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

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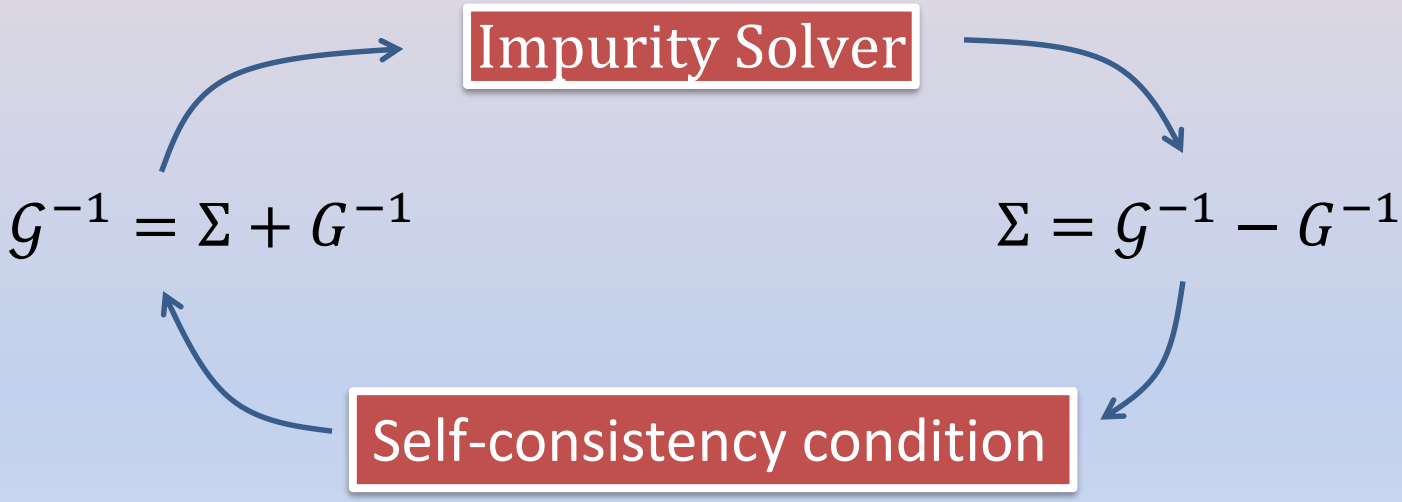
# Self-consistency cycle in DMFT

$$G = G[\mathcal{G}]$$




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

$D=\infty$ 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. ]

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It would be great to combine both worlds!

# The concept of DFT(LDA)+DMFT

*V. I. Anisimov et al., J. Phys. Conden. 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 $H_{\mathrm{LDA}}(\boldsymbol{k})$.

2. Complement $H_{\mathrm{LDA}}(\boldsymbol{k})$ 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 $\hat{H}_{\mathrm{LDA}} + \hat{H}_I^{local} - \hat{H}_{dc}$ 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: $\phi_i = \phi_{a,l,m}(\boldsymbol{r} - \boldsymbol{r_a} - \boldsymbol{R_i})\ (i = \boldsymbol{R}, a, l, m)$

From the DFT(LDA) calculation, one gets

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

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

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:

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

$$\sum_{alm;a\prime l\prime m\prime} H^{\mathrm{LDA}}_{alm,a\prime l\prime m\prime}(\mathbf{k}) \hat{c}_{a,l,m,\sigma}(\mathbf{k})^{\dagger} \hat{c}_{a\prime,l\prime,m\prime,\sigma\prime}(\mathbf{k})$$

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

Adding the multi-orbital Hubbard-type interaction:

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

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

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

# The modified "realistic Hamiltonian"

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

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

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

$$\hat{H}_{dc} = \bar{U}\hat{n}_d - \bar{U}/2$$

(*U, V, J* can be calculated using constrained LDA, constrained RPA, or linear response method)

Finally solve the Hamiltonian:

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

using DMFT !

