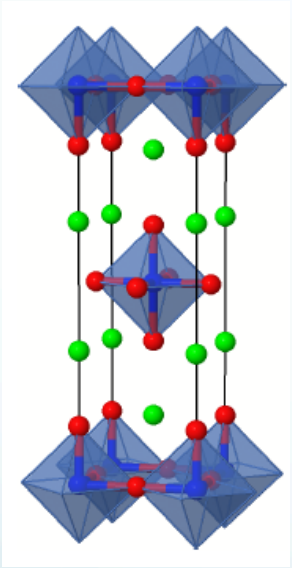
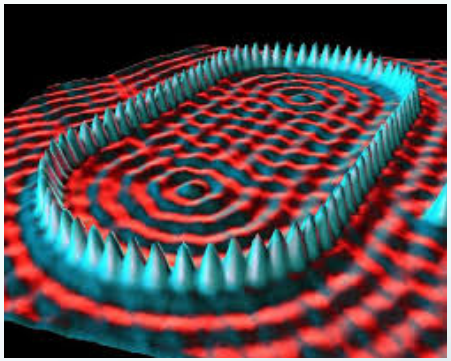
Projectors, Hubbard *U*, Charge Self-Consistency and Double Counting

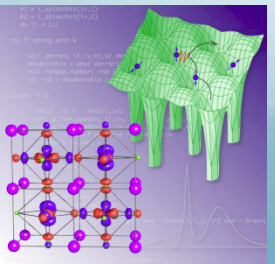


Tim Wehling

University of Bremen

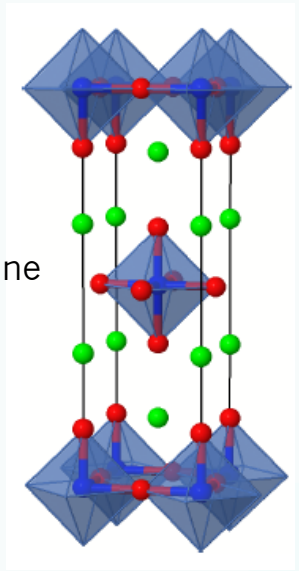


FOR 1346



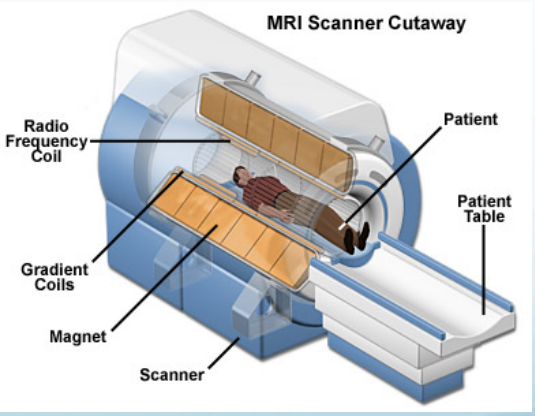
Correlated electron materials

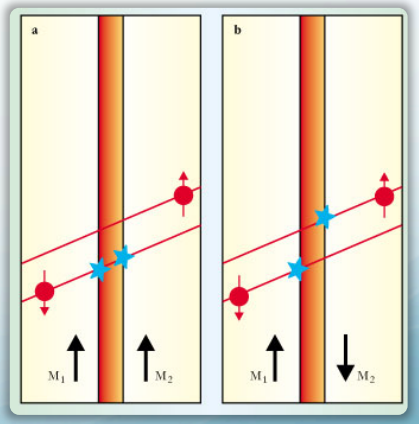
- huge resistivity changes (VO_2, ...)
- colossal magnetoresistance ($La_{1-x}Sr_xMnO_3$, ...)
- high-Tc superconductivity (YBCO, ...)
- novel phenomena at surfaces/interfaces ($LaTiO_3$ /$SrTiO_3$, ...)



Potential for technological applications

- sensors, switches,...
- spintronics
- high-T_c superconductors
- photovolatics





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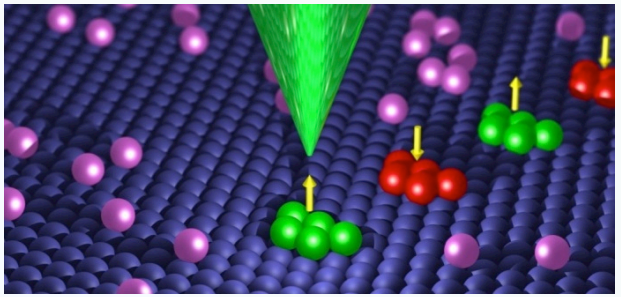
- high-Tc superconductivity (YBCO, ...)
- novel phenomena at surfaces/interfaces ($LaTiO_3$ / $SrTiO_3$, ...)

Potential for technological applications

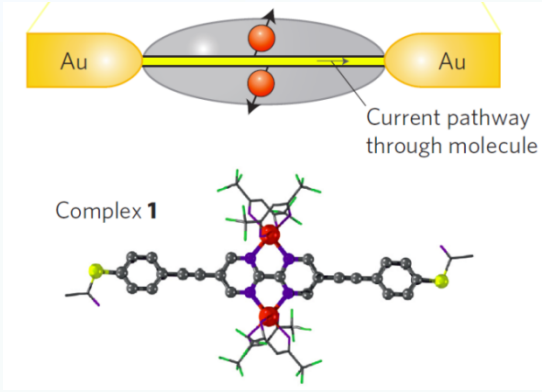
- Kondo effect: $T_K \sim De^{-1/2J\rho_0}$
- Superconductivity: $T_c \sim \omega_D e^{-1/V\rho_0}$

Nanosystems and electronic correlations



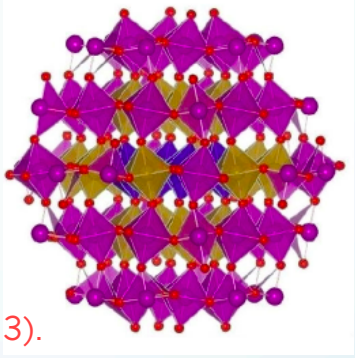
- Atomically thin materials
- Clusters and artificially created nanosystems on surfaces
- Nanoscopic “Mott systems”

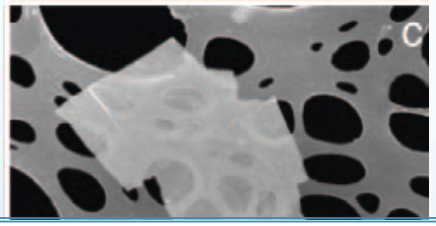
TM clusters on graphene, Cu (111) and CuN



Co_2-complex junction

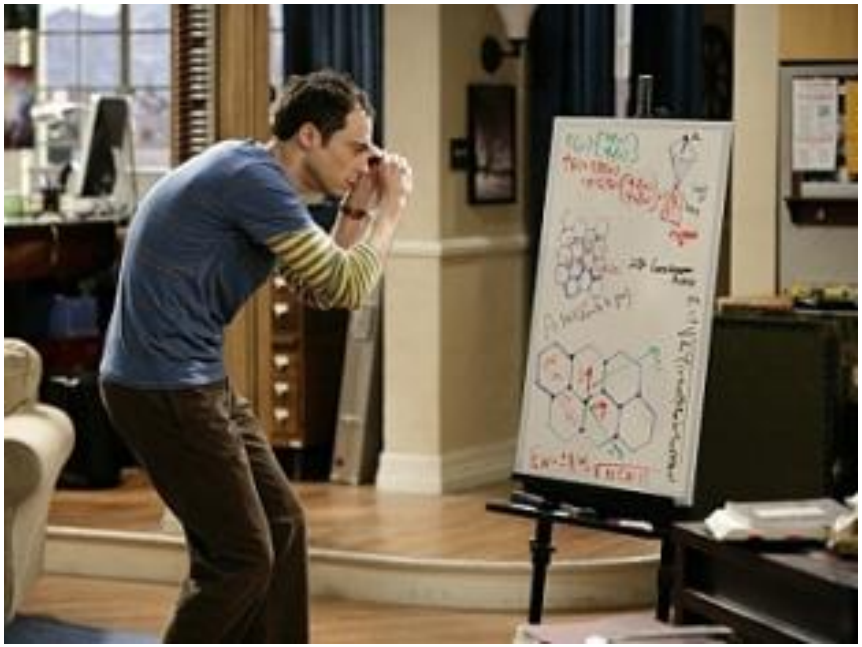
S. Wagner et al., Nature Nanotech. (2013).





Material specific theoretical approaches for correlated materials?

theory



c.f Nobel Lecture: K. S. Novoselov
Rev. Mod. Phys. 83, 837 (2011)

$\ldots, r_N)$

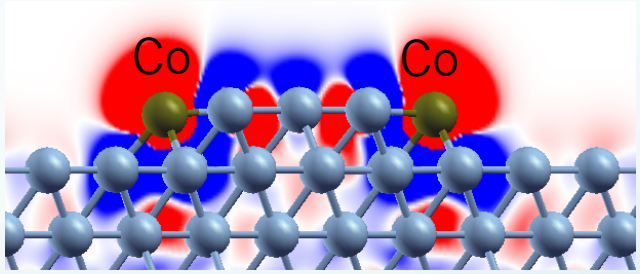
$N \sim 10^{23}$

$+ V_{\text{K-K}}(\underline{\mathbf{R}})$

$v_{\text{e-e}}(\mathbf{r}_i - \mathbf{r}_j)$

$$v_{\text{e-e}}(\mathbf{r} - \mathbf{r}') = \frac{e^2}{|\mathbf{r} - \mathbf{r}'|}$$

$k c_l$



- Flexibility of wave function → large number *M* of single particle states ϕ_i required
- Dimension of Fock space: 2^M
- Dimension of *N* particle subspace: $\begin{pmatrix} M \\ N \end{pmatrix} = \frac{M!}{N!(M-N)!}$

Density functional theory

P. Hohenberg and W. Kohn (1964)

W. Kohn and L. Sham (1965)

For |GS> use density *n(r)* instead of $\Psi(r_1, r_2, \ldots, r_N)$

Energy functional:

$$E[n(\vec{r})] = \langle \Psi | H | \Psi \rangle = \int n(\vec{r}) V_{ext}(\vec{r}) d\vec{r} + F[n(\vec{r})]$$

Universal Functional:

$$F[n] = T[n] + U[n]$$

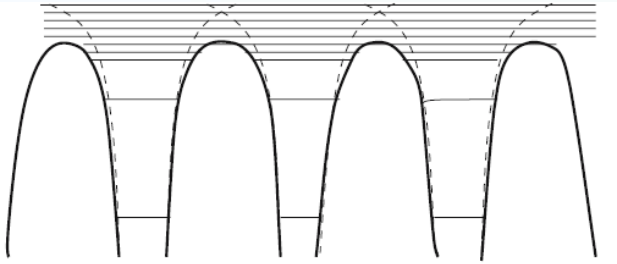
Extremal principle:

$$E[n_0] < E[n] \Rightarrow \frac{\delta E[n]}{\delta n(r)} = \mu$$

DFT: Kohn-Sham Theory

Non interacting auxiliary system:

$$(-\frac{\hbar^2}{2m}\nabla^2 - V_{eff}(\vec{r}))\psi_i(\vec{r}) = \varepsilon_i\psi_i(\vec{r})$$



Density:

$$n(\vec{r}) = \sum_i^N |\psi_i(\vec{r})|^2$$

Energy functional:

$$E[n] = T_s[n] + V_H[n] + \int n(\vec{r})V_{ext}(\vec{r})d\vec{r} + E_{xc}[n]$$

All many body effects in E_{xc}.

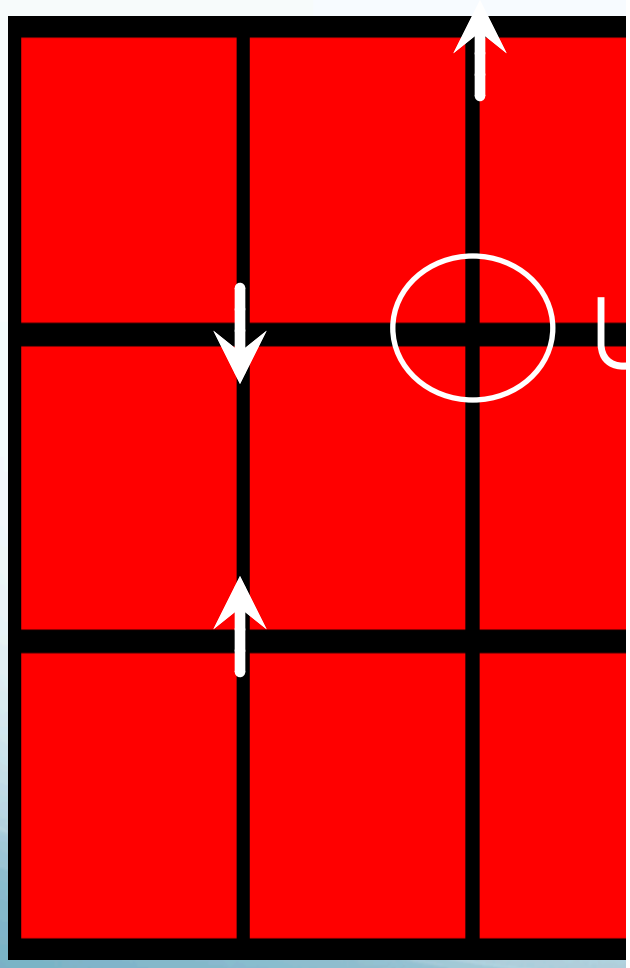
Assumption: KS-auxiliary particles = Quasiparticle excitations

