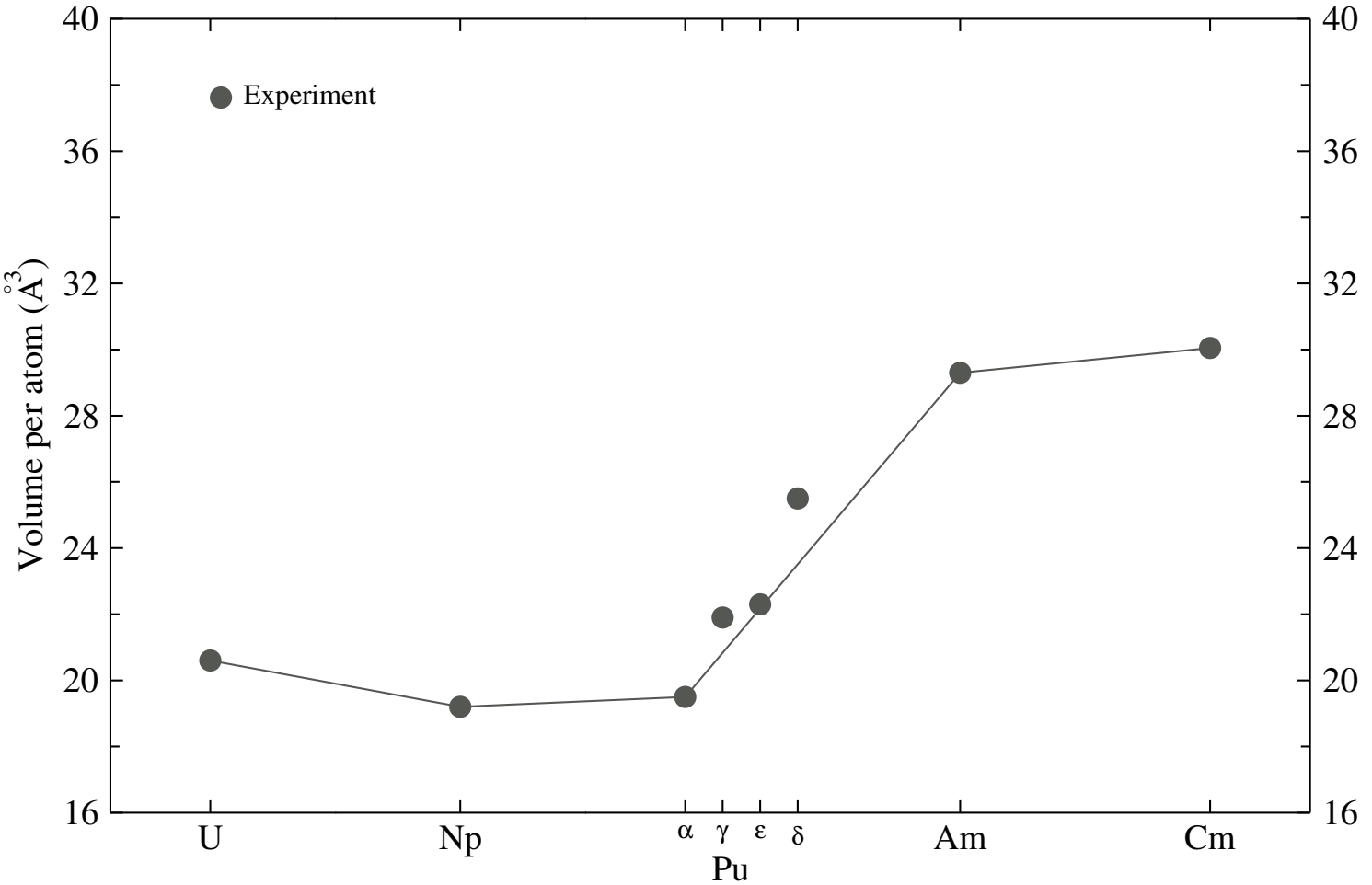
Chen and Millis PRB 2016



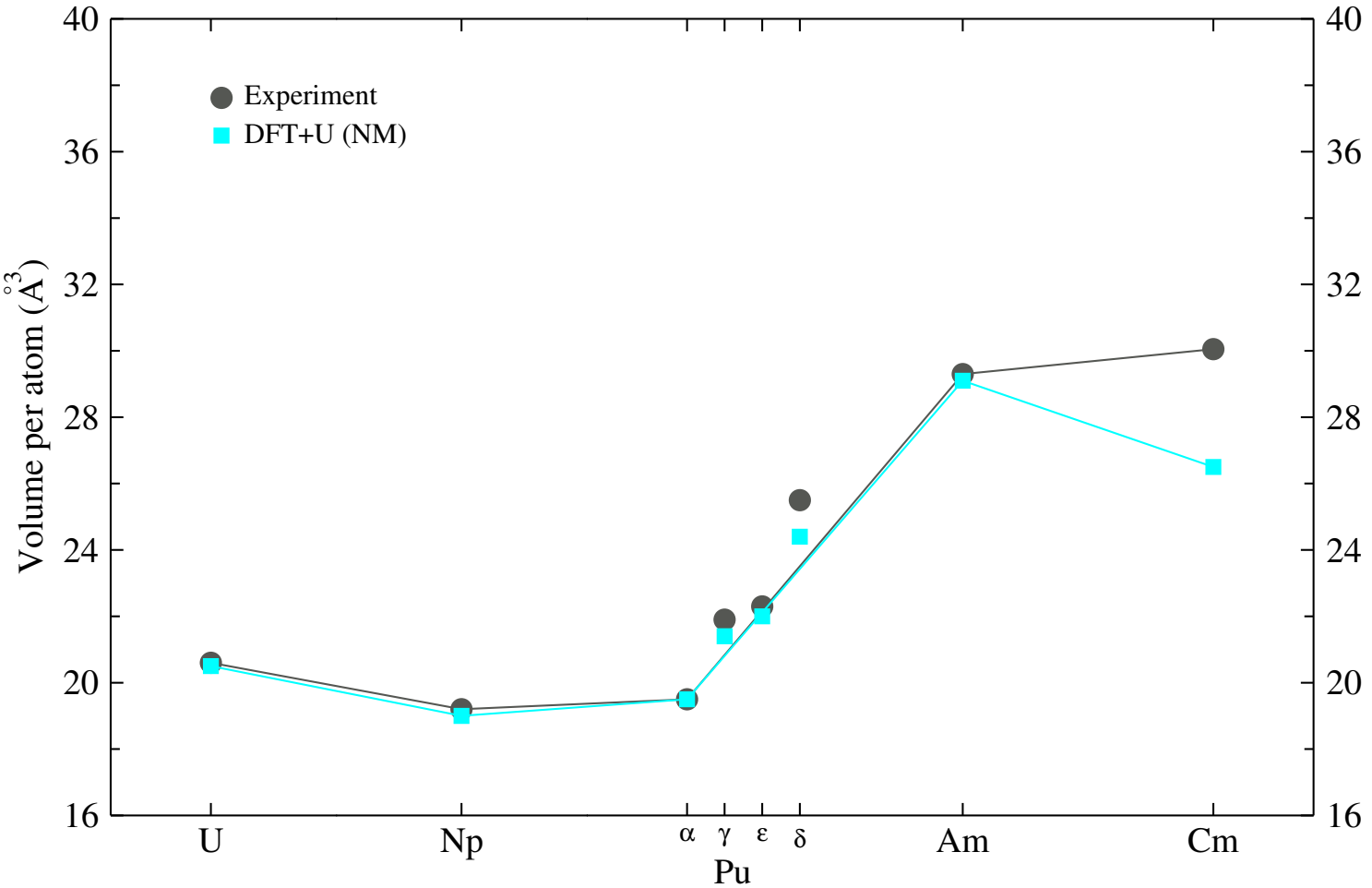
ABINIT (this calculation)

- Experimental magnetic moment (2.2) is overestimated by sPBE+*U* .
- The role of *J* is more physical in nsPBE+*U* : it stabilizes ferromagnetism and increases magnetic moment.

