U.S. DEPARTMENT OF ENERGY | Office of Science

COMSCOPE
from codes to spectroscopies

Gutzwiller Rotationally Invariant Slave-Boson method and its combination with Density Functional Theory

Yongxin Yao, Sangkook Choi, Andrey Kutepov,
Nicola Lanata and Gabriel Kotliar

THE Ames Laboratory
Creating Materials & Energy Solutions
U.S. DEPARTMENT OF ENERGY

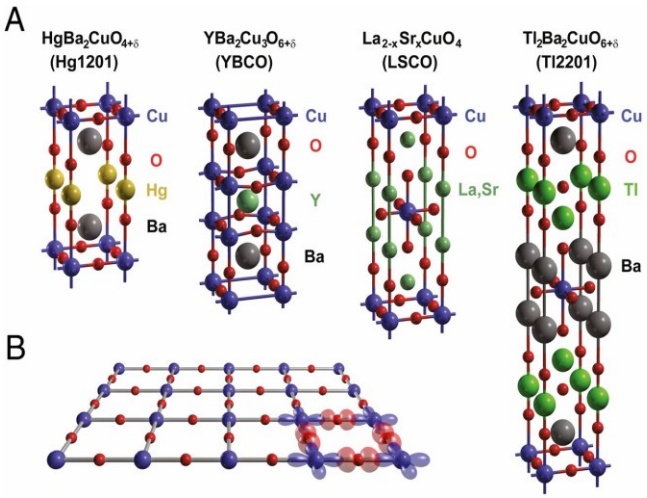
IOWA STATE UNIVERSITY

AARHUS UNIVERSITET

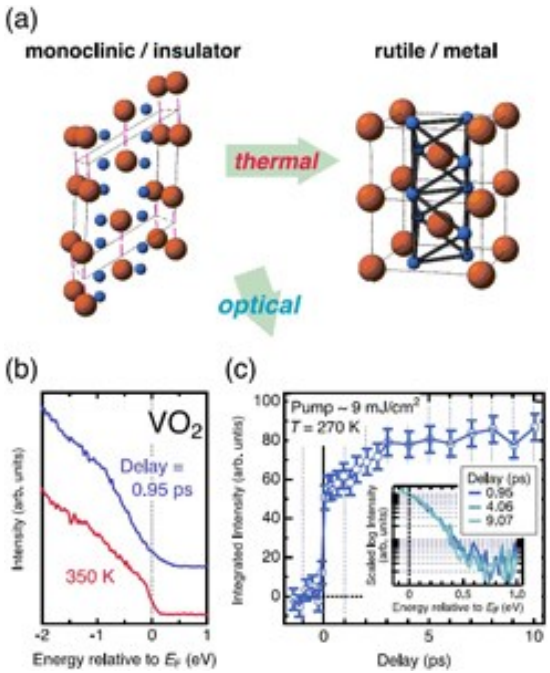
RUTGERS
THE STATE UNIVERSITY OF NEW JERSEY

BROOKHAVEN NATIONAL LABORATORY

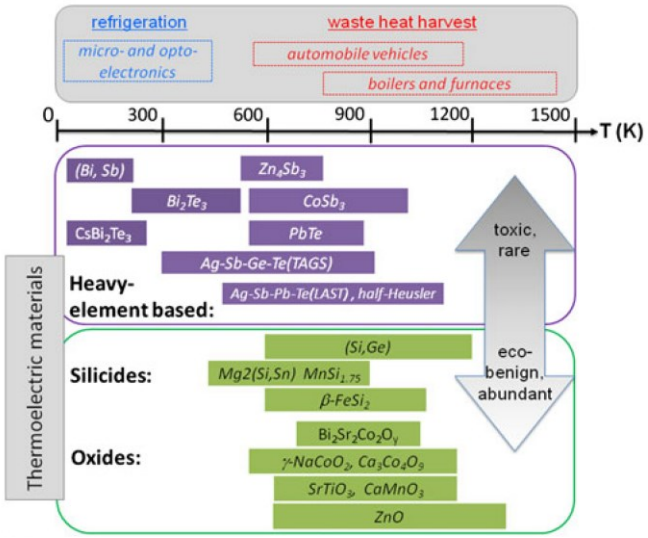
Correlated Functional Quantum Materials



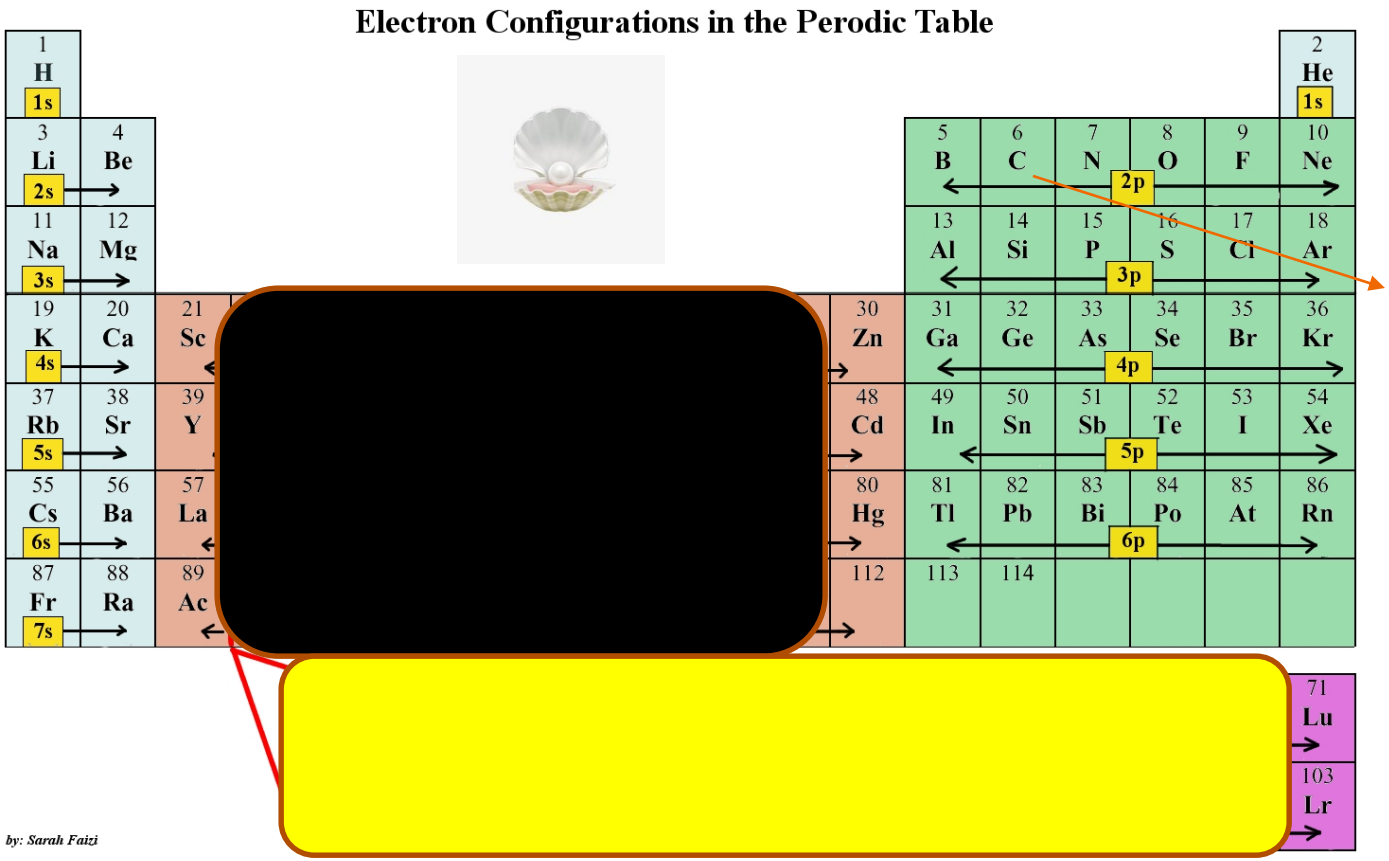
Highest-Tc superconductors

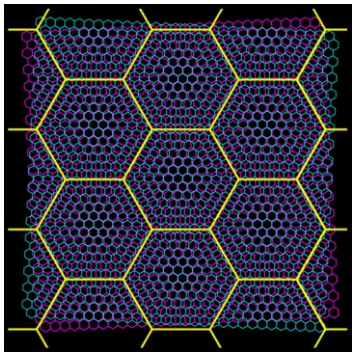


Fastest optical switches

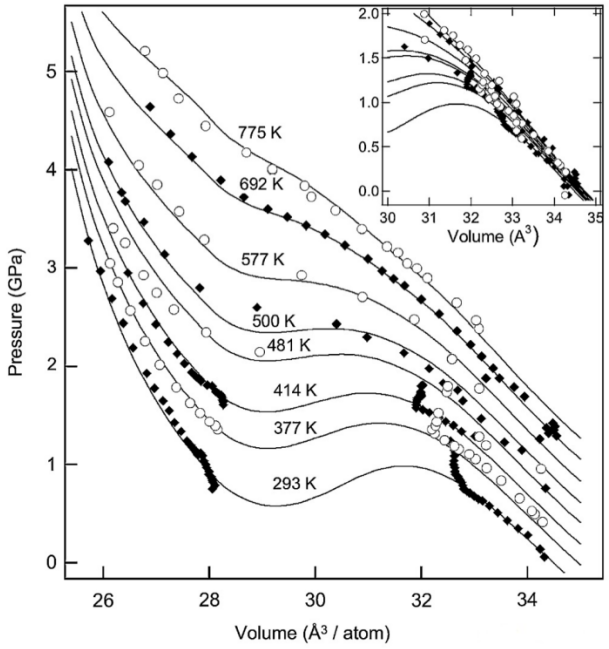


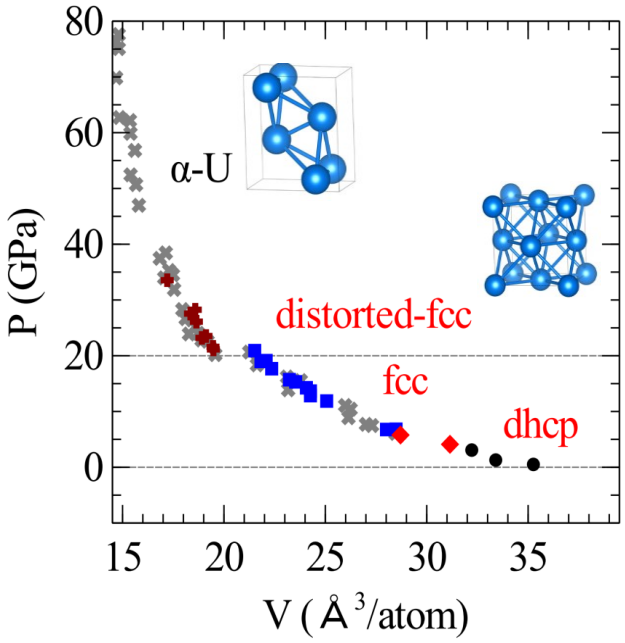
Elements with electron correlation effects

