MAX DRIVING THE EXASCALE TRANSITION

JÜLICH Forschungszentrum

Hubbard *U* parameters from constrained random-phase approximation

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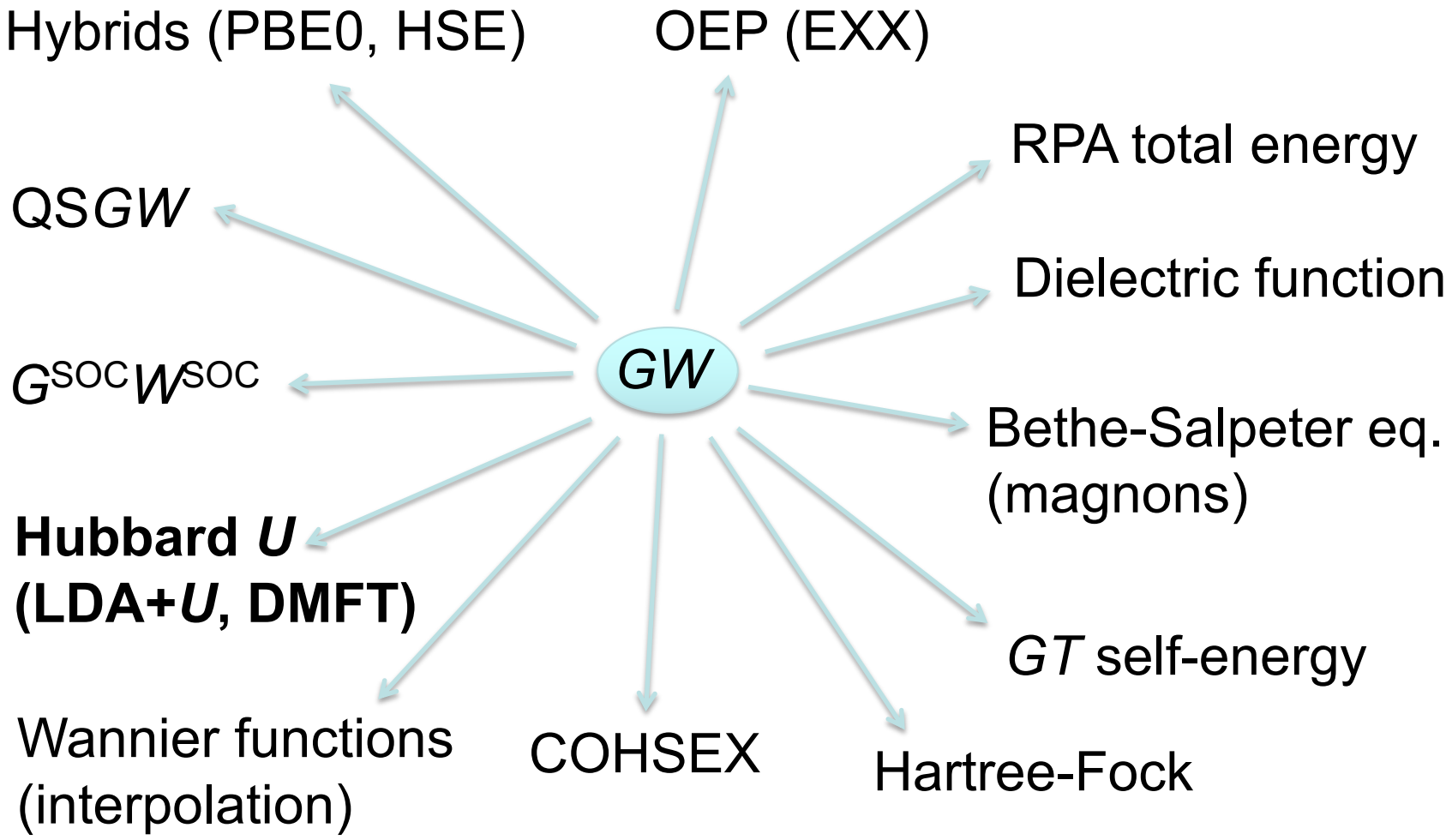
SPEX


Hybrids (PBE0, HSE)
OEP (EXX)
RPA total energy
QS*GW*
Dielectric function
$G^{SOC}W^{SOC}$
GW
Bethe-Salpeter eq. (magnons)
Hubbard *U* (LDA+*U*, DMFT)
GT self-energy
Wannier functions (interpolation)
COHSEX
Hartree-Fock