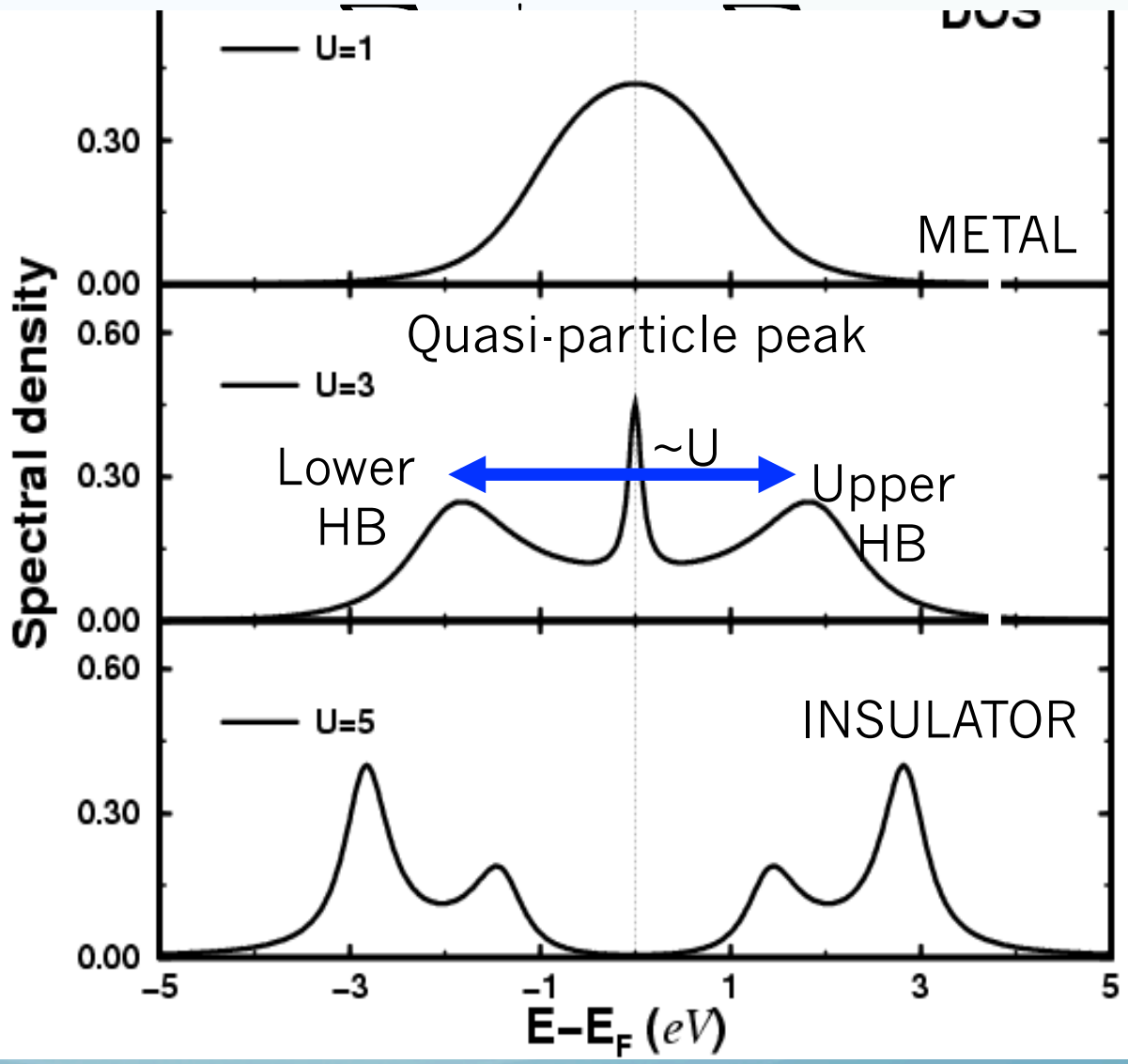
$\Sigma(r,r',\omega) = V_{eff}(r)$

Effective potential:

$$V_{eff}(\vec{r}) = V_{ext}(\vec{r}) + e^2\int d\vec{r}'\frac{n(\vec{r}')}{|\vec{r}-\vec{r}'|} + \frac{\delta E_{xc}[n]}{\delta n(\vec{r})}$$

Correlated electrons: The Hubbard model

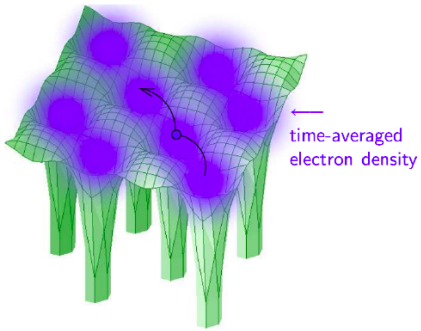


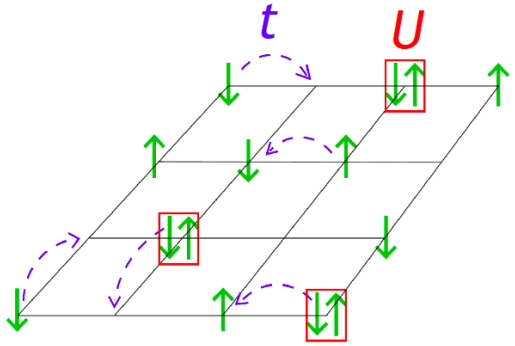


A.Georges, G.Kotliar, W.Krauth and M.Rozenberg, Rev. Mod. Phys. '96

The DFT++ approach

DFT/LDA		Model Hamiltonians	
+	material specific: "ab initio"	−	input parameters unknown: unrealistic
−	fails for strong correlations	+	systematic many-body approach





Material-specific input
Density functional theory (LDA/GGA) or GW

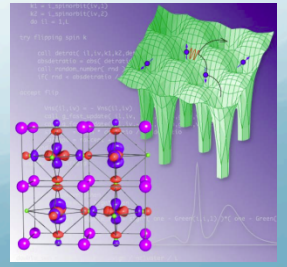
⟷

Electronic correlations
Many-body theory

Slide from D. Vollhardt

V. I. Anisimov et al., J. Phys. Condes. Matter (1997)

A. I Lichtenstein and M. I. Katsnelson, PRB (1998)



FOR 1346

Outline

- **Correlated subspaces and projectors**
  - Quantum impurity problems
  - Projector formalism in LDA+DMFT
- **Interaction terms: Hubbard** U **and beyond**
  - The constrained random phase approximation
  - Non-local Coulomb interactions
- **Double counting and charge self-consistency**

Correlated subspaces and projectors

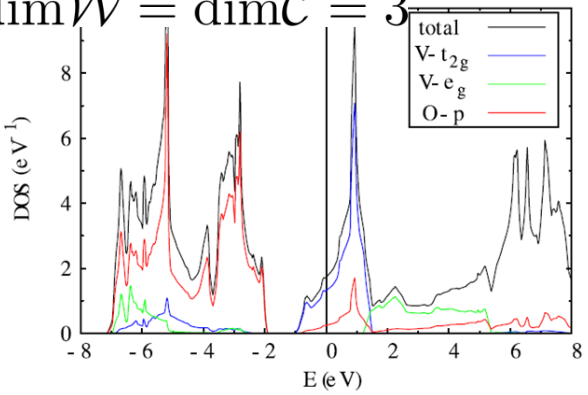
DFT++ Hamiltonian $H = \underbrace{\sum_k \epsilon_k c_k^\dagger c_k}_{H_K} \underbrace{- \mu_{DC} \sum_m d_m^\dagger d_m}_{H_{DC}} + \underbrace{\frac{1}{2} \sum_{m \ldots m'''} U_{m \ldots m'''} d_m^\dagger d_{m'}^\dagger d_{m''} d_{m'''}}_{H_U}$

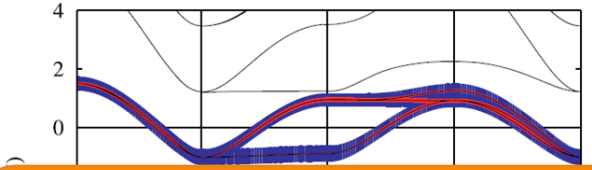
Bloch space $\mathcal{W} = \mathrm{span}\{|k\rangle\}$

Correlated subspace $\mathcal{C} = \mathrm{span}\{|m\rangle\}$

(a) $SrVO_3$

$\dim\mathcal{W} = \dim\mathcal{C} = 3$

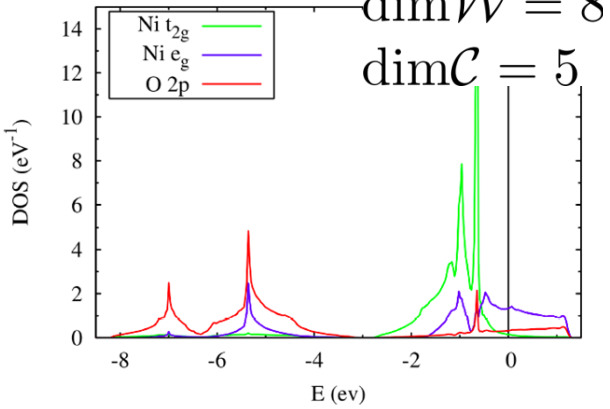


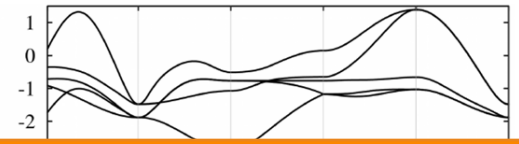


(b) NiO

$\dim\mathcal{W} = 8$

$\dim\mathcal{C} = 5$

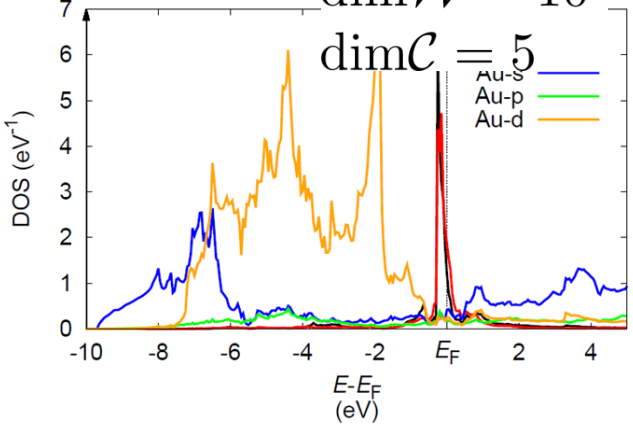


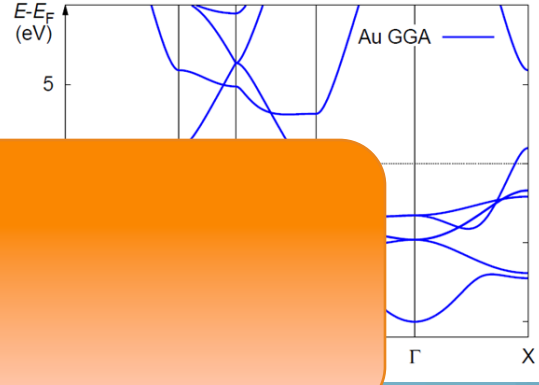


(c) Fe in Au

$\dim\mathcal{W} \sim 10^3$

$\dim\mathcal{C} = 5$





Central quantity to connect DFT and model Hamiltonians:

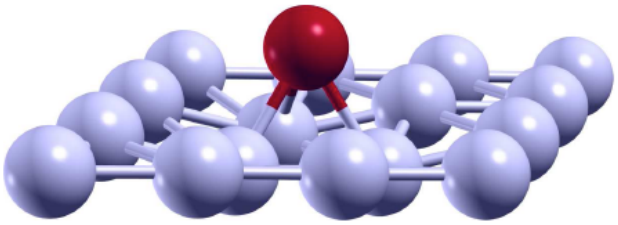
Projection <k|m>

Electronic density of states (DOS) and band structures as obtained from DFT

Magnetic impurity systems

$$H = \sum_k \epsilon_k c_k^\dagger c_k - \mu_{DC} \sum_m d_m^\dagger d_m + \frac{1}{2} \sum_{m \ldots m'''} U_{m \ldots m'''} d_m^\dagger d_{m'}^\dagger d_{m''} d_{m'''}$$

$$\langle k|m\rangle \neq 0$$



Anderson impurity model

$$H_{\mathrm{AIM}} = \sum_{\tilde{k}} \epsilon_{\tilde{k}} c_{\tilde{k}}^\dagger c_{\tilde{k}} + \sum_{\tilde{k},m} (V_{\tilde{k}m} c_{\tilde{k}}^\dagger d_m + h.c.)$$

$$+ \sum_m (\epsilon_m - \mu_{DC}) d_m^\dagger d_m + \frac{1}{2} \sum_{m \ldots m'} U_{m \ldots m'''} d_m^\dagger d_{m'}^\dagger d_{m''} d_{m'''}$$

Effective local action

$$S_{\mathrm{eff}}(d^*, d) = -\int_0^\beta \mathrm{d}\tau \int_0^\beta \mathrm{d}\tau' \sum_{m,m'} d_m^*(\tau) (\mathcal{G}_0^{-1}(\tau - \tau'))_{mm'} d_{m'}(\tau') + \int_0^\beta \mathrm{d}\tau H_{\mathrm{U}}(d^*, d)(\tau)$$

Magnetic impurity systems

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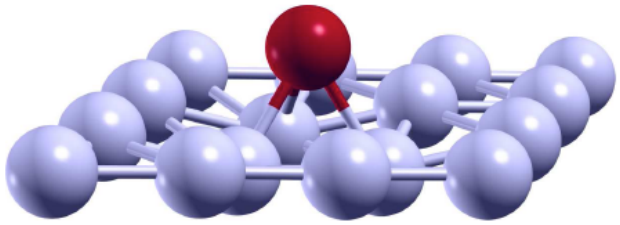

