Generalization of the cRPA scheme : implementation and use. Role of interactions on oxides p orbitals.

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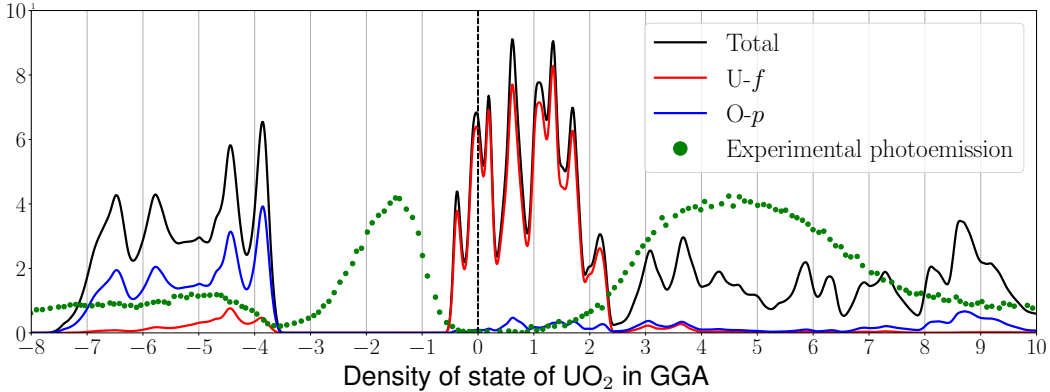
DFT gives a bad description of strongly correlated systems



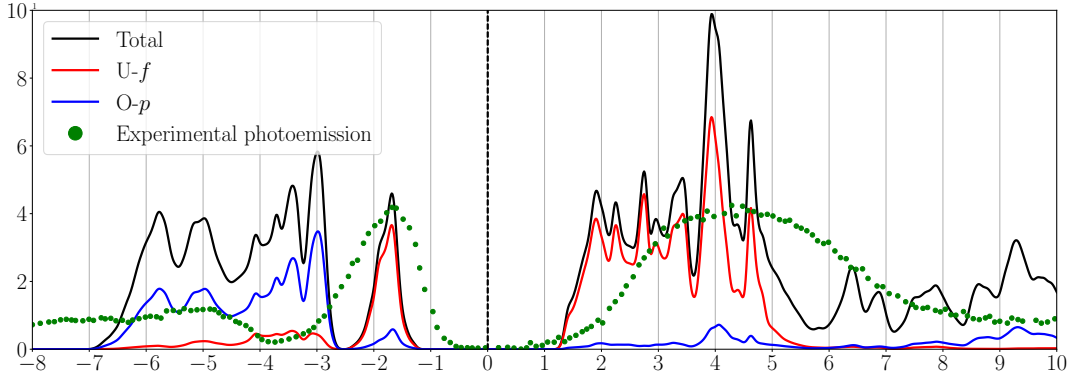
Y. Baer and J. Schoenes. In: *Solid State Communications* 33.8 (1980)

Developed by Anisimov *et al.*
Vladimir I. Anisimov, Jan Zaanen, and Ole K. Andersen. In: *Phys. Rev. B* 44 (1991)
A. I. Liechtenstein, V. I. Anisimov, and J. Zaanen. In: *Phys. Rev. B* 52 (1995)

- Strongly correlated electrons are located on specific orbitals (eg. d or f)
- Explicit interaction limited to those orbitals
- Static (DFT+U) or dynamical (DFT+DMFT) mean field
- Strength of interactions are calculated using U and J

For more on UO_2 see :
S. L. Dudarev et al. In: *physica status solidi (a)* 166.1 (1998)
Boris Dorado et al. In: *Phys. Rev. B* 79 (2009)
Jindřich Kolorenč, Alexander B. Shick, and Alexander I. Lichtenstein. In: *Phys. Rev. B* 92 (2015)