# Projection to "correlated space": "Downfolding"

$$W_{n\boldsymbol{k}}(\boldsymbol{r}) = \sum_{E_1 < E_{m\boldsymbol{k}} < E_2} \psi_{m\boldsymbol{k}}(\boldsymbol{k}) U_{mn}(\boldsymbol{k})$$

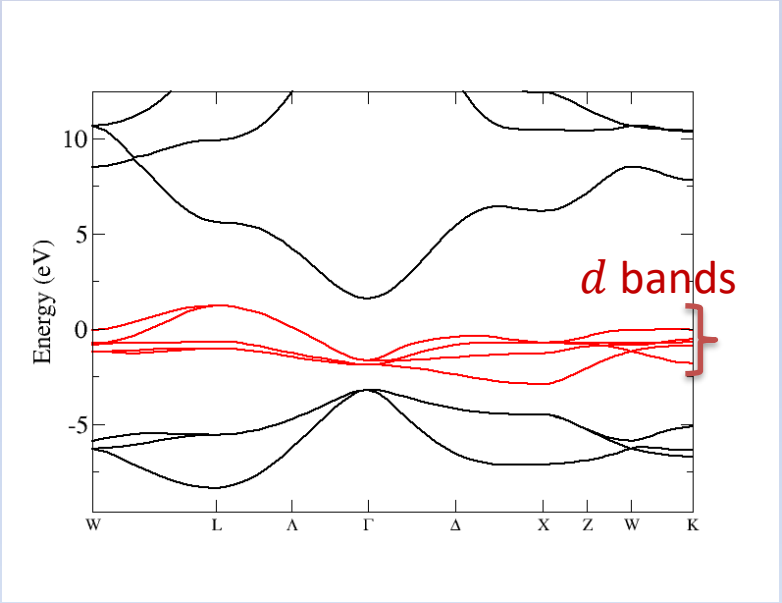
$\{|W_{n\boldsymbol{k}}\rangle\}$ expands the "correlated subspace"

LDA Hamiltonian in the "correlated subspace":

$$H_{mn}(\boldsymbol{k}) = \langle W_{m\boldsymbol{k}} | \hat{H}_{\mathrm{LDA}} | W_{n\boldsymbol{k}} \rangle$$

$$= \sum_l \epsilon_{l\boldsymbol{k}} \langle W_{m\boldsymbol{k}} | \psi_{l\boldsymbol{k}} \rangle \langle \psi_{l\boldsymbol{k}} | W_{n\boldsymbol{k}} \rangle$$

$$= \sum_l \epsilon_{l\boldsymbol{k}} U^*_{lm}(\boldsymbol{k}) U_{ln}(\boldsymbol{k})$$




Finally solve the Hamiltonian:

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

using DMFT !

# The LDA+DMFT self-consistency loop

On-site Green function:

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

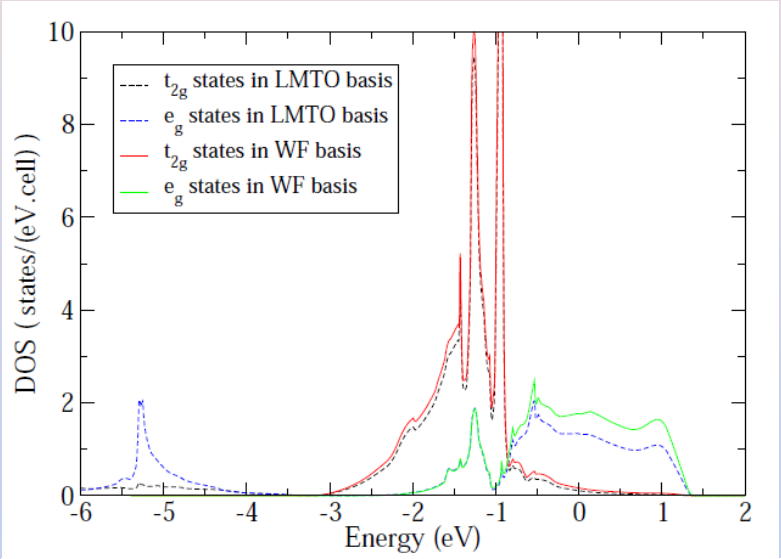
$$\Sigma_{alm,a'l'm'}(i\omega) = \begin{cases} \Sigma_{m,m'}(i\omega), & \text{if } a = a' = a_d, l = l' = d \\ 0, & \text{else} \end{cases}$$

