

Lecture on First-principles Computations (26): The LDA+DMFT scheme

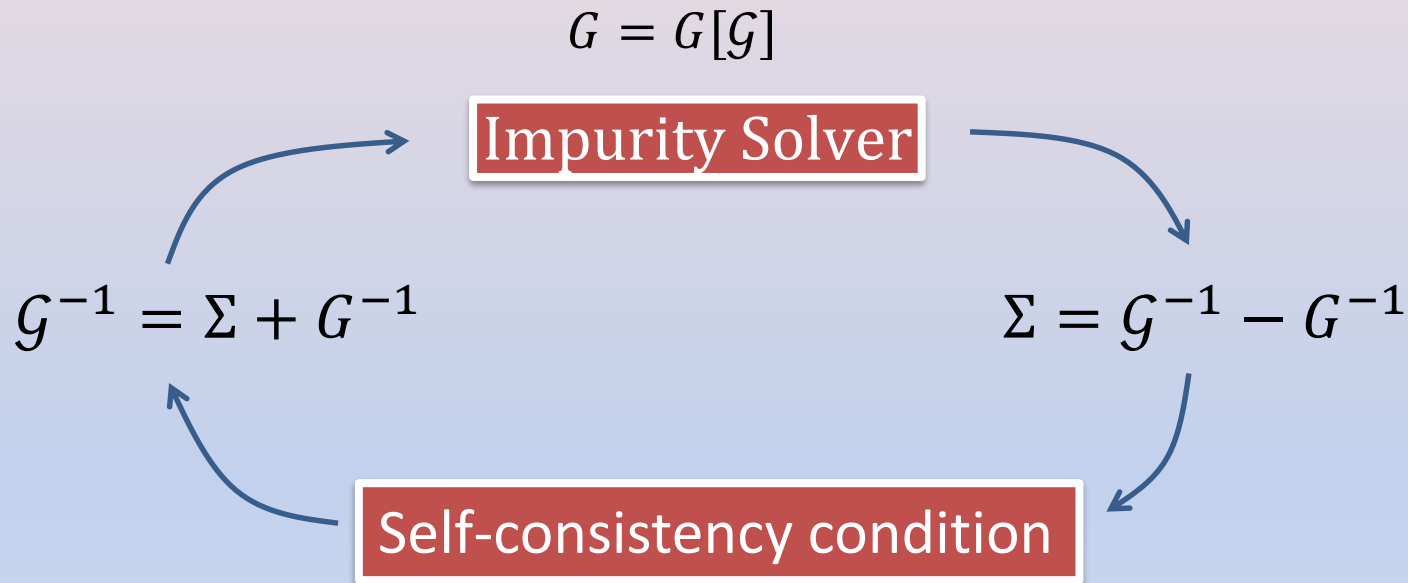
任新国 (Xinguo Ren)

中国科学技术大学
量子信息重点实验室

Key Laboratory of Quantum Information, USTC

Hefei, 2017.12.20

Self-consistency cycle in DMFT



$$G(i\omega_n) = \sum_k \frac{1}{i\omega_n + \mu - \epsilon_k - \Sigma(i\omega_n)}$$

$D=\infty$ Hubbard model = Single-impurity Anderson model
+ self-consistent condition

First-principles approaches versus model Hamiltonian approach

First-principles approach [DFT (LDA/GGAs)]

- + Material-specific properties are accessible
- Good for weakly correlated metals, but **generally fails for strongly correlated systems**
- + fast, computationally efficient software package available

Model Hamiltonian approach [Hubbard, Anderson, Kondo, Heisenberg, t - J , etc.]

- require input model parameters
- + Good for **a qualitative understanding of strongly correlated systems**
- computationally demanding, [e.g., ED, NRG, QMC, DMRG, **DMFT**, etc.]

First-principles approaches versus model Hamiltonian approach

First-principles approach
[DFT (LDA/GGAs)]

Model Hamiltonian approach
[Hubbard, Anderson, Kondo,
Heisenberg, t - J , etc.]

+ Materials
are a

It would be great to combine both worlds!

eters

- Good for weakly correlated metals, but **generally fails for strongly correlated systems**

+ fast, computationally efficient software package available

- Good for a **quantitative understanding of strongly correlated systems**

- computationally demanding, [e.g., ED, NRG, QMC, DMRG, **DMFT**, etc.]

The concept of DFT(LDA)+DMFT

V. I. Anisimov et al., J. Phys. Condens. Matter **9**, 7359 (1997)

A. I. Lichtenstein and M. I. Katsnelson, Phys. Rev. B **57**, 6884 (1998).

Solving the interacting many-body problem in three steps:

1. Perform a DFT(LDA) calculation for a real material, and obtain the DFT(LDA) band structure, or in general the noninteracting KS Hamiltonian $H_{\text{LDA}}(\mathbf{k})$.
2. Complement $H_{\text{LDA}}(\mathbf{k})$ with an explicit Hubbard-type local interaction term for **the physically relevant orbitals**, and subtract the “double-counting (dc)” term (that is already included in LDA)
3. Solve the modified Hamiltonian $\hat{H}_{\text{LDA}} + \hat{H}_I^{\text{local}} - \hat{H}_{dc}$ within DMFT.