Correlation strength

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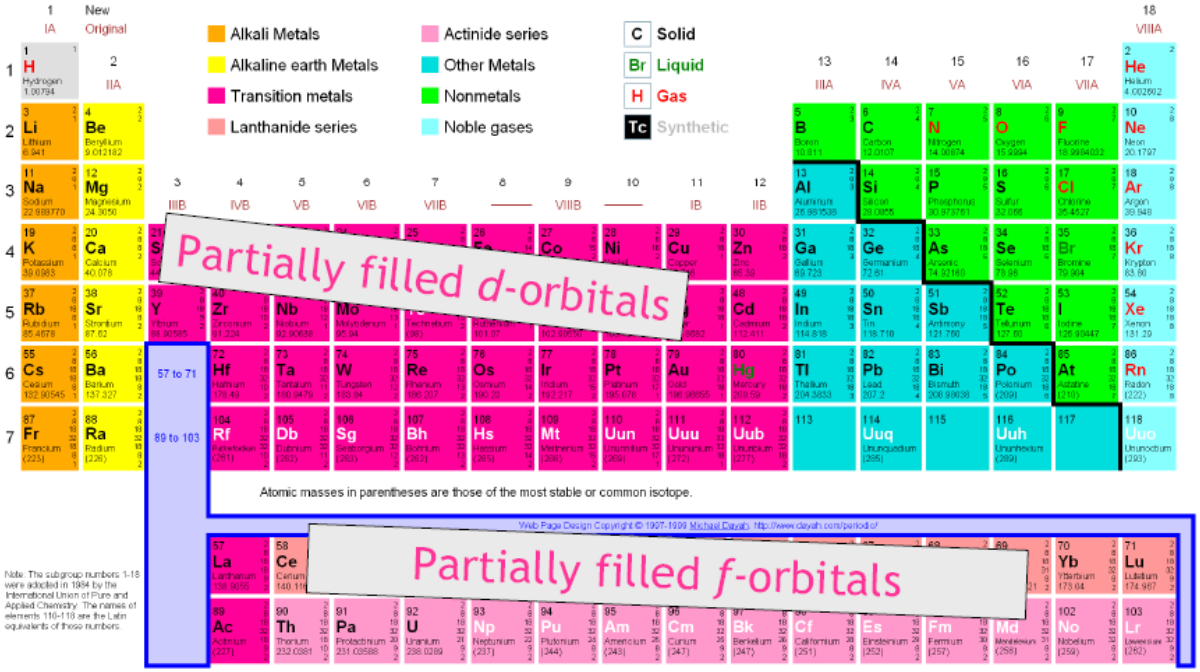
0.1 — 1.0 — 10 — U/t

Alkali metals	Transition metals (TMs)	Semiconductors	TM-Oxides	Rare Earths
Li, Na, K …	Fe, Co, Ni, FePd, …	GaAs, GaN, …	MnO, FeO, …	Gd, Eu, EuO, …

DFT-LDA
***GW*, …….**

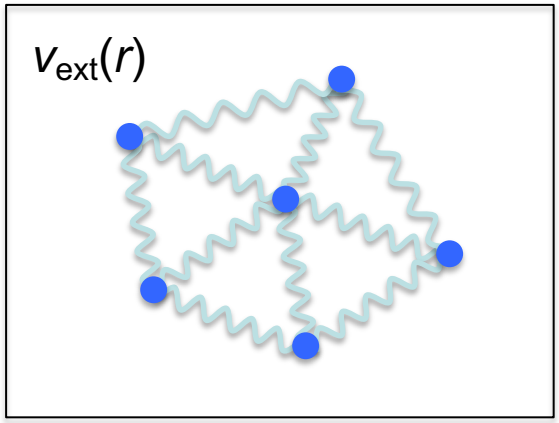
Hubbard Model
LDA+*U*, Hubbard I, LDA+DMFT



Approximation or Downfolding

$$\left[-\frac{1}{2}\sum_i \nabla^2_{\mathbf{r}_i} + \sum_i v_{\text{ext}}(\mathbf{r}_i) + \frac{1}{2}\sum_{i,j}\frac{1}{\mathbf{r}_i - \mathbf{r}_j}\right]\Psi_n(\mathbf{r}_1, \mathbf{r}_2, ...) = E_n\Psi_n(\mathbf{r}_1, \mathbf{r}_2, ...)$$