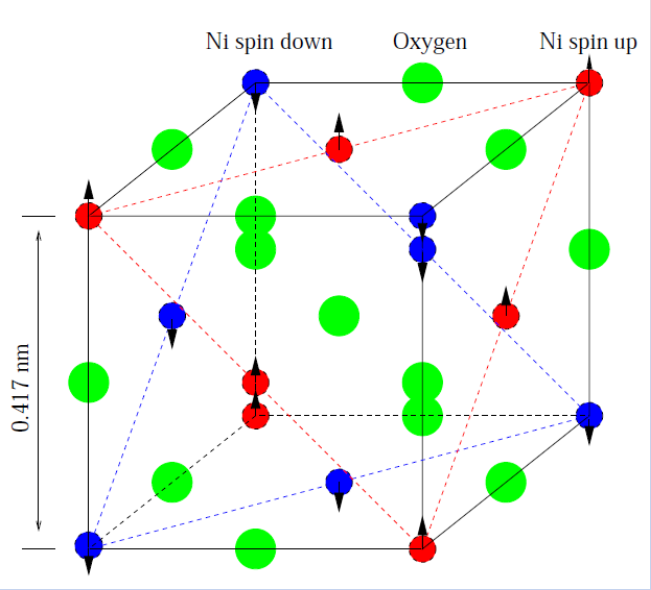
Impurity Green function: $G^{imp}_{mm'}(i\omega_n) = G_{a_d dm,a_d dm'}(i\omega_n)$

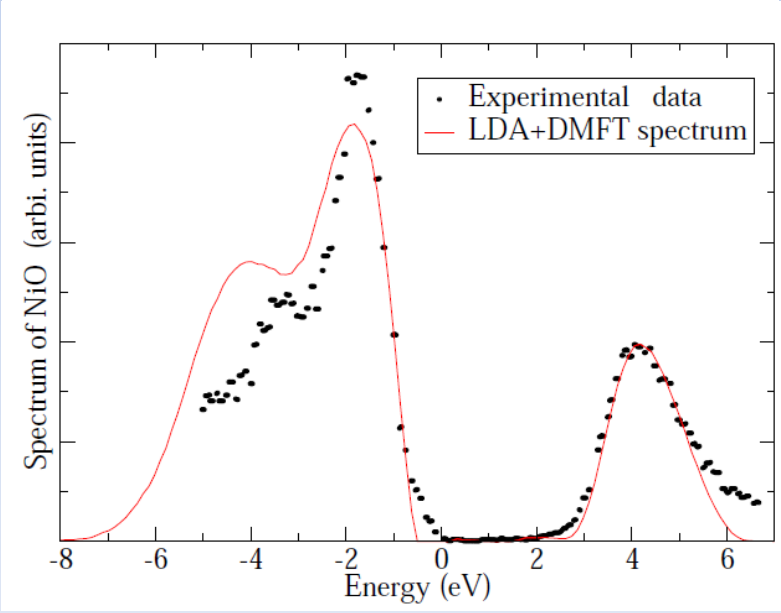
Bath Green function: $\mathcal{G}_{m,m'}(i\omega_n) = \left[\left(G^{imp}(i\omega_n)\right)^{-1} + \Sigma(i\omega_n)\right]^{-1}_{mm'}$