The modified realistic “model Hamiltonian”

$$\hat{H} = \sum_{ij} h_{ij}^0 \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + \frac{1}{2} \sum_{ijkl,\sigma,\sigma'} V_{ijkl} \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma'}^\dagger \hat{c}_{l\sigma'} \hat{c}_{k\sigma}$$

Local-orbital Basis: $\phi_i = \phi_{a,l,m}(\mathbf{r} - \mathbf{r}_a - \mathbf{R}_i)$ ($i = \mathbf{R}, a, l, m$)

From the DFT(LDA) calculation, one gets

$$\hat{H}_{\text{LDA}} = \sum_{n,\mathbf{k},\sigma} \epsilon_{n,\mathbf{k},\sigma} \hat{c}_{n,\mathbf{k},\sigma}^\dagger \hat{c}_{n,\mathbf{k},\sigma} = \sum_{alm;a'l'm'} H_{alm;a'l'm'}^{\text{LDA}}(\mathbf{k}) \hat{c}_{a,l,m,\sigma}^\dagger(\mathbf{k}) \hat{c}_{a',l',m',\sigma'}(\mathbf{k})$$

The most relevant orbitals are those across the Fermi level (e.g. d orbitals for transition metals and their oxides)

$$a = a_d; l = 2; m = xy, xz, yz, x^2 - y^2, 3z^2 - r^2$$

The modified “realistic Hamiltonian”

From the DFT(LDA) calculation:

$$\hat{H}_{\text{LDA}} = \sum_{n,\mathbf{k},\sigma} \epsilon_{n,\mathbf{k},\sigma} \hat{c}_{n,\mathbf{k},\sigma}^\dagger \hat{c}_{p,\mathbf{k},\sigma} =$$

$$\sum_{alm;a'l'm'} H_{alm;a'l'm'}^{\text{LDA}}(\mathbf{k}) \hat{c}_{a,l,m,\sigma}(\mathbf{k})^\dagger \hat{c}_{a',l',m',\sigma'}(\mathbf{k})$$

$$a = a_d; l = 2; m = xy, xz, yz, x^2 - y^2, 3z^2 - r^2$$

Adding the multi-orbital Hubbard-type interaction:

$$\hat{H}_I = U \sum_{m\sigma} \hat{n}_{m\sigma} \hat{n}_{m\bar{\sigma}} + \frac{V}{2} \sum_{m \neq m', \sigma} \hat{n}_{m\sigma} \hat{n}_{m'\bar{\sigma}} + \frac{(V-J)}{2} \sum_{m \neq m', \sigma} \hat{n}_{m\sigma} \hat{n}_{m'\sigma}$$

$$\hat{H}_{dc} = \bar{U} \hat{n}_d - \bar{U}/2 \quad \bar{U} = \frac{U + (m-1)V + (m-1)(V-J)}{(2m-1)}$$

$$\hat{n}_d = \sum_{m\sigma} \hat{n}_{m\sigma}$$

The modified “realistic Hamiltonian”

$$\hat{H}_{\text{LDA}} = \sum_{alm;a'l'm'} H_{alm;a'l'm'}^{\text{LDA}}(\mathbf{k}) \hat{c}_{a,l,m,\sigma}(\mathbf{k})^\dagger \hat{c}_{a',l',m',\sigma'}(\mathbf{k})$$

$$a = a_d; l = 2; m = xy, xz, yz, x^2 - y^2, 3z^2 - r^2$$

$$\hat{H}_I = U \sum_{m\sigma} \hat{n}_{m\sigma} \hat{n}_{m\bar{\sigma}} + \frac{V}{2} \sum_{m \neq m', \sigma} \hat{n}_{m\sigma} \hat{n}_{m'\bar{\sigma}} + \frac{(V-J)}{2} \sum_{m \neq m', \sigma} \hat{n}_{m\sigma} \hat{n}_{m'\sigma}$$

$$\hat{H}_{dc} = \bar{U} \hat{n}_d - \bar{U}/2 \quad (U, V, J \text{ can be calculated using constrained LDA, constrained RPA, or linear response method})$$

Finally solve the Hamiltonian:

$$\hat{H} = \hat{H}_{\text{LDA}} - \hat{H}_{dc} + \hat{H}_I = \hat{H}_0 + \hat{H}_I$$

using DMFT !