Experimental volumes

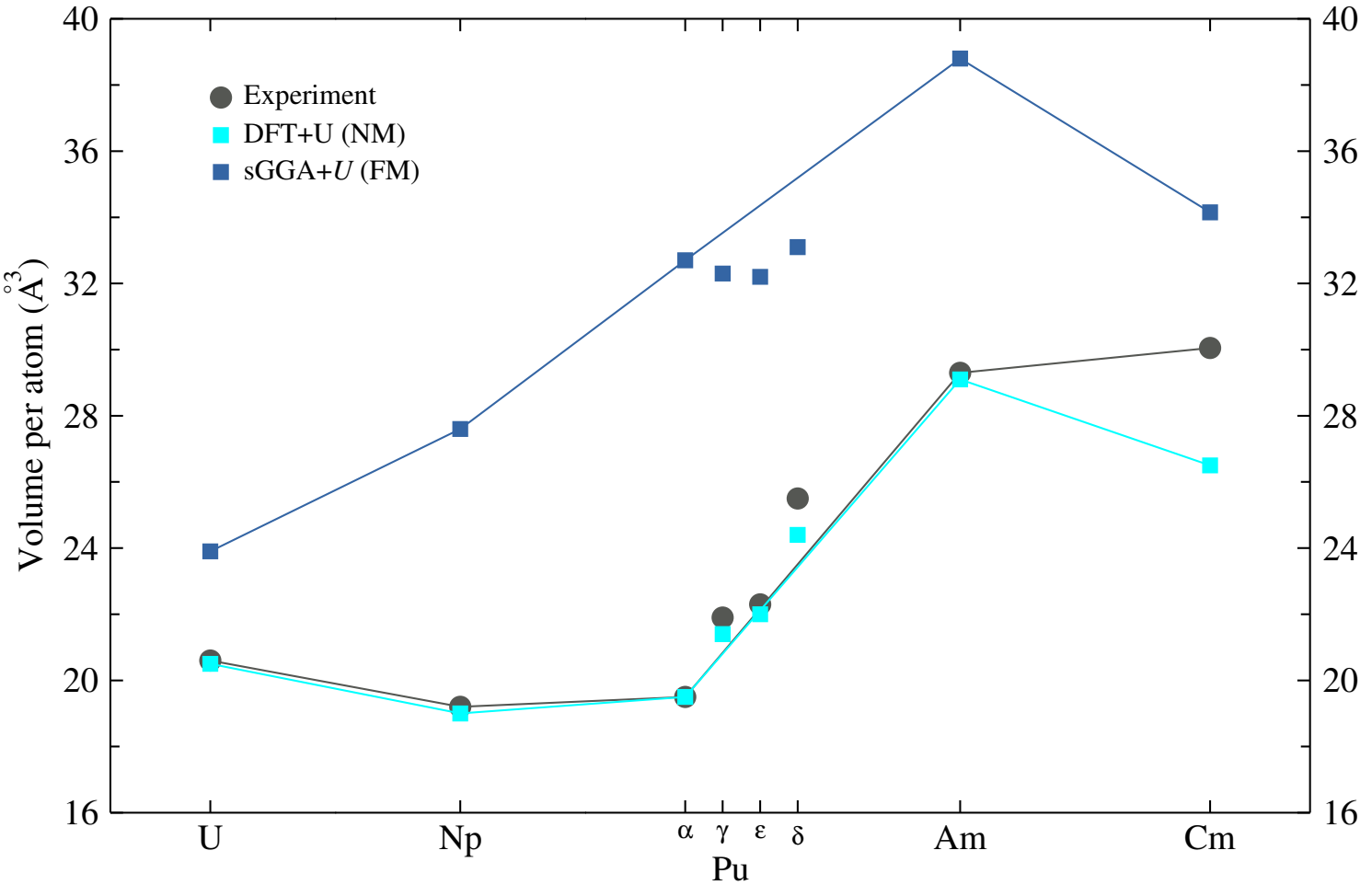


NM DFT+U calculations with cRPA values of U



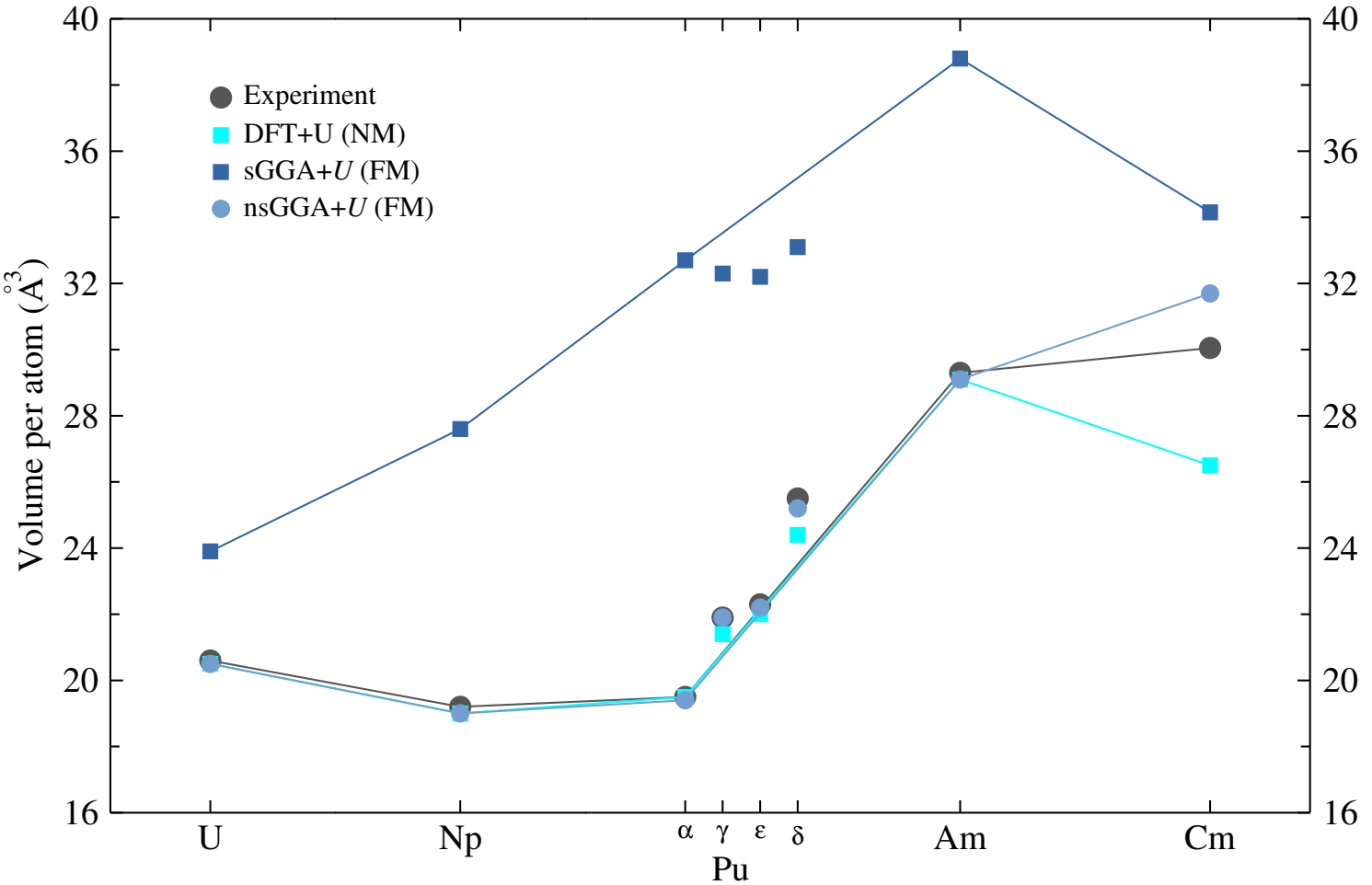
B. Amadon and B. Dorado JPCM 2018

Magnetism



The ground state of sDFT+*U* does not describe well the structure.