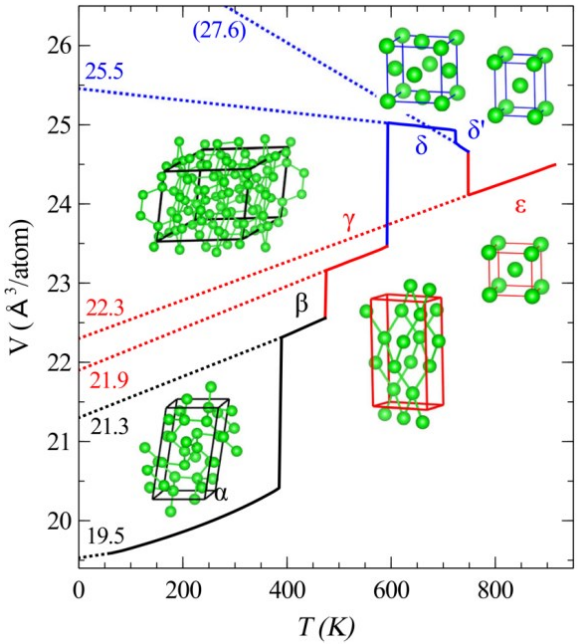




Electron correlation in single element phase







FCC-Ce

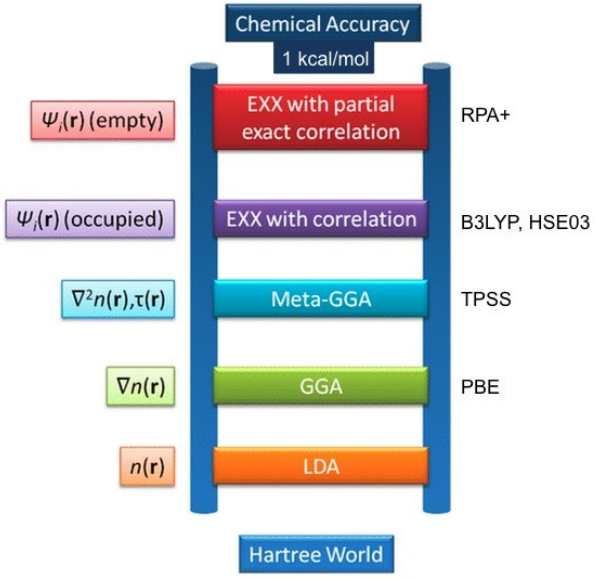
Pr

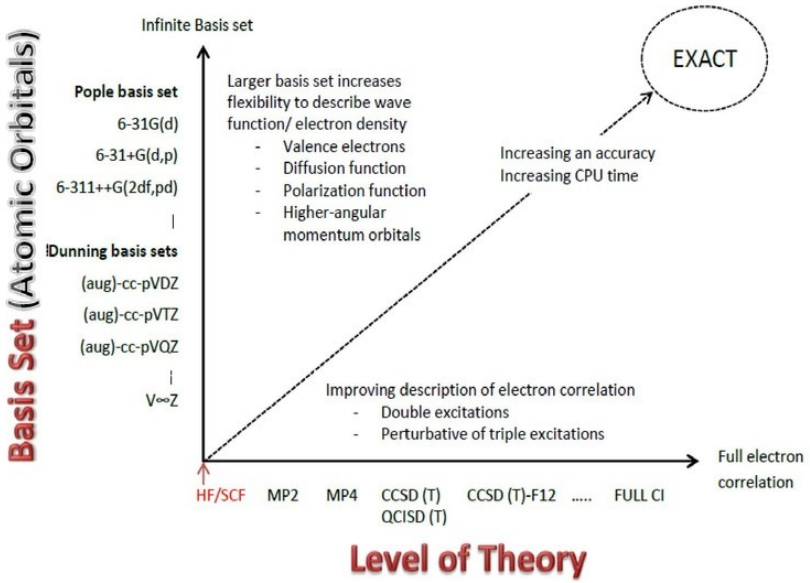
Pu

Many-electron problem

$$H = \sum_i -\frac{\hbar^2}{2m}\nabla_i^2 + \sum_{il}\left(-\frac{e^2}{4\pi\varepsilon_0^2}\frac{Z_l}{|\vec{r}_i - \vec{R}_l|}\right) + \frac{1}{2}\sum_{i\neq j}\left(\frac{e^2}{4\pi\varepsilon_0^2}\frac{1}{|\vec{r}_i - \vec{r}_j|}\right)$$

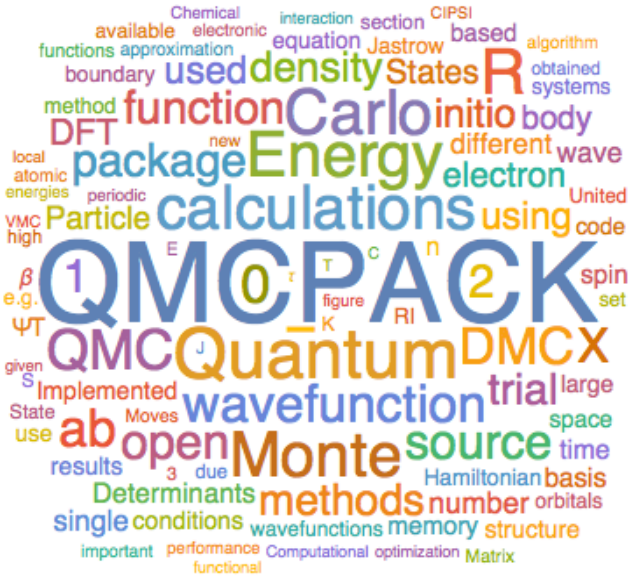
Approaches:

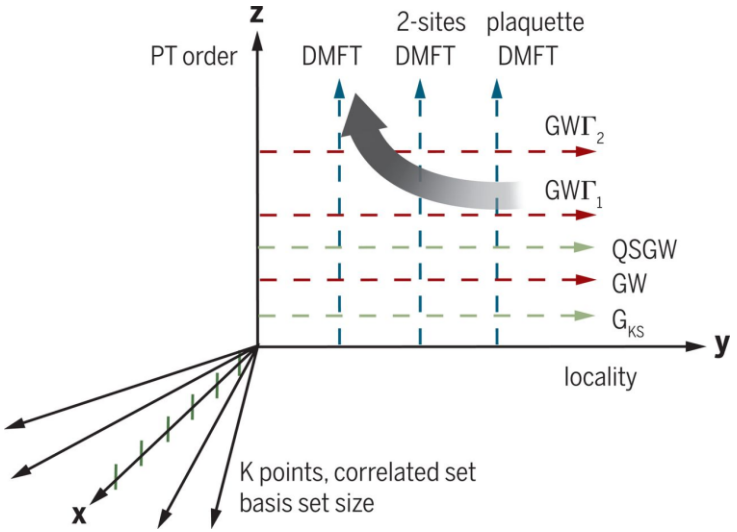




Toward a predictive theory of correlated materials

$$\Psi\left(\{\vec{R}\}\right) = \sum_{i=1}^{M} c_i D_i^{\uparrow} D_i^{\downarrow} e^{J(\{\vec{R}\})}$$

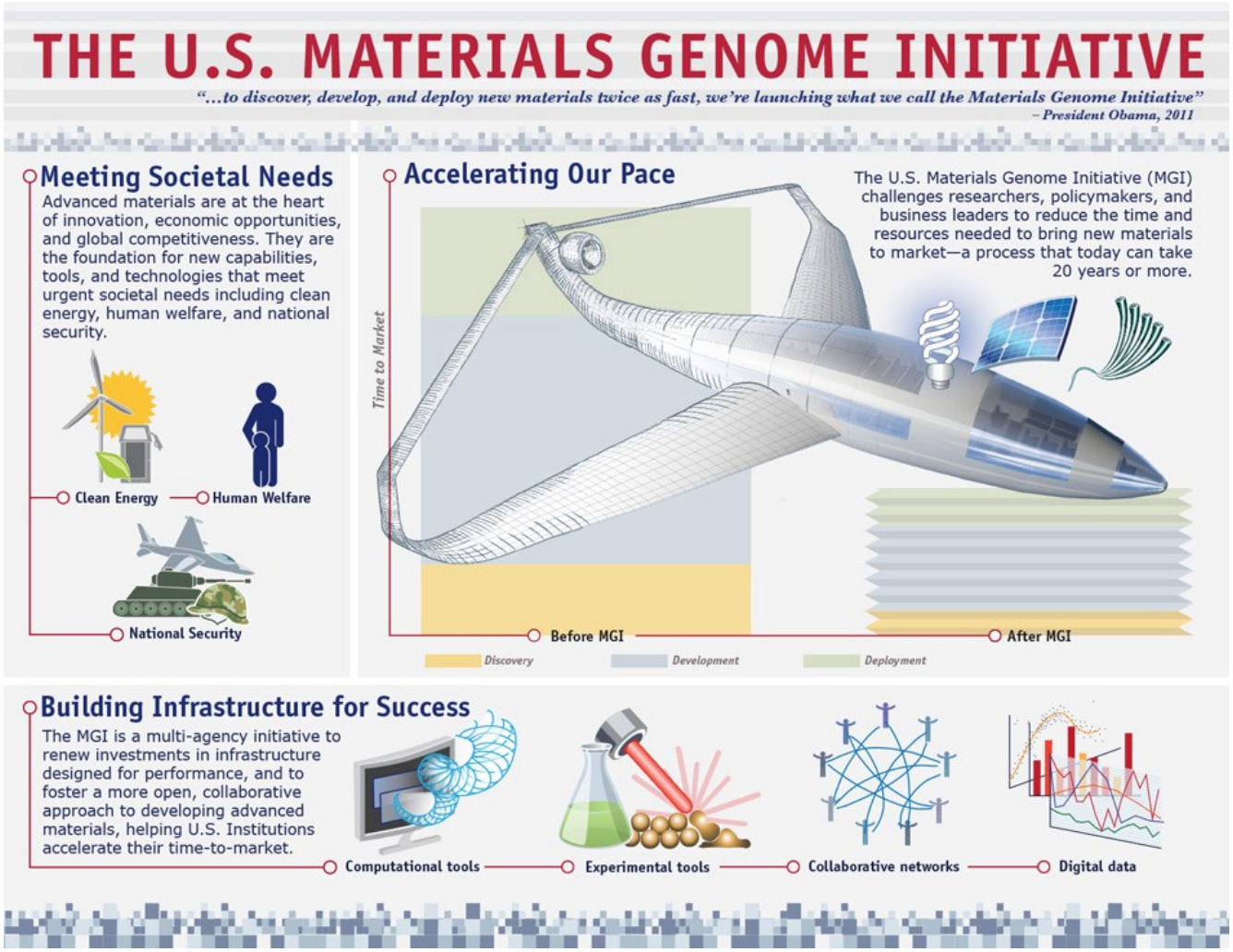




$$G(\vec{r},\vec{r'},\omega) = \frac{1}{\omega + \nabla^2 + \mu - V_{\mathrm{H}} - V_{\mathrm{cryst}} - \Sigma(\omega)}$$

Empowered by the massively parallel high-performance computations.

Efficiency matters



Features of G-RISB approach

Complementary to DMFT (CTQMC@T):

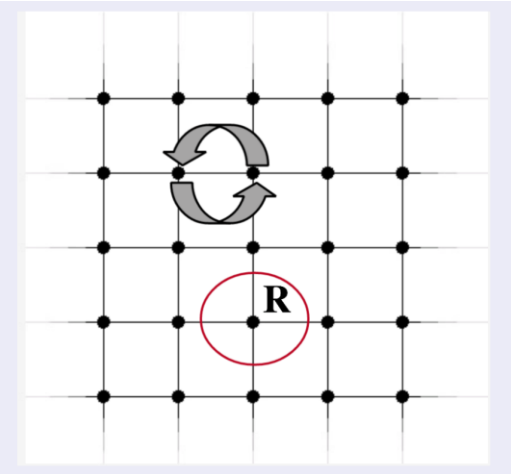
- Comparable accuracy for ground state properties (0K).
- Over two orders of magnitude greater efficiency.
- Able to treat any symmetry breaking (spatial, spin, etc.) without further approximations.
- Rotational/gauge invariance can be fully implemented.
- Semi-analytical, physically more transparent.