Approximate Hamiltonian



Downfold Hamiltonian
(correlated subspace and rest)

HF, DFT (LSDA, GGA), *GW*

LDA+*U*, LDA+DMFT, LDA+Gutzwiller

Downfolding

First quantization

$$\left[-\frac{1}{2}\sum_i \nabla^2_{\mathbf{r}_i} + \sum_i v_{\text{ext}}(\mathbf{r}_i) + \frac{1}{2}\sum_{i,j}\frac{1}{\mathbf{r}_i - \mathbf{r}_j}\right]\Psi_n(\mathbf{r}_1, \mathbf{r}_2, ...) = E_n\Psi_n(\mathbf{r}_1, \mathbf{r}_2, ...)$$

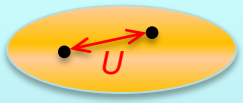
Second quantization

$$\left[-\sum_{\substack{\mathbf{R}\mathbf{R}'\\ ab,\sigma}} t^{ab\sigma}_{\mathbf{R}\mathbf{R}'}\hat{c}^\dagger_{\mathbf{R}a\sigma}\hat{c}_{\mathbf{R}'b\sigma} + \frac{1}{2}\sum_{\substack{\mathbf{R}\mathbf{R}'\mathbf{R}''\mathbf{R}'''\\ abcd,\sigma\sigma'}} V^{abcd}_{\mathbf{R}\mathbf{R}'\mathbf{R}''\mathbf{R}'''}\hat{c}^\dagger_{\mathbf{R}a\sigma}\hat{c}^\dagger_{\mathbf{R}'b\sigma'}\hat{c}_{\mathbf{R}'''d\sigma'}\hat{c}_{\mathbf{R}''c\sigma}\right]\Psi_n = E_n\Psi_n$$

Downfolding → One-band Hubbard model

$$\left[-t\sum_{\langle\mathbf{R},\mathbf{R}'\rangle}\hat{c}^\dagger_{\mathbf{R}}\hat{c}_{\mathbf{R}'} + \epsilon_0\sum_{\mathbf{R}}\hat{c}^\dagger_{\mathbf{R}}\hat{c}_{\mathbf{R}} + U\sum_{\mathbf{R}}\hat{n}_{\mathbf{R}\uparrow}\hat{n}_{\mathbf{R}\downarrow}\right]\Psi_n = E_n\Psi_n$$

Screening



Hubbard *U* parameter: electron-electron interaction and screening by the other electrons

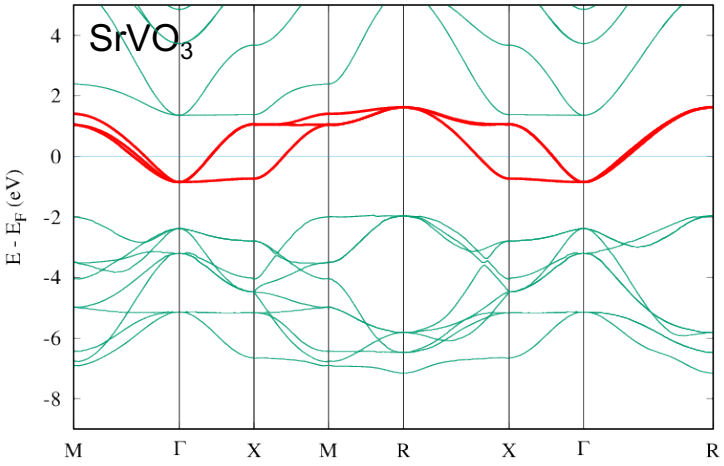
Combining DFT and many-body methods

Correlated Hilbert space

Devide Hilbert space into two parts:

- **Localized Hilbert space (*l* space)**
- **Rest Hilbert space (*r* space)**

DFT (LDA)