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$$+ \sum_m (\epsilon_m - \mu_{DC}) d_m^\dagger d_m + \frac{1}{2} \sum_{m \ldots m'} U_{m \ldots m'''} d_m^\dagger d_{m'}^\dagger d_{m''} d_{m'''}$$

Action

$$S_{\mathrm{AIM}}(c^*, c, d*, d) = \int_0^\beta \mathrm{d}\tau \sum_{\tilde{k}} c_{\tilde{k}}^*(\tau) \partial_\tau c_{\tilde{k}}(\tau) + \sum_m d_m^*(\tau) \partial_\tau d_m(\tau) + H_{\mathrm{AIM}}(c^*, c, d^*, d)(\tau)$$

Effective local action

$$S_{\mathrm{eff}}(d^*, d) = -\int_0^\beta \mathrm{d}\tau \int_0^\beta \mathrm{d}\tau' \sum_{m,m'} d_m^*(\tau) (\mathcal{G}_0^{-1}(\tau - \tau'))_{mm'} d_{m'}(\tau') + \int_0^\beta \mathrm{d}\tau H_{\mathrm{U}}(d^*, d)(\tau)$$

Magnetic impurity systems

Wanted

Projection onto Correlated Subspace

$$\mathcal{G}_0(i\omega_n) = \mathcal{P}_{\mathcal{C}} G_{KS}(i\omega_n) \mathcal{P}_{\mathcal{C}}$$

$$\mathcal{P}_{\mathcal{C}} = \sum_m |m\rangle\langle m|$$

Effective local action

$$S_{\text{eff}}(d^*, d) = -\int_0^\beta \mathrm{d}\tau \int_0^\beta \mathrm{d}\tau' \sum_{m,m'} d_m^*(\tau)(\mathcal{G}_0^{-1}(\tau - \tau'))_{mm'} d_{m'}(\tau') + \int_0^\beta \mathrm{d}\tau H_{\text{U}}(d^*, d)(\tau)$$

Noninteracting impurity GF:

$$\mathcal{G}_0(i\omega_n)_{mm'} = \sum_k \frac{\langle m|k\rangle\langle k|m'\rangle}{i\omega_n + \mu - \epsilon_k}$$

Kohn Sham GF:

$$G_{KS}(i\omega_n) = \sum_k \frac{|k\rangle\langle k|}{i\omega_n + \mu - \epsilon_k}$$

DFT++ implementation

Idea: Use of projections of DFT wave functions $|K\rangle$ onto localized orbitals $|L\rangle$ to obtain hybridization functions

Local Green function

$$\hat{G}_0^{\mathrm{loc}}(\omega) = \sum_{K,L,L'} |L\rangle \frac{\langle L|K\rangle\langle K|L'\rangle}{\omega + i0^+ - \epsilon_K} \langle L'|$$

Projector augmented wave basis

$$|K\rangle = |\tilde{K}\rangle + \sum_i (|\phi_i\rangle - |\tilde{\phi}_i\rangle)\langle \tilde{p}_i|\tilde{K}\rangle$$

$$|L\rangle = |\phi_i\rangle$$

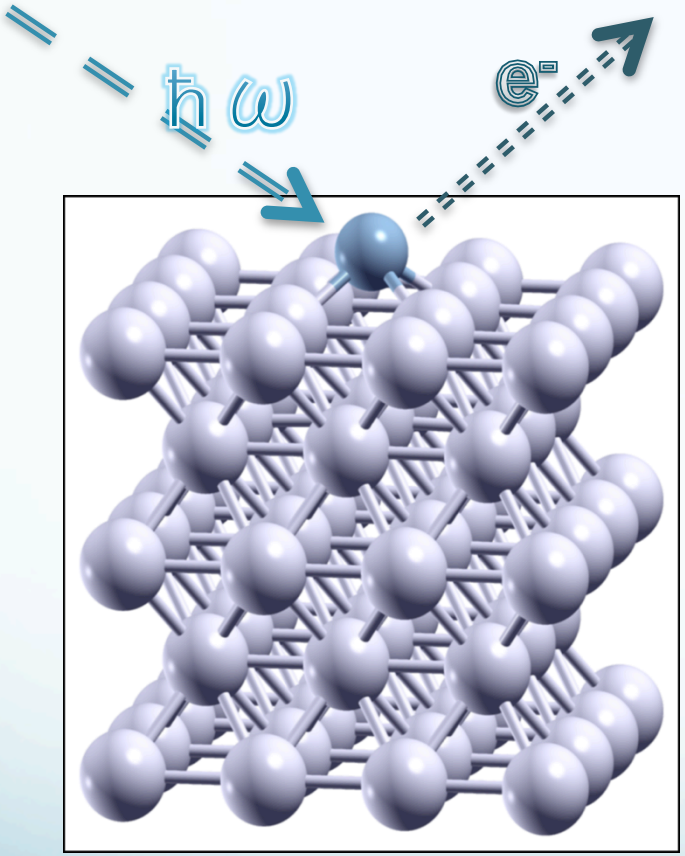
$$\langle L|K\rangle = \sum_{\nu'} \langle L|\phi_{\nu'}\rangle\langle \tilde{p}_{\nu'}|\tilde{K}\rangle$$

Implementation with VASP code

PRB **77**, 205112 (2008), PRB **81**, 085413 (2010), J. Phys.: Condens Matter 23, 085601 (2011).

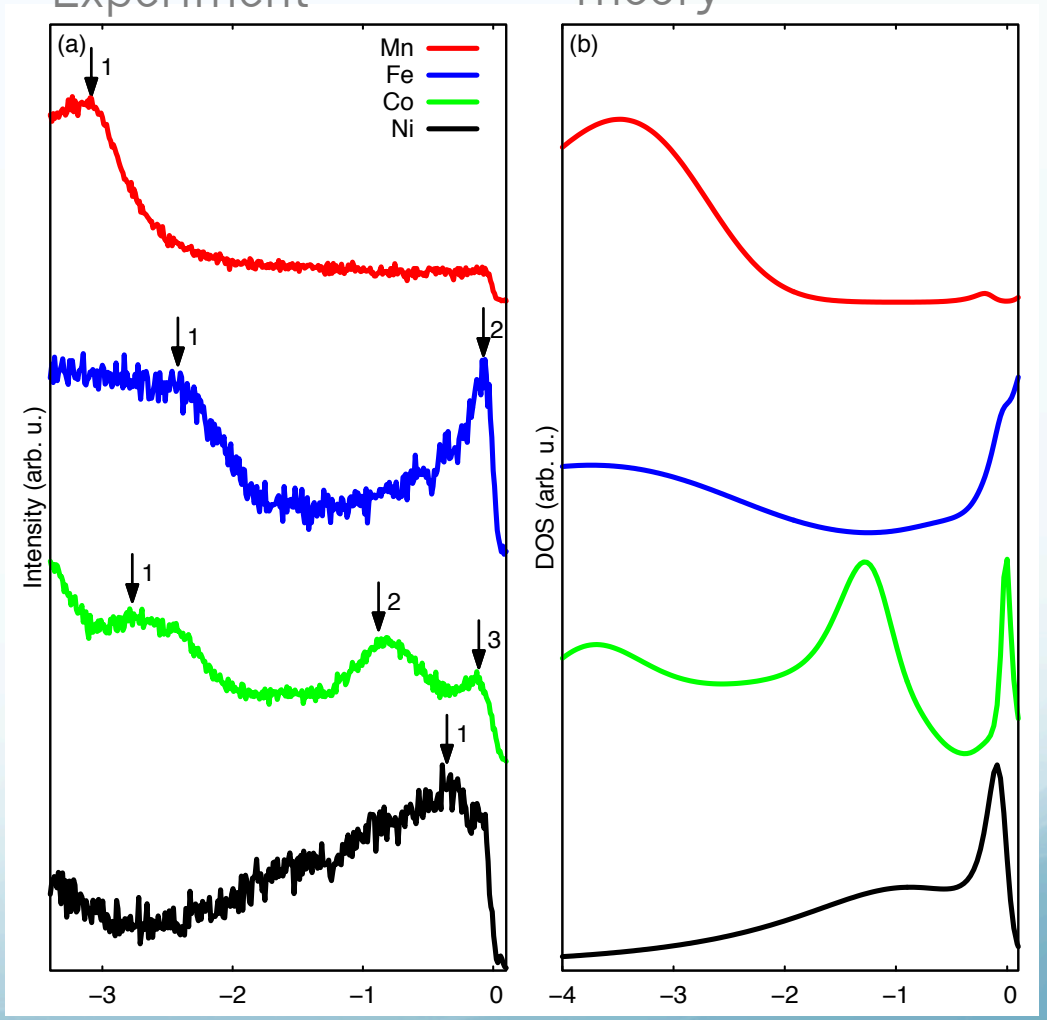
Transition Metal Adatoms on Ag(100)

Photoemission spectroscopy + First-principles theory

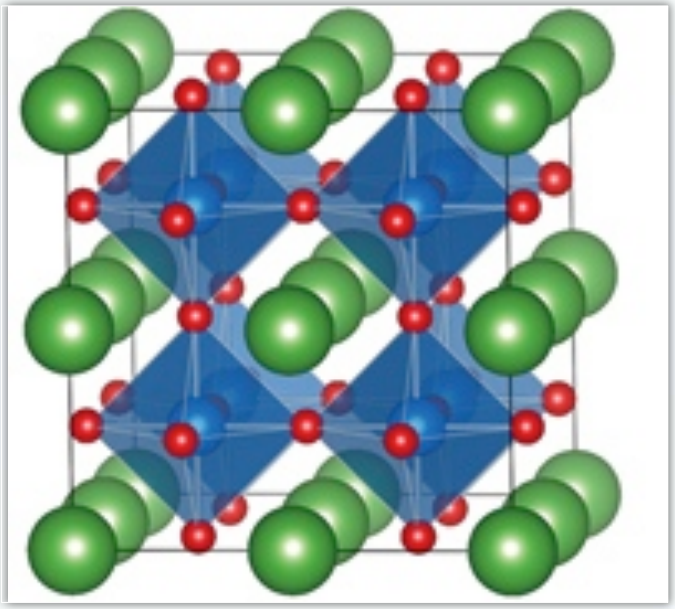


Experiment

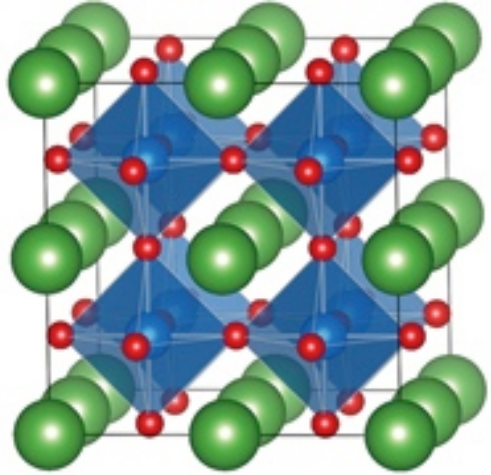
Theory



S. Gardonio, TW, et al., PRL **110**, 186404 (2013)



Projector formalism in LDA +DMFT



LDA+DMFT

$$H = \sum_k \epsilon_k c_k^\dagger c_k - \mu_{DC} \sum_{\mathbf{R},m} d_{\mathbf{R},m}^\dagger d_{\mathbf{R},m} + \frac{1}{2} \sum_{\mathbf{R},m\ldots m'''} U_{m\ldots m'''} d_{\mathbf{R},m}^\dagger d_{\mathbf{R},m'}^\dagger d_{\mathbf{R},m''} d_{\mathbf{R},m'''}$$

DFT part

from charge density $\rho(r)$ construct

$\hat{V}_{\rm KS} = \hat{V}_{\rm ext} + \hat{V}_{\rm H} + \hat{V}_{\rm xc}$

$\left[-\frac{\nabla^2}{2} + \hat{V}_{\rm KS}\right] |\psi_{k\nu}\rangle = \varepsilon_{k\nu} |\psi_{k\nu}\rangle$

update $\{|\chi_{Rm}\rangle\}$

DMFT prelude

build $\hat{G}_{\rm KS} = \left[i\omega_n + \mu + \frac{\nabla^2}{2} - \hat{V}_{\rm KS}\right]^{-1}$

construct initial $\hat{\mathcal{G}}_0$

DMFT loop

impurity solver

$G^{\rm imp}_{mm'}(\tau - \tau') = -\langle \hat{T}\, \hat{d}_{m\sigma}(\tau) \hat{d}^\dagger_{m'\sigma'}(\tau') \rangle_{S_{\rm imp}}$

$\hat{\mathcal{G}}_0^{-1} = \hat{G}_{\rm loc}^{-1} + \hat{\Sigma}_{\rm imp}$

$\hat{\Sigma}_{\rm imp} = \hat{\mathcal{G}}_0^{-1} - \hat{G}_{\rm imp}^{-1}$

$G_{imp}(i\omega_n) = G^{\rm loc}_{\mathbf{R}}(i\omega_n)$

self-consistency condition: construct $\hat{G}_{\rm loc}$

$\hat{G}_{\rm loc} = \hat{P}_R^{(\mathcal{C})} \left[\hat{G}_{\rm KS}^{-1} - \left(\hat{\Sigma}_{\rm imp} - \hat{\Sigma}_{\rm dc}\right)\right]^{-1} \hat{P}_R^{(\mathcal{C})}$