Single-band Hubbard Model

Hamiltonian:

$$H = \sum_{\mathbf{RR}'}{}' \sum_{\sigma} t_{\mathbf{RR}'} c^{\dagger}_{\mathbf{R}\sigma} c_{\mathbf{R}'\sigma} + \sum_{\mathbf{R}} H_{loc,\mathbf{R}}$$

$$= \sum_{\mathbf{RR}'}{}' \sum_{\sigma} t_{\mathbf{RR}'} c^{\dagger}_{\mathbf{R}\sigma} c_{\mathbf{R}'\sigma} + \sum_{\mathbf{R}} \left(\sum_{\sigma} \boldsymbol{\varepsilon}_{\sigma} c^{\dagger}_{\mathbf{R}\sigma} c_{\mathbf{R}\sigma} + U c^{\dagger}_{\mathbf{R}\uparrow} c_{\mathbf{R}\uparrow} c^{\dagger}_{\mathbf{R}\downarrow} c_{\mathbf{R}\downarrow} \right)$$



$\mathbf{R}$: site index
σ: spin index = ↑, ↓
U: Hubbard interaction strength
$t_{\mathbf{RR}'}$: hopping amplitude
$\boldsymbol{\varepsilon}_{\sigma}$: orbital level

$$\mathcal{H}_F = \otimes_{\mathbf{R}} \mathcal{H}_F^{\mathbf{R}};\ \ \mathcal{H}_F^{\mathbf{R}} = \{|0;\mathbf{R}\rangle, |\uparrow;\mathbf{R}\rangle, |\downarrow;\mathbf{R}\rangle, |\uparrow\downarrow;\mathbf{R}\rangle\}$$
$$= \{|0\rangle, c^{\dagger}_{\mathbf{R}\uparrow}|0\rangle, c^{\dagger}_{\mathbf{R}\downarrow}|\mathbf{0}\rangle, c^{\dagger}_{\mathbf{R}\uparrow} c^{\dagger}_{\mathbf{R}\downarrow}|0\rangle\}$$

Single-band Hubbard Model

Gutzwiller wave-function:

$$|\Psi_G\rangle = \mathcal{P}|\Psi_0\rangle = \prod_{\mathbf{R}} \mathcal{P}_{\mathbf{R}}|\Psi_0\rangle = \prod_{\mathbf{R}} \left(\sum_{An} \lambda_{An} |A;\mathbf{R}\rangle\langle n;\mathbf{R}| \right) |\Psi_0\rangle$$

- A, n span the same local Hilbert space of dimension 2^M, however, they are not necessarily the same.
- $[\lambda_{An}]$, which is directly related with the local many-body density matrix, commutes with the local symmetry operations.
- For the single-band HM, $[\lambda_{An}]$ commutes with $\{N, S_z\}$ $\longrightarrow$Simple form for the projector: $\mathcal{P}_{\mathbf{R}}$=$\sum_F \lambda_F |F;\mathbf{R}\rangle\langle F;\mathbf{R}|$, with $\{|F;\mathbf{R}\rangle\} = \{|0;\mathbf{R}\rangle, |\uparrow;\mathbf{R}\rangle, |\downarrow;\mathbf{R}\rangle, |\uparrow\downarrow;\mathbf{R}\rangle\}$

Single-band Hubbard Model

Local onsite exact treatment: able to recover the atomic limit.

$$\langle \Psi_G | H_{loc,\mathbf{R}} | \Psi_G \rangle = \sum_F p_{\mathbf{R},F} \langle F;\mathbf{R} | H_{loc,\mathbf{R}} | F;\mathbf{R} \rangle$$

Gutzwiller approximation:

$$\langle \Psi_G | c^\dagger_{\mathbf{R}\sigma} c_{\mathbf{R}'\sigma} | \Psi_G \rangle = z_{\mathbf{R},\sigma} z_{\mathbf{R}',\sigma} \langle \Psi_0 | c^\dagger_{\mathbf{R}\sigma} c_{\mathbf{R}'\sigma} | \Psi_0 \rangle$$

Hopping renormalization factor:

$$z_{\mathbf{R},\sigma} = \frac{\left(\sum_{FF'} \sqrt{p_F p_{F'}} \left| \langle F;\mathbf{R} | c^\dagger_{\mathbf{R}\sigma} | F';\mathbf{R} \rangle \right| \right)}{\sqrt{n_{\mathbf{R}\sigma}(1-n_{\mathbf{R}\sigma})}}$$

Gutzwiller constraints:

$$\sum_F p_{\mathbf{R},F} = 1; \quad \sum_F p_{\mathbf{R},F} \langle F;\mathbf{R} | n_{\mathbf{R}\sigma} | F;\mathbf{R} \rangle = n_{\mathbf{R}\sigma}$$

Single-band Hubbard Model

Total energy:

$$\langle \Psi_G | H | \Psi_G \rangle = \sum_{\mathbf{R}\mathbf{R}'}{}' \sum_{\sigma} z_{\mathbf{R},\sigma} z_{\mathbf{R}',\sigma} t_{\mathbf{R}\mathbf{R}'} \langle \Psi_0 | c^{\dagger}_{\mathbf{R}\sigma} c_{\mathbf{R}'\sigma} | \Psi_0 \rangle + \sum_{F} p_{\mathbf{R},F} \langle F; \mathbf{R} | H_{loc,\mathbf{R}} | F; \mathbf{R} \rangle$$

Gutzwiller constraints:

$$\sum_{F} p_{\mathbf{R},F} = 1; \quad \sum_{F} p_{\mathbf{R},F} \langle F; \mathbf{R} | n_{\mathbf{R}\sigma} | F; \mathbf{R} \rangle = n_{\mathbf{R}\sigma}$$

$$[H_{loc,\mathbf{R}}] = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & \varepsilon_{\uparrow} & 0 & 0 \\ 0 & 0 & \varepsilon_{\downarrow} & 0 \\ 0 & 0 & 0 & \varepsilon_{\uparrow} + \varepsilon_{\downarrow} + U \end{pmatrix}$$

A constrained minimization problem.

Generic (Kohn-Sham) Hubbard Hamiltonian

$$\hat{H} = \sum_{\mathbf{k}} \sum_{ij} \sum_{\alpha\beta} \epsilon^{\alpha\beta}_{\mathbf{k},ij} \, c^{\dagger}_{\mathbf{k}i\alpha} c_{\mathbf{k}j\beta} + \sum_{\mathbf{R}i \in \mathrm{corr}} \hat{H}^{\mathrm{loc}}_{i} [\{c^{\dagger}_{\mathbf{R}i\alpha}\}, \{c_{\mathbf{R}i\alpha}\}]$$

$$\epsilon_{\mathbf{k}} = \left[\begin{array}{c|c|c} \text{i=0} & * & * \\ \hline * & \text{i=1} & * \\ \hline * & * & \text{i=2} \end{array}\right]$$

$\mathbf{R}$ = label unit cell

$\mathbf{k}$ = Fourier-conjugate of $\mathbf{R}$

Π_i = DMFT projector

$\sum_{\mathbf{k}} \epsilon_{\mathbf{k},ii} = 0 \quad \forall\, i$ correlated

α, β = orbitals in $\mathbf{R}, i$

(Example: $i = 0$: uncorrelated orbitals; $i = 1$: d orbitals atom 1
$i = 2$: f orbitals atom 2 ...)

Variational energy

$$\hat{H} = \sum_{\mathbf{k}} \sum_{\alpha\beta} \boxed{\epsilon^{\alpha\beta}_{\mathbf{k},ij}} c^{\dagger}_{\mathbf{k}i\alpha} c_{\mathbf{k}j\beta} + \sum_{\mathbf{R}i} \boxed{\hat{H}^{\mathrm{loc}}_{\mathbf{R}i}[\{\hat{c}^{\dagger}_{\mathbf{R}i\alpha}\}, \{\hat{c}_{\mathbf{R}i\alpha}\}]}$$

$$z \longrightarrow \mathcal{R}$$

$$\sqrt{p} \longrightarrow \phi$$

$$\mathcal{E}[\Psi_0, \{\phi_i\}] = \frac{1}{\mathcal{N}} \sum_{\mathbf{k},ij} \sum_{ab} \left[\mathcal{R}_i \, \epsilon_{\mathbf{k},ij} \mathcal{R}^{\dagger}_j\right]_{ab} \langle \Psi_0 | f^{\dagger}_{\mathbf{k}ia} f_{\mathbf{k}jb} | \Psi_0 \rangle + \sum_{i \in \mathrm{corr}} \mathrm{Tr}\left[\phi_i \, \phi^{\dagger}_i \, H^{\mathrm{loc}}_i\right]$$

where

$$[\mathcal{R}_i]_{a\alpha} = \sum_b \mathrm{Tr}\left[\phi^{\dagger}_i \, F^{\dagger}_{i\alpha} \, \phi_i \, F_{ib}\right] [\Delta_{pi}(1 - \Delta_{pi})]^{-\frac{1}{2}}_{ba}$$

to be minimized satisfying the Gutzwiller contraints:

$$\mathrm{Tr}\left[\phi^{\dagger}_i \phi_i\right] = \langle \Psi_0 | \Psi_0 \rangle = 1$$

$$\mathrm{Tr}\left[\phi^{\dagger}_i \phi_i \, F^{\dagger}_{ia} F_{ib}\right] = \langle \Psi_0 | f^{\dagger}_{\mathbf{R}ia} f_{\mathbf{R}ib} | \Psi_0 \rangle \equiv \Delta_{pi}$$

Lagrange function with linearization

promoting Δ_p , R to independent variables using Lagrange multipliers.

$$\mathcal{L}[\Psi_0, E; \phi, E^c;\ \mathcal{R}, \mathcal{R}^\dagger, \lambda;\ \mathcal{D}, \mathcal{D}^\dagger, \lambda^c;\ \Delta_p] =$$

$$\frac{1}{\mathcal{N}} \langle \Psi_0 | \hat{H}_G^{\rm qp}[\mathcal{R}, \mathcal{R}^\dagger; \lambda] | \Psi_0 \rangle + E(1 - \langle \Psi_0 | \Psi_0 \rangle) +$$

$$\sum_i \mathrm{Tr}\left[\phi_i\ \phi_i^\dagger\ H_i^{\rm loc} + \sum_{a\alpha} \left([\mathcal{D}_i]_{a\alpha}\ \phi_i^\dagger\ F_{i\alpha}^\dagger\ \phi_i\ F_{ia} + \mathrm{H.c.} \right) + \sum_{ab} [\lambda_i^c]_{ab}\ \phi_i^\dagger \phi_i\ F_{ia}^\dagger F_{ib} \right] +$$

$$\sum_i E_i^c \left(1 - \mathrm{Tr}\left[\phi_i^\dagger \phi_i \right] \right)$$

$$- \sum_i \left[\sum_{ab} \left([\lambda_i]_{ab} + [\lambda_i^c]_{ab} \right) [\Delta_{pi}]_{ab} + \sum_{ca\alpha} \left([\mathcal{D}_i]_{a\alpha}\ [\mathcal{R}_i]_{c\alpha}\ [\Delta_{pi}(1 - \Delta_{pi})]^{\frac{1}{2}}_{c\alpha} + \mathrm{c.c.} \right) \right]$$

where

$$\hat{H}_G^{\rm qp}[\mathcal{R}, \mathcal{R}^\dagger; \lambda] \equiv \sum_{k,ij} \sum_{ab} \left[\mathcal{R}_i\ \epsilon_{k,ij} \mathcal{R}_j^\dagger \right]_{ab} f_{kia}^\dagger f_{kjb} + \sum_{Ri,ab} [\lambda_i]_{ab}\ f_{Ria}^\dagger f_{Rib}$$

Now *quadratic* functional of ϕ (linear in ϕ and $\phi^\dagger$)!