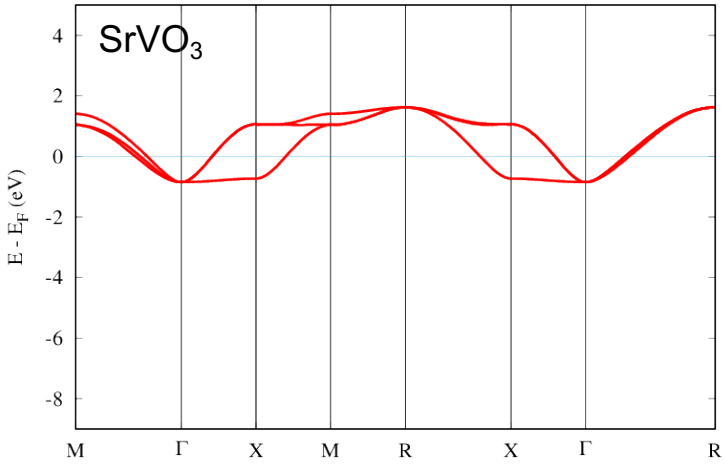


r space

l space

r space

Model Hamiltonian



***l* space:** V3*d* (t_{2g}) → **DFT** ✗

***r* space:** O2*p* + V3*d* (e_g) + ... → **DFT** ✓

Hubbard *U* from first principles

Constrained local-density approximation (cLDA)

[Anisimov and Gunnarsson, PRB 43, 7570 (1991); Cococchioni and de Gironcoli, PRB 71, 035105 (2005)]

$$U = \frac{\partial^2 E}{\partial n_d^2} - \frac{\partial^2 E^{\mathrm{KS}}}{\partial n_d^2}$$

- Easy to implement.
- Cheap computation.
- BUT: not general.

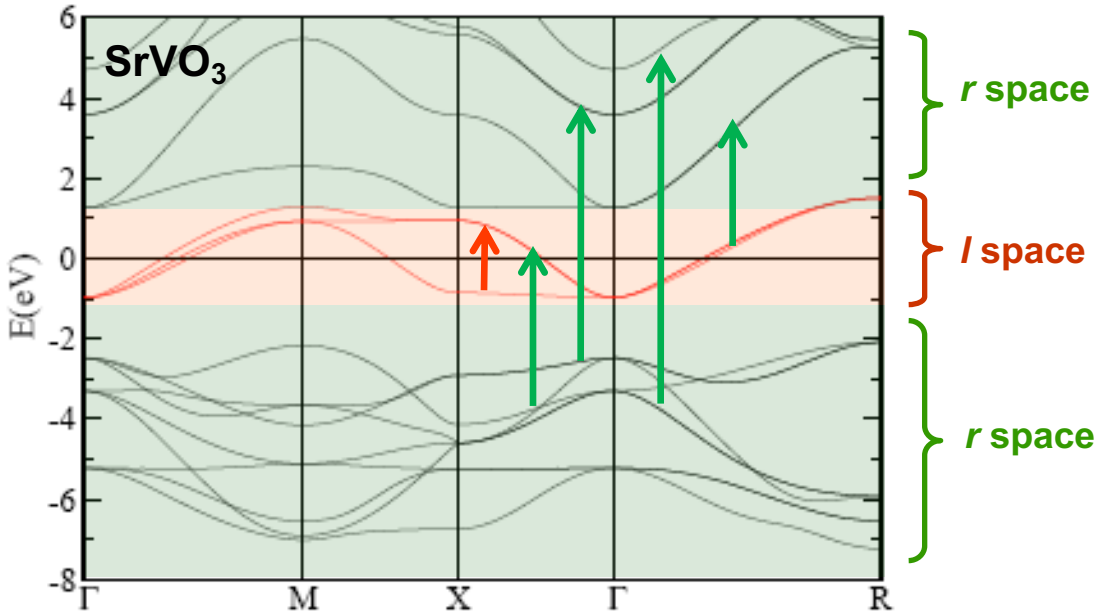
Constrained random-phase approximation (cRPA)

[Springer and Aryasetiawan *et al.*, PRB 57, 4364 (1998); Kotani, J. Phys. Condens. Matter 12, 2413 (2000)]

- Formulation in many-body perturbation theory.
- Frequency dependence $U(\omega)$ accessible.
- Individual matrix elements (*U*, *J*, off-site *U*).
- BUT: more expensive.

