TRIQS Summer School 2023

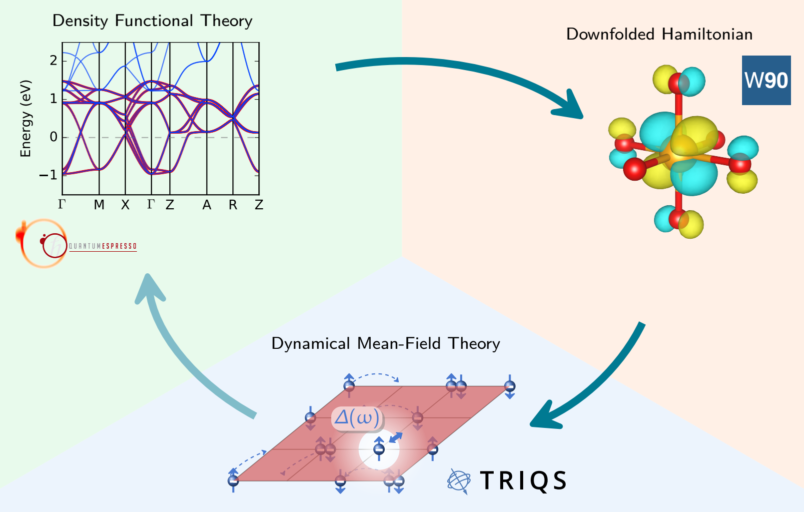
Ab initio description of strongly correlated materials: combining density functional theory plus and dynamical mean-field theory

Sophie Beck

31st August 2023

FLATIRON INSTITUTE
Center for Computational Quantum Physics

Density Functional Theory + Dynamical Mean-Field Theory



Outline

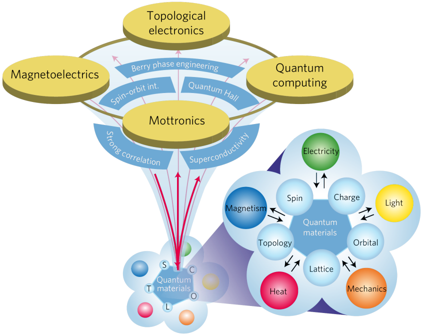
1. Introduction

2. DFT+DMFT
   - DMFT recap
   - Ab initio electronic structure
   - DFT+DMFT ingredients
   - Impurity solvers
   - Charge self-consistency
   - Post-processing

3. Summary

Strongly correlated materials - next generation electronics?

FLATIRON INSTITUTE
Center for Computational Quantum Physics



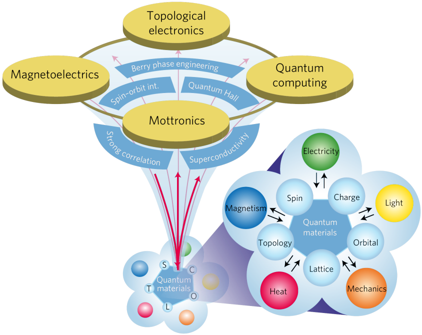
REVIEW ARTICLES **NATURE PHYSICS** DOI: 10.1038/NPHYS4274

Table 1 | Summary of various emergent functions discussed in this article.

Emergent functions	Key concept	Control parameter	Bottleneck/key experiment	Target industry
Mottronics	Electron correlation	Band-filling Bandwidth	E-field switching at RT Above-RT superconductor	Low-energy-cost electronics Energy harvesting/saving
Magnetoelectrics	Spin–orbit interaction	Broken symmetries both in space and time	E-field switching at RT Ultrafast photo-switching	Low-energy-cost electronics Information technology
Topological electronics	Berry phase	Band structure design Spin texture	Zero-field edge current at RT Skyrmionic circuit	Information technology Energy harvesting
Quantum computing	Quantum coherence	Nanomaterials design Topological protection	Qubit/photon interface Quantum simulator	Quantum computer Information security

Y. Tokura, M. Kawasaki, and N. Nagaosa, Nat. Phys. 13, 1056 (2017)

Strongly correlated materials - next generation electronics?



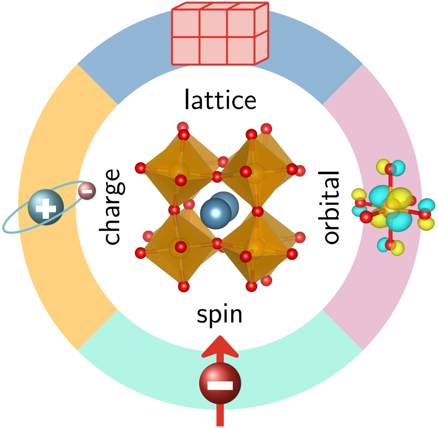
REVIEW ARTICLES NATURE PHYSICS DOI: 10.1038/NPHYS4274

Table 1 | Summary of various emergent functions discussed in this article.

Emergent functions	Key concept	Control parameter	Bottleneck/key experiment	Target industry
Mottronics	Electron correlation	Band-filling Bandwidth	E-field switching at RT Above-RT superconductor	Low-energy-cost electronics Energy harvesting/saving
Magnetoelectrics	Spin–orbit interaction	Broken symmetries both in space and time	E-field switching at RT Ultrafast photo-switching	Low-energy-cost electronics Information technology
Topological electronics	Berry phase	Band structure design Spin texture	Zero-field edge current at RT Skyrmionic circuit	Information technology Energy harvesting
Quantum computing	Quantum coherence	Nanomaterials design Topological protection	Qubit/photon interface Quantum simulator	Quantum computer Information security

Y. Tokura, M. Kawasaki, and N. Nagaosa, Nat. Phys. 13, 1056 (2017)

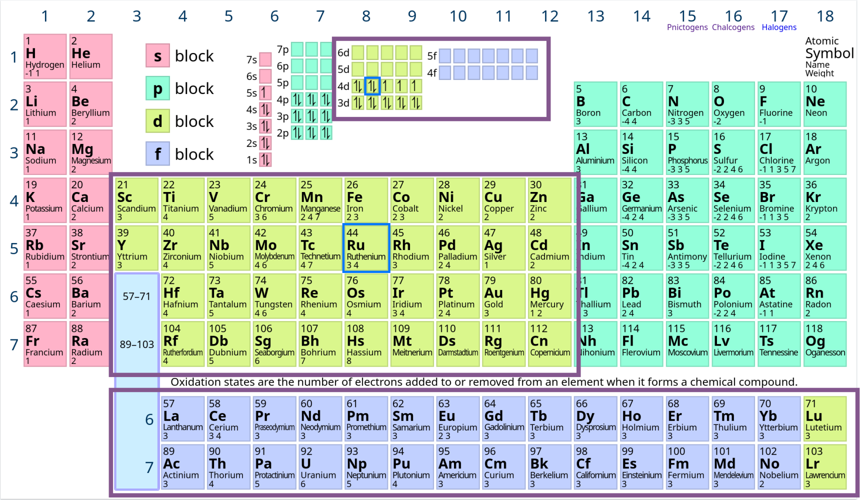
Exotic phenomena



- sensitive to small changes in external parameters:
  - temperature
  - pressure
  - doping
  - ...
- emerging phenomena:
  - high T_C superconductivity
  - colossal magnetoresistance
  - Mott physics
  - ...

Correlated d-/f-shells

FLATIRON INSTITUTE
Center for Computational Quantum Physics



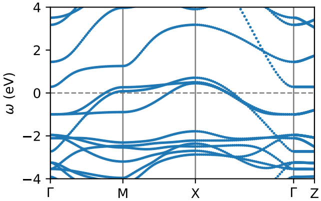
https://ptable.com

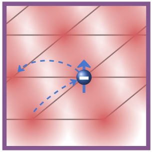
Weak versus strong correlation

effective single-particle picture

- weakly correlated systems
- density functional theory
- Fermi liquid theory

$$\Psi(\mathbf{x}_1, \mathbf{x}_2) = \frac{1}{\sqrt{2}}\{\chi_1(\mathbf{x}_1)\chi_2(\mathbf{x}_2) - \chi_1(\mathbf{x}_2)\chi_2(\mathbf{x}_1)\}$$
$$= \frac{1}{\sqrt{2}}\begin{vmatrix} \chi_1(\mathbf{x}_1) & \chi_2(\mathbf{x}_1) \\ \chi_1(\mathbf{x}_2) & \chi_2(\mathbf{x}_2) \end{vmatrix},$$





Weak versus strong correlation

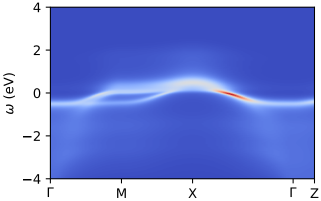
effective single-particle picture

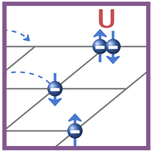
- weakly correlated systems
- density functional theory
- Fermi liquid theory

$$\Psi(\mathbf{x}_1,\mathbf{x}_2) = \frac{1}{\sqrt{2}}\{\chi_1(\mathbf{x}_1)\chi_2(\mathbf{x}_2) - \chi_1(\mathbf{x}_2)\chi_2(\mathbf{x}_1)\} = \frac{1}{\sqrt{2}}\begin{vmatrix}\chi_1(\mathbf{x}_1) & \chi_2(\mathbf{x}_1)\\ \chi_1(\mathbf{x}_2) & \chi_2(\mathbf{x}_2)\end{vmatrix},$$

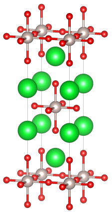
strongly correlated systems

- breakdown of single-particle picture
- strong local Coulomb interaction U
- between ionic localization and itinerant behavior

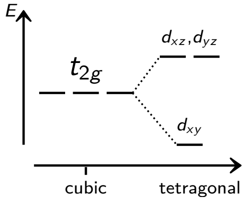


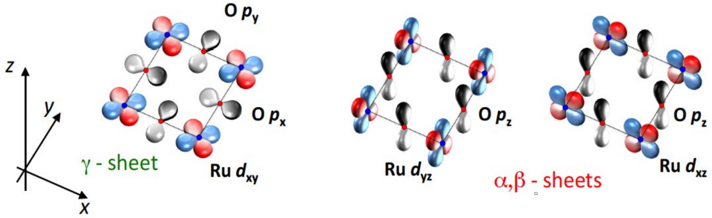


Case study: Fermi surface of Sr_2RuO_4



- strong correlations ($U = 2.3$ eV)
- Hund physics ($J = 0.4$ eV)
- spin-orbit coupling ($\lambda = 0.1 - 0.2$ eV)
- Fermi liquid ($T_{\mathrm{FL}} \approx 25$ K)
- superconductivity ($T_{\mathrm{C}} \approx 1.5$ K)
- Van Hove singularity close to E_{F}