Projection to “correlated space”: “Downfolding”

$$W_{nk}(\mathbf{r}) = \sum_{E_1 < E_{mk} < E_2} \psi_{mk}(\mathbf{k}) U_{mn}(\mathbf{k})$$

$\{|W_{nk}\rangle\}$ expands the “correlated subspace”

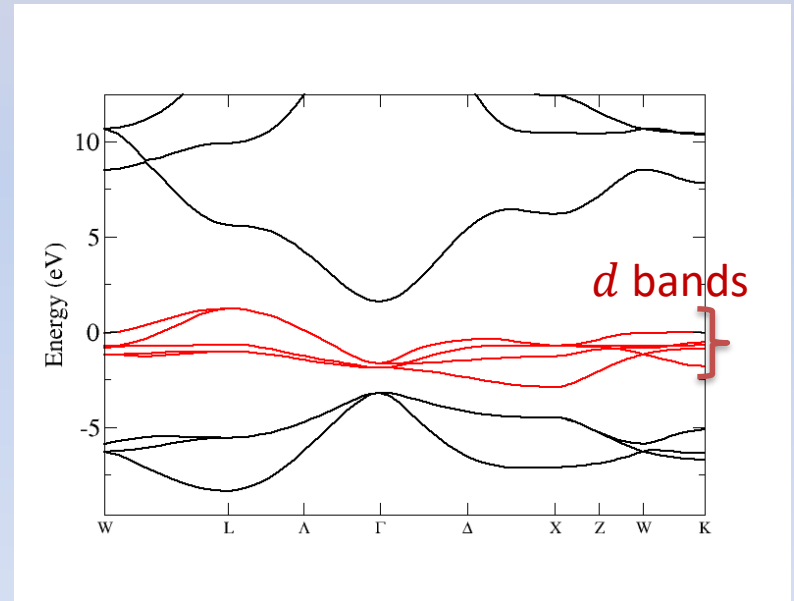
LDA Hamiltonian in
the “correlated subspace”:

$$\begin{aligned} H_{mn}(\mathbf{k}) &= \langle W_{mk} | \hat{H}_{\text{LDA}} | W_{nk} \rangle \\ &= \sum_l \epsilon_{lk} \langle W_{mk} | \psi_{lk} \rangle \langle \psi_{lk} | W_{nk} \rangle \\ &= \sum_l \epsilon_{lk} U_{lm}^*(\mathbf{k}) U_{ln}(\mathbf{k}) \end{aligned}$$

Finally solve the Hamiltonian:

$$\hat{H} = \sum_{\mathbf{k}} H_{mn}(\mathbf{k}) - \hat{H}_{dc} + \hat{H}_I$$

using DMFT !



The LDA+DMFT self-consistency loop

On-site Green function:

$$G_{alm,a'l'm'}(i\omega_n) = \frac{1}{\Omega_B} \int d\mathbf{k} [(i\omega + \mu) - H_0(\mathbf{k}) - \Sigma(i\omega_n)]_{alm,a'l'm'}^{-1}$$

$$\Sigma_{alm,a'l'm'}(i\omega) = \begin{cases} \Sigma_{m,m'}(i\omega), & \text{if } a = a' = a_d, l = l' = d \\ 0, & \text{else} \end{cases}$$

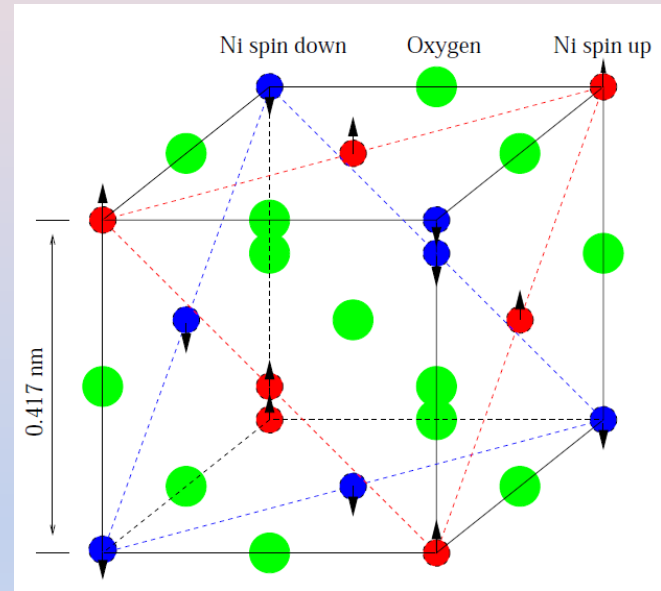
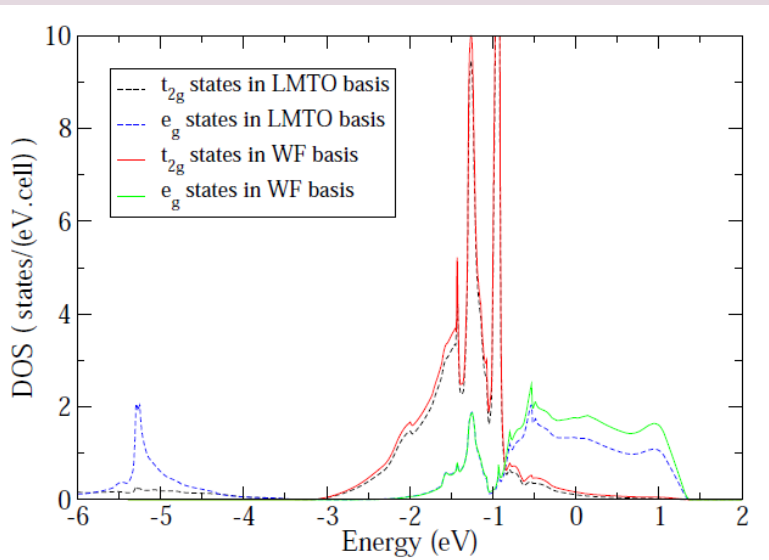
Impurity Green function: $G_{mm'}^{imp}(i\omega_n) = G_{a_d d m, a_d d m'}(i\omega_n)$