Hubbard *U* parameters from constrained random-phase approximation (cRPA)

$$P(\mathbf{r},\mathbf{r}';\omega) = \sum_{m}^{\text{occ}} \sum_{m'}^{\text{unocc}} \phi_m(\mathbf{r})\phi^*_{m'}(\mathbf{r})\phi^*_m(\mathbf{r}')\phi_{m'}(\mathbf{r}') \left[\frac{1}{\omega - \epsilon_{m'} + \epsilon_m + i\eta} - \frac{1}{\omega + \epsilon_{m'} - \epsilon_m - i\eta} \right]$$

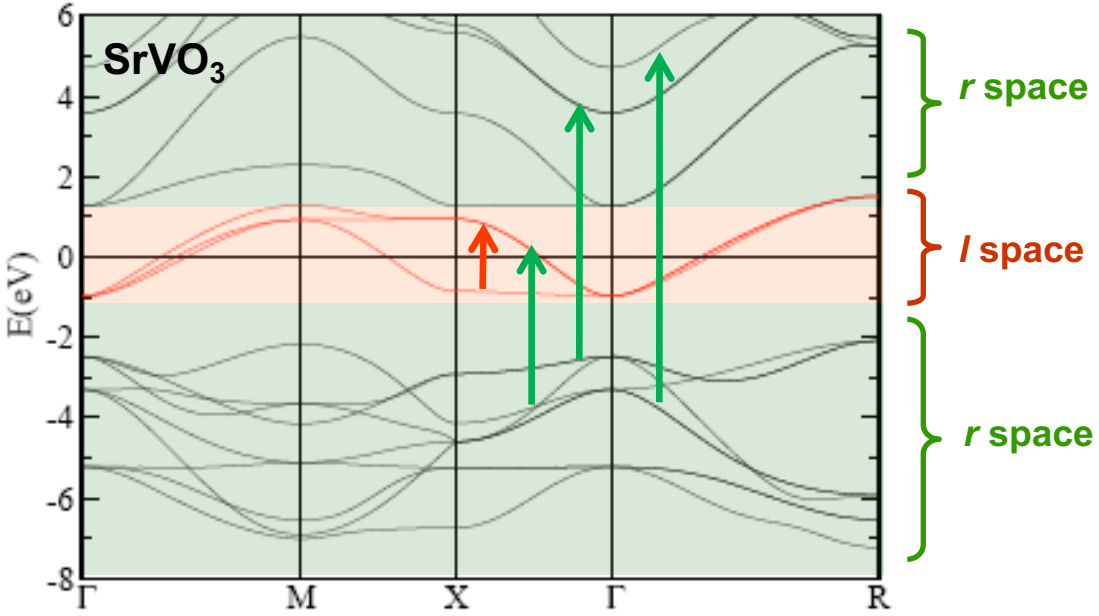


$$P = P_l + P_r$$

$$\sum_{l}^{\text{occ}} \sum_{l'}^{\text{unocc}}$$

$$\sum_{r}^{\text{occ}} \sum_{r'}^{\text{unocc}} + \sum_{r}^{\text{occ}} \sum_{l'}^{\text{unocc}} + \sum_{l}^{\text{occ}} \sum_{r'}^{\text{unocc}}$$

Hubbard *U* parameters from constrained random-phase approximation (cRPA)



$$P = P_l + P_r$$

$$U(\omega) = \frac{v}{1 - vP_r(\omega)}$$

→ projected onto Wannier functions:

$$w_{\mathbf{R}a}(\mathbf{r}) = \frac{1}{N} \sum_{\mathbf{k}m} T^{\mathbf{k}m}_{\mathbf{R}a} \phi_{\mathbf{k}m}(\mathbf{r})$$

transformation matrix *T* defined such that Wannier functions are (maximally) localized

Constrained RPA (cRPA)

- *U* is basis independent
- *U, J,* and off-site *U* easy to calculate
- *U*(ω) accessible
- subspace screening easy to eliminate if bands are **disentangled (!)**