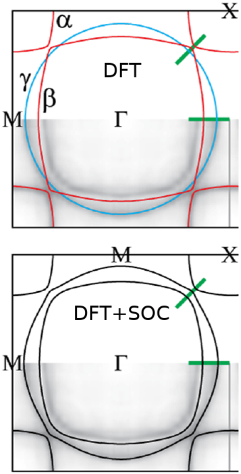




A. Georges, lecture notes (2017)

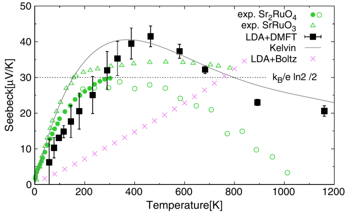
Where DFT may be insufficient

Fermi surface



M. W. Haverkort *et al.*, Phys. Rev. Lett. 101, 026406 (2008)

Seebeck

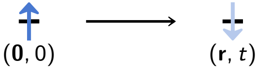


- also: mass enhancement, orbital occupations, optics, SOC, ...
- **more obvious**: local-moment paramagnet (**Mott insulator**) versus (anti-)ferromagnet or non-magnetic metal in DFT

J. Mravlje, A. Georges, Phys. Rev. Lett. 117, 036401 (2016)

Spectral function $A(\mathbf{k}, \omega)$

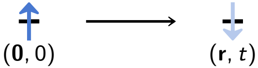
$$A(\mathbf{k}, \omega) = \frac{1}{\pi} \, \mathrm{Im} \int \mathrm{d}\mathbf{r} \int \mathrm{d}t \, e^{\mathrm{i}(\mathbf{k}\mathbf{r} - \omega t)} \underbrace{\mathrm{i}\theta(t) \left\langle \left[\Psi(\mathbf{r}, t), \Psi^\dagger(\mathbf{0}, 0) \right] \right\rangle}_{G^R(\mathbf{r}, t)}$$

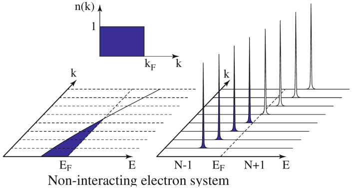


A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. 75, 473 (2003)

Spectral function $A(\mathbf{k},\omega)$ - non-interacting

$$A(\mathbf{k},\omega) = \frac{1}{\pi}\,\mathrm{Im}\int \mathrm{d}\mathbf{r}\int \mathrm{d}t\, e^{\mathrm{i}(\mathbf{kr}-\omega t)}\, \underbrace{\mathrm{i}\theta(t)\left\langle[\Psi(\mathbf{r},t),\Psi^\dagger(\mathbf{0},0)]\right\rangle}_{G^R(\mathbf{r},t)}$$





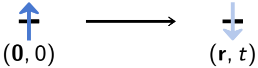
$$G(\mathbf{k},\omega) = \frac{1}{\omega - \epsilon_{\mathbf{k}} + \mathrm{i}\eta}$$

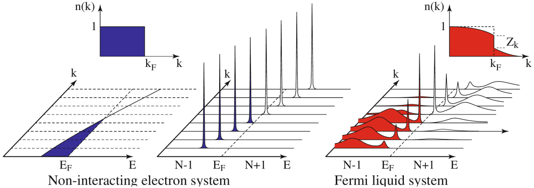
$$A(\mathbf{k},\omega) = -\frac{1}{\pi}\delta(\omega - \epsilon_{\mathbf{k}})$$

A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. 75, 473 (2003)

Spectral function $A(\mathbf{k}, \omega)$ - interacting

$$A(\mathbf{k},\omega) = \frac{1}{\pi}\,\mathrm{Im}\int \mathrm{d}\mathbf{r}\int \mathrm{d}t\, e^{\mathrm{i}(\mathbf{kr}-\omega t)}\, \underbrace{\mathrm{i}\theta(t)\left\langle\left[\Psi(\mathbf{r},t),\Psi^\dagger(\mathbf{0},0)\right]\right\rangle}_{G^R(\mathbf{r},t)}$$





$$G(\mathbf{k},\omega) = \frac{1}{\omega - \epsilon_{\mathbf{k}} - \Sigma(\omega)}$$

$$\Sigma(\omega) = \Sigma'(\omega) + \mathrm{i}\Sigma''(\omega)$$

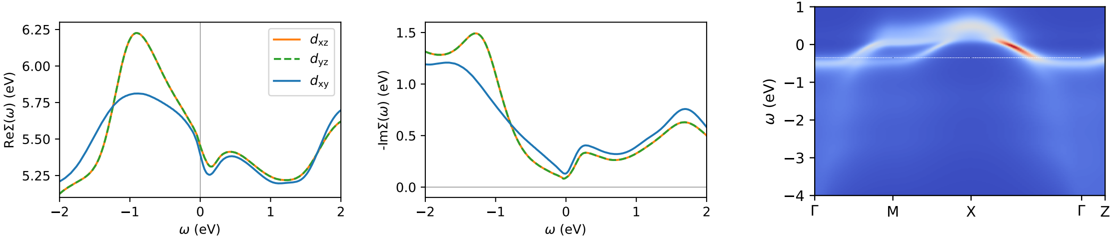
$$A(\mathbf{k},\omega) = -\frac{1}{\pi}\frac{\Sigma''(\omega)}{(\omega-\epsilon_{\mathbf{k}}-\Sigma'(\omega))^2+\Sigma''(\omega)^2}$$

A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. 75, 473 (2003)

Quasiparticles and Fermi liquid theory

$$G(\mathbf{k},\omega) = \frac{1}{\omega - \epsilon_{\mathbf{k}} - \Sigma(\omega)}$$

with $\Sigma(\omega) = \Sigma'(\omega) + \mathrm{i}\Sigma''(\omega)$



Quasiparticles and Fermi liquid theory

$$G(\mathbf{k},\omega) = \frac{Z(\epsilon^*_{\mathbf{k}})}{\omega - \epsilon^*_{\mathbf{k}} - \mathrm{i}\Gamma(\epsilon^*_{\mathbf{k}})} + G_{\text{incoh}}$$

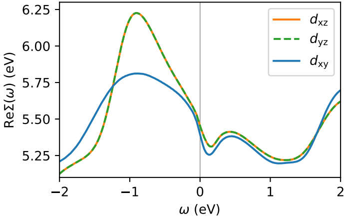
if $\Sigma''(\omega)$ not too large: quasiparticles

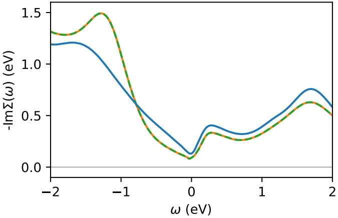
- $\epsilon^*_{\mathbf{k}}$ quasiparticle dispersion
- Z quasiparticle renormalization
- Γ scattering rate/inverse lifetime

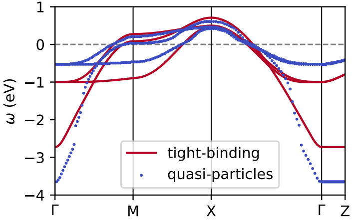
$$\epsilon^*_{\mathbf{k}} = \epsilon_{\mathbf{k}} + \Sigma'(\epsilon^*_{\mathbf{k}})$$

$$Z(\omega) = [1 - \frac{\partial \Sigma'(\omega)}{\partial \omega}]^{-1}$$

$$\Gamma(\omega) = -Z(\omega)\Sigma''(\omega)$$







Quasiparticles and Fermi liquid theory

$$G(\mathbf{k},\omega) = \frac{Z}{\omega - \epsilon^*_{\mathbf{k}} - i\Gamma} + G_{\text{incoh}}$$

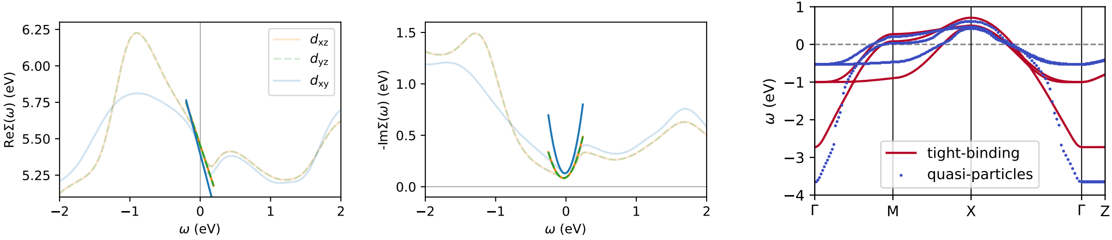
if $\Sigma''(\omega)$ not too large and near $\omega = 0$

- $\epsilon^*_{\mathbf{k}}$ quasiparticle dispersion
- Z quasiparticle renormalization
- Γ scattering rate/inverse lifetime

$$\epsilon^*_{\mathbf{k}} = Z(\epsilon_{\mathbf{k}} + \Sigma'(0))$$

$$Z = [1 - \frac{\partial \Sigma'(\omega)}{\partial \omega}|_{\omega=0}]^{-1} = \frac{m}{m^*}$$

$$\Gamma = -Z\Sigma''(0)$$



Quasiparticles and Fermi liquid theory

$$G(\mathbf{k},\omega) = \frac{Z}{\omega - \epsilon^*_{\mathbf{k}} - \mathrm{i}\Gamma} + G_{\text{incoh}}$$

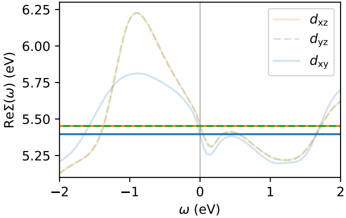
if $\Sigma''(\omega)$ not too large and near $\omega = 0$

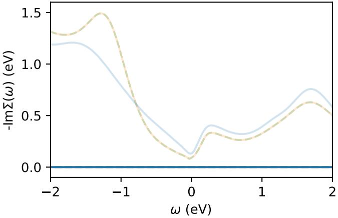
- $\epsilon^*_{\mathbf{k}}$ quasiparticle dispersion
- Z quasiparticle renormalization
- Γ scattering rate/inverse lifetime