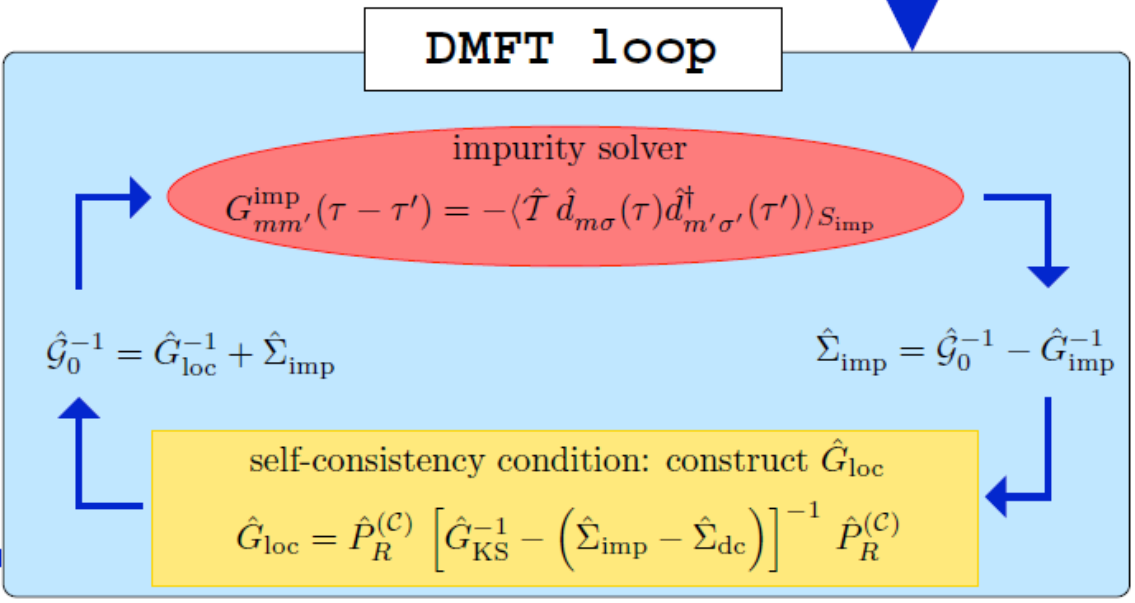
ρ update

compute new chemical potential μ

$\rho(r) = \rho_{\rm KS}(r) + \Delta\rho(r)$

(Appendix A)

F. Lechermann et al. PRB 2006



LDA+DMFT

Matrix representations of Green functions

Bloch basis

$$H_{\rm KS}(\mathbf{k})_{\alpha\alpha'} = \sum_k$$

$$\Sigma_{\alpha\alpha'}(\mathbf{k}, i\omega_n) =$$

$$G_{\alpha\alpha'}(\mathbf{k}, i\omega_n) =$$

Correlated s

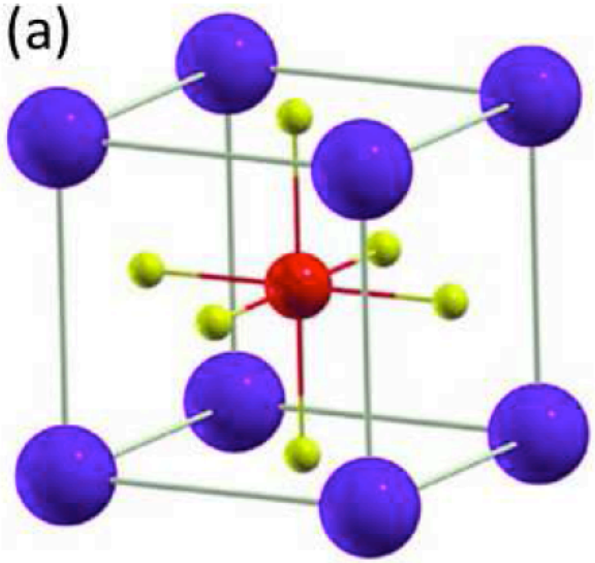
$$G^{\rm loc}_{\rm R}(i\omega_n)_{mm'} =$$

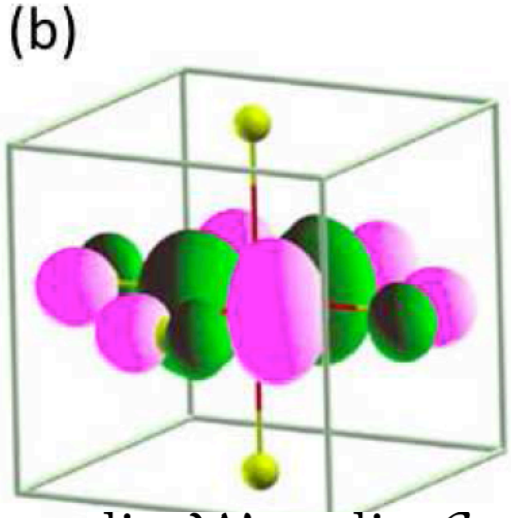
$$\mathbf{k},\alpha,$$

Required

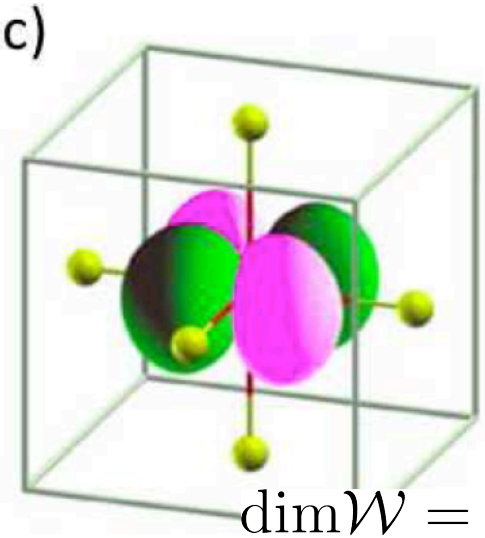
1. Choice of **correlated subspace basis** |Rm>, e.g. localized orbitals from basis set or MLWFs
2. Choice of **Bloch basis** $|B_{\mathbf{k}\alpha}\rangle$, e.g. KS eigenstates |k>
3. **Projections** $\langle Rm|B_{\mathbf{k}\alpha}\rangle$