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

# Application: NiO



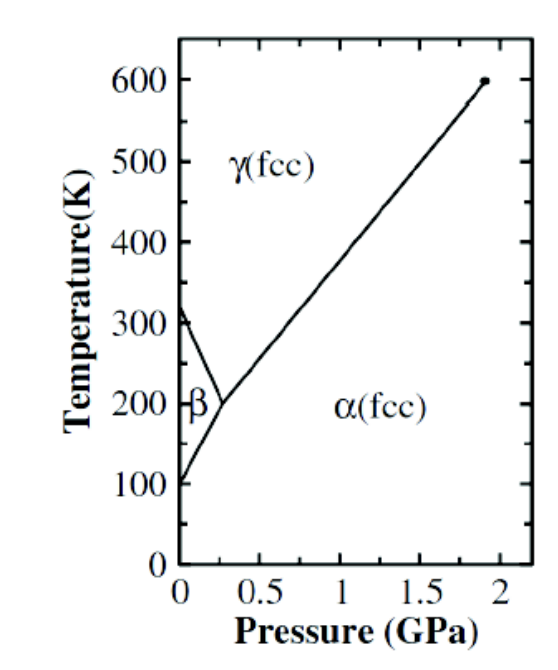




LDA+DMFT can describe the Mott insulator!

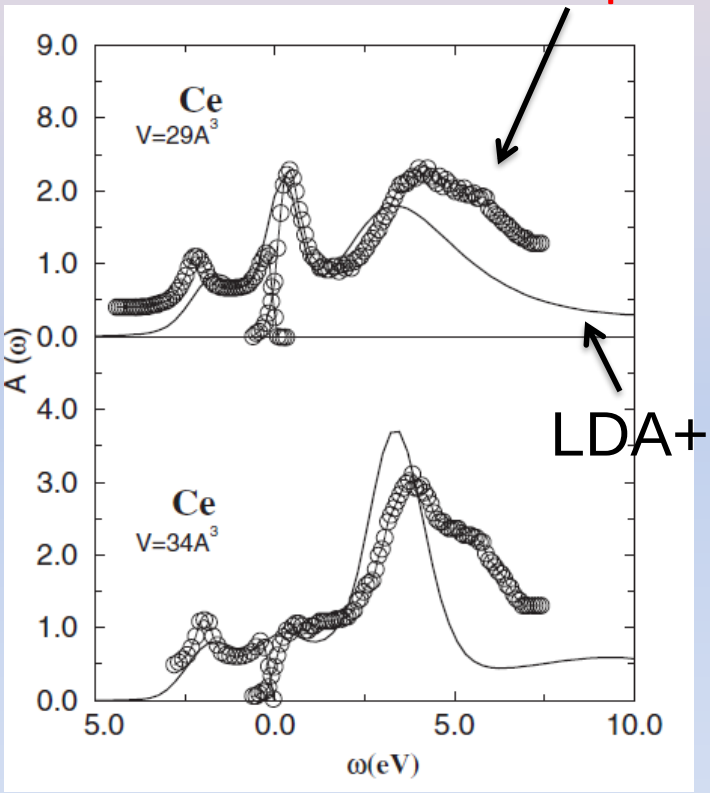
*X. Ren et al., Phys. Rev. B,* ***74****, 195114 (2006)*