Entangled bands

Projection method

[Sasioglu *et al.*, PRB 83, 121101 (2011)]

$$P(\mathbf{r},\mathbf{r}';\omega) = \sum_{m}^{\text{occ}} \sum_{m'}^{\text{unocc}} \phi_m(\mathbf{r})\phi^*_{m'}(\mathbf{r})\phi^*_m(\mathbf{r}')\phi_{m'}(\mathbf{r}') \left[\frac{1}{\omega - \epsilon_{m'} + \epsilon_m + i\eta} - \frac{1}{\omega + \epsilon_{m'} - \epsilon_m - i\eta} \right] \quad (*)$$

$$P(\mathbf{r},\mathbf{r}';\omega) = \sum_{m}^{\text{occ}} \sum_{m'}^{\text{unocc}} p_m p_{m'} \phi_m(\mathbf{r})\phi^*_{m'}(\mathbf{r})\phi^*_m(\mathbf{r}')\phi_{m'}(\mathbf{r}') \left[\frac{1}{\omega - \epsilon_{m'} + \epsilon_m + i\eta} - \frac{1}{\omega + \epsilon_{m'} - \epsilon_m - i\eta} \right]$$

Disentanglement method

[Miyake *et al.*, PRB 80, 155134 (2009)]

Hybridization between subspace and rest switched off.
→ Bands are disentangled.
→ Equation (*) is applicable.

Effective parameters

Example: Parameters for d states

$$U^{\sigma\sigma'}_{m_1,m_2,m_3,m_4}(\omega)$$

Output file ”spex.cou”

- 625 different matrix elements

“Hubbard-Hund“ parameters (full d shell)

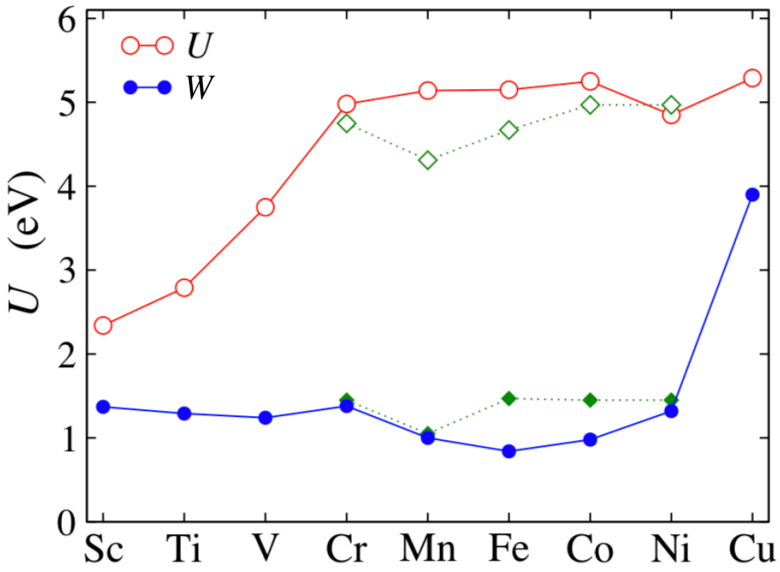
- 15 different matrix elements (or 8)
- 3 are independent

Kanamori parameters (t_{2g} or e_g Hamiltonian)

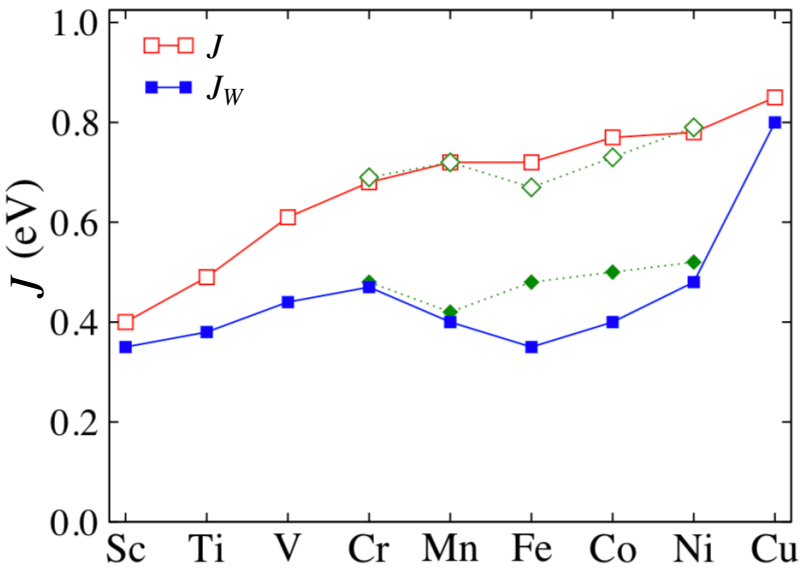
- 3 different matrix elements
- 2 are independent