Bath Green function: $\mathcal{G}_{m,m'}(i\omega_n) = [(G^{imp}(i\omega_n))^{-1} + \Sigma(i\omega_n)]_{mm'}^{-1}$

$\mathcal{G}_{m,m'}(i\omega_n)$ → Impurity Solver → $\Sigma^{new}(i\omega)$

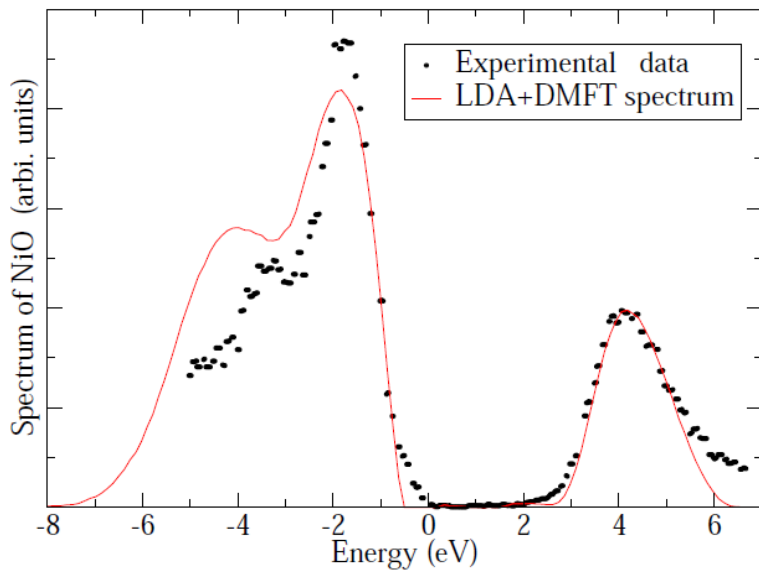


Application: NiO



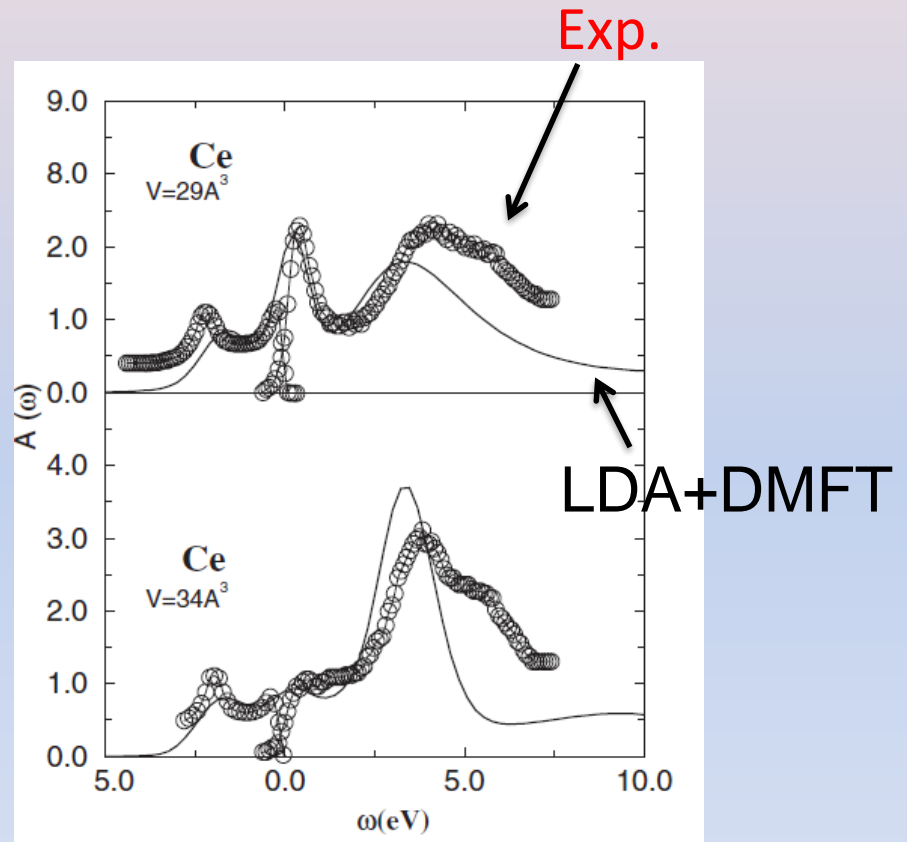
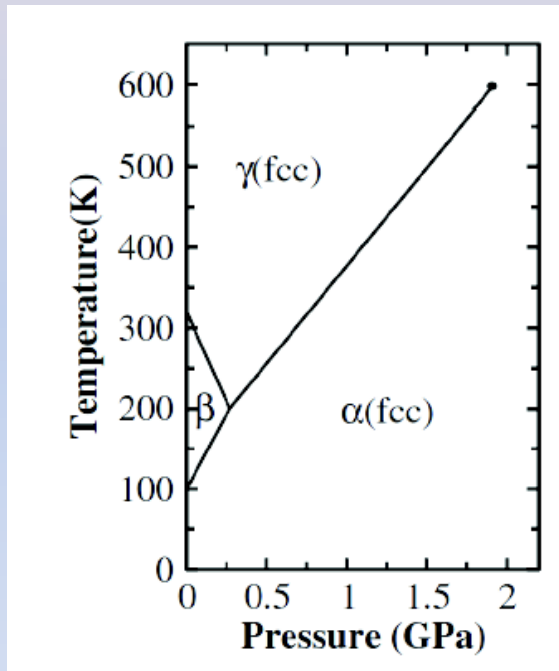
LDA+DMFT can describe
the Mott insulator!