# Application: phase transition in Ce

Phase diagram



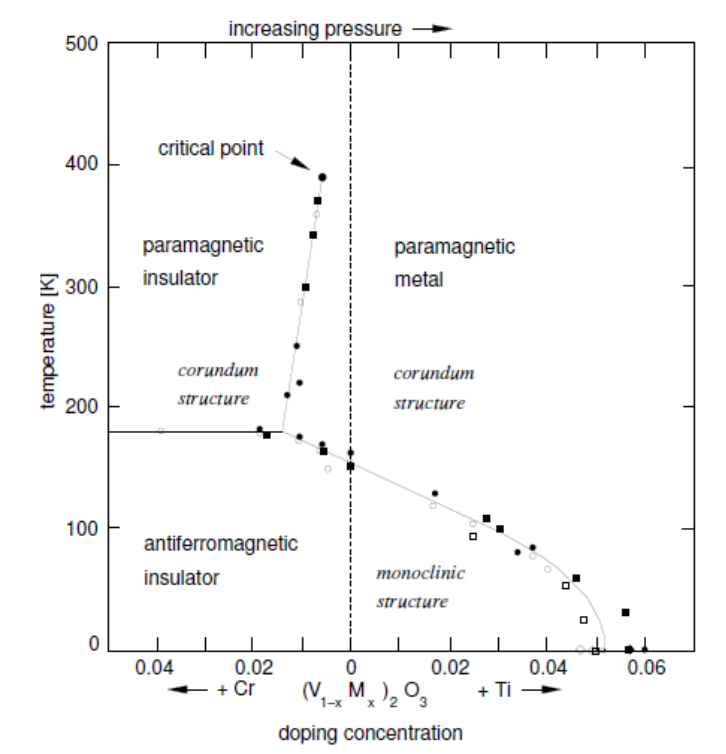

Exp.