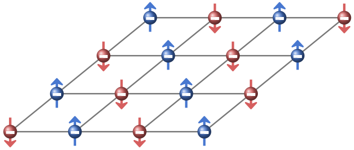
$\epsilon^*_{\mathbf{k}} = \epsilon_{\mathbf{k}} + \Sigma'(0)$

$Z = [1 - \frac{\partial \Sigma'(0)}{\partial \omega}]^{-1} = 1$

$\Gamma \to 0$





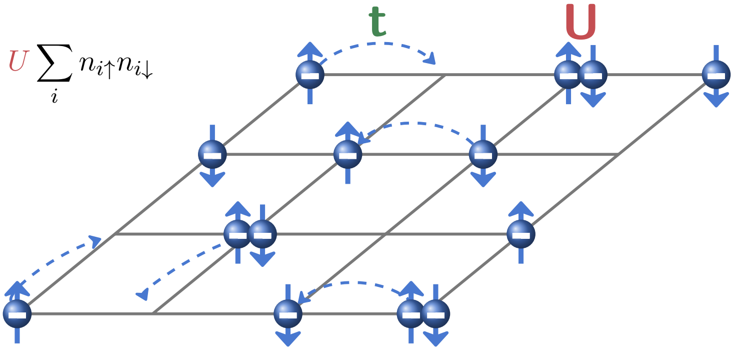


DFT+DMFT

- **situation:** complex physics arising from strong local Coulomb interaction in partially filled orbitals in strongly correlated materials
- **goal:** ab-initio, material-realistic description
- **challenge:** combining localized, atomic-like and itinerant electronic behavior
- **ansatz:** DFT+DMFT, downfolding & embedding
- **ingredients:** hoppings t and Coulomb repulsion U for downfolded model, projector functions P to transform from/to full system
- **example:** Fermi surface of Sr_2RuO_4

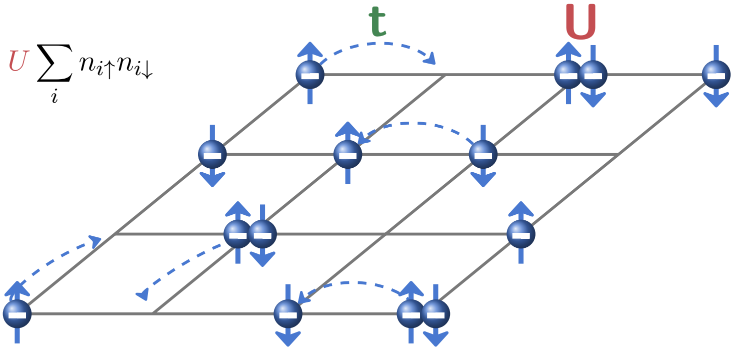
recap: the Hubbard model

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



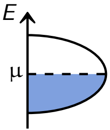
recap: the Hubbard model

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



In the limits:

$t \gg U$:

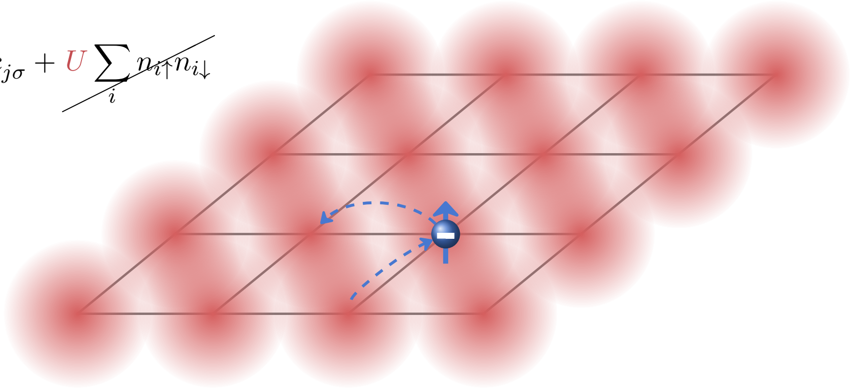


metallic

recap: the Hubbard model

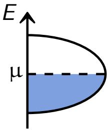
FLATIRON INSTITUTE
Center for Computational Quantum Physics

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



In the limits:

$t \gg U$:

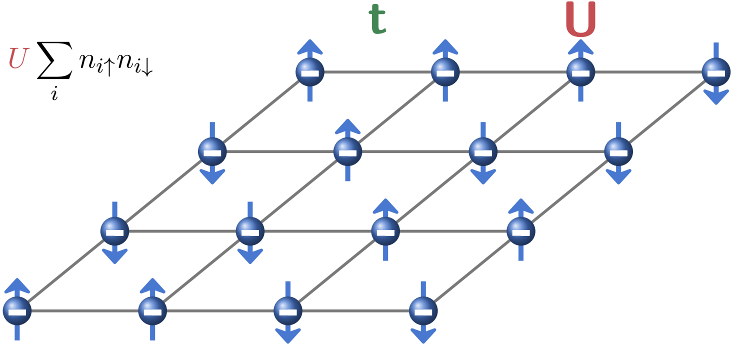


metallic

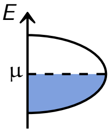
recap: the Hubbard model

FLATIRON INSTITUTE
Center for Computational Quantum Physics

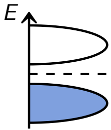
$$H = -\sum_{ij,\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



In the limits:

$t \gg U$: 

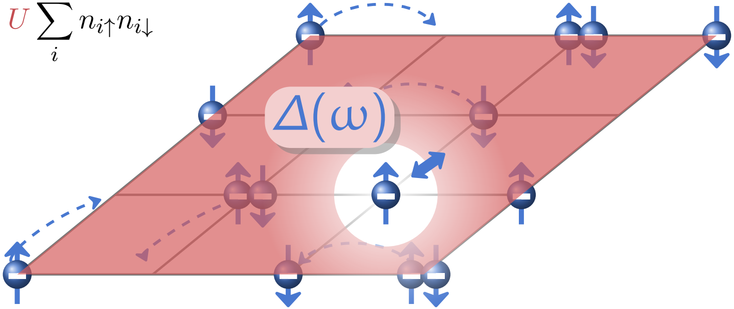
metallic

$t \ll U$: 

insulating

recap: Dynamical Mean Field Theory

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



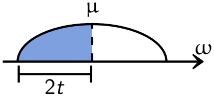
- map lattice to effective impurity model (AIM) embedded in bath
- impurity-bath coupling $\Delta(\omega)$ determined self-consistently
- basic ingredients: t, U

W. Metzner and D. Vollhardt, Phys. Rev. Lett. 62, 3 (1989)
A. Georges and G. Kotliar, Phys. Rev. B 45, 12 (1992)

DMFT self-consistency - example: Bethe lattice

FLATIRON INSTITUTE
Center for Computational Quantum Physics

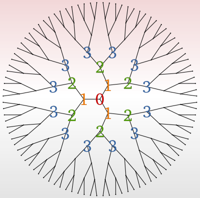
$z \to \infty$: $\quad \rho(\omega) = \frac{1}{2\pi t^2}\sqrt{4t^2 - \omega^2}$



impurity

$$G_{\text{imp}}(\mathrm{i}\omega_n) = \langle c^\dagger(\mathrm{i}\omega_n) c(\mathrm{i}\omega_n)\rangle_{\mathcal{G}_0}$$

$$\mathcal{G}_0^{-1} = [\mathrm{i}\omega_n - t^2 G_{\text{imp}}]^{-1}$$



$$\Sigma = \mathcal{G}_0^{-1} - G_{\text{imp}}^{-1}$$

lattice

$$G(\mathrm{i}\omega_n) = \sum_{\mathbf{k}}[G_0(\mathbf{k}, \mathrm{i}\omega_n)^{-1} - \Sigma(\mathrm{i}\omega_n)]^{-1}$$

G_0

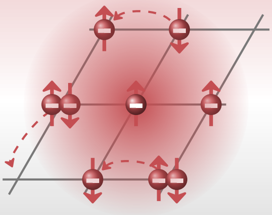
DMFT self-consistency

FLATIRON INSTITUTE
Center for Computational Quantum Physics

impurity

$$G_{\mathrm{imp}}(i\omega_n) = \langle c^\dagger(i\omega_n) c(i\omega_n) \rangle_{\mathcal{G}_0}$$

$$\mathcal{G}_0^{-1} = G^{-1} + \Sigma$$



$$\Sigma = \mathcal{G}_0^{-1} - G_{\mathrm{imp}}^{-1}$$

$$G(i\omega_n) = \sum_{\mathbf{k}} [G_0(\mathbf{k}, i\omega_n)^{-1} - \Sigma(i\omega_n)]^{-1}$$

G_0

lattice

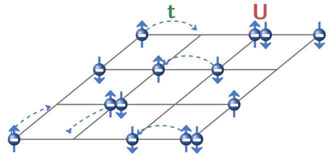
- basic ingredients: t, U, and P

From many-body to effective one-body problem

FLATIRON INSTITUTE
Center for Computational Quantum Physics

electronic Schrödinger equation:

$$\hat{H}\Psi(\mathbf{r}_1, \cdots, \mathbf{r}_N) = \epsilon\Psi(\mathbf{r}_1, ..., \mathbf{r}_N)$$



with

$$\hat{H} = -\sum_i \frac{\hbar^2 \nabla_i^2}{2m} + \sum_{i<j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} + \sum_i^N v_{\text{ext}}(\mathbf{r}_i) = T + U + V_{\text{ext}}$$

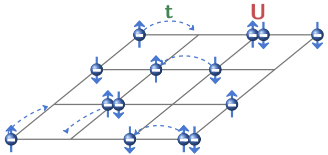
in second quantization:

$$\hat{H} = \sum_{ij} t_{ij} c_i^\dagger c_j + \sum_{ijkl} U_{ijkl} c_i^\dagger c_j^\dagger c_l c_k$$

From many-body to effective one-body problem

electronic Schrödinger equation:

$$\hat{H}\Psi(\mathbf{r}_1, \cdots, \mathbf{r}_N) = \epsilon\Psi(\mathbf{r}_1, ..., \mathbf{r}_N)$$



with

$$\hat{H} = -\sum_i \frac{\hbar^2\nabla_i^2}{2m} + \sum_{i<j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} + \sum_i^N v_{\text{ext}}(\mathbf{r}_i) = T + U + V_{\text{ext}}$$

in second quantization:

$$\hat{H} = \sum_{ij} t_{ij} c_i^\dagger c_j + \sum_{ijkl} U_{ijkl} c_i^\dagger c_j^\dagger c_l c_k \rightarrow \hat{H}_{\text{DFT}} = \sum_{ij} \tilde{t}_{ij} c_i^\dagger c_j$$