X. Ren et al., Phys. Rev. B,
74, 195114 (2006)



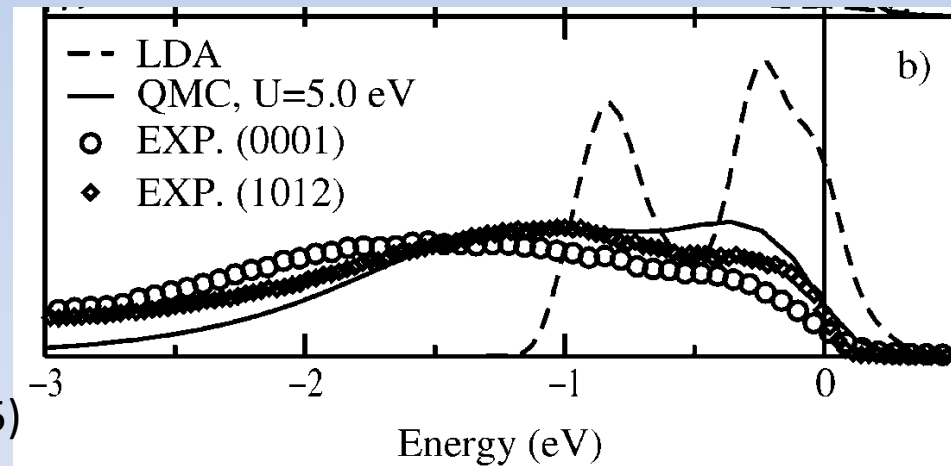
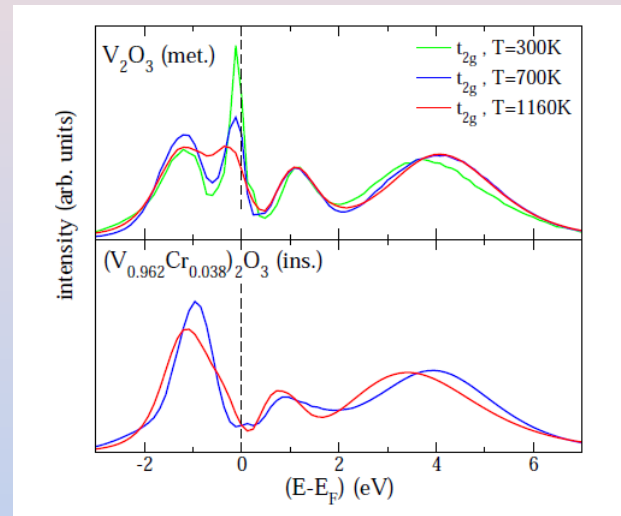
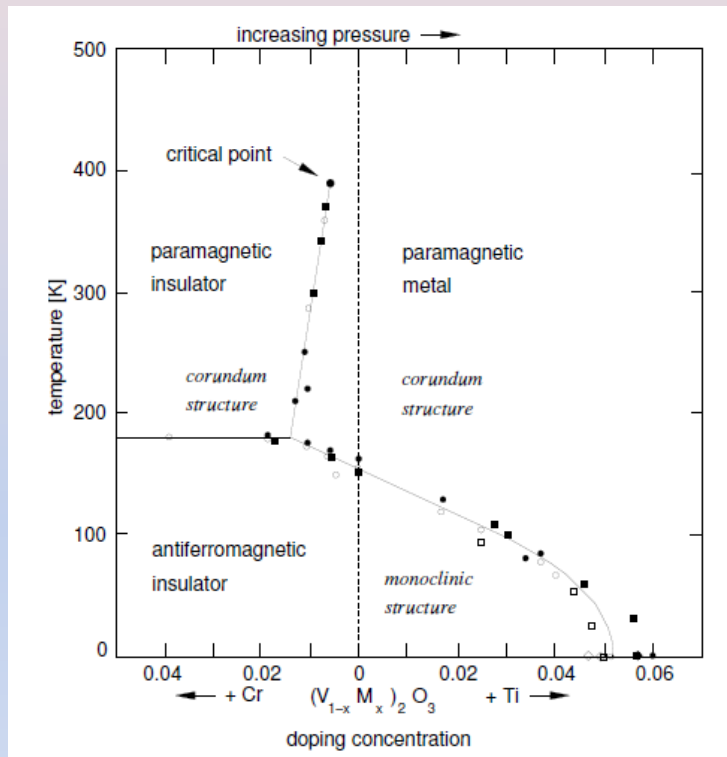
Application: phase transition in Ce