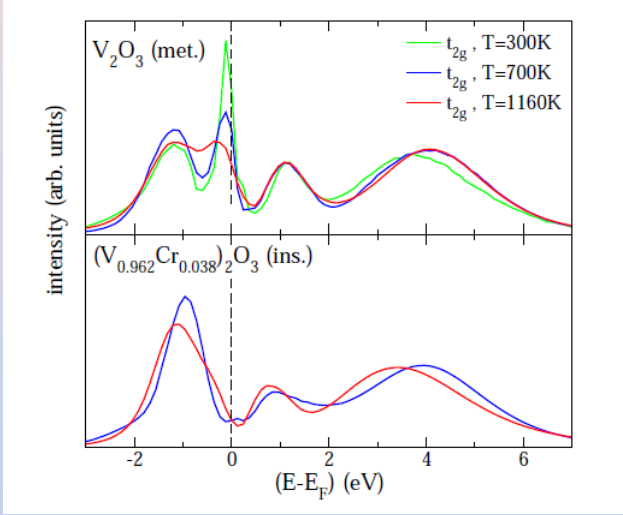


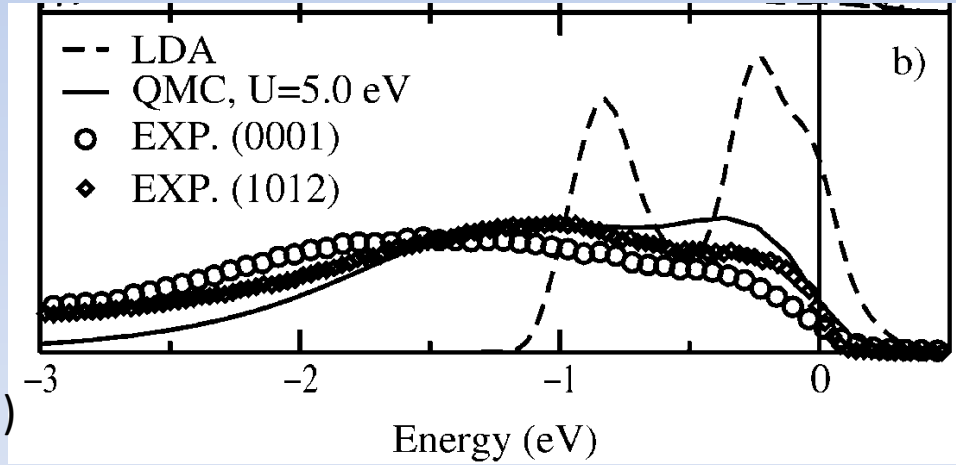

LDA+DMFT

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

# Metal insulator transition in $V_2O_3$



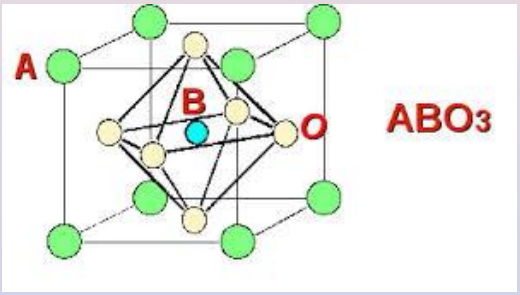