Example: $SrVO_3$



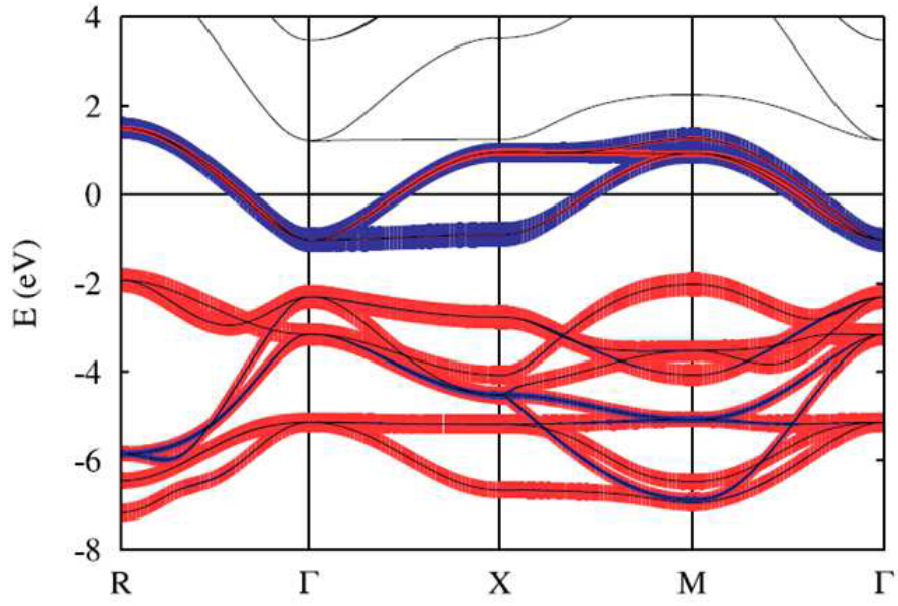


$\dim\mathcal{W} = \dim\mathcal{C} = 3$

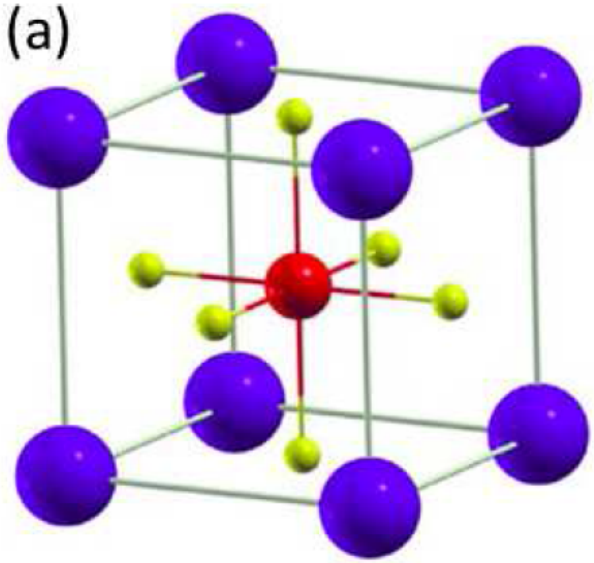


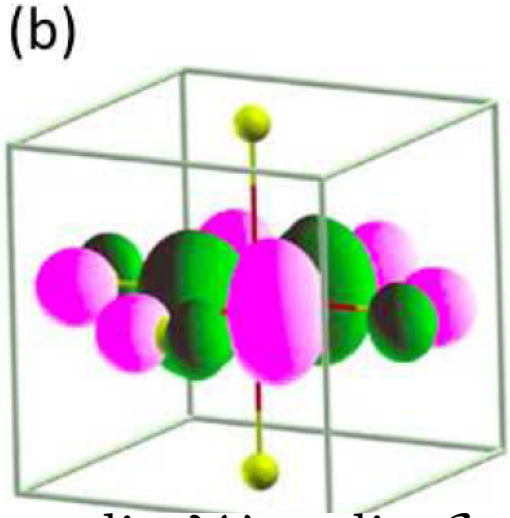
$\dim\mathcal{W} = 14$

$\dim\mathcal{C} = 5$

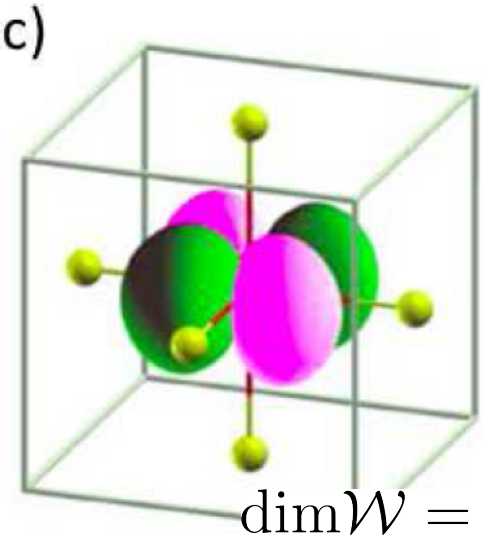


Example: $SrVO_3$



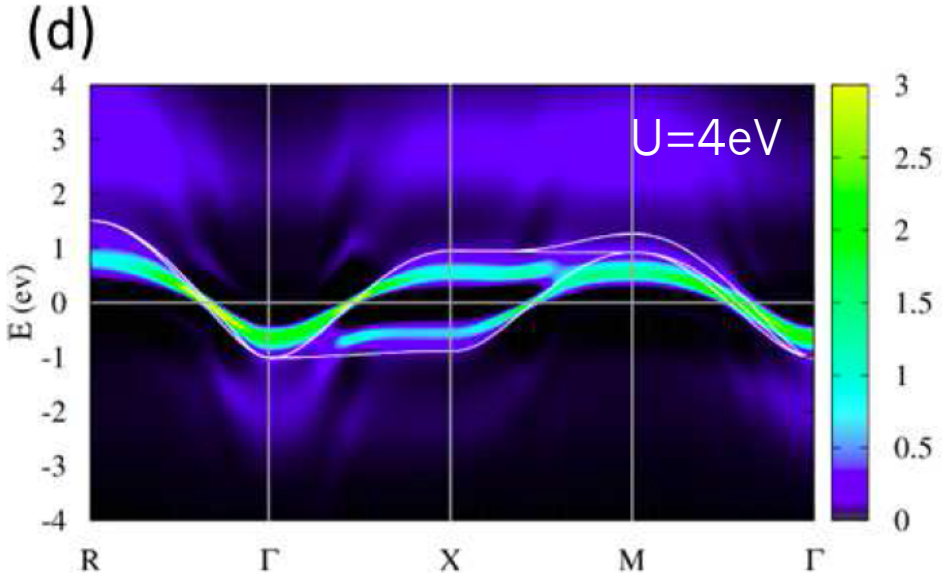


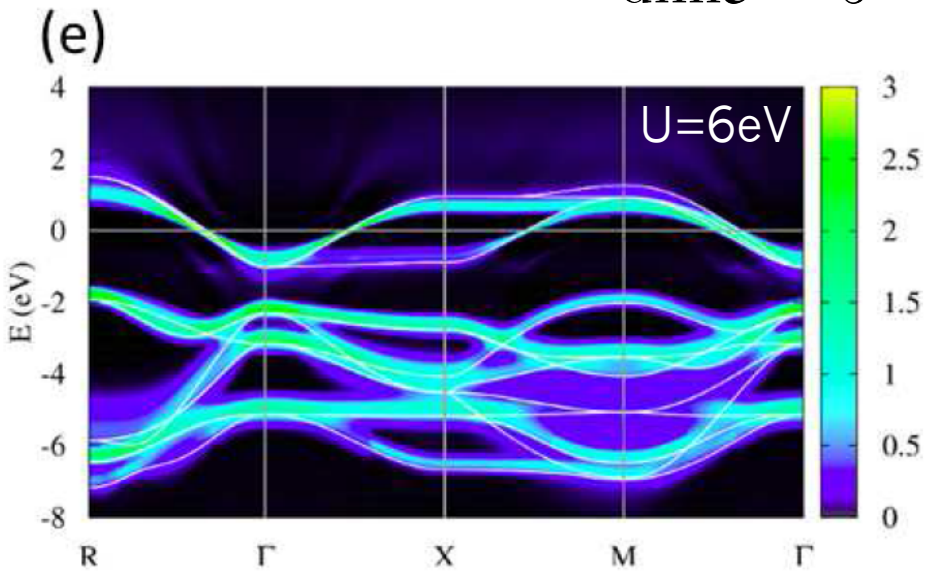
$\dim\mathcal{W} = \dim\mathcal{C} = 3$



$\dim\mathcal{W} = 14$

$\dim\mathcal{C} = 5$





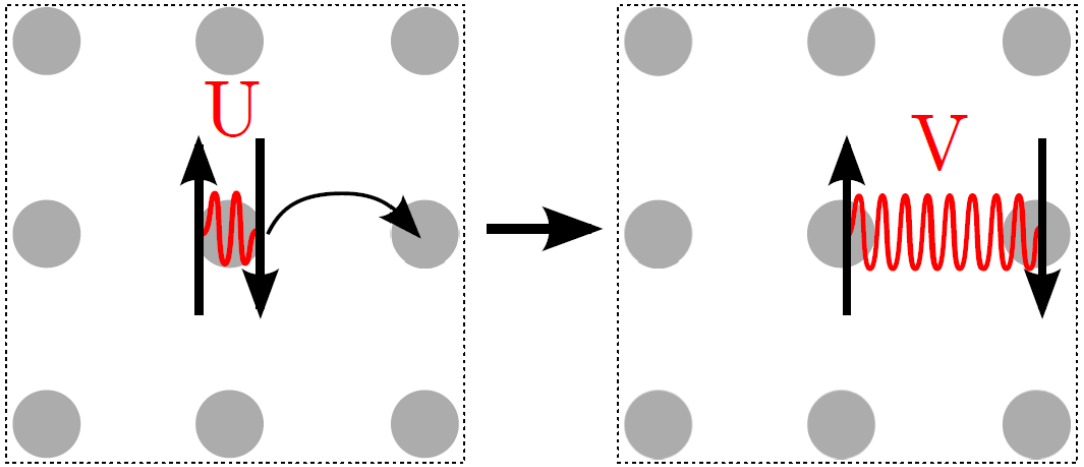
Interaction terms: Hubbard U and beyond

$$H = \sum_k \epsilon_k c_k^\dagger c_k - \mu_{DC} \sum_{\mathbf{R},m} d_{\mathbf{R},m}^\dagger d_{\mathbf{R},m}$$
$$+ \frac{1}{2} \sum_{\mathbf{R},m\ldots m'''} U_{m\ldots m'''} d_{\mathbf{R},m}^\dagger d_{\mathbf{R},m'}^\dagger d_{\mathbf{R},m''} d_{\mathbf{R},m'''}$$

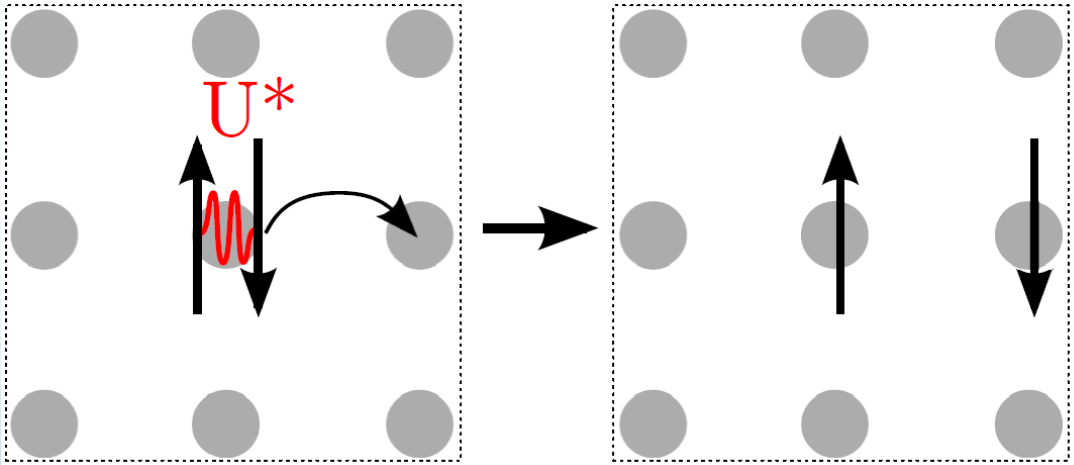
$$U_{m\ldots m'''} \stackrel{?}{=} \langle \mathbf{R}m | \langle \mathbf{R}m' | \frac{e^2}{\hat{\mathbf{r}} - \hat{\mathbf{r}}'} | \mathbf{R}m'' \rangle | \mathbf{R}m''' \rangle$$

Interaction local and restricted to correlated subspace!

A gedanken experiment



- full model
- energy gain $U - V$



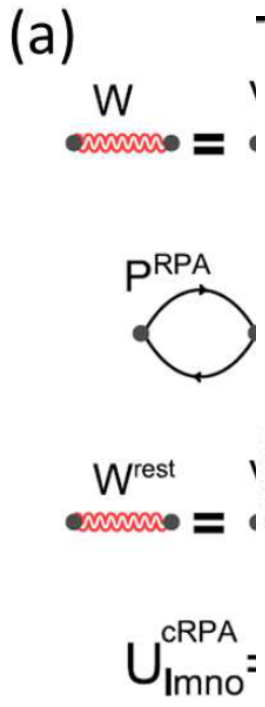
- effective model
- energy gain U^*

- equivalent for $U^* = U - V$

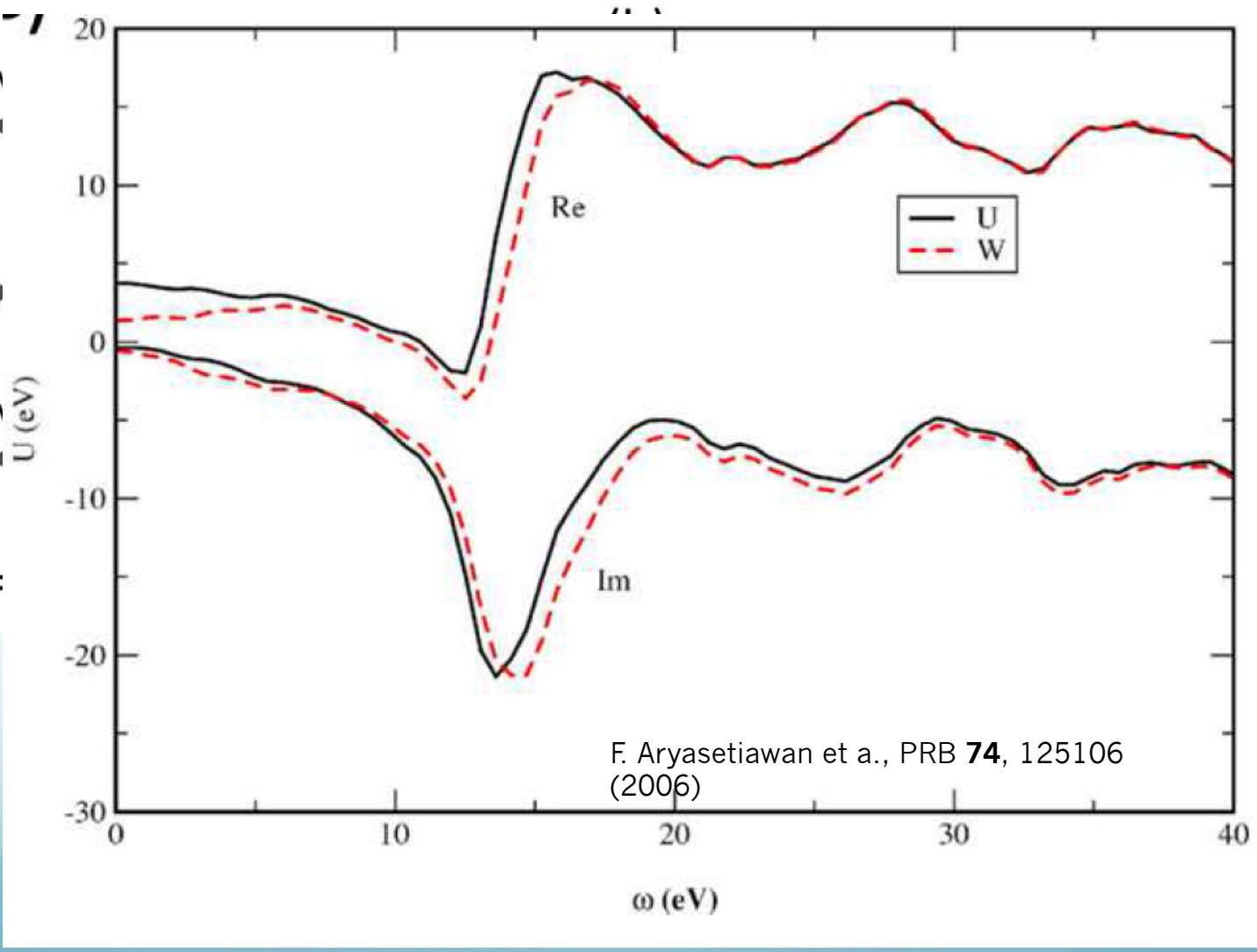
DFT++ Hamiltonian includes only local interactions within correlated subspace but all electrons provide screening

Partially screened interaction in DFT++ Hamiltonian → cLDA, cRPA

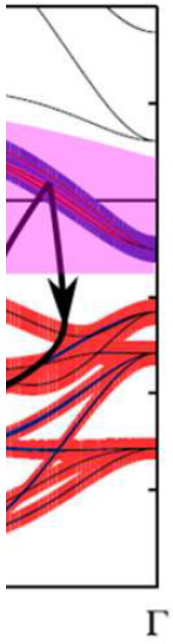
The constrained random phase approximation (cRPA)



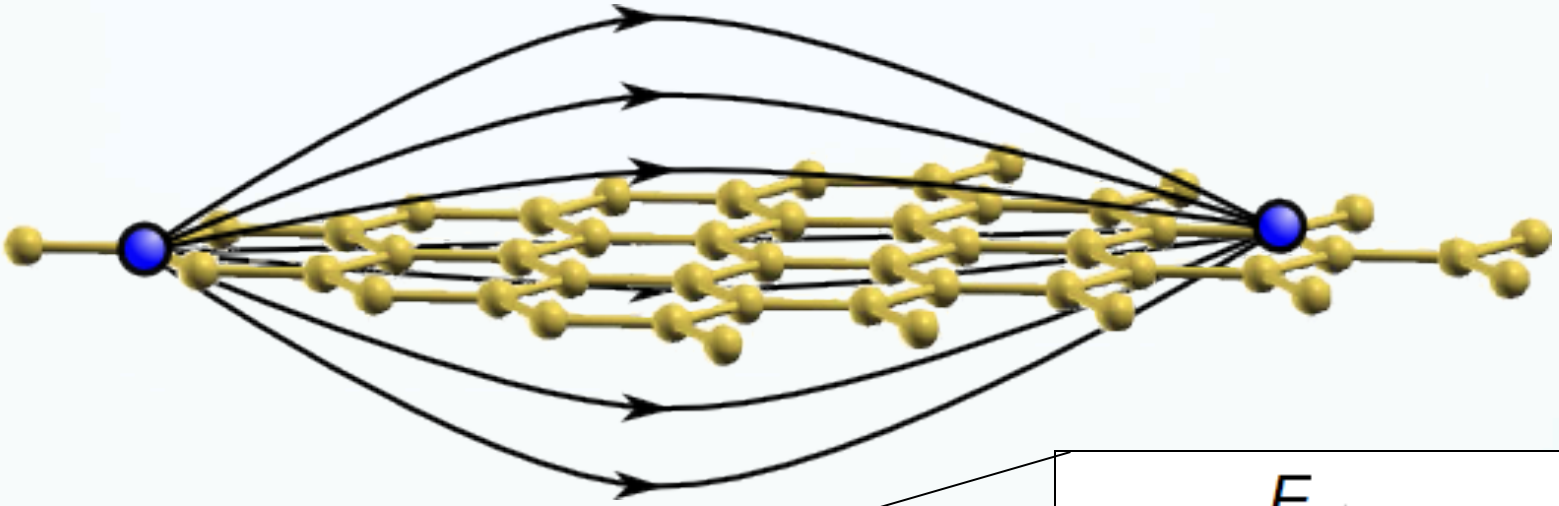
P. Hansmann et

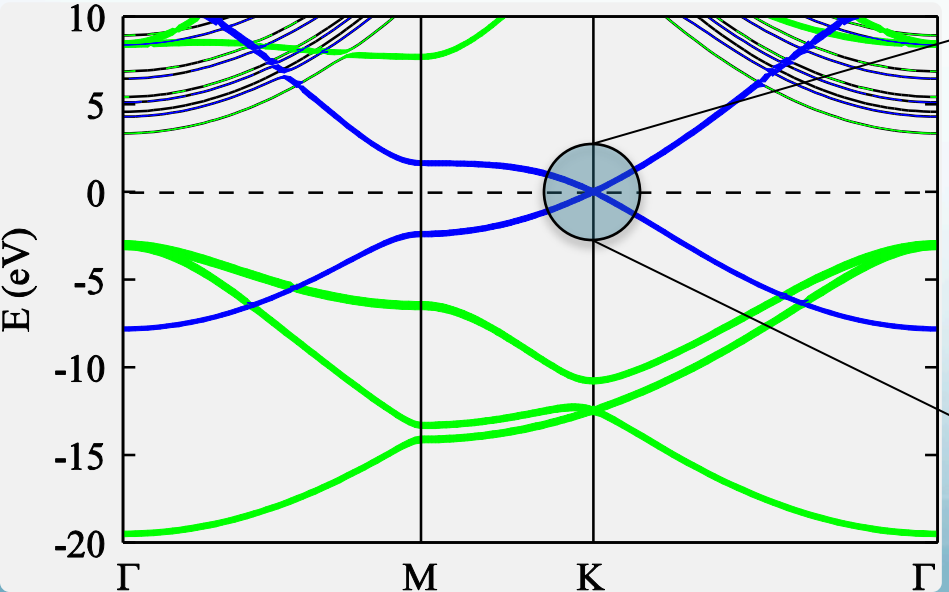


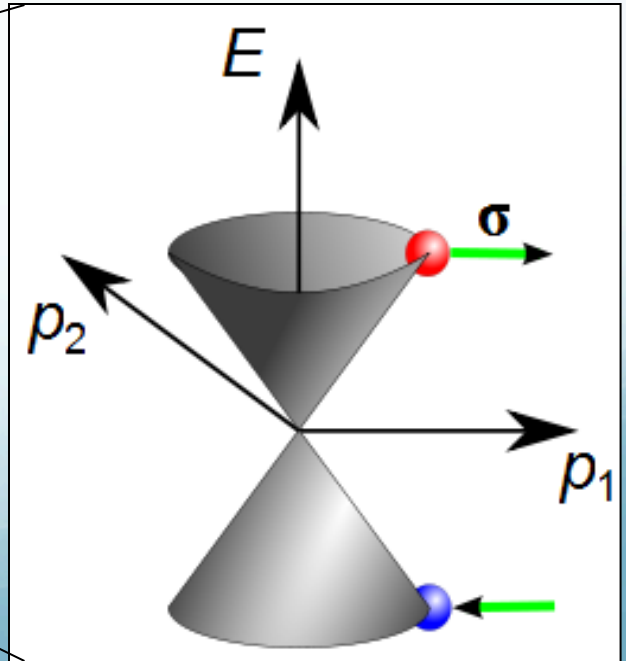
F. Aryasetiawan et a., PRB **74**, 125106 (2006)