Phase diagram



Held et al., Phys. Rev. Lett. **85**, 373 (2000)

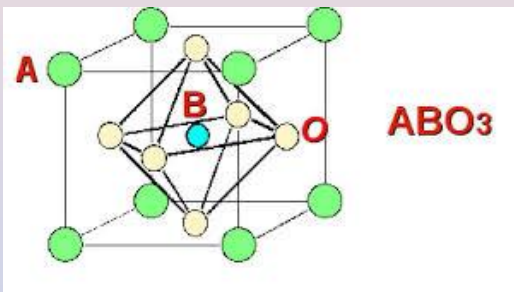
Metal insulator transition in V_2O_3



K. Held et al., Phys. Rev. Lett. 86, 5345 (2001).

G. Keller, PhD thesis (Augsburg University, 2015)

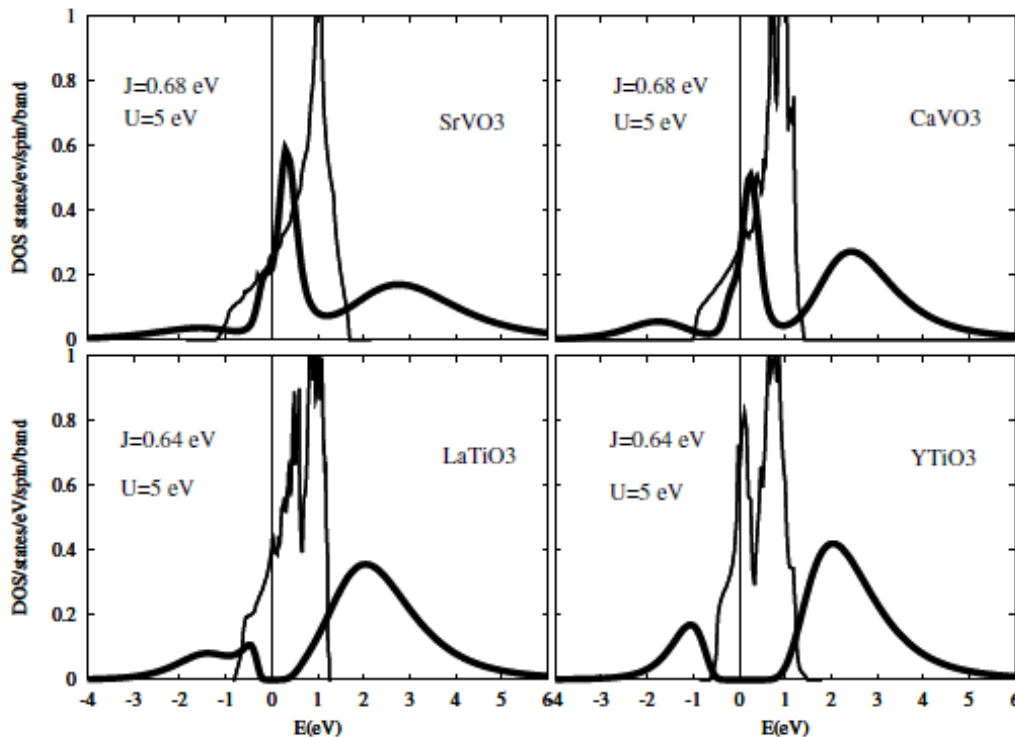
Mott transition in perovskite oxides



ABO_3 (钙钛矿结构)