Magnetism



nsDFT+*U* (”charge-only-DFT”+*U*) does describe well the structure.

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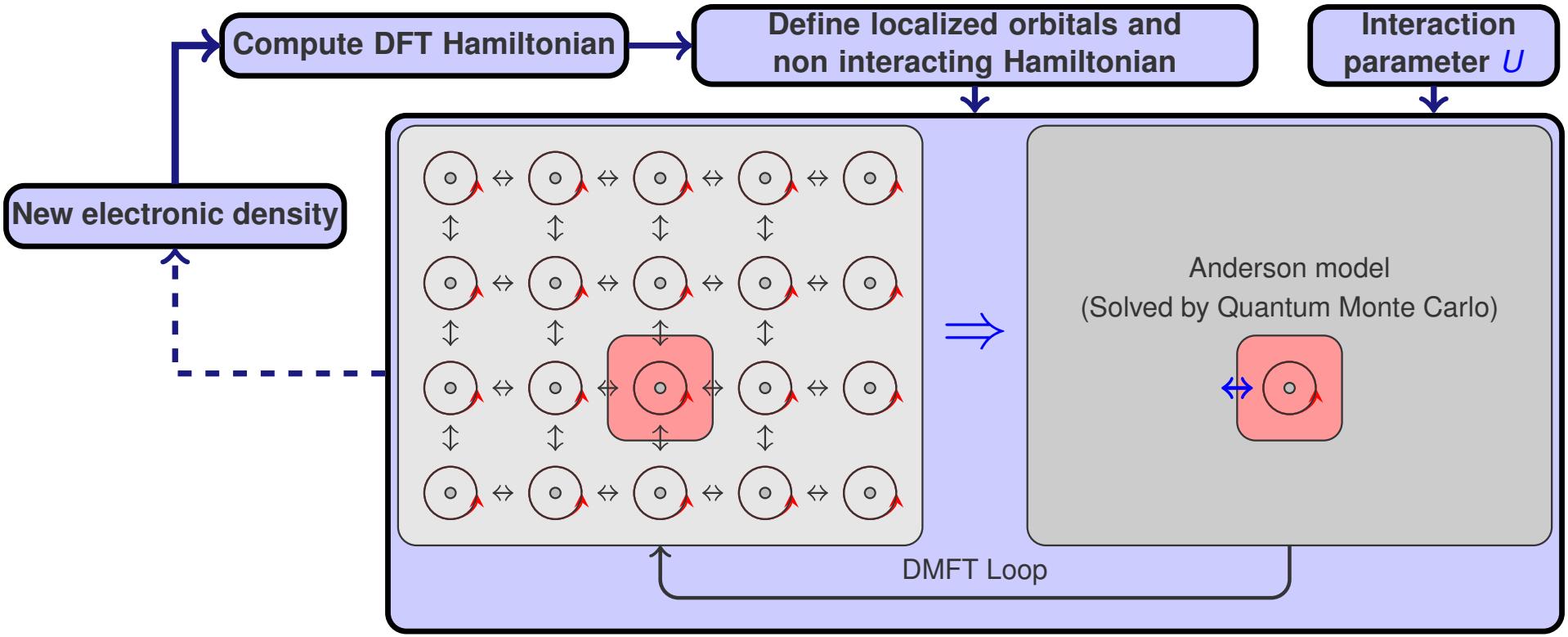
2. k-resolved spectral function