Density Functional Theory

1. Hohenberg-Kohn theorem: the external potential (and total energy) is a unique functional of the electron density: $\Psi(\mathbf{r}_1, ..., \mathbf{r}_N) \rightarrow \rho(\mathbf{r})$

$$\rho(\mathbf{r}) = N \int \mathrm{d}^3\mathbf{r}_2 \cdots \int \mathrm{d}^3\mathbf{r}_N \, |\Psi(\mathbf{r}, \mathbf{r}_2, \cdots, \mathbf{r}_N)|^2$$

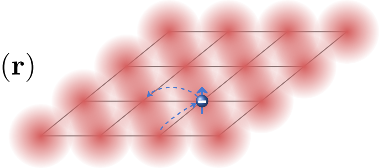
2. Hohenberg-Kohn theorem: the ground-state charge density ρ_0 minimises the energy functional, i.e. yielding the ground-state energy E_0

$$E[\rho_0] \leq E[\rho] = \min_{\Psi \rightarrow \rho_0} \langle \Psi | T + U + V_{\text{ext}} | \Psi \rangle$$

Effective single-particle picture

Recast full system into a ficticious, auxiliary system of separable Kohn-Sham orbitals $\{\psi_n\}$, that generates the same density as the original one

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + v_{\text{eff}}(\mathbf{r})\right]\psi_n(\mathbf{r}) = \epsilon_n \psi_n(\mathbf{r})$$



$$v_{\text{eff}}(\mathbf{r}) = v_{\text{H}}[\rho](\mathbf{r}) + \frac{\delta E_{\text{XC}}[\rho]}{\delta\rho(\mathbf{r})} + v_{\text{ext}}(\mathbf{r})$$

- solution is found self-consistently
- exchange-correlation potential is the only unknown
- Kohn-Sham orbital energies have little physical meaning

$$\rightarrow \hat{H}_{\text{KS}} = \sum_{ij} \tilde{t}_{ij} c_i^\dagger c_j$$

DFT+DMFT ingredients: target bands t

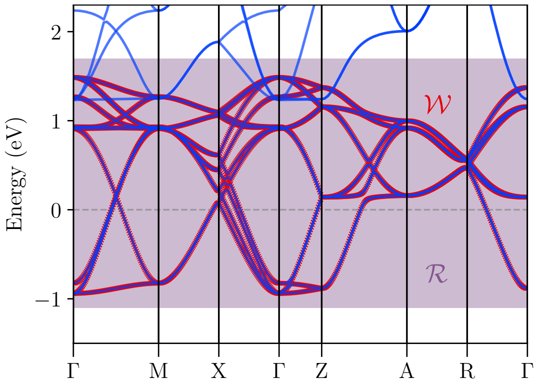
- partitioning of the system
- maximally localized Wannier functions $|\mathbf{R}j\rangle$ from Kohn-Sham states $|\psi_{n\mathbf{k}}\rangle$:

$$\left|\psi_{j\mathbf{k}}^{\mathrm{W}}\right\rangle = \sum_n U_{\mathbf{k},nj} \left|\psi_{n\mathbf{k}}\right\rangle$$

$$|\mathbf{R}j\rangle = \frac{V}{(2\pi)^3} \int_{\mathrm{BZ}} d\mathbf{k}\, \mathrm{e}^{-\mathrm{i}\mathbf{k}\mathbf{R}} \left|\psi_{j\mathbf{k}}^{\mathrm{W}}\right\rangle$$

hopping elements:

$$t_{ij}(\mathbf{R}) = \left\langle 0i \right| \hat{H}^{\mathrm{KS}} \left| \mathbf{R}j \right\rangle$$



N. Marzari, and D. Vanderbilt, Phys. Rev. B 56, 20 (1997)

DFT+DMFT ingredients: projector functions P

lattice Green's function:

$$\hat{G}(\mathbf{k}, i\omega_n) = \sum_{mn} \left[i\omega_n + \mu - \hat{\epsilon}(\mathbf{k}) - \Delta\hat{\Sigma}(\mathbf{k}, i\omega_n) \right]^{-1}_{mn} |\psi_{m\mathbf{k}}\rangle \langle \psi_{n\mathbf{k}}|$$

downfolding:

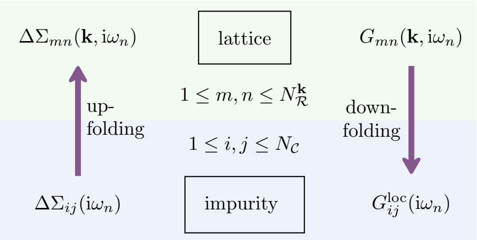
$$G^{\mathrm{loc}}_{ij,\mathcal{R}}(i\omega_n) = \sum_{\mathbf{k},mn} P^{\mathcal{R}}_{im}(\mathbf{k}) G_{mn}(\mathbf{k}, i\omega_n) P^{\mathcal{R}*}_{nj}(\mathbf{k})$$

with projector onto orbital j at atomic site $\mathcal{R}$:

$$P^{\mathcal{R}}_{jn}(\mathbf{k}) = \left\langle \psi^{\mathrm{W}}_{\mathcal{R}_j \mathbf{k}} \middle| \psi_{n\mathbf{k}} \right\rangle$$

upfolding:

$$\Delta\Sigma_{mn}(\mathbf{k}, i\omega_n) = \sum_{\mathcal{R},ij} P^{\mathcal{R}*}_{mi}(\mathbf{k}) \Delta\Sigma^{\mathcal{R}}_{ij}(i\omega_n) P^{\mathcal{R}}_{jn}(\mathbf{k})$$



- basis transformation
- entanglement
- local symmetries

Double counting

- E_U is a functional of the orbital occupations, but E_{XC} is a non-linear functional of the total electron density
- ill-posed problem due to the formally incompatible footing: diagrammatic vs. non-perturbative
- different analytic, *phenomenological* expressions have been proposed: FLL, AMF, ANI, Kunes, nominal...
- remedy: GW+DMFT

$$\Delta\Sigma^{\mathcal{R}}_{ij}(\mathrm{i}\omega_n) = \Sigma^{\mathcal{R}}_{ij}(\mathrm{i}\omega_n) - \Sigma_{\mathrm{DC}}$$

$$E_{\mathrm{DFT}+U}[\rho] = E_{\mathrm{DFT}}[\rho] + E_U[n^{\sigma}_{ij}] - E_{\mathrm{DC}}$$

$$E_{\mathrm{XC}} \approx E^{\mathrm{LDA}}_{\mathrm{XC}}[\rho] = \int \mathrm{d}\mathbf{r}\, \epsilon^{\mathrm{hom}}_{\mathrm{XC}}[\rho(\mathbf{r})]\rho(\mathbf{r})$$

$$E_{\mathrm{XC}}[n^{\sigma}_{ij}]\,?$$

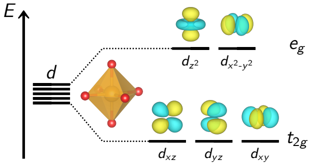
DFT+DMFT ingredients: interaction Hamiltonian U

FLATIRON INSTITUTE
Center for Computational Quantum Physics

$$\hat{H}_{\text{int}} = \frac{1}{2} \sum_{ijkl}^{\text{at } \mathcal{R}} U_{ijkl} c_i^\dagger c_j^\dagger c_l c_k$$

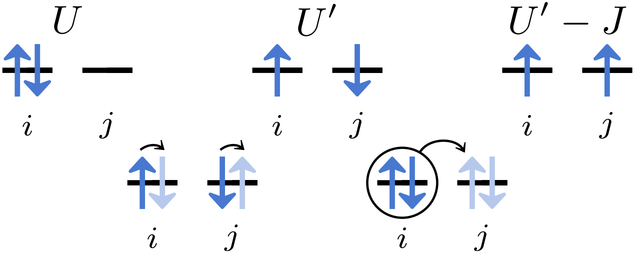
$$V_{ijkl} = \int \mathrm{d}^3\mathbf{r}\, \mathrm{d}^3\mathbf{r}' w_i^*(\mathbf{r}) w_j^*(\mathbf{r}') \frac{e^2}{|\mathbf{r}-\mathbf{r}'|} w_l(\mathbf{r}') w_k(\mathbf{r})$$

- complicated 4-rank tensor
- use symmetries to reduce complexity
- for cubic systems: Hubbard-Kanamori parametrization
- for spherical systems: Slater parametrization



J. Kanamori, Prog. Theor. Exp. Phys. 30 (1963)

DFT+DMFT ingredients: interaction Hamiltonian U

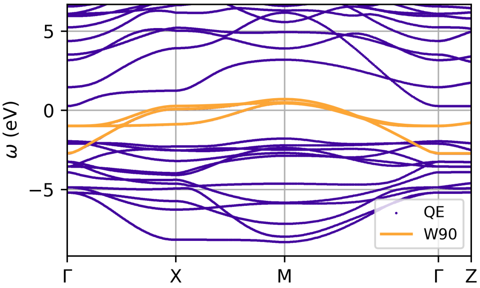


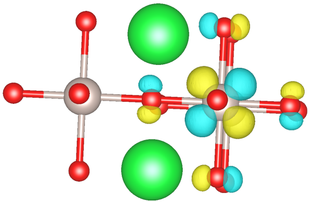
$$\hat{H}_U = U \sum_i n_{i\uparrow} n_{i\downarrow} + U' \sum_{i\neq j} n_{i\uparrow} n_{j\downarrow} + (U' - J) \sum_{i<j,\sigma} n_{i\sigma} n_{j\sigma}$$
$$- J \sum_{i\neq j} c^\dagger_{i\uparrow} c_{i\downarrow} c^\dagger_{j\downarrow} c_{j\uparrow} + J \sum_{i\neq j} c^\dagger_{i\uparrow} c^\dagger_{i\downarrow} c_{j\downarrow} c_{j\uparrow}$$

J. Kanamori, Prog. Theor. Exp. Phys. 30 (1963)

DFT+DMFT ingredients: energy window

t_{2g} model





pros:

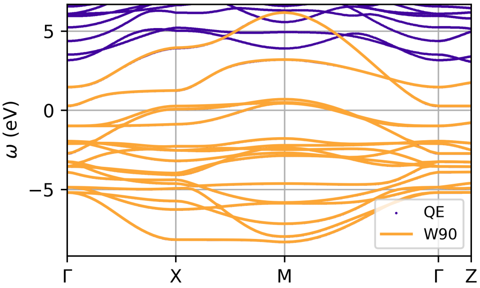
- no DC
- nominal occupations
- less work for impurity solver

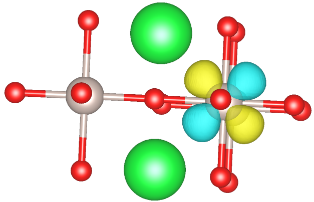
cons:

- smaller U, more frequency-dependent
- larger spread Ω, oxygen tails $\rightarrow$ less localized
- no information on e_g states...