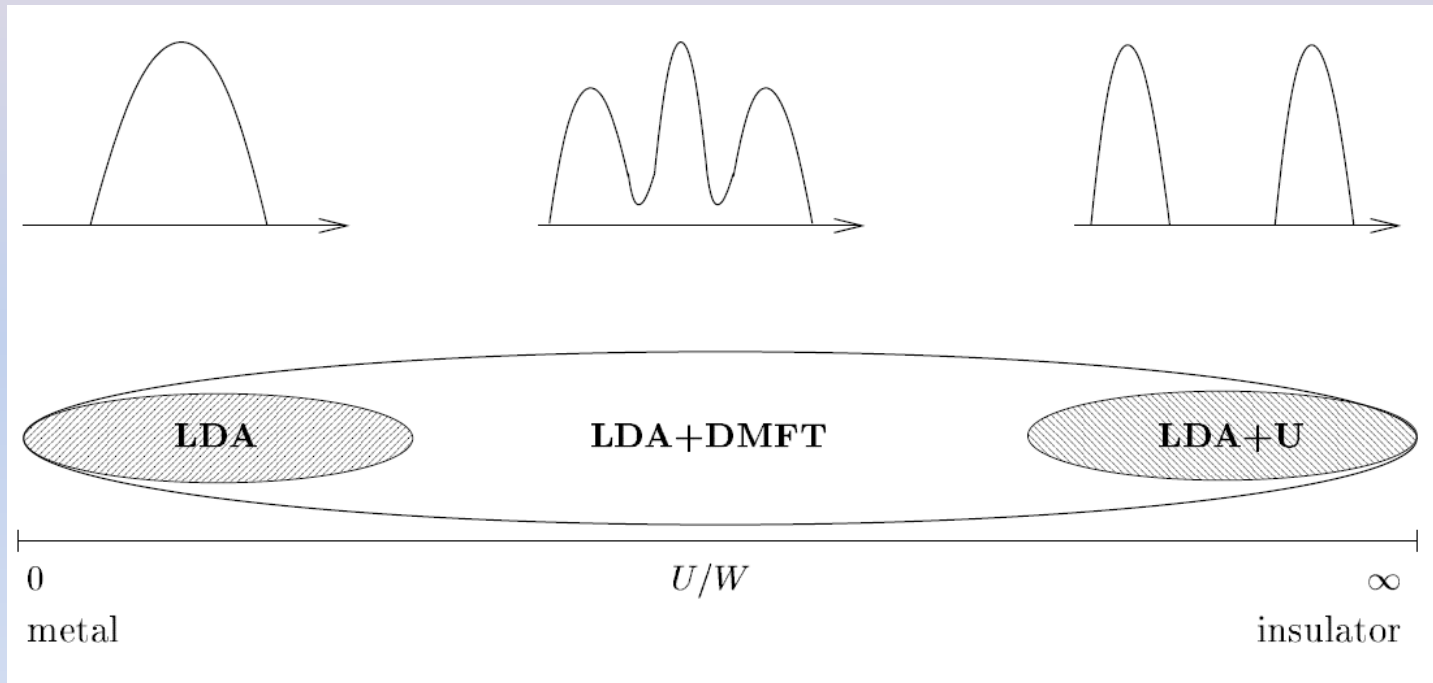
Periodic Table of Elements

1	2	3	4	5	6	7	8	9	10	11	12	13	14				
1 H Hydrogen 1.00794	2 He Helium 4.002602											3 Li Lithium 6.941	4 Be Beryllium 9.012182				
5 B Boron 10.811	6 C Carbon 12.0107											7 N Nitrogen 14.00643	8 O Oxygen 15.999				
9 F Fluorine 18.9984032	10 Ne Neon 20.1797											11 Na Sodium 22.98976928	12 Mg Magnesium 24.304				
13 Al Aluminum 26.9815386	14 Si Silicon 28.08558											15 P Phosphorus 30.973761998	16 S Sulfur 32.06				
17 Cl Chlorine 35.453	18 Ar Argon 39.948											19 K Potassium 39.0983	20 Ca Calcium 40.078				
21 Sc Scandium 44.955912	22 Ti Titanium 47.88	23 V Vanadium 50.9415	24 Cr Chromium 51.9961	25 Mn Manganese 54.938044	26 Fe Iron 55.845	27 Co Cobalt 58.933195	28 Ni Nickel 58.6934	29 Cu Copper 63.546	30 Zn Zinc 65.38	31 Ga Gallium 69.723	32 Ge Germanium 72.64						
33 As Arsenic 74.9216	34 Se Selenium 78.96	35 Br Bromine 79.904	36 Kr Krypton 83.80	37 Rb Rubidium 85.4678	38 Sr Strontium 87.62	39 Y Yttrium 88.90584	40 Zr Zirconium 91.224	41 Nb Niobium 92.90638	42 Mo Molybdenum 95.94	43 Tc Technetium 97.9072	44 Ru Ruthenium 101.07	45 Rh Rhodium 102.9055	46 Pd Palladium 106.42	47 Ag Silver 107.8682	48 Cd Cadmium 112.411	49 In Indium 114.818	50 Sn Tin 118.710
51 Sb Antimony 121.757	52 Te Tellurium 127.6	53 I Iodine 126.90547	54 Xe Xenon 131.29	55 Cs Cesium 132.90545196	56 Ba Barium 137.327	57-71 La-Lu Lanthanides 89-103	72 Hf Hafnium 178.49	73 Ta Tantalum 180.94788	74 W Tungsten 183.84	75 Re Rhenium 186.207	76 Os Osmium 190.23	77 Ir Iridium 192.222	78 Pt Platinum 195.084	79 Au Gold 196.966569	80 Hg Mercury 200.59	81 Tl Thallium 204.3833	82 Pb Lead 207.2
83 Bi Bismuth 208.980389	84 Po Polonium [209]	85 At Astatine [210]	86 Rn Radon [222]	87 Fr Francium [223]	88 Ra Radium [226]	89-103 La-Lu Lanthanides	104 Rf Rutherfordium [261]	105 Db Dubnium [262]	106 Sg Seaborgium [263]	107 Bh Bohrium [264]	108 Hs Hassium [265]	109 Mt Meitnerium [266]	110 Ds Darmstadtium [271]	111 Rg Roentgenium [272]	112 Uub Ununbium [277]	113 Uut Ununtrium [284]	114 Uuq Ununquadium [285]