end of standard output

3d transition metals



E. Sasioglu, Spring School 2014

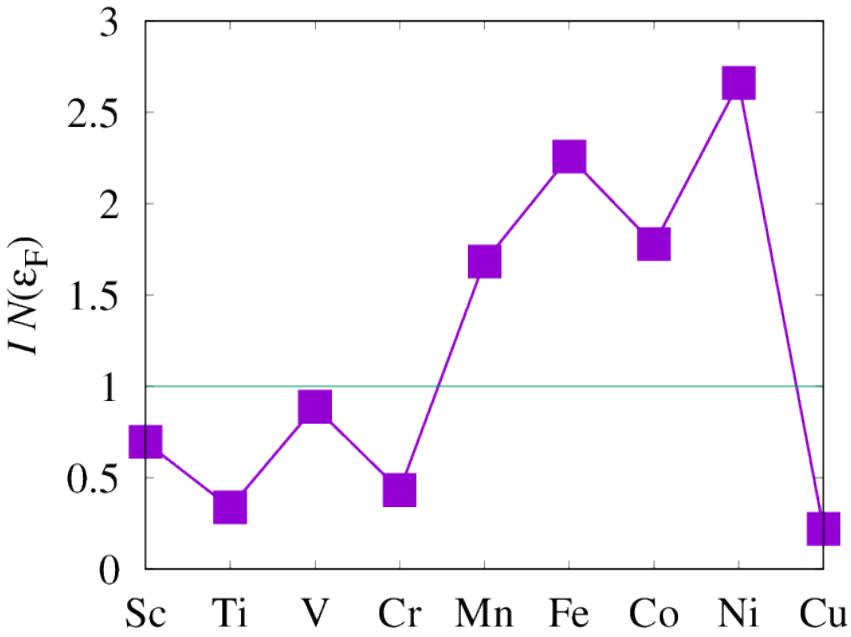


Stoner criterion $I \cdot N(\epsilon_{\mathrm{F}}) > 1$

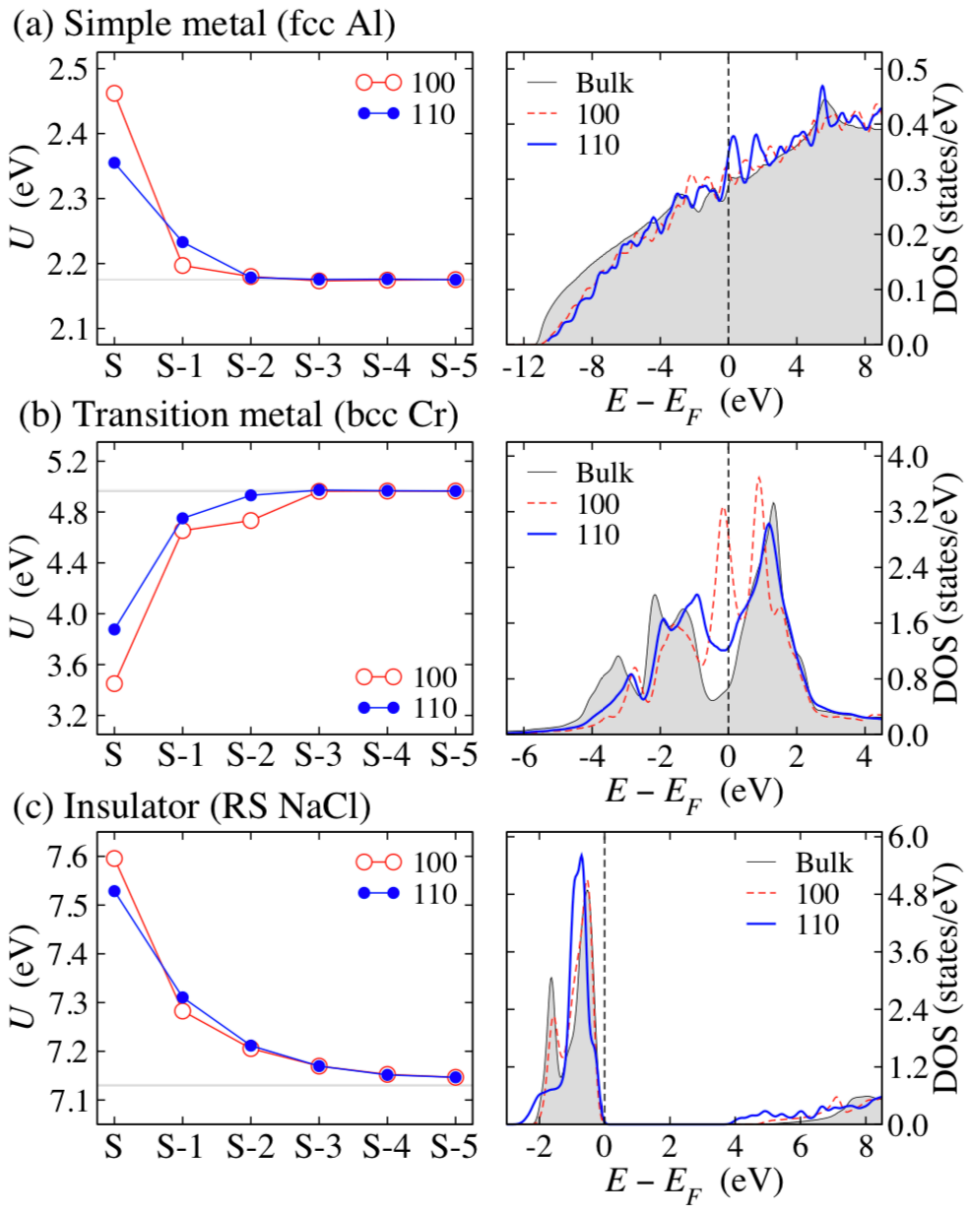
Stollhoff *et al.*, PRB 41, 7028:

$$I = (U + 6J)/5$$

(40% reduction due to correlation)



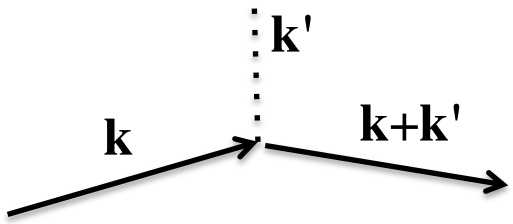
Hubbard *U* at surfaces



Sasioglu *et al.*, PRL 109, 146401 (2012)

Computational procedure
One-Shot *GW*

- FLEUR: Self-consistent field calculation
  ➔ Density, Exchange-correlation potential
- SPEX: Generate special equidistant k-point set
  k, **k'**, **k+k'**, and **0** must be elements

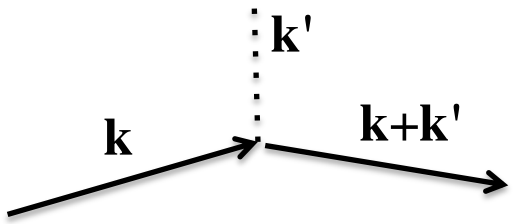


- FLEUR: Diagonalize Hamiltonian on new k points (non iterative)
  ➔ Kohn-Sham energies and wavefunctions
- SPEX: *GW* calculation
  ➔ Quasiparticle energies

`spex.inp:` JOB GW FULL X:(1-4)

Computational procedure
Hubbard *U* (cRPA)

- FLEUR: Self-consistent field calculation
  ➔ Density, Exchange-correlation potential
- SPEX: Generate special equidistant k-point set
  k, **k'**, **k+k'**, and **0** must be elements



- FLEUR: Diagonalize Hamiltonian on new k points (non iterative)
  ➔ Kohn-Sham energies and wavefunctions
- SPEX: cRPA calculation
  - Construction of Wannier orbitals (Wannier90 library used for MLWFs)
  - Calculation of P_r
  - Calculation of *U* and projection onto Wannier basis

  ➔ Hubbard *U* parameters

`spex.inp:` JOB SCREENW {0}

Summary

- For strongly correlated systems, methods like LDA+*U* or LDA+DMFT, which are based on the Hubbard Hamiltonian of a correlated subspace, might be more appropriate than DFT (LDA, GGA) or *GW*.
- These methods require an effective interaction parameter, the Hubbard *U* parameter, which incorporates the screening processes of the electrons that are not included in the correlated subspace.
- The constrained random-phase-approximation (cRPA) is a first-principles method to determine the Hubbard *U* parameter.
- Spex has an implementation of cRPA. Correlated subspace spanned in Wannier basis. Possibility of treating entangled bands.