($\frac{\delta \mathcal{L}}{\delta \phi} = 0$ same complexity of ground-state eigenvalue problem ...)

Functional formulation

$$\mathcal{L}[\boxed{\Phi}, E^c;\ \mathcal{R}, \mathcal{R}^\dagger, \lambda;\ \mathcal{D}, \mathcal{D}^\dagger, \lambda^c;\ \Delta_p] = \frac{\mathcal{T}}{\mathcal{N}} \sum_{k,\omega} \mathrm{Tr}\log\left(\frac{1}{i\omega - \mathcal{R}\epsilon_k\mathcal{R}^\dagger - \lambda}\right) e^{i\omega 0^+} +$$

$$\boxed{\sum_i \left[\langle\Phi_i|\, \mathcal{H}_i^{\mathrm{emb}}[\mathcal{D}_i, \mathcal{D}_i^\dagger; \lambda_i^c]\, |\Phi_i\rangle + E_i^c(1 - \langle\Phi_i|\Phi_i\rangle)\right]}$$

$$-\sum_i \left[\sum_{ab}\left([\lambda_i]_{ab} + [\lambda_i^c]_{ab}\right)[\Delta_{pi}]_{ab} + \sum_{ca\alpha}\left([\mathcal{D}_i]_{a\alpha}\,[\mathcal{R}_i]_{c\alpha}\,[\Delta_{pi}(1-\Delta_{pi})]^{\frac{1}{2}}_{c\alpha} + \mathrm{c.c.}\right)\right]$$

where:

$$\mathcal{H}_i^{\mathrm{emb}}[\mathcal{D}_i, \mathcal{D}_i^\dagger; \lambda_i^c] \equiv \hat{H}_i^{\mathrm{loc}}[\{\hat{c}_{i\alpha}^\dagger\}, \{\hat{c}_{i\alpha}\}] + \sum_{a\alpha}\left([\mathcal{D}_i]_{a\alpha}\,\hat{c}_{i\alpha}^\dagger \hat{f}_{ia} + \mathrm{H.c.}\right) + \sum_{ab}[\lambda_i^c]_{ab}\hat{f}_{ib}\hat{f}_{ia}^\dagger$$

Stationary equations

$$(\mathcal{R},\lambda) \longrightarrow \frac{1}{\mathcal{N}}\left[\sum_k \Pi_i f\left(\mathcal{R}\epsilon_k\mathcal{R}^\dagger+\lambda\right)\Pi_i\right]_{ba} = [\Delta_{pi}]_{ab} \tag{1}$$

$$\frac{1}{\mathcal{N}}\left[\frac{1}{\mathcal{R}_i}\sum_k \Pi_i\,\mathcal{R}\epsilon_k\mathcal{R}^\dagger f\left(\mathcal{R}\epsilon_k\mathcal{R}^\dagger+\lambda\right)\Pi_i\right]_{\alpha a} = \sum_c [\mathcal{D}_i]_{c\alpha}\,[\Delta_{ip}\,(1-\Delta_{ip})]^{\frac{1}{2}}_{ac} \tag{2}$$

$$\left[\sum_{cb\alpha}\frac{\partial}{\partial d^p_{is}}\,[\Delta_{pi}\,(1-\Delta_{pi})]^{\frac{1}{2}}_{cb}\,[\mathcal{D}_i]_{b\alpha}[\mathcal{R}_i]_{c\alpha}+\text{c.c.}\right]+\left[l+l^c\right]_{is}=0 \tag{3}$$

$$\mathcal{H}^{\text{emb}}_i[\mathcal{D}_i,\mathcal{D}_i^\dagger;\lambda^c_i]\,|\Phi_i\rangle = E^c_i\,|\Phi_i\rangle \tag{4}$$

$$\left[\mathcal{F}^{(1)}_i\right]_{\alpha a} \equiv \langle\Phi_i|\,\hat{c}^\dagger_{i\alpha}\hat{f}_{ia}\,|\Phi_i\rangle - \sum_c [\Delta_{ip}\,(1-\Delta_{ip})]^{\frac{1}{2}}_{ca}\,[\mathcal{R}_i]_{c\alpha}=0 \tag{5}$$

$$\left[\mathcal{F}^{(2)}_i\right]_{ab} \equiv \langle\Phi_i|\hat{f}_{ib}\hat{f}^\dagger_{ia}\,|\Phi_i\rangle - [\Delta_{pi}]_{ab}=0 \tag{6}$$

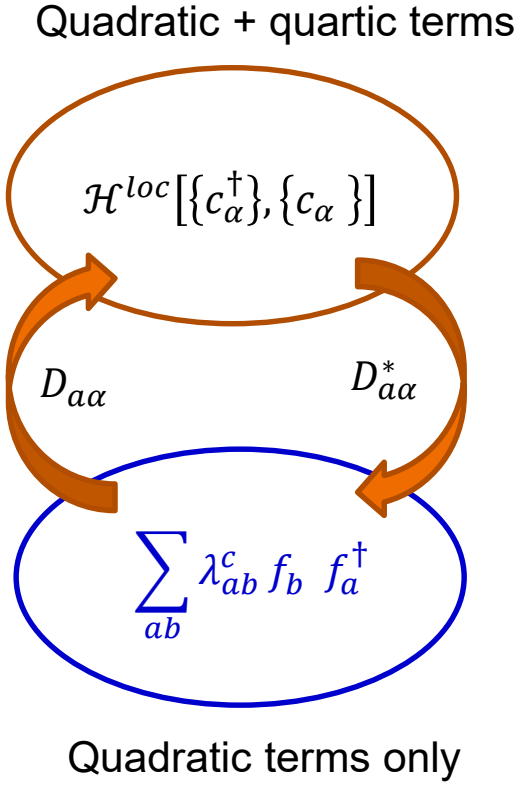
where:

$$\Delta_{pi}=\sum_s d^p_{is}\,{}^t h_{is}\,;\quad \lambda^c_i=\sum_s l^c_{is}\,h_{is}\,;\quad \lambda_i=\sum_s l_{is}\,h_{is}\,;\quad \mathcal{R}_i=\sum_s r_{is}\,h_{is}$$

Embedding Hamiltonian

$$\hat{\mathcal{H}}^{\mathrm{emb}}[\mathcal{D},\mathcal{D}^{\dagger};\lambda^{c}] \equiv \hat{\mathcal{H}}^{\mathrm{loc}}[\{c_{\alpha}^{\dagger}\},\{c_{\alpha}\}] + \sum_{a\alpha}\left(\mathcal{D}_{a\alpha}\, c_{\alpha}^{\dagger} f_{a} + \mathrm{H.c.}\right) + \sum_{ab}\lambda_{ab}^{c}\, f_{b}\, f_{a}^{\dagger}$$

- The number of bath orbitals is exactly equal to the number of impurity orbitals.
- Recursively solve the grounds state of the model at "half-filling" subject to different $\{D_{a\alpha}, \lambda_{ab}^{c}\}$. Only one-particle density matrix is required in the calculations.
- Methods:
    1. Exact diagonalization.
    2. Quantum chemistry methods.
    3. DMRG.



DFT(LDA)+G-RISB energy functional

$$\mathcal{L}\left[N^{loc}, V^{dc}; \rho, \mathcal{J};\ \Phi, E^{c}; R, R^{\dagger}, \lambda; D, D^{\dagger}, \lambda^{c}; \Delta_{p}\right]$$

$$= -\lim_{T\to 0}\frac{T}{N}\sum_{\omega} Tr\log\left(\frac{1}{i\omega + R\left(\hat{\Delta}^{hop} - \hat{J}^{hop}\right)R^{\dagger} - \lambda + \mu}\right)$$

$$+ \sum_{i}\left(\left\langle\Phi_{i}\middle|H_{i}^{emb}[D_{i}, D_{i}^{\dagger}, \lambda^{c}]\middle|\Phi_{i}\right\rangle + E_{i}^{c}\left(1 - \langle\Phi_{i}|\Phi_{i}\rangle\right)\right)$$

$$+ \sum_{i}\left(\sum_{ab}\left([\lambda_{i}]_{ab} + [\lambda_{i}^{c}]_{ab}\right)\left[\Delta_{pi}\right]_{ab} + \sum_{ca\alpha}\left([D_{i}]_{a\alpha}[R_{i}]_{c\alpha}\left[\Delta_{pi}\left(1 - \Delta_{pi}\right)\right]_{ca}^{1/2} + c.c.\right)\right)$$

$$- \int d\boldsymbol{r}\mathcal{J}(\boldsymbol{r})\rho(\boldsymbol{r}) + E_{Hxc}^{LDA}[\rho] + E_{ion}[\rho] + E_{ion-ion}$$

$$+ \sum_{i}\left(V_{i}^{dc}Tr\left(\Delta_{pi}\right) + E_{i}^{dc}\left[N_{i}^{loc}\right] - V_{i}^{dc}N_{i}^{loc}\right)$$

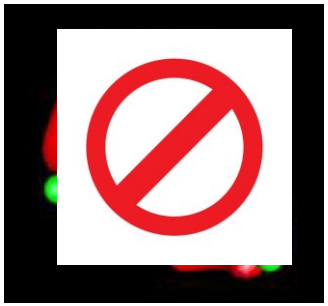
Nicola Lanatà, Yongxin Yao, Xiaoyu Deng, Vladimir Dobrosavljević, and Gabriel Kotliar. Phys. Rev. Lett. **118**, 126401 (2017)
Nicola Lanatà, Yongxin Yao, Cai-Zhuang Wang, Kai-Ming Ho, and Gabriel Kotliar
Phys. Rev. X **5**, 011008 (2015)

Local Projector

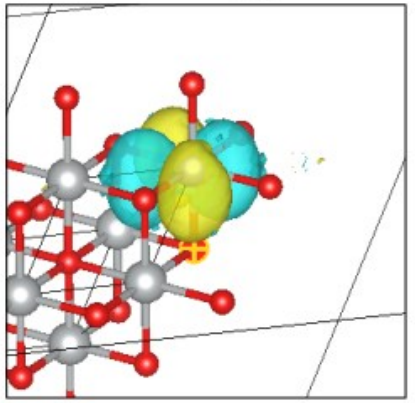
The choice of local projectors can be crucial!