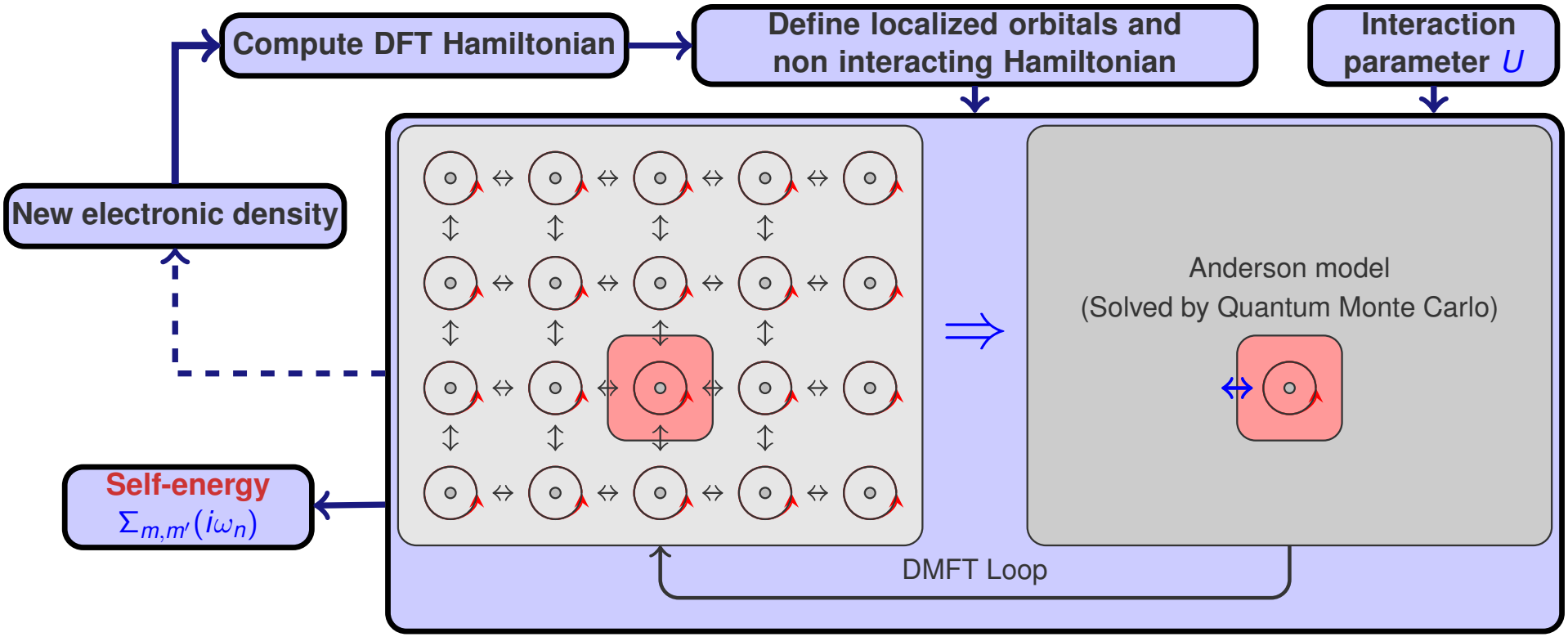
DFT+DMFT: describes correlations



- Scheme implemented in ABINIT

L.V. Pourovskii, B. Amadon, S. Biermann, A. Georges Phys. Rev. B **76**, 235101 (2007)
B. Amadon, F. Lechermann, A. Georges, F. Jollet, T. Wehling and A. I. Lichtenstein Phys. Rev. B **77**, 205112 (2008)
B. Amadon, Journal of Physics: Condensed Matter **24**, 075604 (2012).

DFT+DMFT: describes correlations



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From the local self-energy to the spectral function

- First step: Analytical continuation using Maximum Entropy Method.
  Self-energy $\Sigma_{m,m'}(i\omega_n)$ (Imaginary frequencies)

  $\downarrow$ Analytical continuation

  Self-energy $\Sigma_{m,m'}(\omega)$ (Real frequencies)
- Second step: Calculation of the lattice Green's function along a k-point path
  - Build Self-energy in the Bloch Basis

$$\Sigma_{\mathbf{k}\nu\nu'}(\omega) = \sum_{mm'} \langle \Psi_{\mathbf{k}\nu} | \chi_m \rangle \Sigma_{m,m'}(\omega) \langle \chi_{m'} | \Psi_{\mathbf{k}\nu'} \rangle .$$

  - Compute Green's function

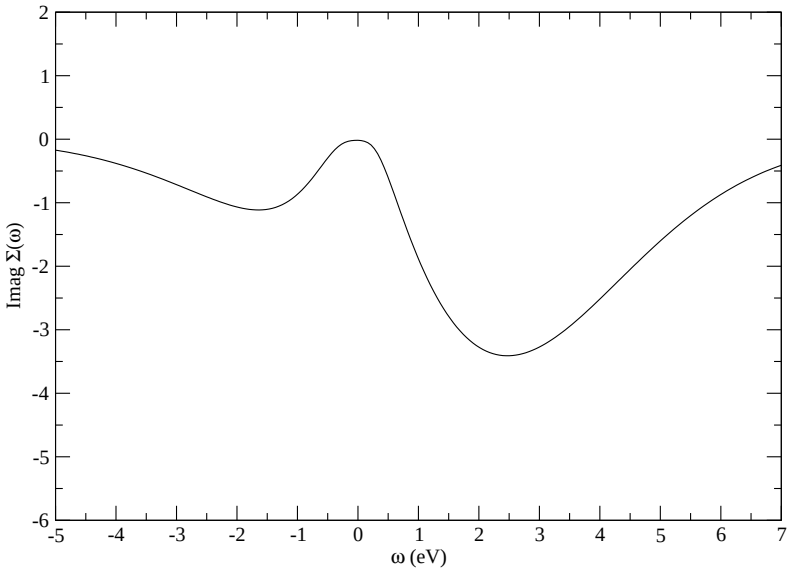
$$G_{\nu\nu'}(\mathbf{k},\omega) = \left[\omega I - H(\mathbf{k}) - \Sigma_{\mathbf{k}}(\omega)\right]^{-1}\Big|_{\nu\nu'}$$