DFT+DMFT ingredients: energy window

dp model





pros:

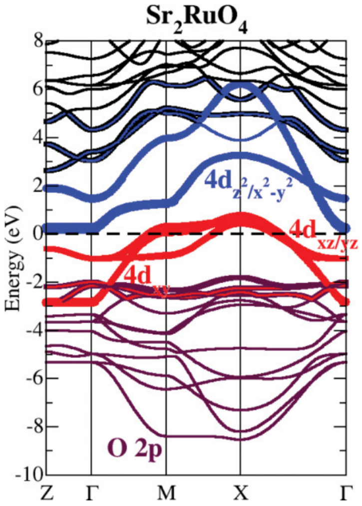
- more localized, DMFT more valid
- larger U and more atomic-like, less frequency-dependent
- renormalizes all states

cons:

- DC, in principle U_{dp}, U_p
- fractional occupations
- heavy for impurity solver

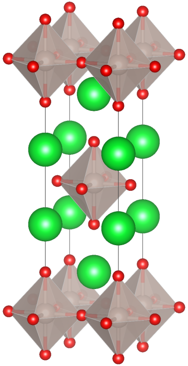
How to determine Coulomb interaction

- V of the order of 11 eV for t_{2g}, i.e. $\gg$ bandwidth ≈ 3.4 eV
- effective Coulomb interaction screened by surrounding electrons
- screened interaction $U(\mathbf{r}, \mathbf{r}')$ in practice:
  - cRPA: screening channels, frequency dependence, Hund J
  - cLDA: only full d shell, static, no Hund J
- $d - dp$: $F^0 = 3.23$ eV, $\bar{U}_{mm} = 4.1$ eV, $t_{2g} - t_{2g}$: $\mathcal{U} = 2.56$ eV



L. Vaugier, H. Jiang, S. Biermann, Phys. Rev. B 86, 165105 (2012)

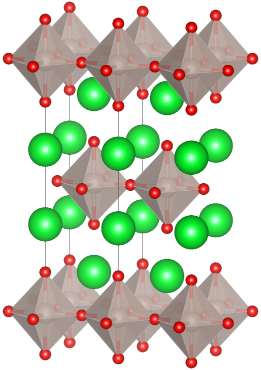
Multi-site DMFT



$$\Sigma = \left(\boxed{\Sigma_{\mathrm{imp}}} \right)$$

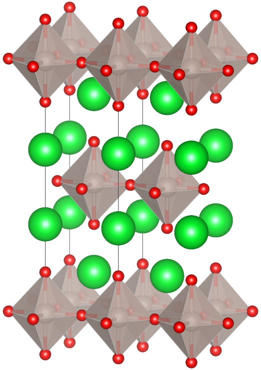
Multi-site DMFT

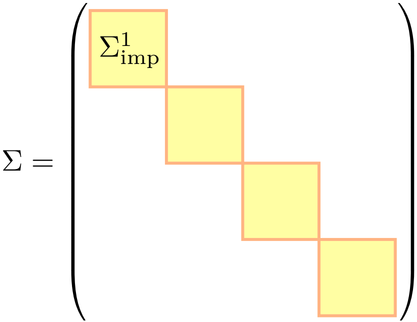
FLATIRON INSTITUTE
Center for Computational Quantum Physics



$$\Sigma = \begin{pmatrix} \Sigma^1_{\text{imp}} & & & \\ & \Sigma^2_{\text{imp}} & & \\ & & \Sigma^3_{\text{imp}} & \\ & & & \Sigma^4_{\text{imp}} \end{pmatrix}$$

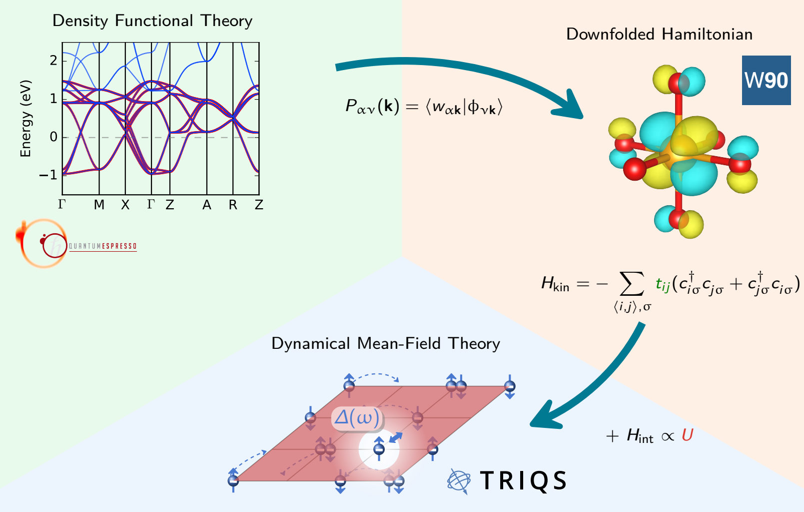
- self-energy approximated as block-diagonal in orbital basis





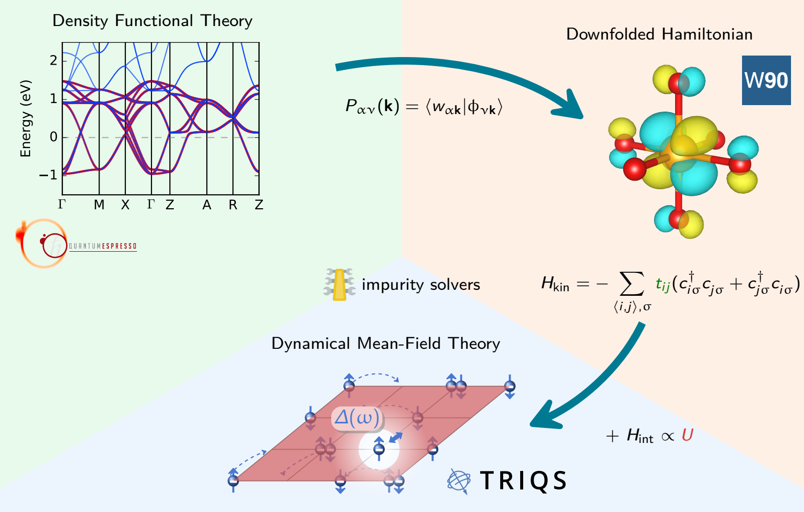
- self-energy approximated as block-diagonal in orbital basis
- map self-energy to symmetry-equivalent sites
- use spin channel for AFM solutions

DFT+DMFT workflow

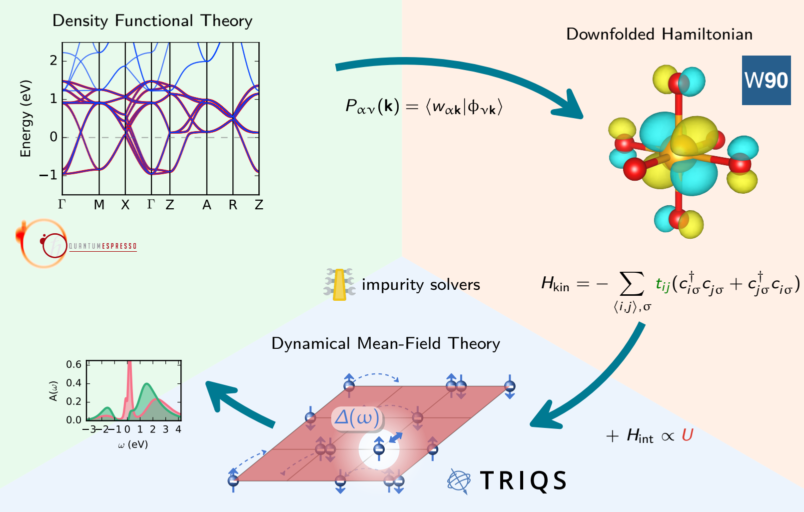


DFT+DMFT workflow

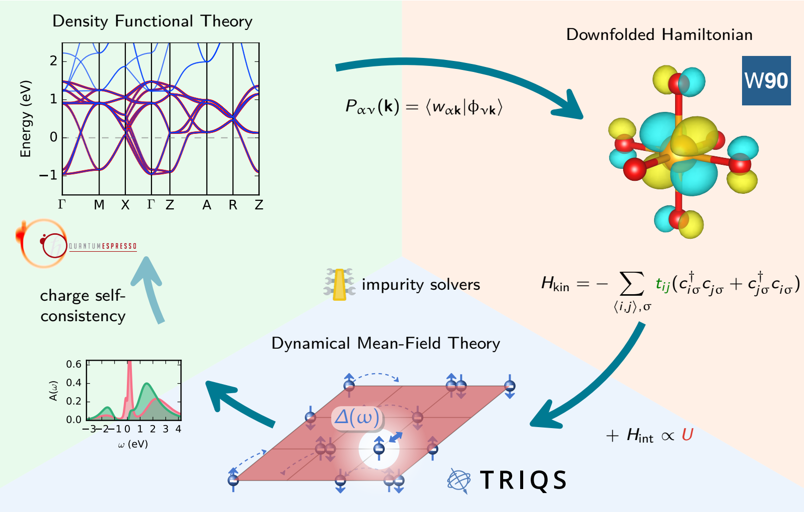
FLATIRON INSTITUTE
Center for Computational Quantum Physics



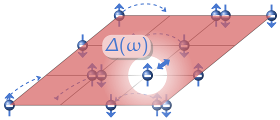
DFT+DMFT workflow



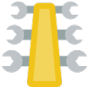
DFT+DMFT workflow



Impurity solvers



$$G^{\text{imp}}_{\sigma}(\tau) = \left\langle T c_{\sigma}(\tau) c^{\dagger}_{\sigma}(0) \right\rangle_{\mathcal{G}_0}$$



approximate solvers:

- Hartree(-Fock)
- Hubbard-I
- Iterated perturbation theory (IPT)
- Slave boson technique
- ...