Candidates:

- Maximally Localized Wannier Function
  a) sufficiently large energy window
  b) optimized initial guess (centered at atoms)
- Projected Wannier Function
- Quasi-Atomic Minimal basis set orbitals



$Ni - d_{xy}$



- Nicola Marzari and David Vanderbilt, Phys. Rev. B **56**, 12847 (1997)
- V. I. Anisimov, D. E. Kondakov, A. V. Kozhevnikov, I. A. Nekrasov, Z. V. Pchelkina, J. W. Allen, S.-K. Mo, H.-D. Kim, P. Metcalf, S. Suga, A. Sekiyama, G. Keller, I. Leonov, X. Ren, and D. Vollhardt. Phys. Rev. B 71, 125119 (2005)
- Kristjan Haule, Chuck-Hou Yee, and Kyoo Kim, Phys. Rev. B **81**, 195107 (2010)
- T.-L. Chan, Y. X. Yao, C. Z. Wang, W. C. Lu, J. Li, X. F. Qian, S. Yip, and K. M. Ho, Phys. Rev. B **76**, 205119 (2007)

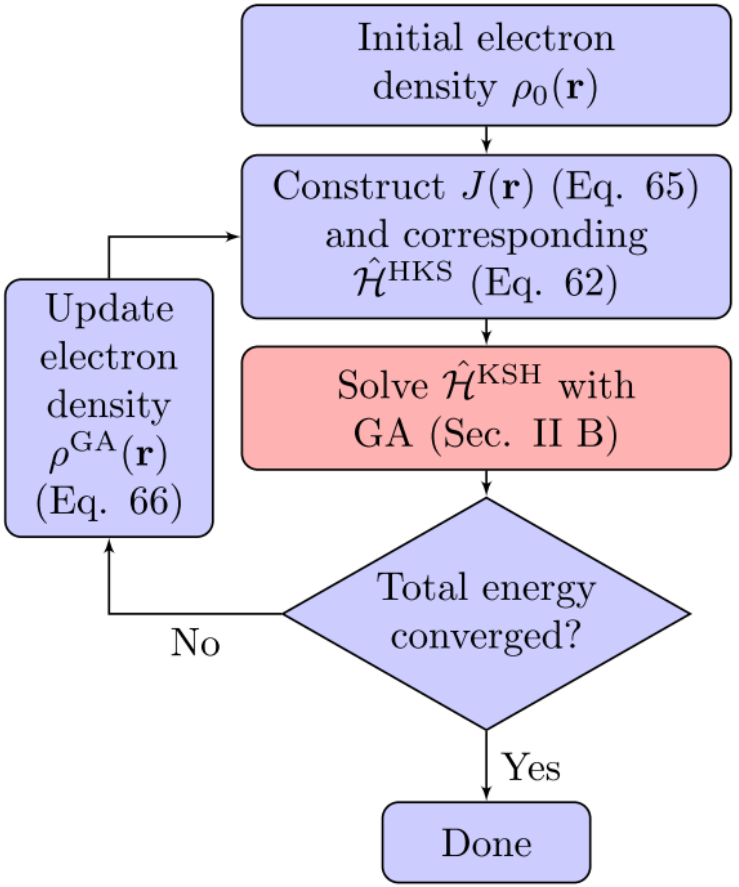
Double counting

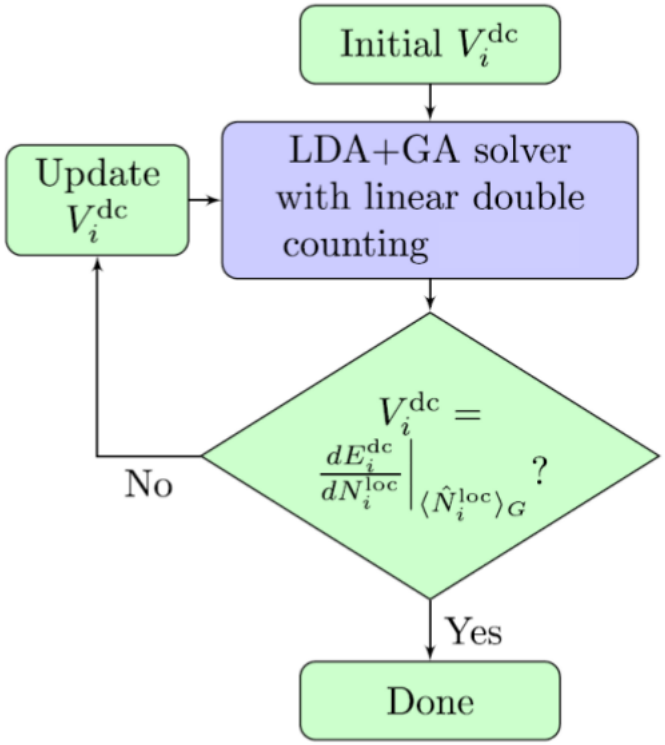
- "Fully Localized Limit" DC $\mu_{dc}^{FLL} = U(N_{\mathrm{imp}} - 1/2) + J(N_{\mathrm{imp}}^{\sigma} - 1/2),$
- "Around Mean Field" DC $\mu_{dc}^{AMF} = \sum_{m'} U_{mm'} n^0 + \sum_{m',m' \neq m} (U_{mm'} - J_{mm'}) n^0$

  $n^0 = \frac{1}{2(2l+1)} \sum_{m,\sigma} n_{m\sigma}$ is the average occupancy
- **Nominal DC** $\mu_{dc}^{FLL} = U(\overline{N_{\mathrm{imp}}} - 1/2) + J(\overline{N_{\mathrm{imp}}^{\sigma}} - 1/2),$
- "Exact" DC

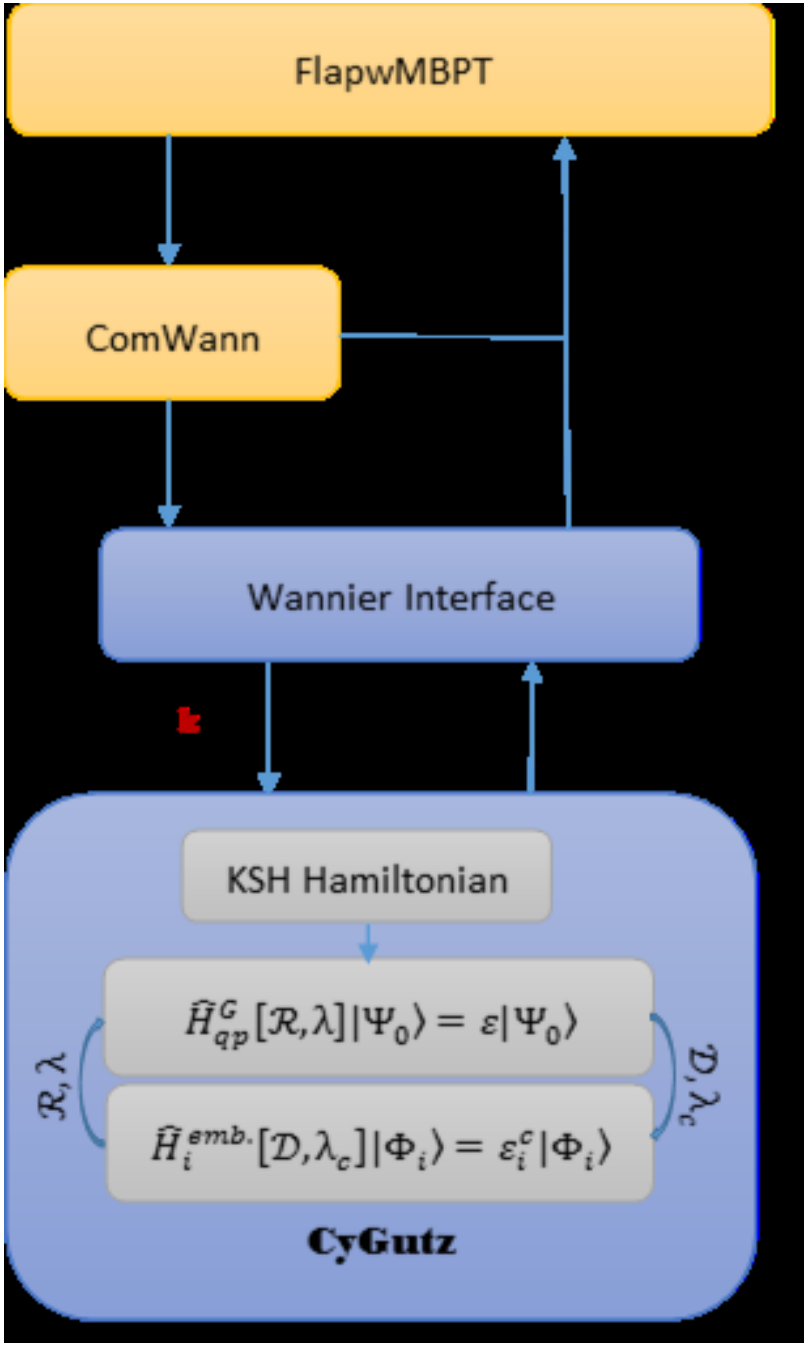
- M. T. Czyżyk and G. A. Sawatzky. Phys. Rev. B **49**, 14211 (1995)
- Kristjan Haule. Phys. Rev. Lett. **115**, 196403 (2015)

Flow chart


Initial electron density $\rho_0(\mathbf{r})$
Construct $J(\mathbf{r})$ (Eq. 65) and corresponding $\hat{\mathcal{H}}^{\mathrm{HKS}}$ (Eq. 62)
Solve $\hat{\mathcal{H}}^{\mathrm{KSH}}$ with GA (Sec. II B)
Total energy converged?
No
Update electron density $\rho^{\mathrm{GA}}(\mathbf{r})$ (Eq. 66)
Yes
Done


Initial V_i^{dc}
LDA+GA solver with linear double counting
$V_i^{\mathrm{dc}} = \frac{dE_i^{\mathrm{dc}}}{dN_i^{\mathrm{loc}}}\Big|_{\langle \hat{N}_i^{\mathrm{loc}} \rangle_G}$?
No
Update V_i^{dc}
Yes
Done

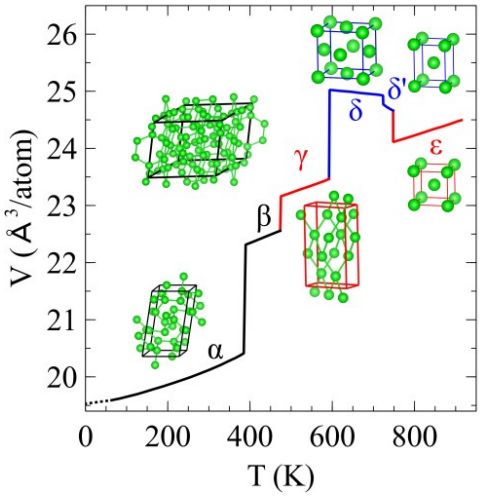
ComRISB structure

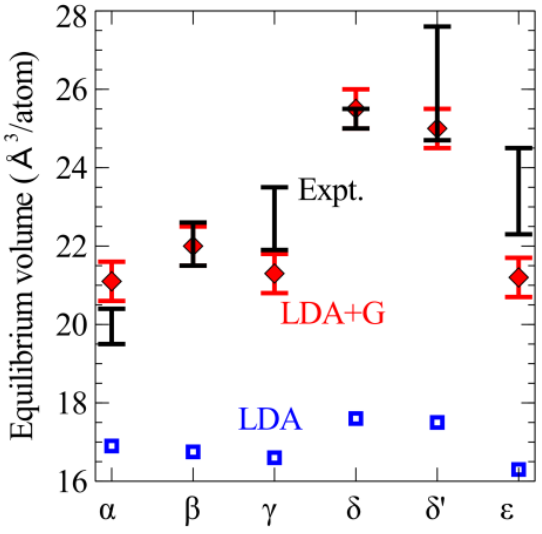


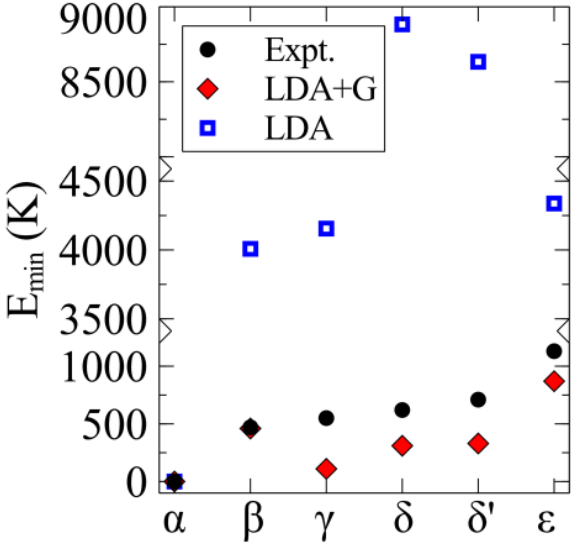
What science can it address?