$$A(\omega,\mathbf{k}) = \sum_{\nu} -\frac{1}{\pi}\mathcal{I}G_{\nu\nu}(\mathbf{k},\omega)$$

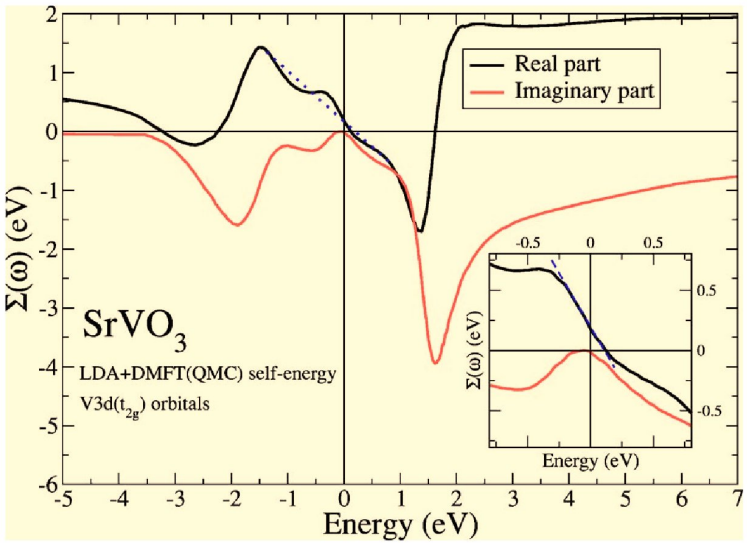
In practice: input and output files

- ABINIT DS1: A DFT calculation
- ABINIT DS2: A usual converged DFT+DMFT calculation should be done.
  Output: O_DS2Self-omega_iatom0001_isppol1
  Output: O_DS2Selfrotformaxent0001_isppol1_iflavor000x
  (Self-energy in the basis that diagonalizes the crystal field)
  Output: O_DS2.UnitaryMatrix_for_DiagLevel (Transformation matrix)
- OmegaMaxent: Analytical continuation using Maximum Entropy
  Input: O_DS2Selfrotformaxent0001_isppol1_iflavor000x
  Output: $\Sigma(\omega) \rightarrow$ O_DS3Self_ra-omega_iatom0001_isppol1
  Output: Freq. Grid $\rightarrow$ I_DS3_spectralfunction_realfrequencygrid
- ABINIT DS3: Calculation of the spectral function
  Input: I_DS3_DEN
  Input: O_DS3Self-omega_iatom0001_isppol1
  Input: O_DS3Self_ra-omega_iatom0001_isppol1
  Input: I_DS3_spectralfunction_realfrequencygrid
  Input: I_DS3.UnitaryMatrix_for_DiagLevel
  Output: O_DS3SpFunc_kresolved_forspectralfunction