Density of state of UO_2 is corrected using GGA+U with U=5eV



Y. Baer and J. Schoenes. In: *Solid State Communications* 33.8 (1980)

S. L. Dudarev et al. In: *physica status solidi (a)* 166.1 (1998)

1.4 Obtaining U

Experimentally fitted

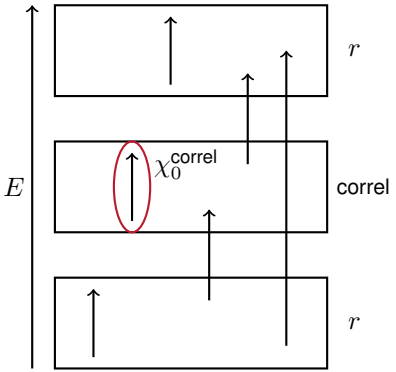
- Limited to known systems
- Not truly *first principle*

Calculated

- constrained LDA
  V. I. Anisimov and O. Gunnarsson. In: *Phys. Rev. B* 43 (1991)
  Matteo Cococcioni and Stefano de Gironcoli. In: *Phys. Rev. B* 71 (2005)
- constrained Random Phase Approximation (**cRPA**)
  F. Aryasetiawan et al. In: *Phys. Rev. B* 70 (2004)

Developed by Aryasetiawan *et al.*

- Screened interaction of correlated electrons
- Electron-hole transition representation
- Separation of χ_0 in two terms
- χ_0^{correl} screening due to correlated electrons
- χ_0^{r} screening due to the rest of the system



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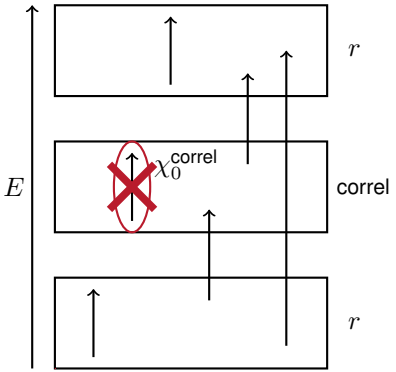


The effective interaction W is calculated using χ_0^r

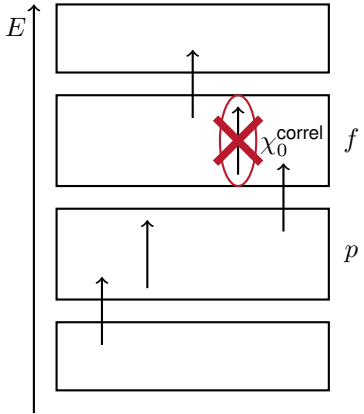
- Need of a local basis to project W because U is a local quantity
- We use the Projected Localized Orbital Wannier functions of `ABINIT`

$$[U]_{m_1,m_2,m_1,m_2} = \langle w_{m_1}^{\mathbf{R}_1 l_1} w_{m_2}^{\mathbf{R}_1 l_1} | W^r(\mathbf{r}, \mathbf{r'}, \omega) | w_{m_1}^{\mathbf{R}_1 l_1} w_{m_2}^{\mathbf{R}_1 l_1} \rangle$$

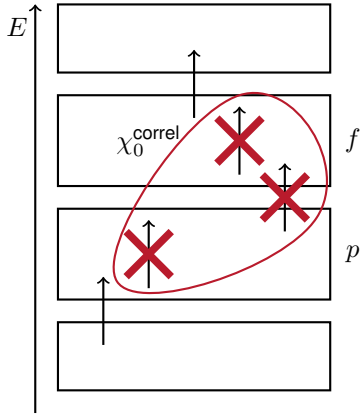
New implementation :

- Enabling any number of effective interaction per system (e.g. U_{ff},U_{pp} and U_{fp} in UO_2)
- Using a separate module for PLO-Wannier
- New data-type for the interaction matrix

Old screening, 1 orbital



New screening, any orbitals



Active screening processes for f and fp cases

Wannier functions are selected by the `plowan` keywords, enabling more than one orbital.