numerically exact solvers:

- Quantum Monte Carlo (QMC)
- exact diagonalization (ED)
- numerical renormalization group (NRG)
- density matrix renormalization group (DMRG)
- tensor-network based approaches (MPS/TTN)

---

overview: see lecture by O. Parcollet, Arnold Sommerfeld School (2017)

Charge self-consistency: spectral density-functional approach

FLATIRON INSTITUTE
Center for Computational Quantum Physics

Method	Physical quantity	Constraining field
Baym-Kadanoff	$G_{\alpha\beta}(\mathbf{k}, i\omega)$	$\Sigma_{\text{int},\alpha\beta}(\mathbf{k}, i\omega)$
DMFT (BL)	$G_{\text{loc},\alpha\beta}(i\omega)$	$\mathcal{M}_{\text{int},\alpha\beta}(i\omega)$
DMFT (AL)	$G_{\text{loc},\alpha\beta}(i\omega)$	$\Delta_{\alpha\beta}(i\omega)$
LDA+DMFT (BL)	$\rho(r)$, $G_{\text{loc},ab}(i\omega)$	$V_{\text{int}}(r)$, $\mathcal{M}_{\text{int},ab}(i\omega)$
LDA+DMFT (AL)	$\rho(r)$, $G_{\text{loc},ab}(i\omega)$	$V_{\text{int}}(r)$, $\Delta_{ab}(i\omega)$
LDA+U	$\rho(r)$, n_{ab}	$V_{\text{int}}(r)$, λ_{ab}
LDA	$\rho(r)$	$V_{\text{int}}(r)$

G. Kotliar *et al.*, Rev. Mod. Phys. 78, 865 (2006)

Occupation updates for DFT

interacting charge density

$$\rho(\mathbf{r}) = \frac{1}{\beta} \sum_{n,\mathbf{k}} \langle \mathbf{r} | \hat{G}(\mathbf{k}, \mathrm{i}\omega_n) | \mathbf{r} \rangle \equiv \rho^{\mathrm{KS}}(\mathbf{r}) + \Delta\rho(\mathbf{r})$$

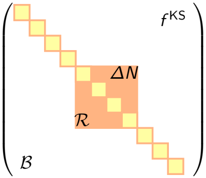
KS charge density:

$$\rho^{\mathrm{KS}}(\mathbf{r}) = \sum_{\mathbf{k}} \sum_{n=1}^{N_{\mathcal{B}}} f^{\mathrm{KS}}_{\nu\mathbf{k}} \langle \mathbf{r} | \psi_{n\mathbf{k}} \rangle \langle \psi_{n\mathbf{k}} | \mathbf{r} \rangle$$

→ compute $\Delta\rho(\mathbf{r})$, feed it back to DFT, compute updated KS charge density $\rho^{\mathrm{KS}}(\mathbf{r})$

$$\Delta\rho(\mathbf{r}) = \frac{1}{\beta} \sum_{n,\mathbf{k}} \langle \mathbf{r} | \hat{G}(\mathbf{k}, \mathrm{i}\omega_n) - \hat{G}^{\mathrm{KS}}(\mathbf{k}, \mathrm{i}\omega_n) | \mathbf{r} \rangle$$
$$\equiv \sum_{\mathbf{k}} \langle \mathbf{r} | \Delta\hat{N}(\mathbf{k}) | \mathbf{r} \rangle$$

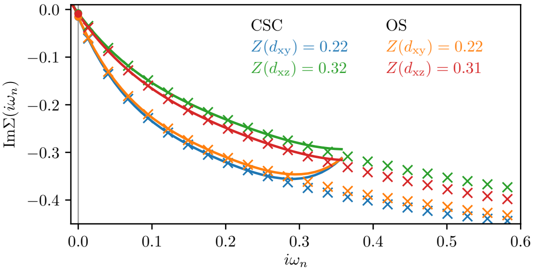
$\hat{N}(\mathbf{k}) =$

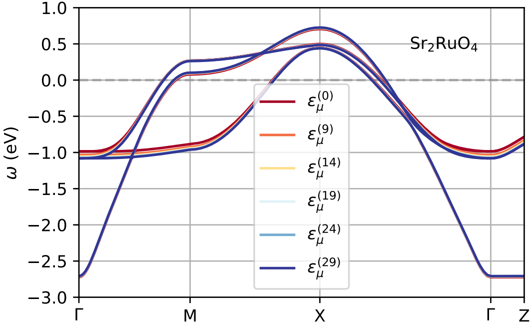


F. Lechermann *et al.*, Phys. Rev. B 74, 125120 (2006)
M. Schüler *et al.*, J. Phys. Condens. Matter 30, 475901 (2018)

Quasiparticle mass renormalization in Sr_2RuO_4

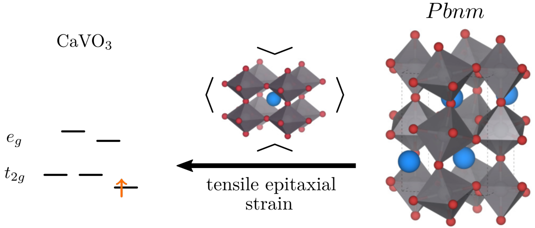
FLATIRON INSTITUTE
Center for Computational Quantum Physics

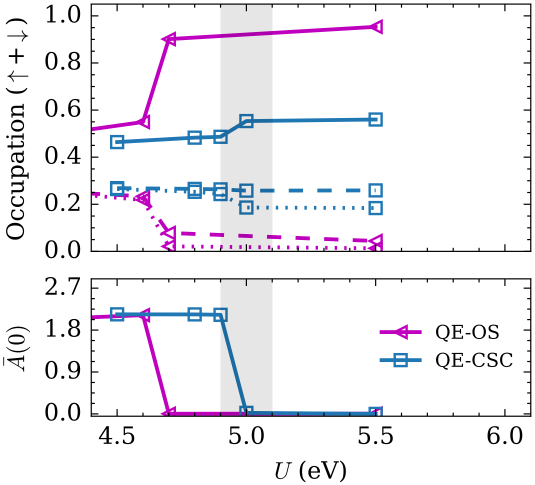




- CT-HYB solver, $\beta = 232$ eV^{-1}
- minimal effect of charge self-consistency

Orbital polarization in $CaVO_3$ (tensile strain)





- CT-HYB solver, $\beta = 40$ eV^{-1}
- charge self-consistency strongly reduces the orbital polarization found in one-shot calculations

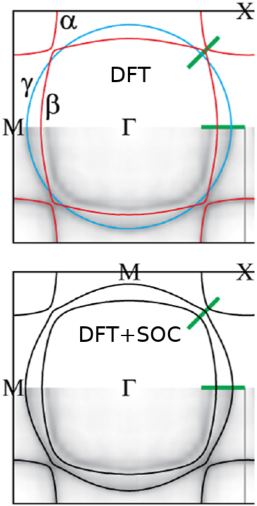
A. Hampel, SB, and C. Ederer, Phys. Rev. Res. 2, 033088 (2020)

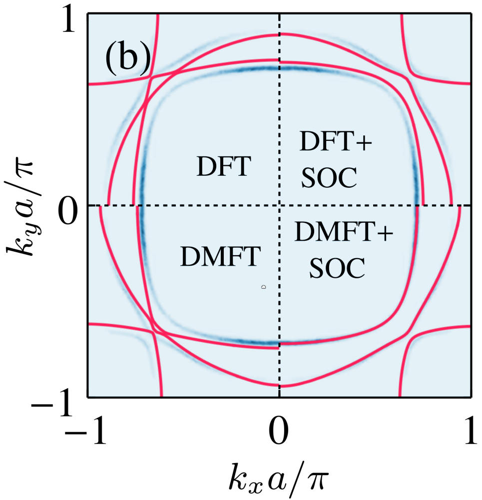
Post-processing

What we can compute:

- spectral properties
- optical and thermal conductivity
- Hall and Seebeck coefficient
- two-particle correlation function (susceptibilities)
- ...
- electronic Raman spectroscopy
- x-ray photoemission and absorption spectroscopy
- resonant inelastic x-ray scattering
- phonon spectra

Back to the experiment





M. W. Haverkort *et al.*, Phys. Rev. Lett. 101, 026406 (2008)

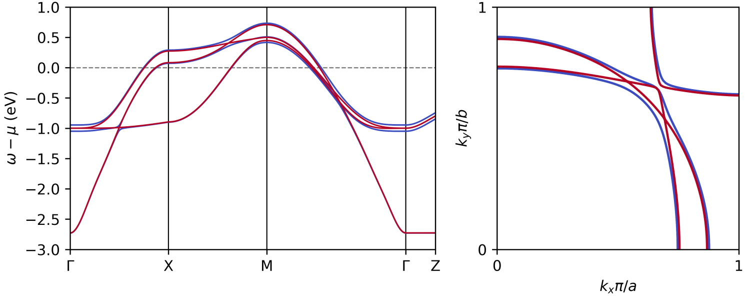
A. Tamai *et al.*, Phys. Rev. X 9, 021048 (2019)

X. Cao *et al.*, Phys. Rev. B 104, 115119 (2021)

Spin-orbit coupling in Sr_2RuO_4

spin-orbit coupling λ

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_{xy} & 0 & 0 & 0 & \frac{\lambda_{xy}}{2} & \frac{i\lambda_{xy}}{2} \\ 0 & \varepsilon_{yz} & -\frac{i\lambda_z}{2} & -\frac{\lambda_{xy}}{2} & 0 & 0 \\ 0 & \frac{i\lambda_z}{2} & \varepsilon_{xz} & -\frac{i\lambda_{xy}}{2} & 0 & 0 \\ 0 & -\frac{\lambda_{xy}}{2} & \frac{i\lambda_{xy}}{2} & \varepsilon_{xy} & 0 & 0 \\ \frac{\lambda_{xy}}{2} & 0 & 0 & 0 & \varepsilon_{yz} & \frac{i\lambda_z}{2} \\ -\frac{i\lambda_{xy}}{2} & 0 & 0 & 0 & -\frac{i\lambda_z}{2} & \varepsilon_{xz} \end{pmatrix}$$

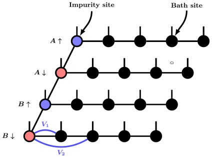


$$\hat{H}_{\lambda}^{\mathrm{SOC}} = \frac{\lambda}{2} \sum_{ij} \sum_{\sigma\sigma'} c_{i\sigma}^{\dagger} (\mathbf{l}_{ij} \cdot \boldsymbol{\sigma}_{\sigma\sigma'}) c_{j\sigma'}$$