interactions
Example: Graphene

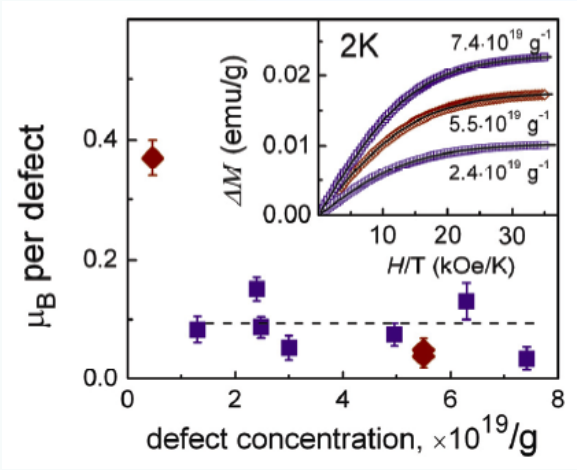






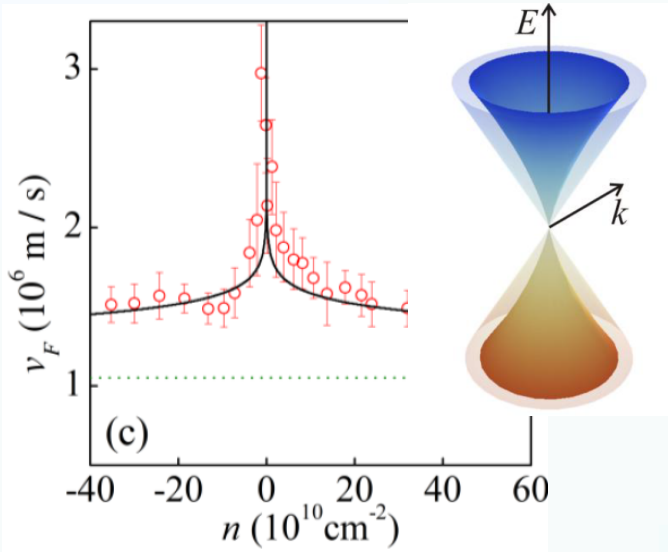
materials

Magnetism



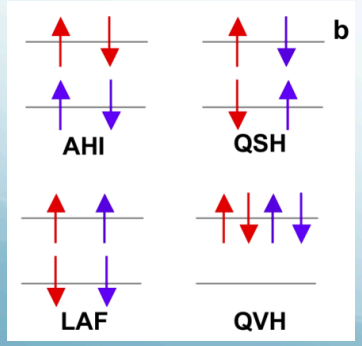
Defect induced magnetism in graphene. Magnetic moment per vacancy.
Nair et al. Nature Phys. 8, 199 (2012).

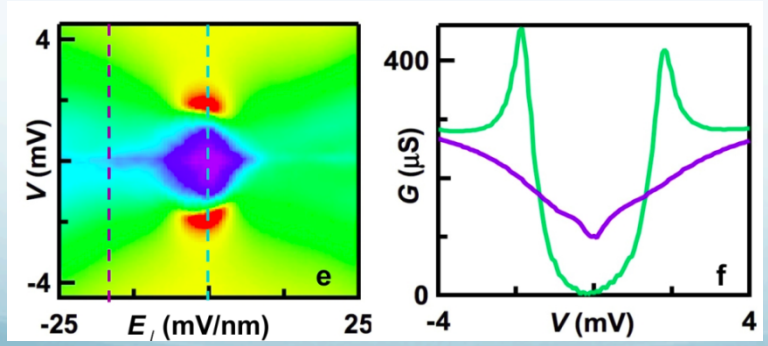
Renormalized Fermi velocity



Elias et al., Nature Phys. 7, 701 (2011)

Symmetry broken ground states in multilayer graphene





J. Velasco Jr et al., Nature Nanotech. 7, 156 (2012).

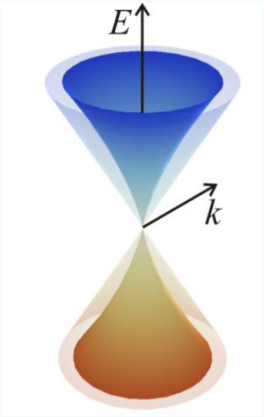
B. Feldman et al., Nature Physics 5, 893 (2009).

R. T. Weitz, et al., Science 330, 812 (2010).

A. S. Mayorov et al., Science 333, 860 (2011).

How to model Coulomb interactions?

Renormalized Fermi velocity



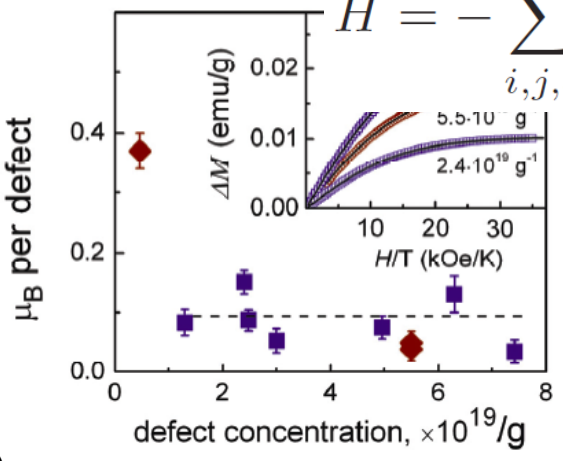
$$H = v_f(p \cdot \sigma)$$

$$V(p) = 2\pi e^2/p$$

Kotov et al.,Rev. Mod. Phys. 84, 1067 (2012).

Magnetism

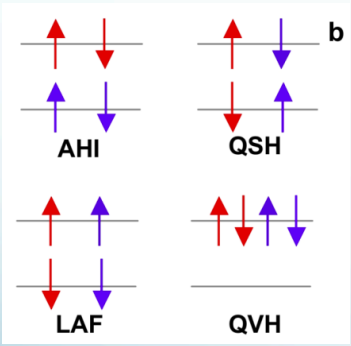
$$H = -\sum_{i,j,\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



U ~ t ~ 3 eV

Yazyev, *Rep. Prog. Phys.* **73** 056501 (2010).

Symmetry broken ground states in multilayer graphene



$$H = -\sum_{i,j,\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{1}{2} \sum_{\substack{i \neq j \\ \sigma,\sigma'}} V_{ij} n_{i\sigma} n_{j\sigma'}$$

Hydrocarbons: PPP model

TABLE II. Integrals over atomic orbitals (ev).

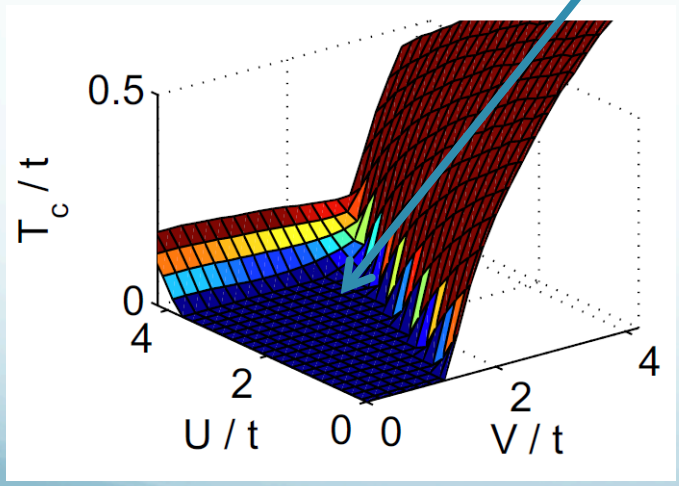
Integral[a]	R (in A)	Value[b]	Molecule[d]
(11|11)cc	0.00	10.53	all
(11|22)cc	1.35	7.38	ethylene
	1.39	7.30	Bz, Py, Pz, Pm
	1 46	7.16	cB, tB
	2.41	5.46	Bz, Py, Pz, Pm, T

R. Pariser and R. G. Parr, J. Chem. Phys. 21, 767 (1953).
J. A. Pople, Proc. Phys. Soc. A 68, 81 (1955).

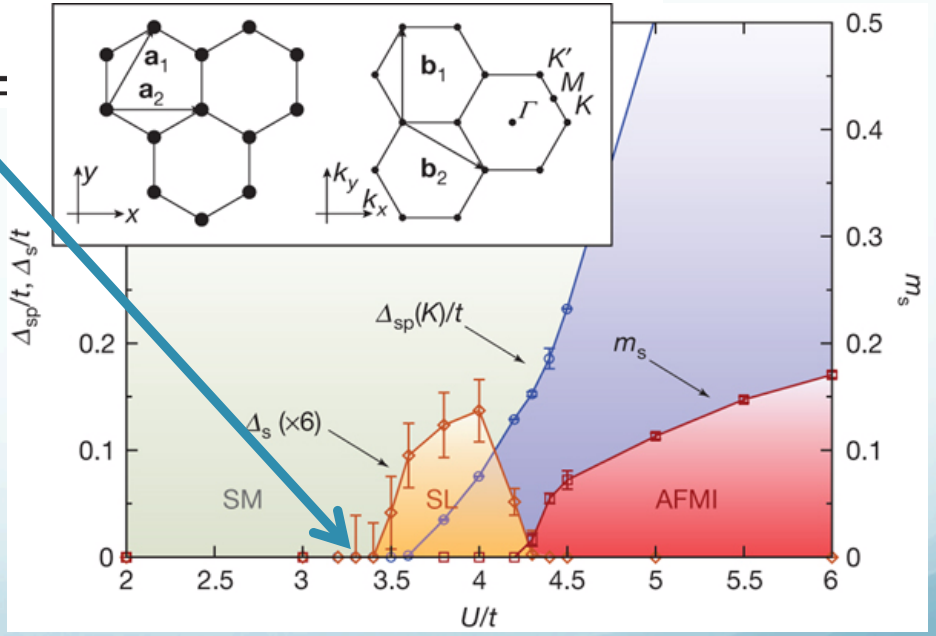
Coulomb interactions in graphene

	Graphene		Graphite	
	Bare	cRPA	Bare	cRPA
$U_{00}^{A \text{ or } B}$ (eV)	17.0	9.3	17.5, 17.7	8.0, 8.1
U_{01} (eV)	8.5	5.5	8.6	3.9
$U_{02}^{A \text{ or } B}$ (eV)	5.4	4.1	5.4, 5.4	2.4, 2.4
U_{03} (eV)	4.7	3.6		

TW et al., PRL **106**, 236805 (2011)



Honerkamp, PRL 100, 146404 (2008)



Z. Y. Meng et al., Nature **464**, 847-851 (2010)

Optimal local Hubbard model for graphene

extended Hubbard model

$$H_{\text{full}} = -\sum_{i,j,\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \frac{1}{2} \sum_{\substack{i \neq j \\ \sigma,\sigma'}} V_{ij} n_{i\sigma} n_{j\sigma'}$$

effective strictly local Hubbard model

$$H_{\text{eff}} = -\sum_{i,j,\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U^{*} \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

U*=?