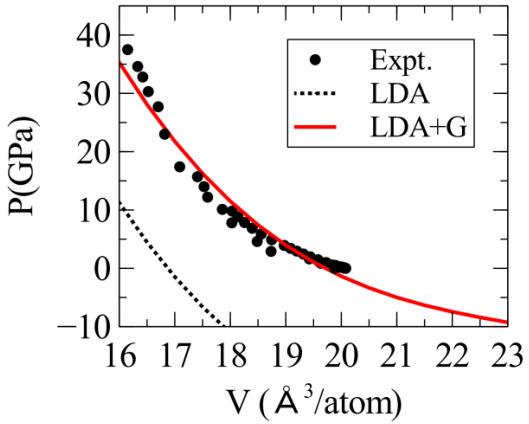
Benchmark: Correlated metal

Plutonium: good description based on correct physics







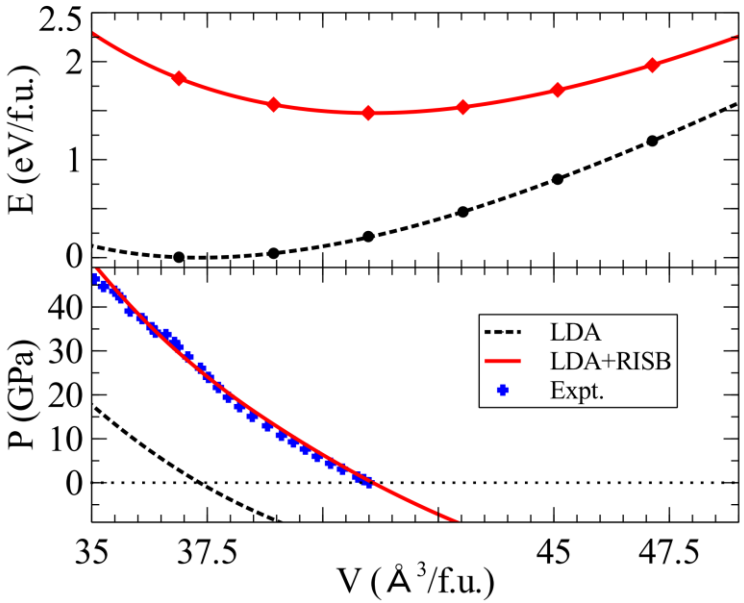


- Good description of all six phases of Pu, including the equilibrium volumes, relative energies etc.
- Correlation effects are crucial to balance the competition between electrostatic Madelung effect and Peierls effect, by renormalizing the quasi-particle bands.
- No artificial symmetry breaking is introduced.

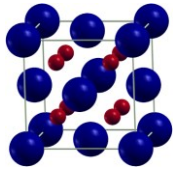
Lanatà, Nicola*, Yongxin Yao*, Cai-Zhuang Wang, Kai-Ming Ho, and Gabriel Kotliar.
Phys. Rev. X **5**, 011008 (2015).

Benchmark:
Mott insulator

UO_2: orbital differentiation and covalent bonding



Mott localized states:



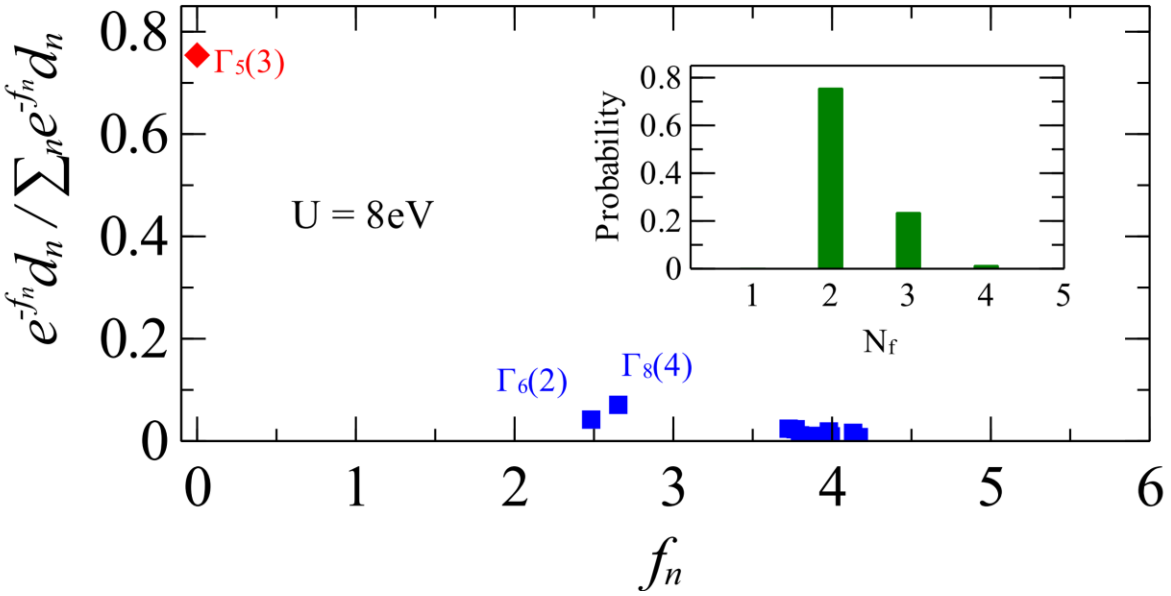
$$|1\rangle \simeq 0.939\,|\Gamma_8^{(1)},5/2,+\rangle + 0.343\,|\Gamma_8^{(2)},7/2,-\rangle$$
$$|2\rangle \simeq 0.939\,|\Gamma_8^{(1)},5/2,-\rangle + 0.343\,|\Gamma_8^{(2)},7/2,+\rangle$$
$$|3\rangle \simeq 0.939\,|\Gamma_8^{(2)},5/2,+\rangle + 0.343\,|\Gamma_8^{(1)},7/2,-\rangle$$
$$|4\rangle \simeq 0.939\,|\Gamma_8^{(2)},5/2,-\rangle + 0.343\,|\Gamma_8^{(1)},7/2,+\rangle$$

- DFT+G reproduces the experimental EOS.
- Mott localized states have considerably mixed J^2 character due to the crystal field effect (not a simple picture of all the J=5/2 orbitals localized).

Nicola Lanata*, Yongxin Yao*, Xiaoyu Deng, Vladimir Dobrosavljevic, and Gabriel Kotliar,
PRL **118**, 126401 (2017)

Multiplet effects

local reduced density matrix $\hat{\rho}_f \equiv e^{-\hat{F}}/\mathrm{Tr}[e^{-\hat{F}}]$

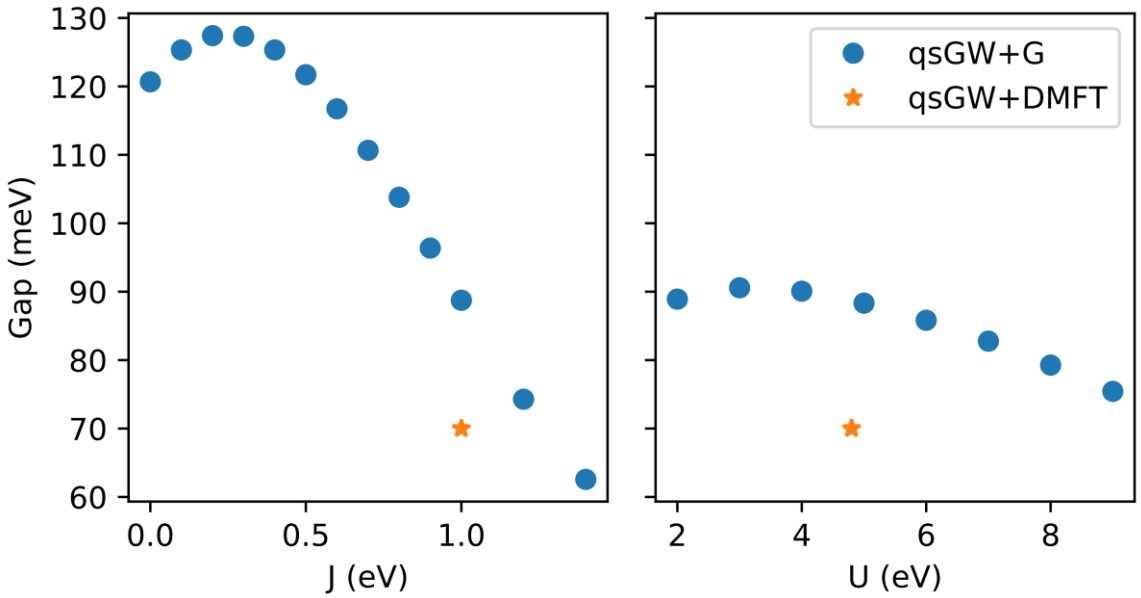


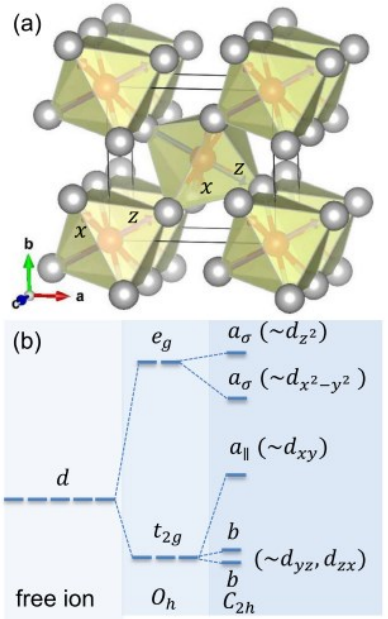
Most probable local configuration $f2\ \Gamma_5$ triplet, consistent with experiment.