- correlation-induced enhancement of crystal-field splitting
- correlation-induced enhancement of effective spin-orbit coupling

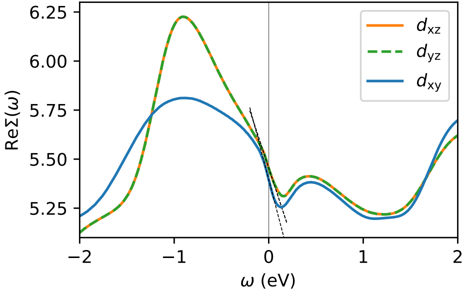
Tensor network real-frequency impurity solver

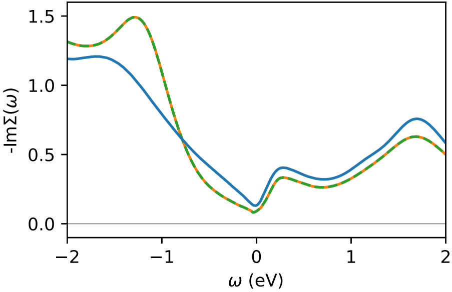
FLATIRON INSTITUTE
Center for Computational Quantum Physics



self-energy from ForkTPS: $\Sigma(\omega) = \Sigma'(\omega) + i\Sigma''(\omega)$

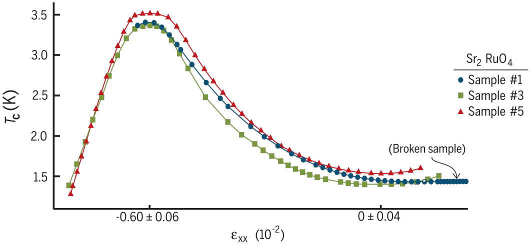
$$A(\mathbf{k},\omega) = \frac{1}{\pi}\frac{\Sigma''(\omega)}{(\omega - \epsilon_{\mathbf{k}} - \Sigma'(\omega))^2 + \Sigma''(\omega)^2}$$



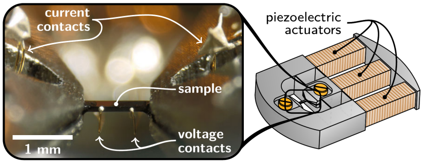


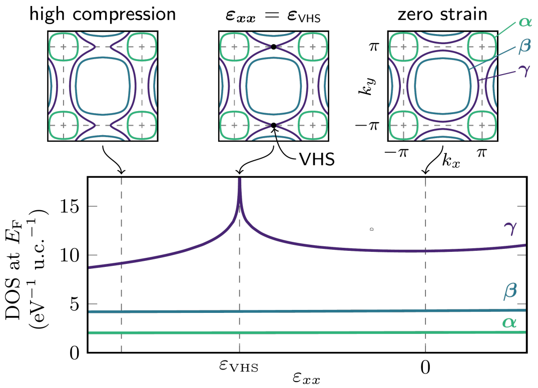
D. Bauernfeind *et al.*, Phys. Rev. X 7, 031013 (2017)

Sr_2RuO_4 under uniaxial pressure



(b)





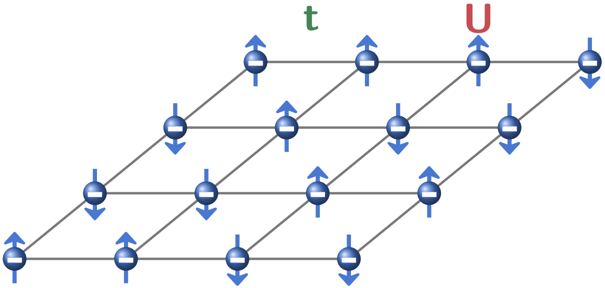
A. Steppke *et al.*, Science 355, eaaf9398 (2017)

M. E. Barber *et al.*, Phys. Rev. Lett. 120, 076602 (2018)

Uniaxial strain experiments

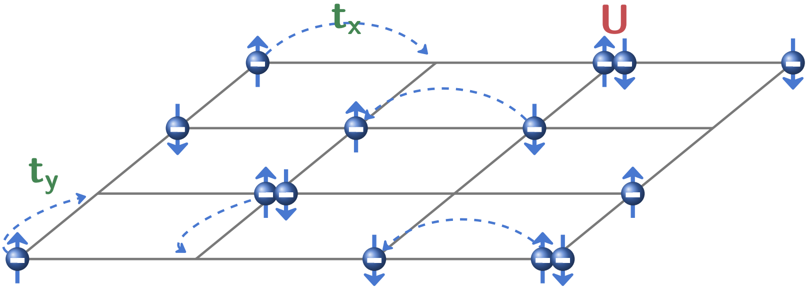
FLATIRON INSTITUTE
Center for Computational Quantum Physics

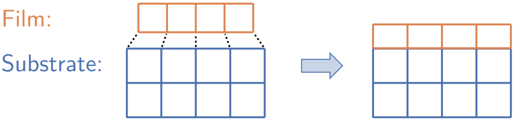
$$H = -\sum_{\langle ij\rangle\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



Uniaxial strain experiments

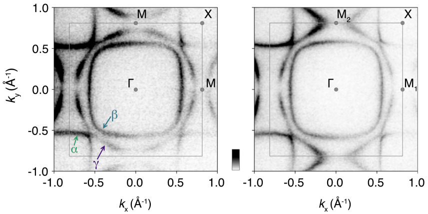
$$H = -\sum_{\langle ij \rangle \sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

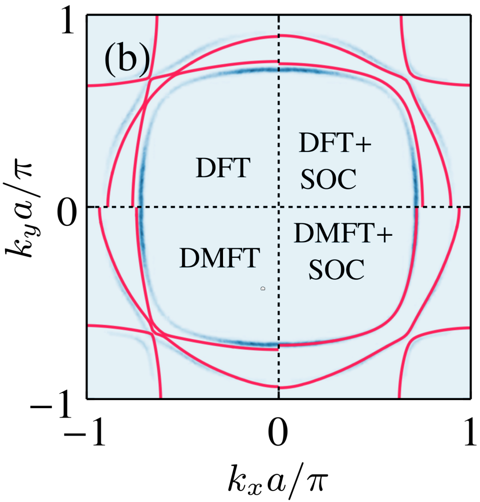




Pressure-driven Lifshitz transition in Sr_2RuO_4

FLATIRON INSTITUTE
Center for Computational Quantum Physics

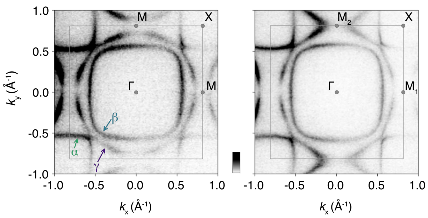




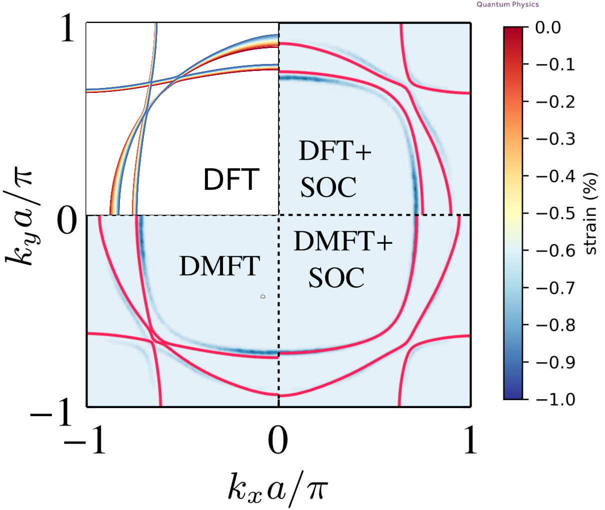
- Lifshitz transition with uniaxial strain
- novel FTPS impurity solver, including spin-orbit coupling
- critical strain $\epsilon_{xx} \approx -0.4$ consistent with experiment

V. Sunko *et al.*, npj Quantum Mater. 4, 46 (2019)
M. E. Barber *et al.*, Phys. Rev. B 100, 245139 (2019)
D. Bauernfeind *et al.*, Phys. Rev. X 7, 031013 (2017)
X. Cao *et al.*, Phys. Rev. B 104, 115119 (2021)

Pressure-driven Lifshitz transition in Sr_2RuO_4



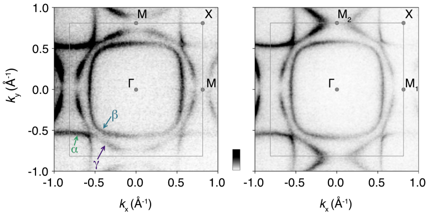
- Lifshitz transition with uniaxial strain
- novel FTPS impurity solver, including spin-orbit coupling
- critical strain $\epsilon_{xx} \approx -0.4$ consistent with experiment

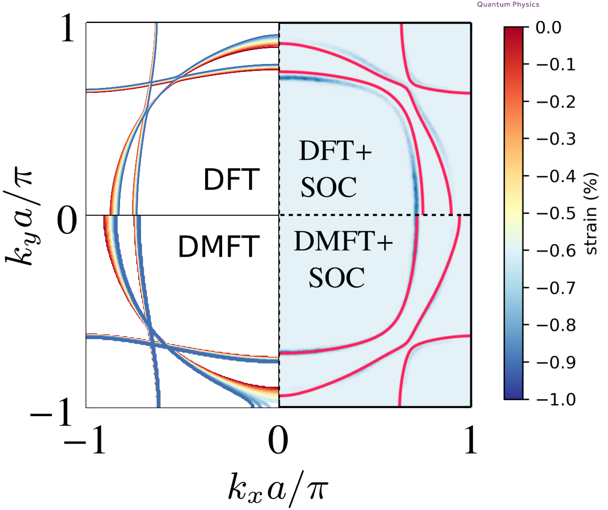


V. Sunko *et al.*, npj Quantum Mater. 4, 46 (2019)
M. E. Barber *et al.*, Phys. Rev. B 100, 245139 (2019)

D. Bauernfeind *et al.*, Phys. Rev. X 7, 031013 (2017)
X. Cao *et al.*, Phys. Rev. B 104, 115119 (2021)

Pressure-driven Lifshitz transition in Sr_2RuO_4

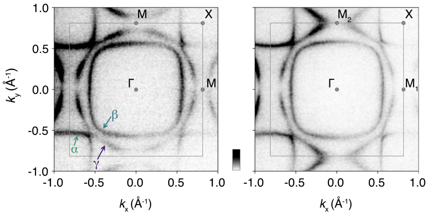


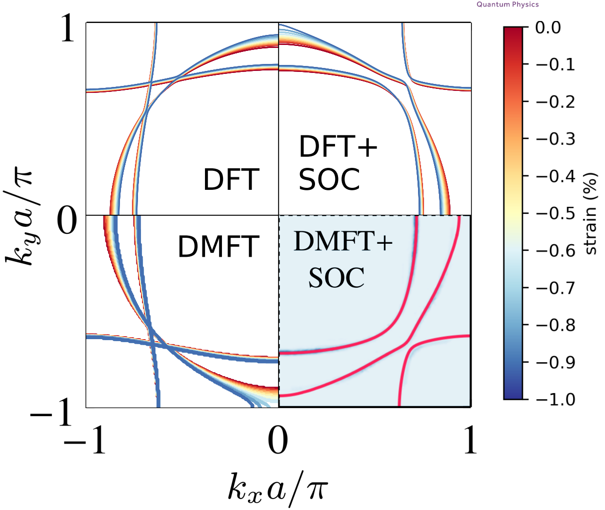


- Lifshitz transition with uniaxial strain
- novel FTPS impurity solver, including spin-orbit coupling
- critical strain $\epsilon_{xx} \approx -0.4$ consistent with experiment