Calculation of Self function in DFT+DMFT in SrVO3

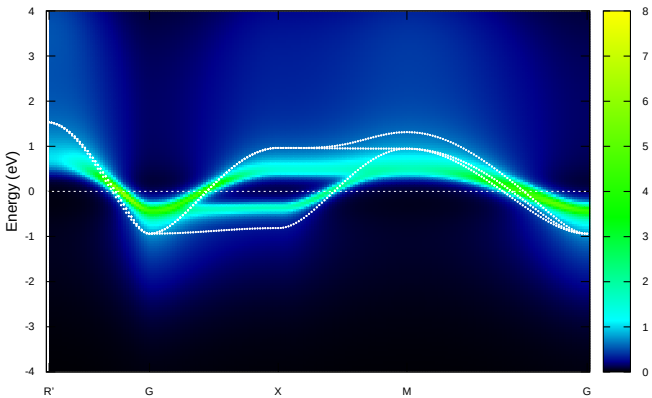


Maxent on Self
This work



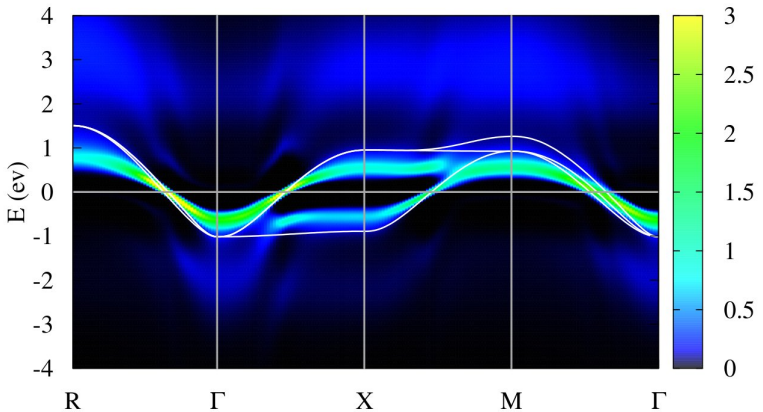
Maxent on G
Nekrassov *et al*
PRB 73 115112 (2006)

Calculation of spectral function in DFT+DMFT



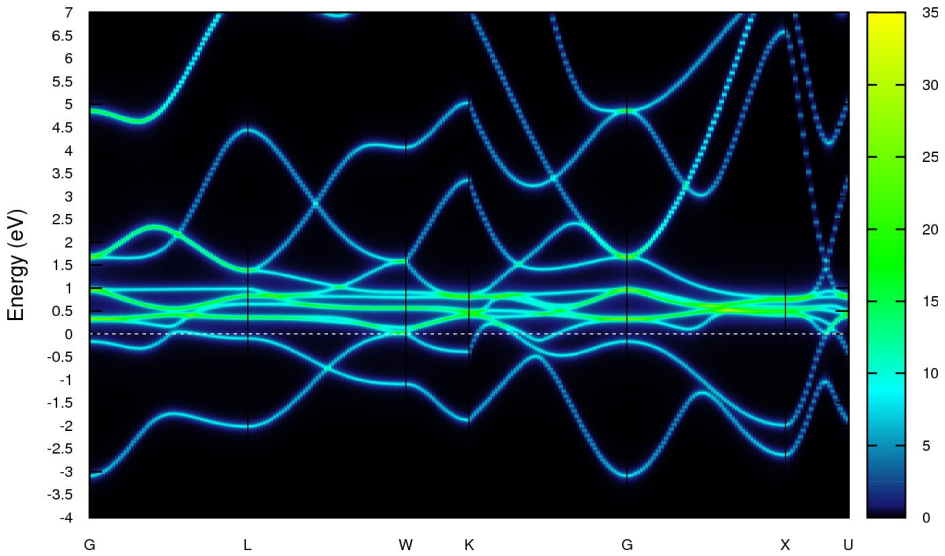
This work

(Same values of U and J)