Nicola Lanata*, Yongxin Yao*, Xiaoyu Deng, Vladimir Dobrosavljevic, and Gabriel Kotliar, *PRL* **118**, 126401 (2017)

Benchmark:
correlated semiconductor and nonlocal correlation effect

GW+G: $FeSb_2$ is a Hund's semiconductor

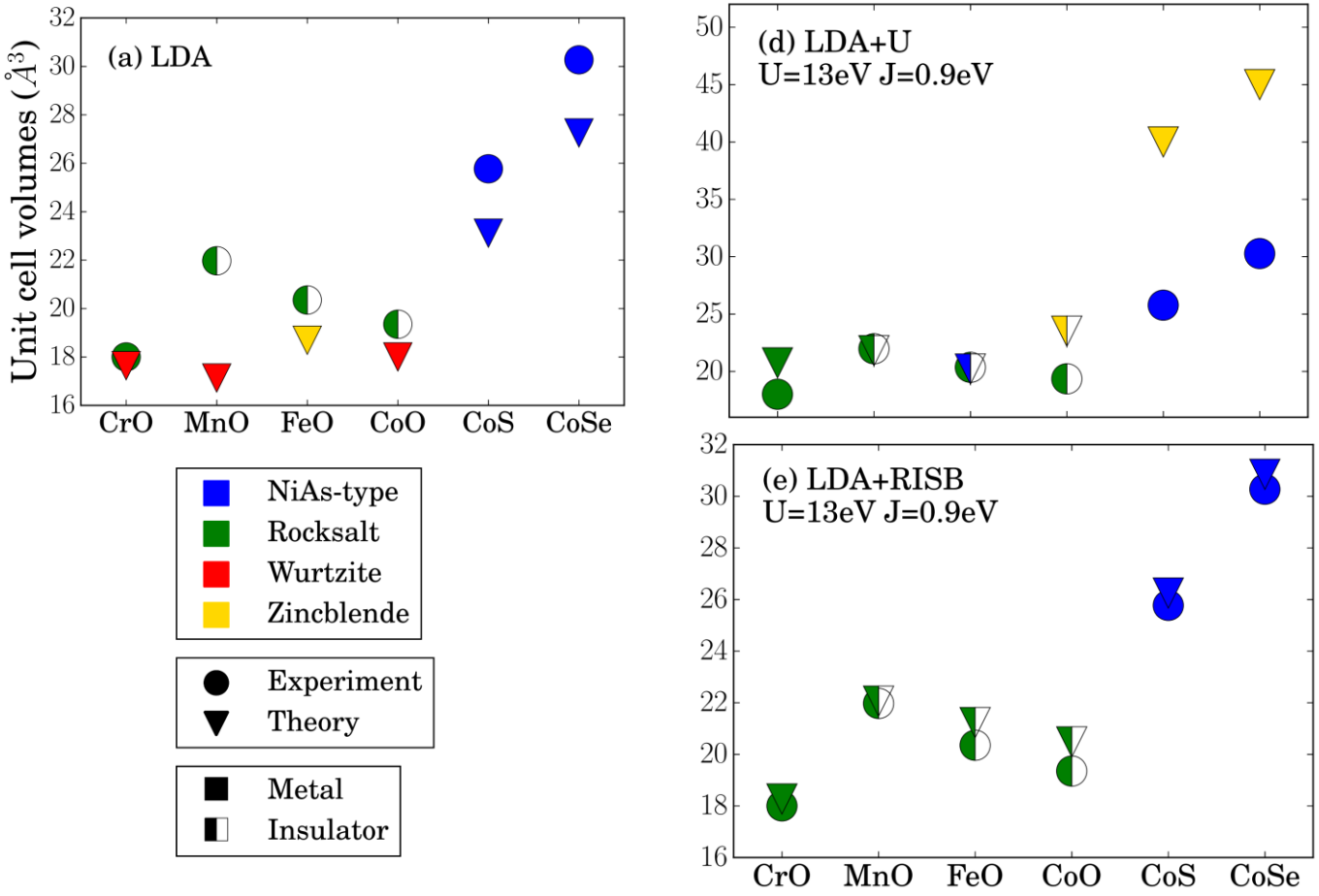




- Band gap size is more sensitive to Hund's coupling J rather than Hubbard U
- qsGW+G produces the band gap size in close agreement with qsGW+DMFT: 88 meV vs. 70 meV, or at elevated $J \approx 1.2$ eV.
- Calculation time ≈ 1 minute/point.

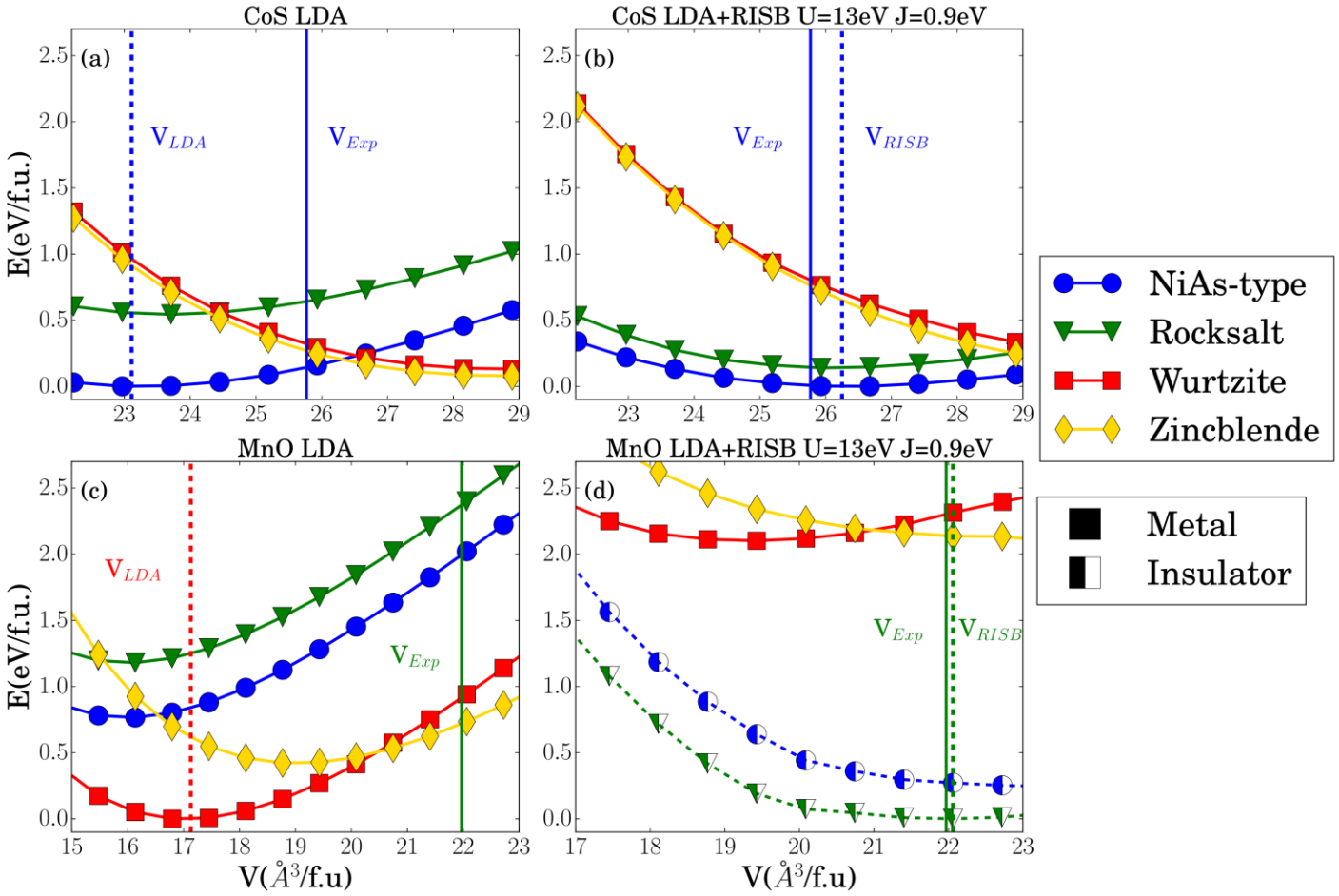
Benchmark:
Correct energetics for a series of transition metal compounds

Critical role of correlation for crystal structures



Lanatà, Nicola, Tsung-Han Lee, Yong-Xin Yao, Vladan Stevanović, and Vladimir Dobrosavljević. "Connection between Mott physics and crystal structure in a series of transition metal binary compounds." *npj Computational Materials (accepted, 2019)*

Correlation induced huge modification in potential energy landscape

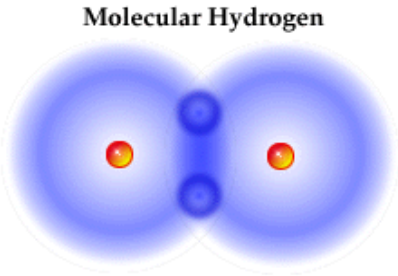


Lanatà, Nicola, Tsung-Han Lee, Yong-Xin Yao, Vladan Stevanović, and Vladimir Dobrosavljević. "Connection between Mott physics and crystal structure in a series of transition metal binary compounds." *npj Computational Materials (accepted, 2019)*

- Some of functionalities presented above are still under development to be user friendly modules in the *Comsuite*.
- We need continuous support from US DOE and you!

Thank You!

Toy H_2 model



$$H_1 = \sum_{\sigma} -t(c^{\dagger}_{1\sigma}c_{2\sigma} + \text{H.c.}) \qquad H_{\mathcal{L}} = \sum_{i=1}^{2}\left(\sum_{\sigma}\varepsilon c^{\dagger}_{i\sigma}c_{i\sigma} + \frac{1}{2}\sum_{\sigma\sigma'}u_0 c^{\dagger}_{i\sigma}c^{\dagger}_{i\sigma'}c_{i\sigma'}c_{i\sigma}\right)$$

Hartree-Fock wavefunction (spin singlet):

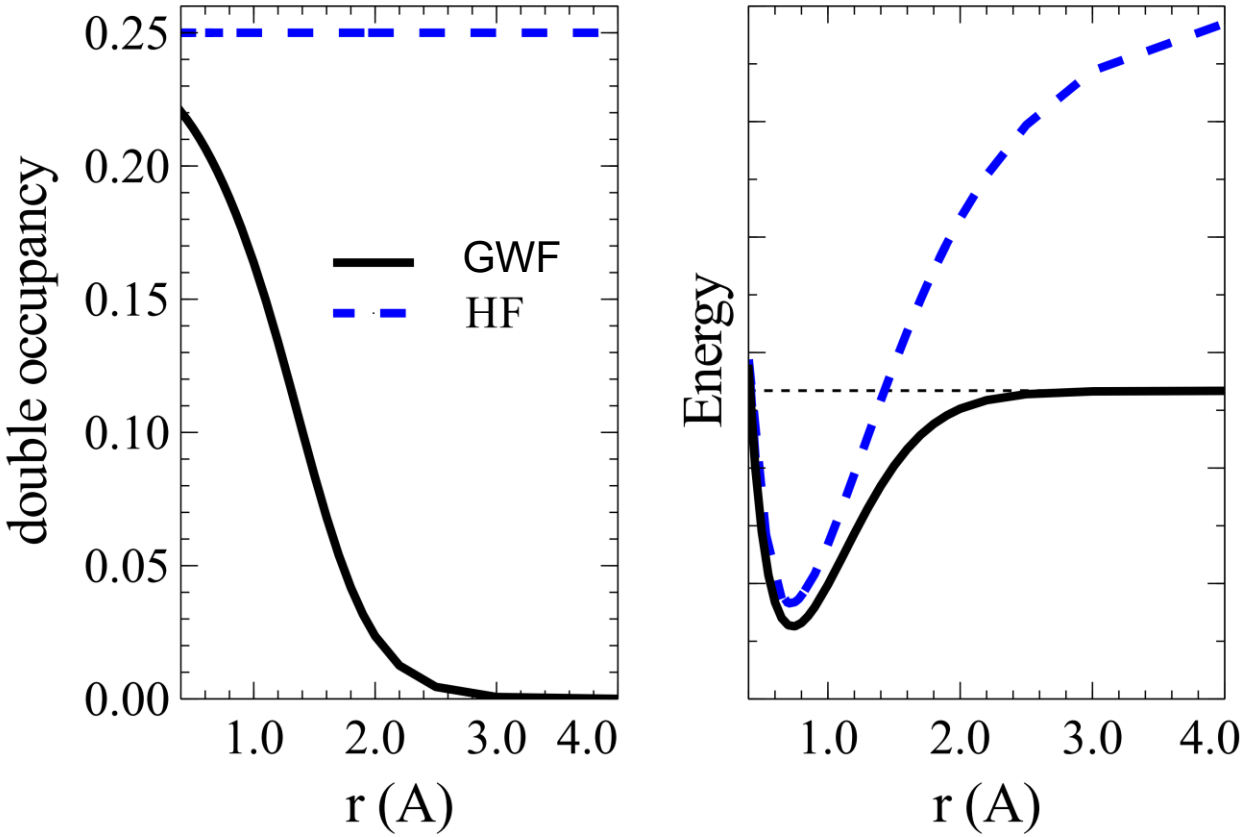
$$|\Psi_0\rangle = \tfrac{1}{2}(c^{\dagger}_{1\uparrow}c^{\dagger}_{1\downarrow}|0\rangle + c^{\dagger}_{2\uparrow}c^{\dagger}_{2\downarrow}|0\rangle + c^{\dagger}_{1\uparrow}c^{\dagger}_{2\downarrow}|0\rangle + c^{\dagger}_{2\uparrow}c^{\dagger}_{1\downarrow}|0\rangle)$$

Gutzwiller wavefunction (with selection rules due to symmetry):

$$|\Psi_{\text{GWF}}\rangle = \sqrt{d}(c^{\dagger}_{1\uparrow}c^{\dagger}_{1\downarrow}|0\rangle + c^{\dagger}_{2\uparrow}c^{\dagger}_{2\downarrow}|0\rangle) + \sqrt{\frac{1}{2} - d}(c^{\dagger}_{1\uparrow}c^{\dagger}_{2\downarrow}|0\rangle + c^{\dagger}_{2\uparrow}c^{\dagger}_{1\downarrow}|0\rangle)$$

Fixed (1/4) vs variationally optimizable double occupancy!