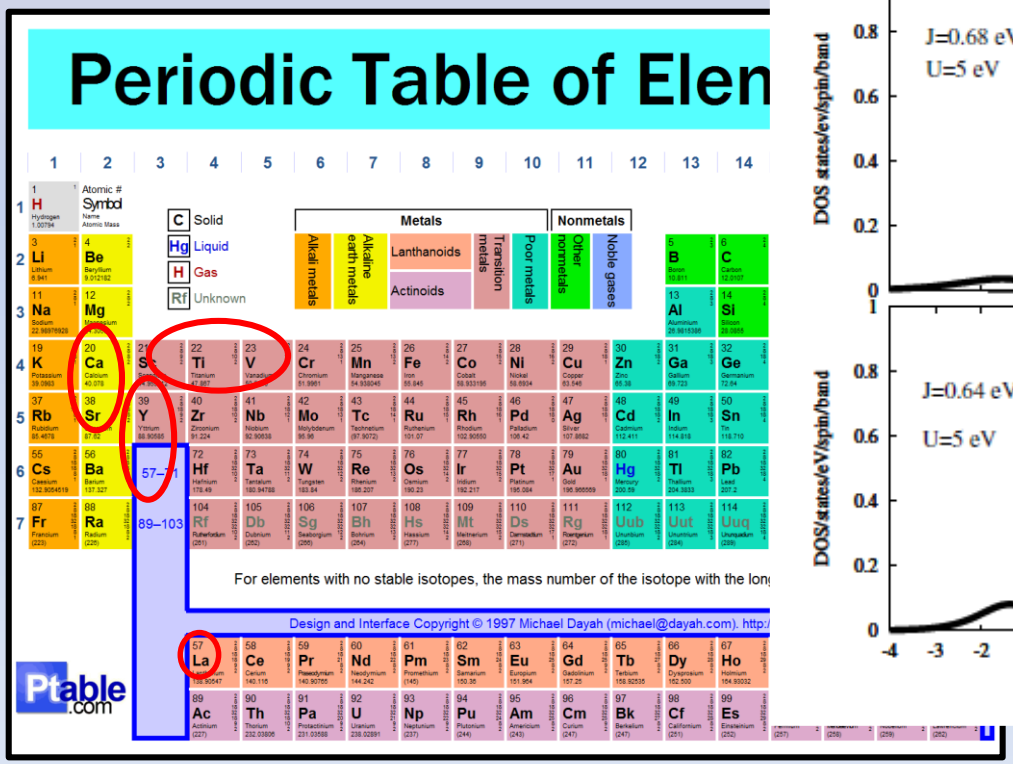

*K. Held et al., Phys. Rev. Lett. 86, 5345 (2001).*

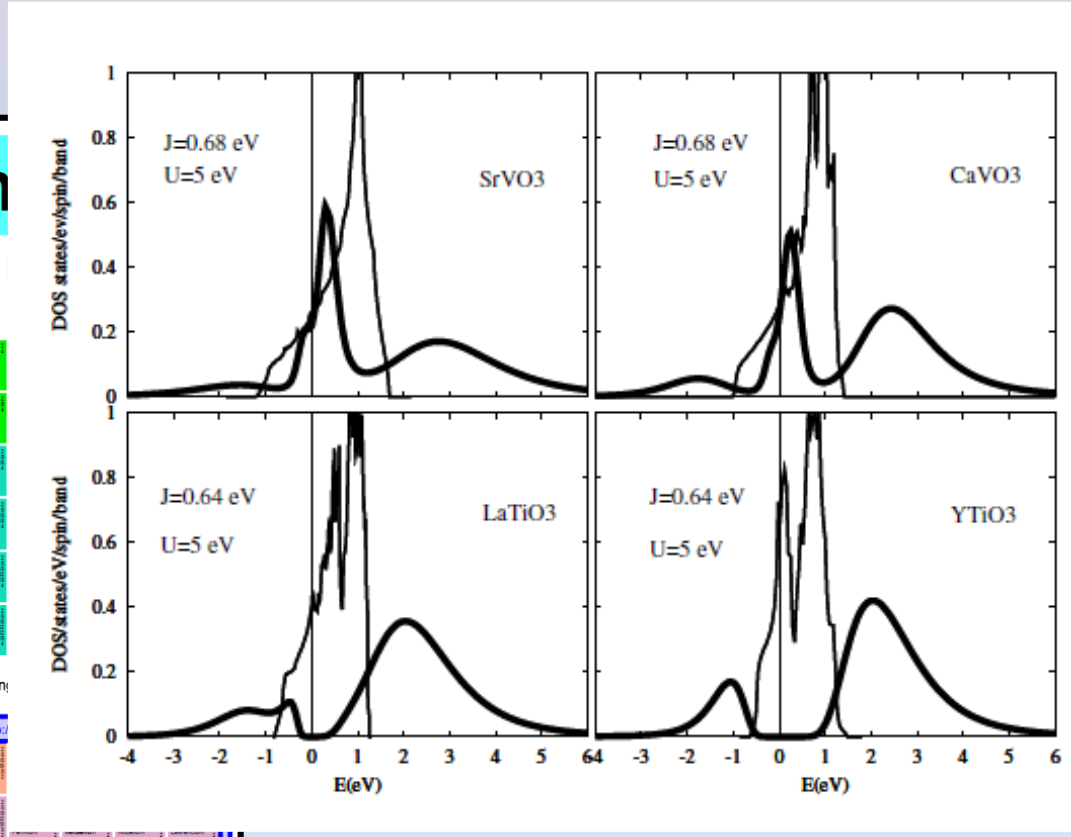
*G. Keller, PhD thesis* (Augsburg University, 2015)