The projection of W^r becomes :

$$[U]^{\mathbf{R}_1,l_1,\mathbf{R_2},l_2}_{m_1,m_2,m_1,m_2} = \langle w^{\mathbf{R}_1 l_1}_{m_1} w^{\mathbf{R_2} l_2}_{m_2} | W^r(\mathbf{r},\mathbf{r}',\omega) | w^{\mathbf{R}_1 l_1}_{m_1} w^{\mathbf{R_2} l_2}_{m_2} \rangle$$

and U is :

$$U = \frac{1}{(2l_1+1)(2l_2+1)} \sum_{m_1=-l_1}^{l_1} \sum_{m_2=-l_2}^{l_2} U_{m_1,m_2,m_1,m_2}$$

In the old implementation :

- $\mathbf{R}_1 = \mathbf{R}_2$ and $l_1 = l_2$ so $m = (-l, +l) \Rightarrow$ (-3,3) for U_{ff}

In the new implementation :

- $\mathbf{R}_1 \neq \mathbf{R}_2$ and as l depend on $\mathbf{R} \Rightarrow l_1 \neq l_2$

The dimension of $[U]$ depend on the values of $l \Rightarrow$ need of nested data-types

The heavy uses of data-types dramatically slow the optimized compilation on Intel compilers

No optimization by default, `HAVE_CRPA_OPTIM` pre-processor macro to activate

Example of UO_2 :

Old implementation

	f	d	p	p'
f	8.56			
d		4.97		
p			9.27	
p'				9.27

$\Rightarrow$

New implementation

	f	d	p	p'
f	8.56	5.73	2.93	2.93
d	5.73	4.97	2.90	2.90
p	2.93	2.90	9.27	2.32
p'	2.93	2.90	2.32	9.27

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Comparison to previous results

	UO_2		
Method	ff	pp	fp
Experimental[1]	4.5		
cRPA fp-model[2]	6.5	6.0	1.9
cRPA fp-model (our work[3])	6.7	5.1	2.3
cRPA f-model (our work[3])	5.0		

[1] Akio Kotani and Takao Yamazaki. In: *Progress of Theoretical Physics Supplement* 108 (1992)
[2] Priyanka Seth et al. In: *Phys. Rev. Lett.* 119 (2017)
[3] Jean-Baptiste Morée, Robinson Outerovitch, and Bernard Amadon. In: *Phys. Rev. B* 103 (2021)

- New implementation of cRPA
  - Available since `ABINIT` v9
  - Increased modularity
- Using generalized cRPA with correlated p orbital
  - Improve description of some system with p orbitals

Perspectives

- Assess the impact of U_{pp} on cRPA
- Full-consistent DFT+U_{ff}+U_{pp}/cRPA
- Implementation of DFT+U with Wannier functions

Any questions ?

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Expression of ϵ

- $\epsilon = 1 - v\chi_0$
- with $\chi_0 = \chi_0^r + \chi_0^{\text{correl}}$
- $\epsilon^{-1} = \frac{1}{1 - v(\chi_0^r + \chi_0^{\text{correl}})}$

Decomposition of ϵ^{-1}

- $\epsilon_r^{-1} = \frac{1}{1 - v\chi_0^r}$
- $\epsilon_{\text{correl}}^{-1} = \frac{1}{1 - W_r \chi_0^{\text{correl}}}$

with $W_r = \frac{v}{1 - v\chi_0^r} = v\epsilon_r^{-1}$

- $|\widetilde{\chi}^{\mathbf{k}}_m\rangle = \sum_{\nu \in W} \langle \Psi^{\mathbf{k}}_\nu | \chi^{\mathbf{k}}_m \rangle |\Psi^{\mathbf{k}}_\nu\rangle$
- Renormalization using overlap matrix
- $|w^{\mathbf{k}}_m\rangle = \sum_\nu C^{\mathbf{k}}_{m\nu} |\Psi^{\mathbf{k}}_\nu\rangle$

A weight w is applied to each transition from ν to ν'

Case n°1 (a) : separate bands

If ν **and** ν' are correlated bands : $w = 0 \Rightarrow$ transition suppressed

Otherwise : $w = 1 \Rightarrow$ transition kept

Case n°2 (b) : intricate bands [4]

The weight is a function of Wannier coefficients $C^{\mathbf{k}}_{m\nu}$

If coefficients are important, then bands are very correlated, $w \approx 0$

If coefficients are low, then bands are not correlated, $w \approx 1$

[4] Ersoy Şaşıoğlu, Christoph Friedrich, and Stefan Blügel. In: *Phys. Rev. B* 83 (2011)