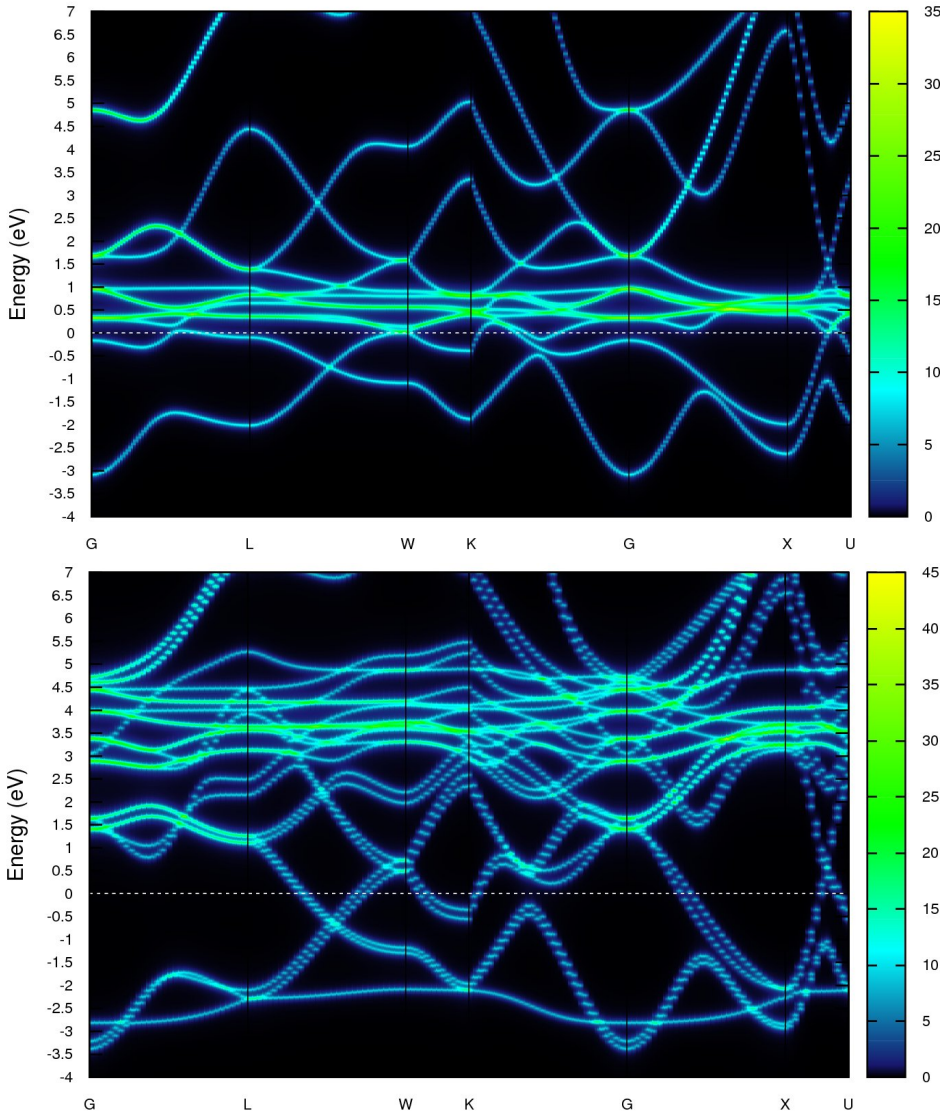


Karolak *et al*
JPCM 23 085601 (2012)

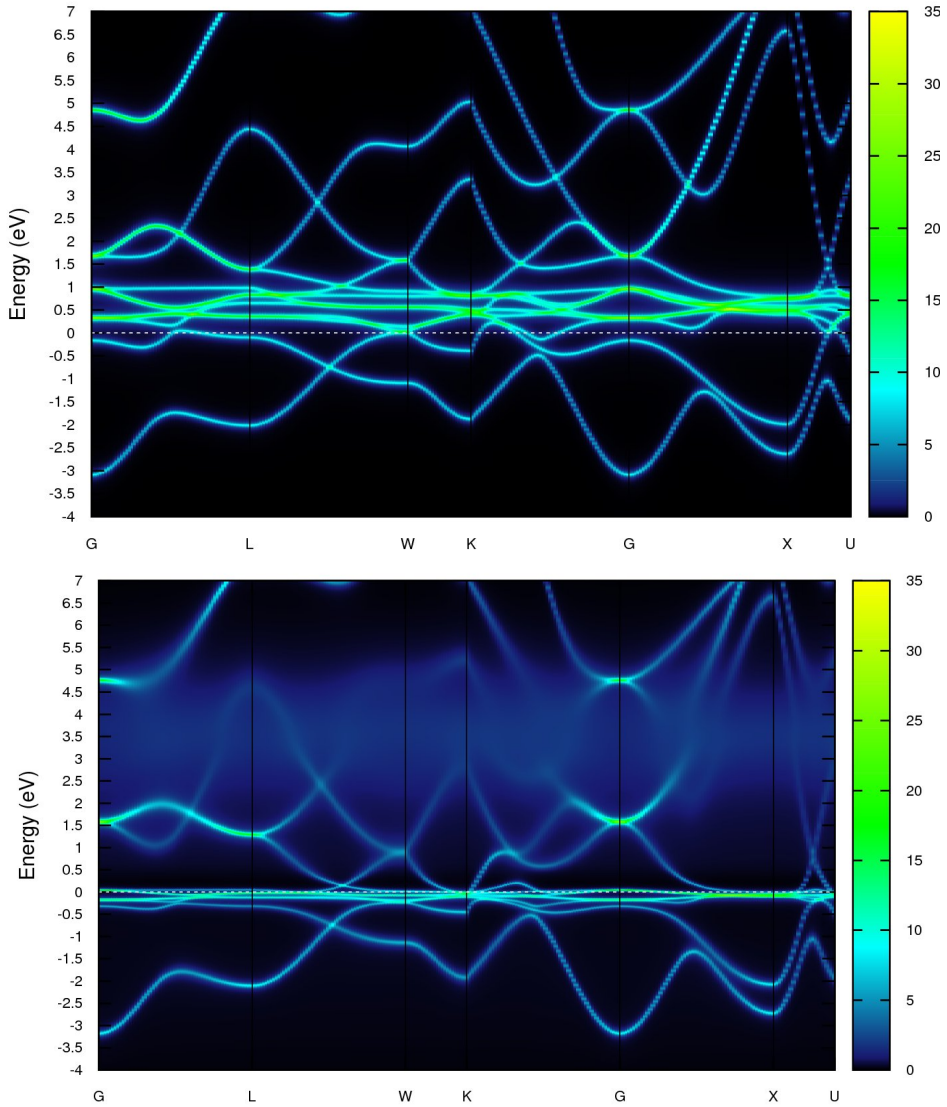
k-resolved spectral function in cerium (LDA)



k-resolved spectral function in cerium LDA and LDA+U



k-resolved spectral function in cerium LDA and LDA+DMFT



Conclusion

- ”charge only DFT”+U can be used in the current version of ABINIT.
- **k**-resolved spectral function is currently in development.
  - Use OmegaMaxent, a code written by Dominic Bergeron (https://www.physique.usherbrooke.ca/MaxEnt/), but other codes are available.
  - Generalization (atoms, spin orbit coupling).
  - Perspective: calculation of frequency dependent conductivity.

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