Peierls-Feynman-Bogoluibov variational principle

Choose U* such that density matrix of effective system approximates density matrix of original system as close as possible

Vary the functional $\tilde{\Phi}[\rho^*]$ w.r.t. U^*

$$\tilde{\Phi}[\rho^*] = \Phi^* + \langle H - H^* \rangle^* \geq \tilde{\Phi}[\rho]$$

- Φ^* is the free energy of the eff. system
- $\langle \cdots \rangle^*$ means expectation value w.r.t. the eff. system

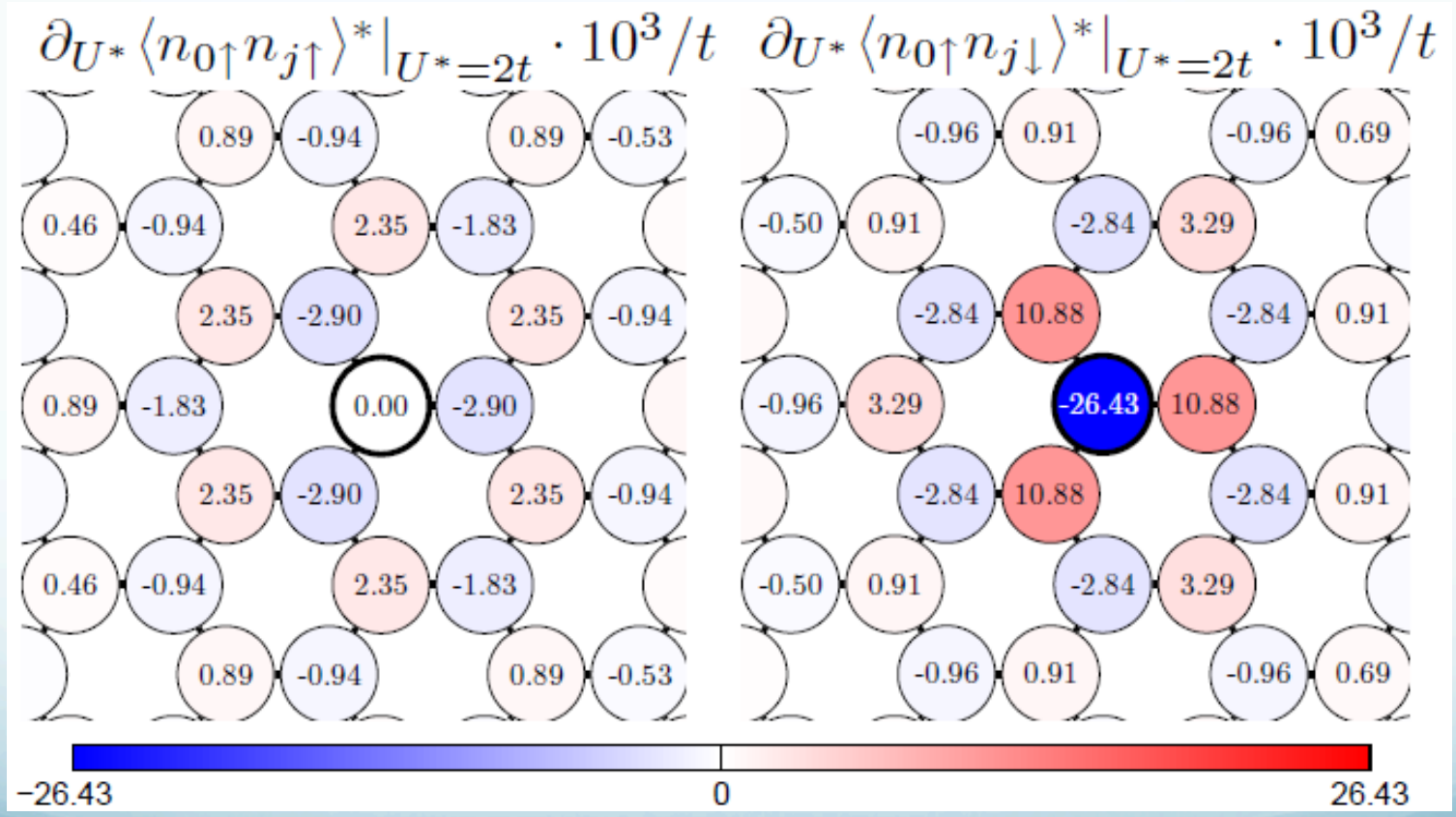
effective local interaction (for translational invariant systems)

$$U^* = U + \sum_{\substack{j \neq 0 \\ \sigma'}} V_{0j} \frac{\partial_{U^*} \langle n_{0\uparrow} n_{j\sigma'} \rangle^*}{\partial_{U^*} \langle n_{0\uparrow} n_{0\downarrow} \rangle^*}$$

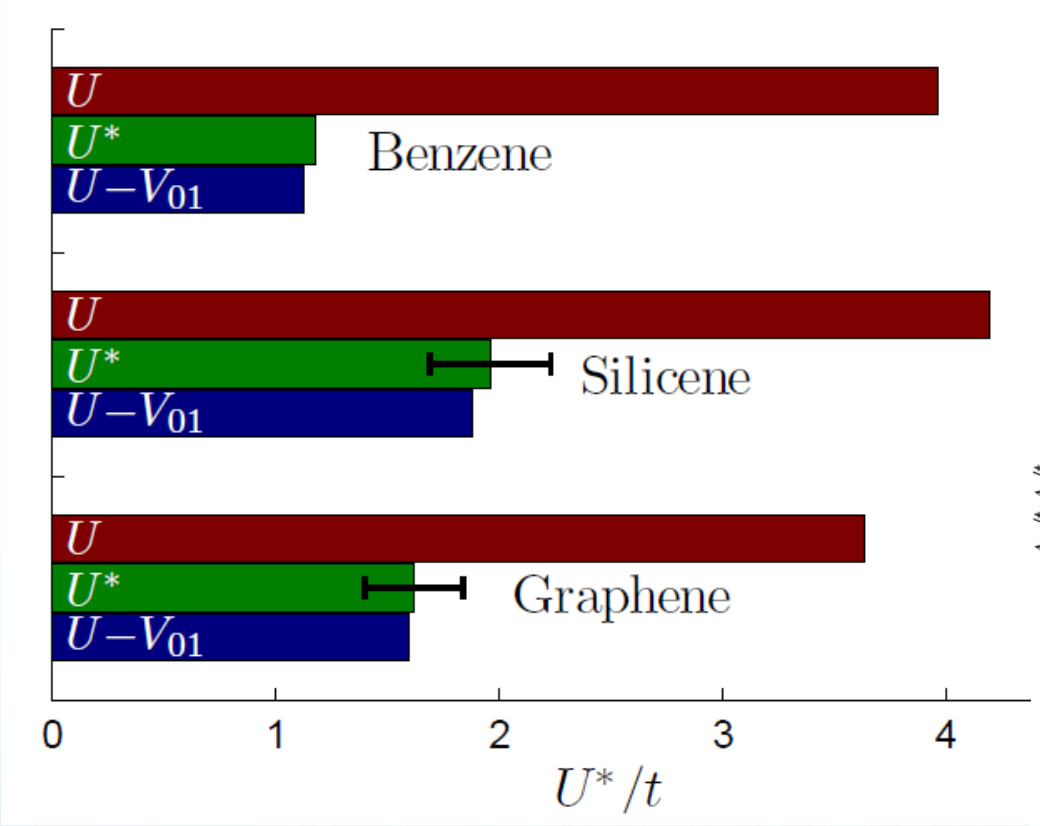
M. Schüler, TW et al., PRL 111, 036601 (20

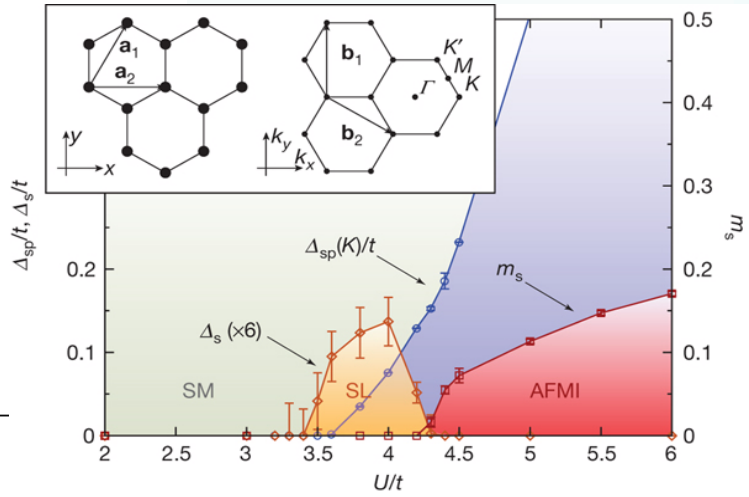
Non local density correlation functions

- Calculations within DQMC (Quest code)



Renormalized local interactions





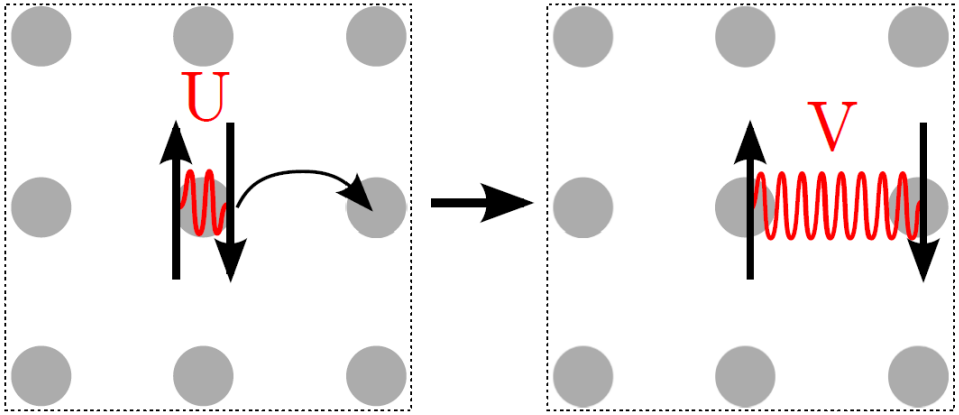
Z. Y. Meng et al., Nature **464**, 847-851 (201

Effective local interaction U* reduced by the non-local interactions

Dirac semimetal phase stabilized against the AFM phase

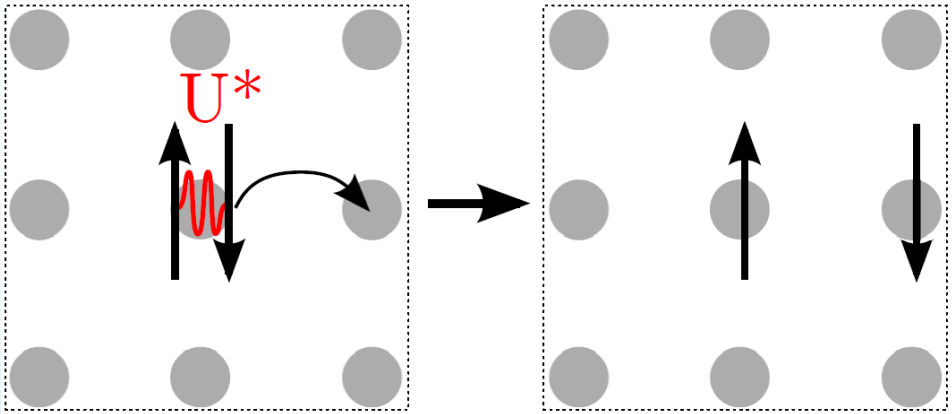
M. Schüler, TW et al., PRL 111, 036601 (201

A gedanken experiment



- full model
- energy gain $U - V$

- equivalent for $U^* = U - V$



- effective model
- energy gain U^*

effective local interaction (for translational invariant systems)

$$U^* = U + \sum_{\substack{j \neq 0 \\ \sigma'}} V_{0j} \frac{\partial_{U^*} \langle n_{0\uparrow} n_{j\sigma'} \rangle^*}{\partial_{U^*} \langle n_{0\uparrow} n_{0\downarrow} \rangle^*}$$

$$\sum_{j\sigma} \langle n_{0\uparrow} n_{j\sigma} \rangle^* = \text{const.}$$

$$\partial_{U^*} \langle n_{0\uparrow} n_{0\downarrow} \rangle^* = - \sum_{j \neq 0, \sigma} \partial_{U^*} \langle n_{0\uparrow} n_{j\sigma} \rangle^*.$$

Model Hamiltonians from first principles

$$H = \sum_k \epsilon_k c_k^\dagger c_k - \mu_{DC} \sum_{\mathbf{R},m} d_{\mathbf{R},m}^\dagger d_{\mathbf{R},m}$$
$$+ \frac{1}{2} \sum_{\mathbf{R},m\ldots m'''} U_{m\ldots m'''} d_{\mathbf{R},m}^\dagger d_{\mathbf{R},m'}^\dagger d_{\mathbf{R},m''} d_{\mathbf{R},m'''}$$

Remaining issues:

- **Double counting μ_{DC}**
- **Charge self-consistency**

$$G_{\mathrm{DMFT}} \rightarrow \rho_{\mathrm{DMFT}}(\mathbf{r}) \neq \rho_{\mathrm{DFT}}(\mathbf{r})$$

Double counting problem

$$H = \sum_k \epsilon_k c_k^\dagger c_k - \mu_{DC} \sum_{\mathbf{R},m} d_{\mathbf{R},m}^\dagger d_{\mathbf{R},m}$$
$$+ \frac{1}{2} \sum_{\mathbf{R},m\ldots m'''} U_{m\ldots m'''} d_{\mathbf{R},m}^\dagger d_{\mathbf{R},m'}^\dagger d_{\mathbf{R},m''} d_{\mathbf{R},m'''}$$

- Interaction "U" added
- Kohn Sham eigenvalues ε_k already include some interaction contribution
- For LDA or GGA xc-functionals not clear which part of exchange and correlation contribution to ε_k corresponds to added interaction "U"
  - Nonlinearity of xc-functionals like LDA or GGA
  - No diagrammatic representation of LDA or GGA

Fully diagrammatic approaches (e.g. GW+DMFT)

Empirical schemes to fix μ_{DC}

Empirical schemes to fix μ_{DC}

- Assumption on how xc-functional includes xc-effects or quantities it describes correctly

  - Correct charge by LDA, GGA:

$$\mathrm{Tr}\ G^{imp}_{mm'}(i\omega_n) \stackrel{!}{=} \mathrm{Tr}\ G^{0,loc}_{mm'}(i\omega_n).$$