Toy H_2 model

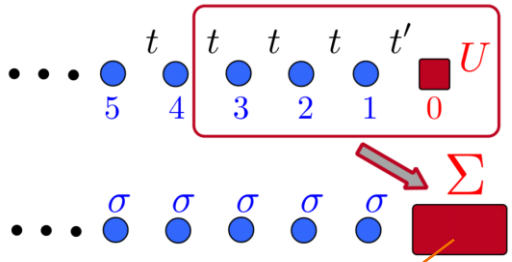

0.25
0.20
0.15
0.10
0.05
0.00
double occupancy
GWF
HF
1.0
2.0
3.0
4.0
r (A)
Energy
1.0
2.0
3.0
4.0
r (A)
$u_0/2$

Improving the accuracy: Gutzwiller renormalization group

$$\hat{H}_{\rm imp} = \frac{U}{2}\left[1 - \sum_{\sigma} c^{\dagger}_{0,\sigma} c_{0,\sigma}\right]^2 - \sum_{\sigma=\pm\frac{1}{2}} t'(c^{\dagger}_{1,\sigma} c_{0,\sigma} + \text{H.c.})$$

$$- \sum_{\sigma=\pm\frac{1}{2}} \sum_{R=1}^{\infty} t(c^{\dagger}_{R,\sigma} c_{R+1,\sigma} + \text{H.c.}). \qquad (1)$$

$$D = 2t, \Gamma = t'^2/t$$



Redefine:

$$\hat{H} = \hat{S}[\{a^{\dagger}_{\Sigma}, a_{\Sigma}\}] + \sum_{bb'} B_{bb'}\, b^{\dagger}_{b} b_{b'}$$

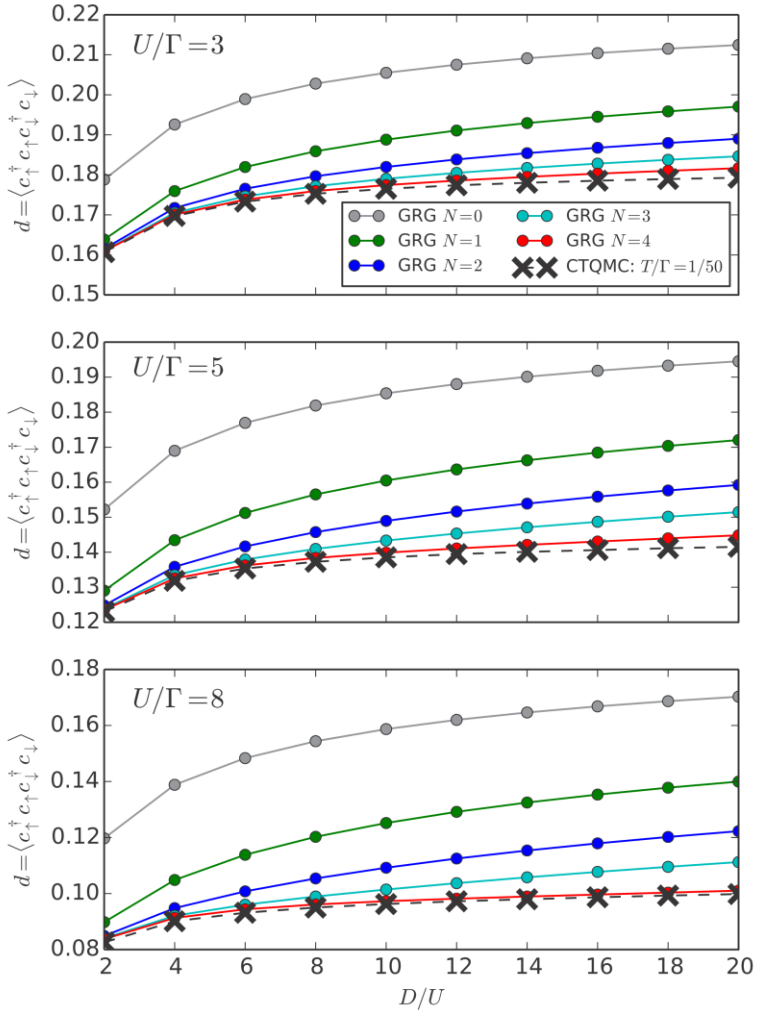
$$+ \sum_{b\Sigma} \left(V^{\dagger}_{b\Sigma}\, b^{\dagger}_{b} a_{\Sigma} + V_{\Sigma b}\, a^{\dagger}_{\Sigma} b_{b} \right) ,$$

$$\hat{S} = \frac{U}{2}\left[1 - \sum_{\sigma} c^{\dagger}_{0,\sigma} c_{0,\sigma}\right]^2 + \sum_{\sigma=\pm 1/2} t' \left(c^{\dagger}_{1,\sigma} c_{0,\sigma} + \text{H.c.} \right)$$

$$+ \sum_{\sigma=\pm 1/2} \sum_{R=1}^{N-1} t \left(c^{\dagger}_{R,\sigma} c_{R+1,\sigma} + \text{H.c.} \right) \equiv \hat{S}_N \, . \qquad (2)$$

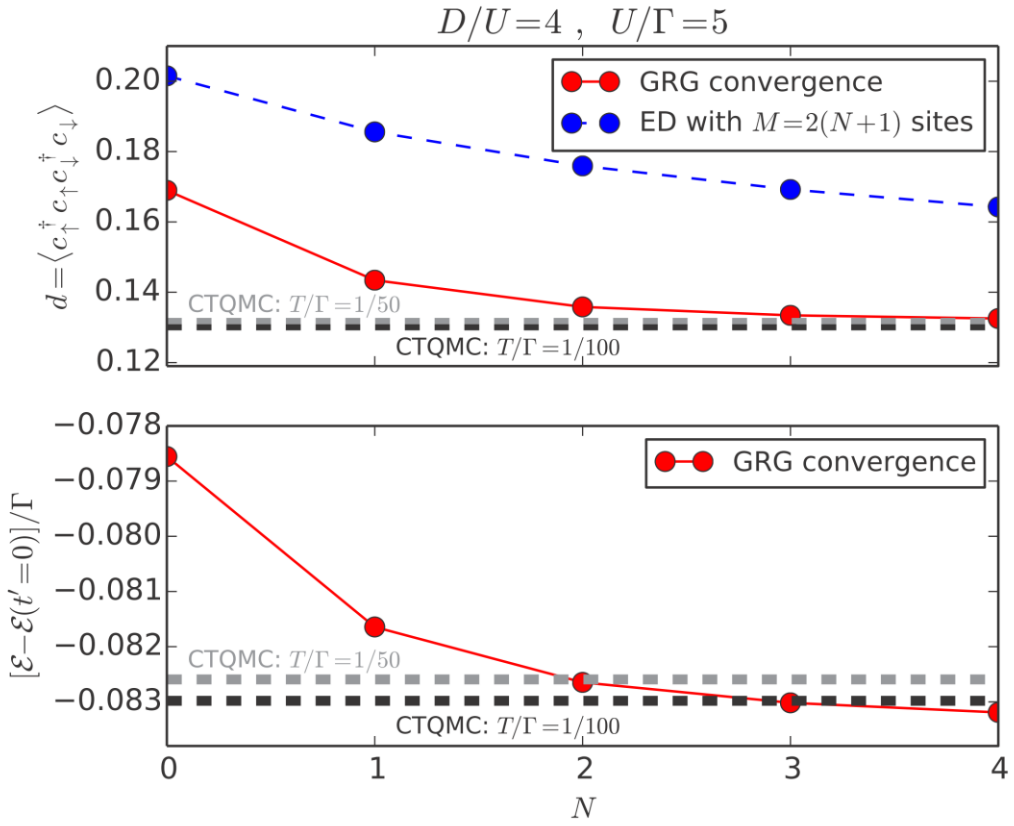
Nicola Lanatà, Yong-Xin Yao, Xiaoyu Deng, Cai-Zhuang Wang, Kai-Ming Ho, and Gabriel Kotliar
Phys. Rev. B **93**, 045103 (2016)

Improving the accuracy: Gutzwiller renormalization group



Nicola Lanatà, Yong-Xin Yao, Xiaoyu Deng, Cai-Zhuang Wang, Kai-Ming Ho, and Gabriel Kotliar
Phys. Rev. B **93**, 045103 (2016)

Improving the accuracy: Gutzwiller renormalization group



Nicola Lanatà, Yong-Xin Yao, Xiaoyu Deng, Cai-Zhuang Wang, Kai-Ming Ho, and Gabriel Kotliar
Phys. Rev. B **93**, 045103 (2016)

Possible improvement

Emergent Bloch Excitations in Mott Matter

Nicola Lanatà,[1] Tsung-Han Lee,[1] Yong-Xin Yao,[2] and Vladimir Dobrosavljević[1]

[1] *Department of Physics and National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32306, USA*

[2] *Ames Laboratory-U.S. DOE and Department of Physics and Astronomy, Iowa State University, Ames, Iowa IA 50011, USA*

(Dated: July 25, 2017)

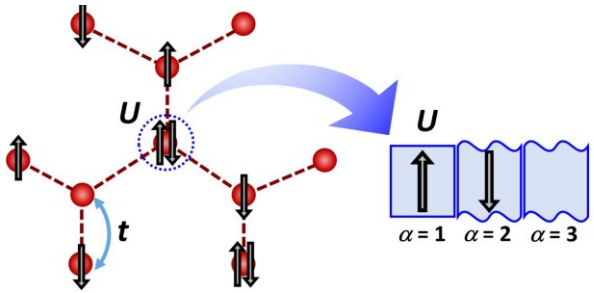


Figure 1: (Color online) Representation of a lattice including 2 ghost orbitals ($\alpha = 2, 3$). The Hamiltonian of the system acts as 0 over the the auxiliary ghost degrees of freedom. In particular, the Hubbard interaction U acts only over the physical orbital $\alpha = 1$.