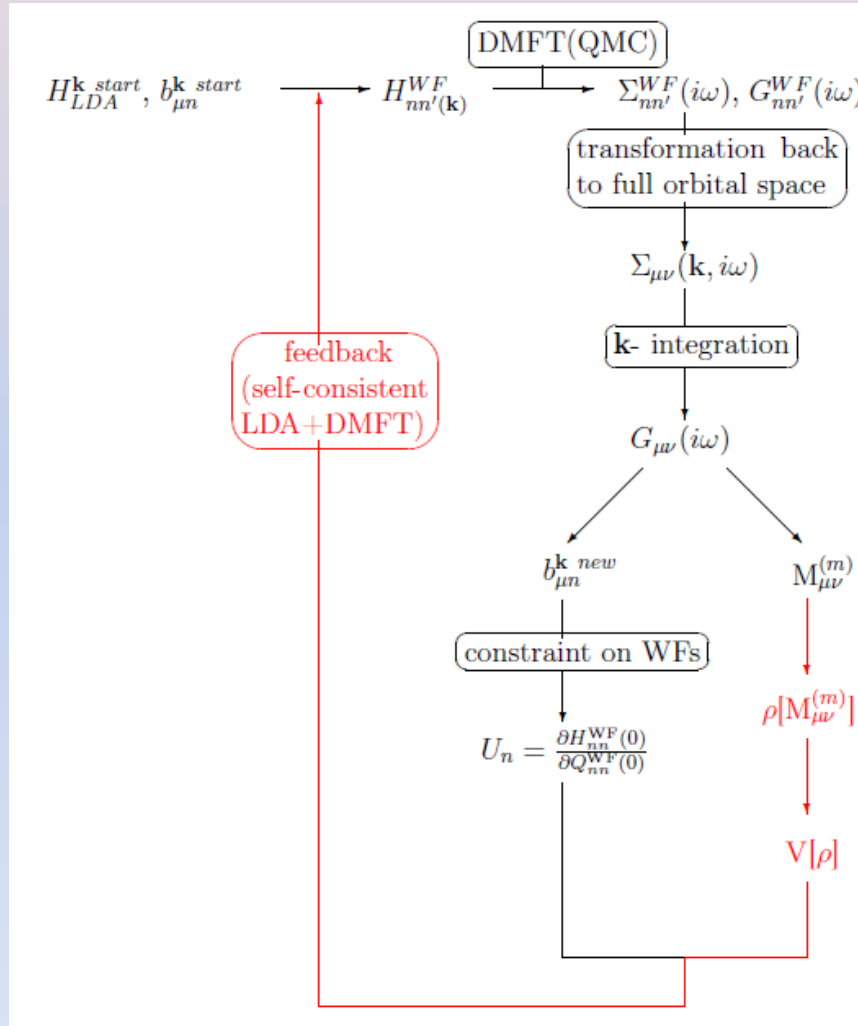


E. Pavarini et al., Phys. Rev. Lett. 92, 176403 (2004)

Comparing LDA+DMFT to LDA and LDA+U



Self-consistent LDA+DMFT



Flow diagram for one possible self-consistent LDA+DMFT implementation

Review papers

K. Held, “*Electronic structure calculations using dynamical mean-field theory*”, Adv. Phys. **56**, 829-926 (2007)

A. Georges, G. Kotliar, W. Krauth, M. J. Rozenberg, “*Dynamical mean-field theory of strongly correlated fermionic systems and the limit of infinite dimensions*”, Rev. Mod. Phys. **68**, 13 (1996).