  - Correct static mean field components of self-energy in LDA

$$\mathrm{Re}\,\mathrm{Tr}\,(\Sigma^{imp}_{mm'}(i\omega_N)) \stackrel{!}{=} 0.$$

  - Correct self-energy at Fermi level in LDA

$$\mathrm{Re}\,\mathrm{Tr}\,(\Sigma^{imp}_{mm'}(i2\pi/\beta)) \stackrel{!}{=} 0.$$

Empirical schemes to fix μ_{DC}

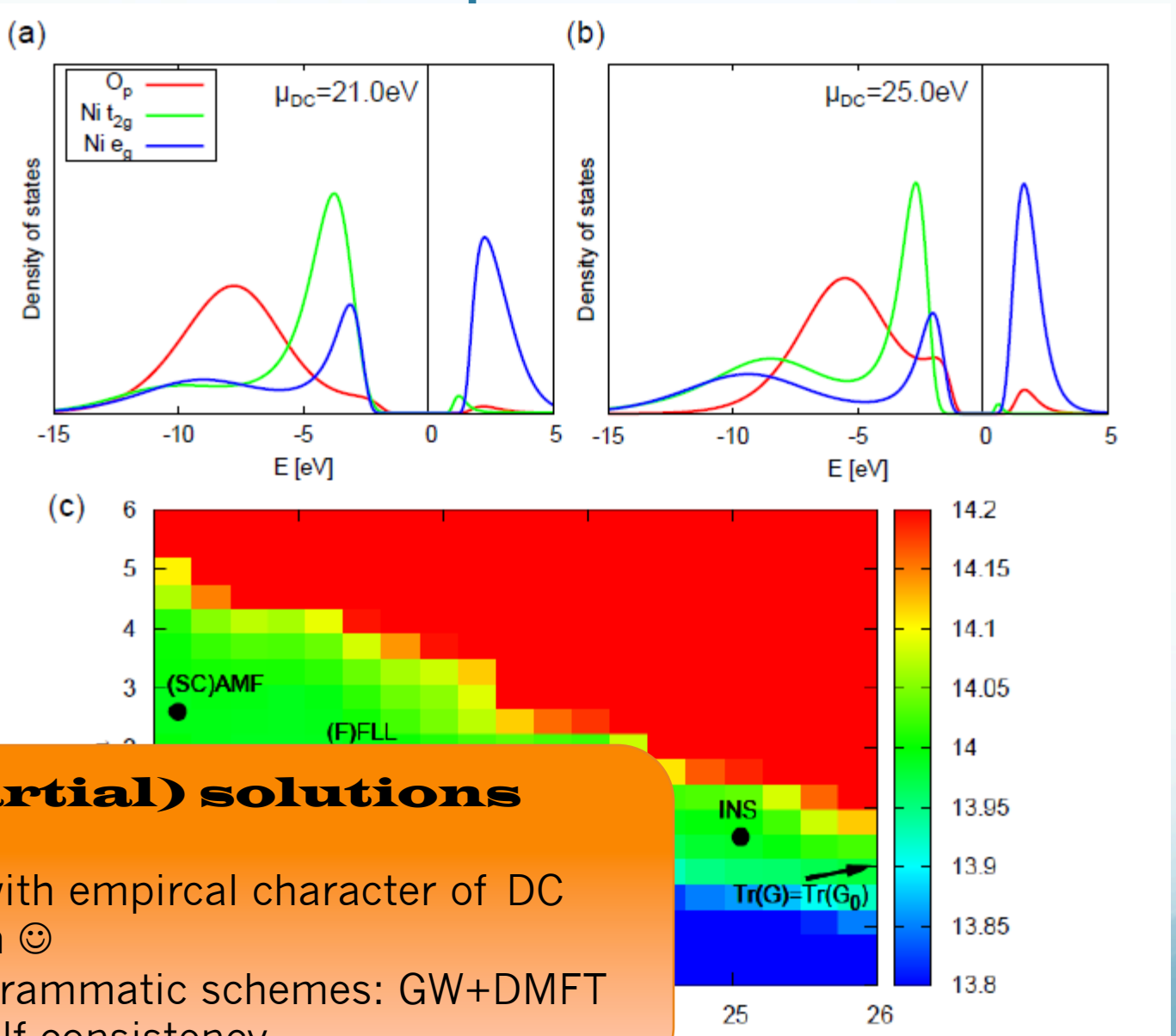
- Around mean field:
  - LDA eigenvalues include xc-effects in spherically / orbitally averaged manner

$$\mu_{dc}^{AMF} = \sum_{m'} U_{mm'} n^0 + \sum_{m',m' \neq m} (U_{mm'} - J_{mm'}) n^0$$

- Fully localized limit
  - LDA / GGA total energies fine for purely integer occupancies

$$\mu_{dc}^{FLL} = U(N_{\mathrm{imp}} - 1/2) + J(N_{\mathrm{imp}}^{\sigma} - 1/2)$$

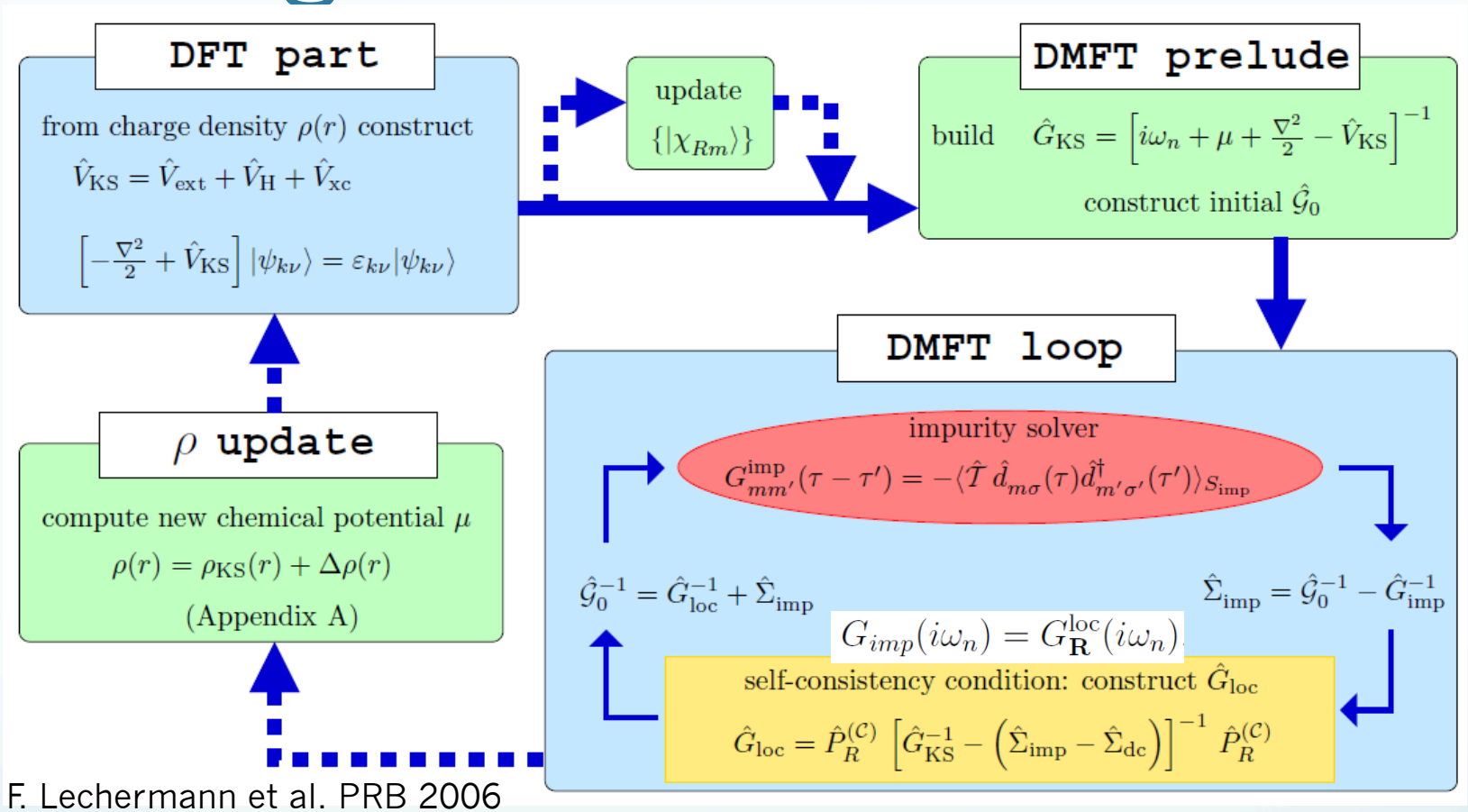
Example: NiO



(Partial) solutions

- Arrange with empircal character of DC correction ☺
- Fully diagrammatic schemes: GW+DMFT
- Charge-self-consistency

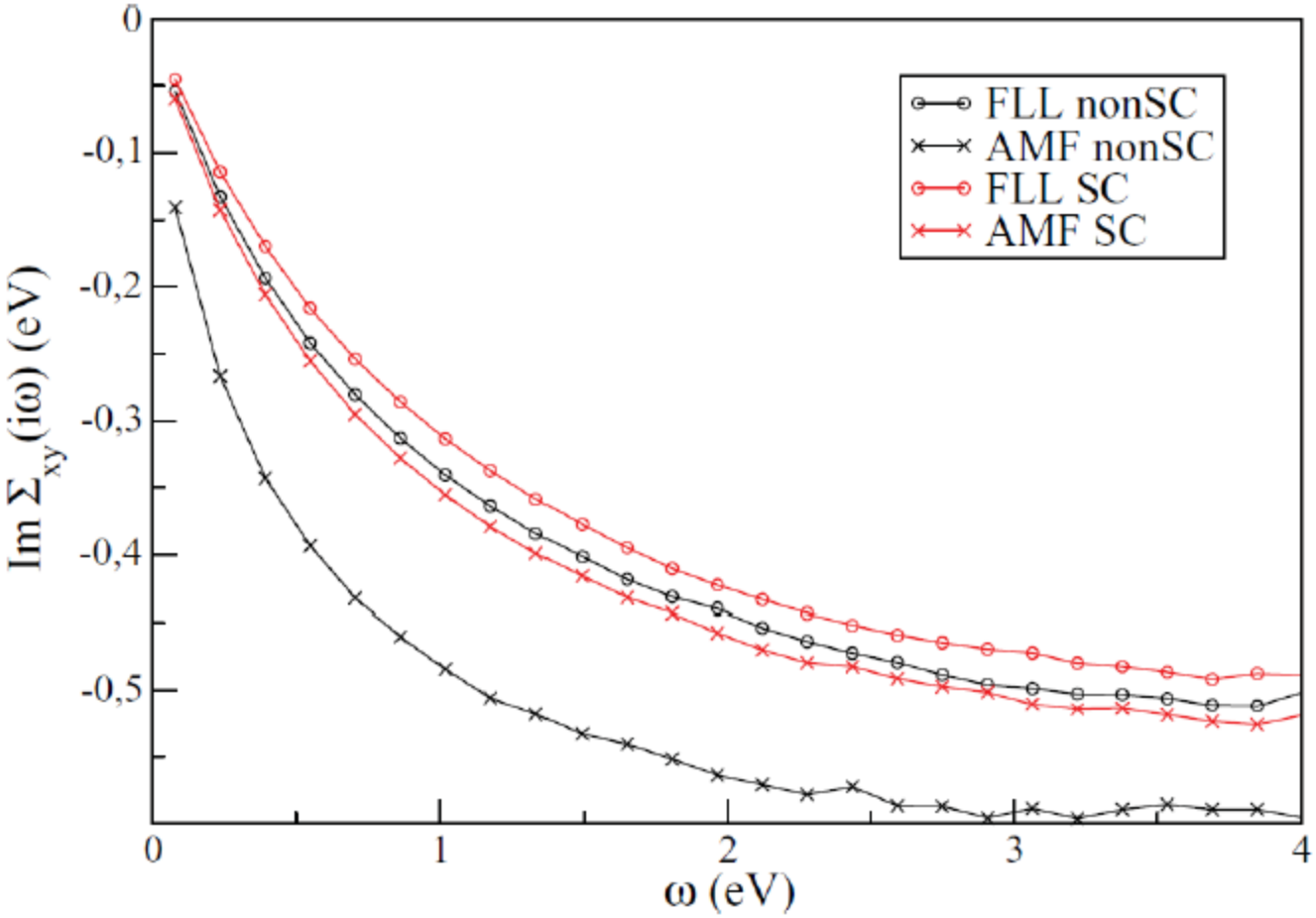
Charge self-consistent LDA



F. Lechermann et al. PRB 2006

- Charge redistribution due to many body part $\rho(r) = \frac{1}{\beta}\sum_{\mathbf{k},\alpha,\alpha',n} \langle r|B_{\mathbf{k}\alpha}\rangle G_{\alpha\alpha'}(\mathbf{k}, i\omega_n)\langle B_{\mathbf{k}\alpha'}|r\rangle$
- „Bare“ part of DFT++ Hamiltonian includes interactions: Hartree and xc potentia
- Hartree terms within DFT++ charge self-consistency counteract charge redistribu
- Scheme less sensitive to double counting correction?

Example: LaFeAsO



[Aichhorn *et al.*, PRB (2011)]

Conclusions

$$H = \sum_k \epsilon_k c_k^\dagger c_k - \mu_{DC} \sum_m d_m^\dagger d_m + \frac{1}{2} \sum_{m \ldots m'''} U_{m \ldots m'''} d_m^\dagger d_{m'}^\dagger d_{m''} d_{m'''}$$

- Number of adjustable parameters in *H* can be significantly reduced by deriving them from first principles → (almost) „ab-initio“ studies of correlated materials
- Useful tools
  - Projectors
  - cRPA approach
- Issues
  - Double counting
  - Non-local interactions