# Mott transition in perovskite oxides



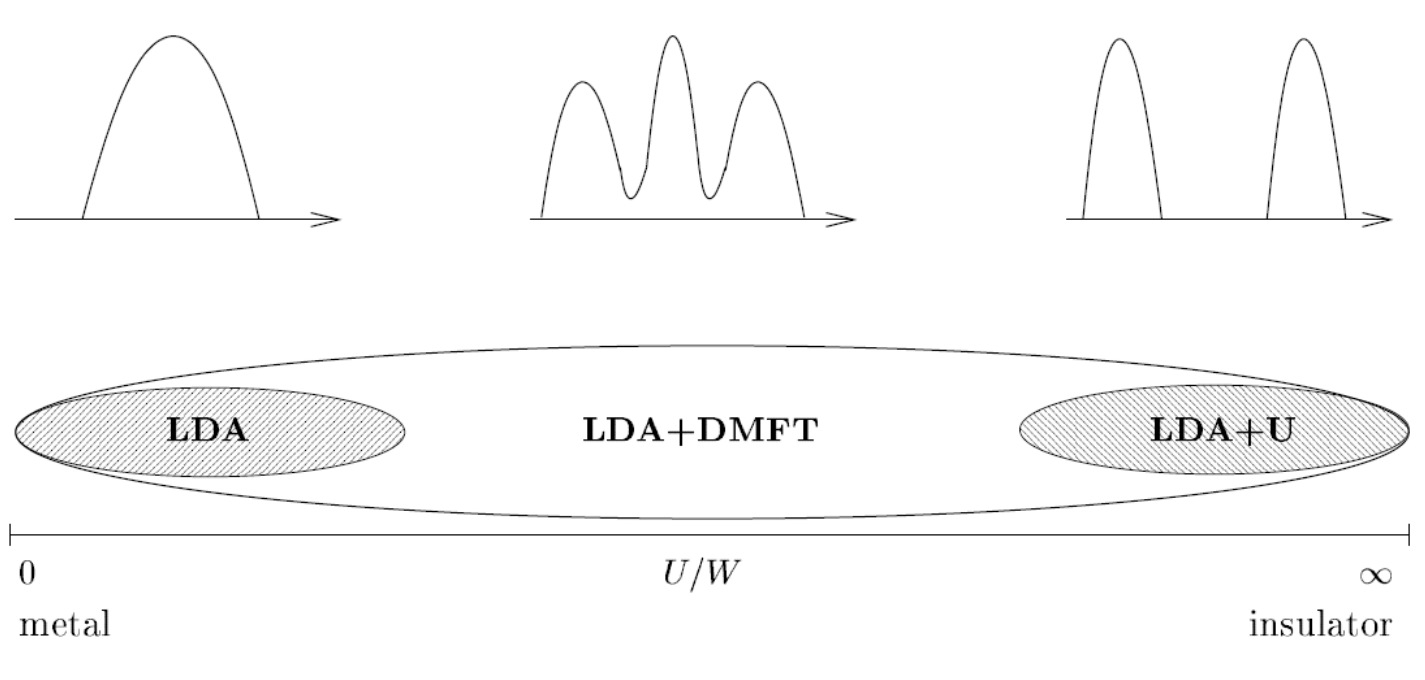

$ABO_3$(钙钛矿结构)



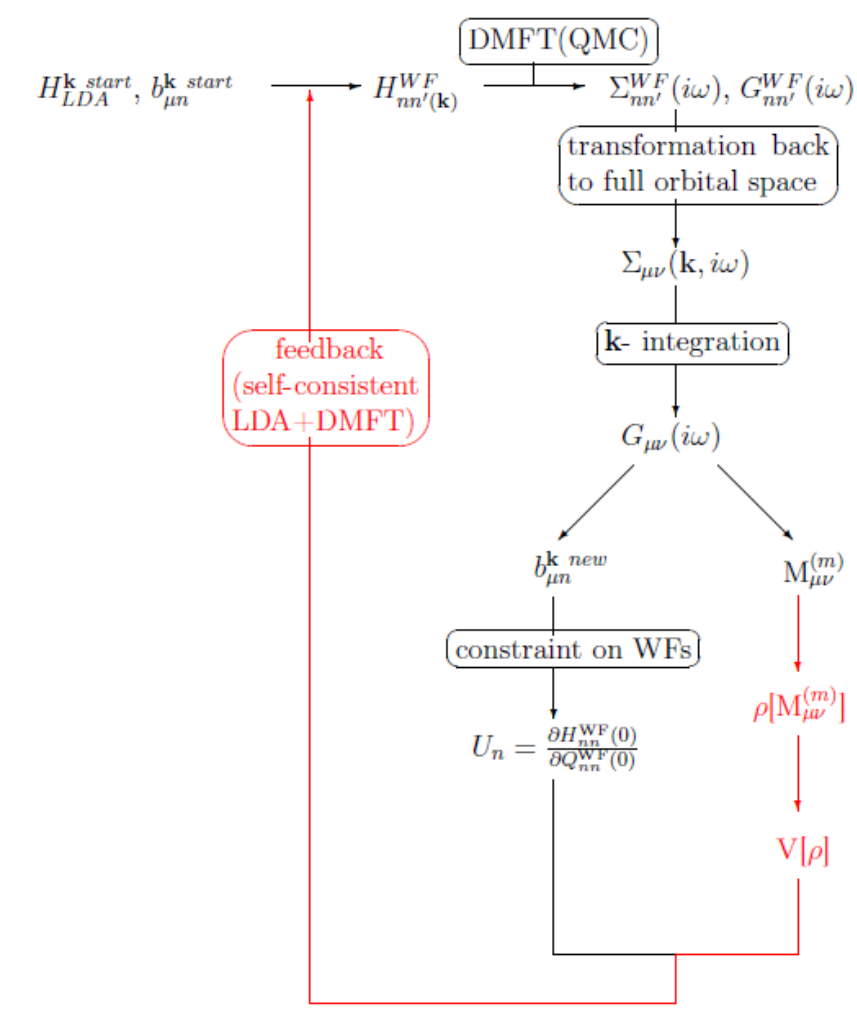




*E. Pavarini et al., Phys. Rev. Lett.* ***92****, 176403 (2004)*

# 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).