V. Sunko *et al.*, npj Quantum Mater. 4, 46 (2019)
M. E. Barber *et al.*, Phys. Rev. B 100, 245139 (2019)
D. Bauernfeind *et al.*, Phys. Rev. X 7, 031013 (2017)
X. Cao *et al.*, Phys. Rev. B 104, 115119 (2021)

Pressure-driven Lifshitz transition in Sr_2RuO_4

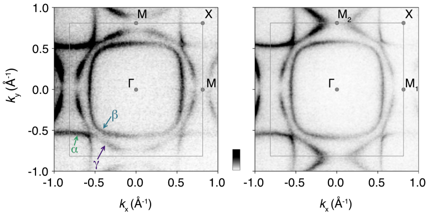




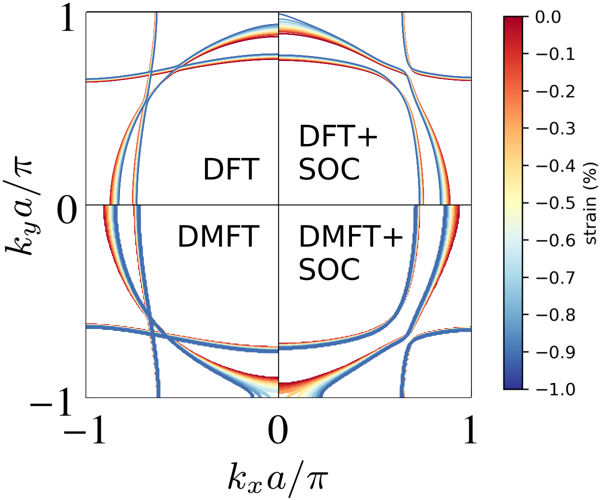
- Lifshitz transition with uniaxial strain
- novel FTPS impurity solver, including spin-orbit coupling
- critical strain $\epsilon_{xx} \approx -0.4$ consistent with experiment

V. Sunko *et al.*, npj Quantum Mater. 4, 46 (2019)
M. E. Barber *et al.*, Phys. Rev. B 100, 245139 (2019)
D. Bauernfeind *et al.*, Phys. Rev. X 7, 031013 (2017)
X. Cao *et al.*, Phys. Rev. B 104, 115119 (2021)

Pressure-driven Lifshitz transition in Sr_2RuO_4



- Lifshitz transition with uniaxial strain
- novel FTPS impurity solver, including spin-orbit coupling
- critical strain $\epsilon_{xx} \approx -0.4$ consistent with experiment



V. Sunko *et al.*, npj Quantum Mater. 4, 46 (2019)

M. E. Barber *et al.*, Phys. Rev. B 100, 245139 (2019)

D. Bauernfeind *et al.*, Phys. Rev. X 7, 031013 (2017)

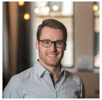
X. Cao *et al.*, Phys. Rev. B 104, 115119 (2021)

Streamlined, robust and efficient screening of materials

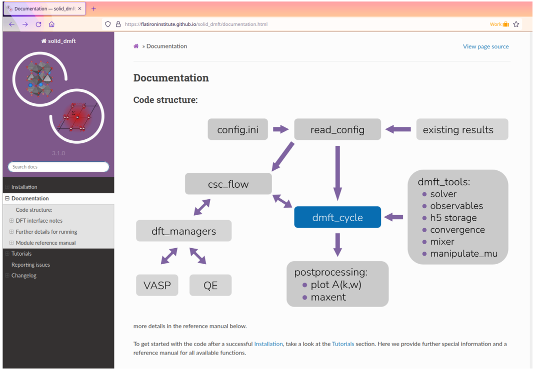
solid_dmft

A versatile python wrapper to perform DFT + DMFT calculations utilizing the TRIQS software library.



M. Merkel (ETHZ) | A. Carta (ETHZ) | S. Beck | A. Hampel

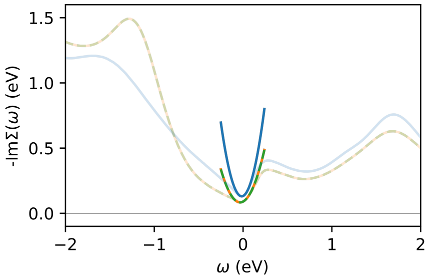


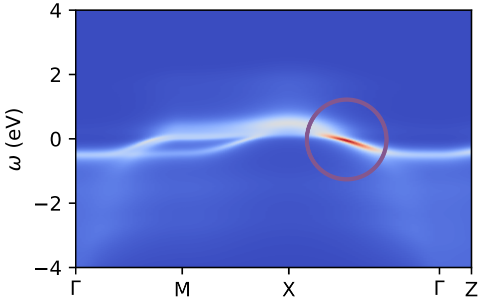
M. Merkel, A. Carta, SB and A. Hampel, J. Open Source Softw. 7(77), 4623 (2022)

Scattering rate and Brillouin zone integrals

FLATIRON INSTITUTE
Center for Computational Quantum Physics

$$\Sigma(\omega, T) \propto -\frac{\mathrm{i}}{Z\alpha}[\omega^2 + (\pi k_{\mathrm{B}} T)^2]$$



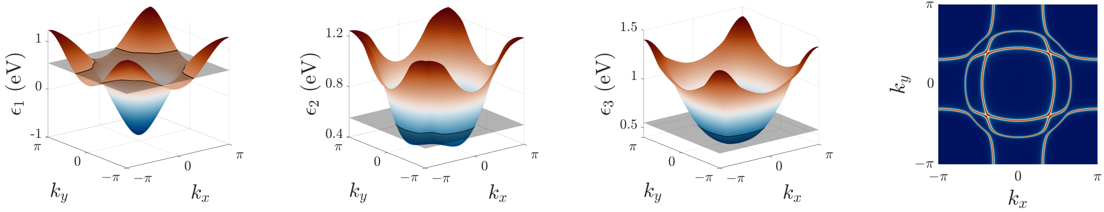


- scattering rate finite but possibly extremely small
- frequency dependence requires adaptivity for momentum integration

Automatic, high-order, adaptive Brillouin zone integration

Task: compute local single-particle Green's function (i.e. DOS)

$$G(\omega) = \int_{\mathsf{BZ}} \mathrm{d}^3\mathbf{k}\, \mathrm{Tr}\left[\left(\omega - H(\mathbf{k}) - \Sigma(\mathbf{k},\omega)\right)^{-1}\right]$$

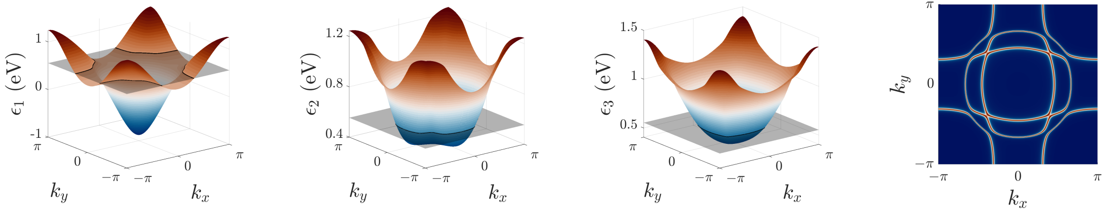


Automatic, high-order, adaptive Brillouin zone integration

Task: compute local single-particle Green's function (i.e. DOS)

$$G(\omega) = \int_{\mathrm{BZ}} \mathrm{d}^3\mathbf{k}\,\mathrm{Tr}\left[(\omega - H(\mathbf{k}) - \Sigma(\mathbf{k},\omega))^{-1}\right]$$

- **Applications**: self-consistency loops in DMFT and post-processing

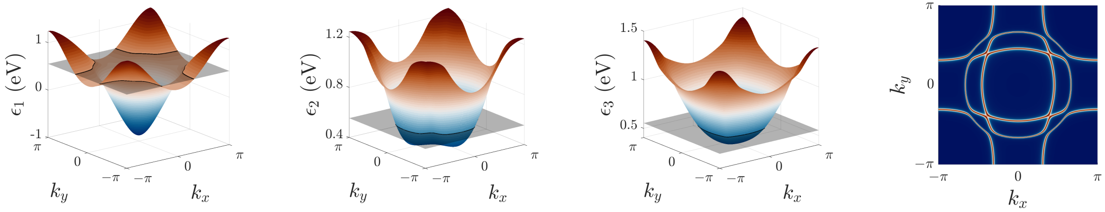


Automatic, high-order, adaptive Brillouin zone integration

Task: compute local single-particle Green's function (i.e. DOS)

$$G(\omega) = \int_{\mathsf{BZ}} \mathrm{d}^3\mathbf{k}\, \mathrm{Tr}\left[\left(\omega - H(\mathbf{k}) - \Sigma(\mathbf{k},\omega)\right)^{-1}\right]$$

- **Applications**: self-consistency loops in DMFT and post-processing
- **Setting**: $H(\mathbf{k})$ obtained from a Wannier Hamiltonian $H(\mathbf{R})$, $\Sigma(\mathbf{k},\omega) = \mathrm{i}\eta$



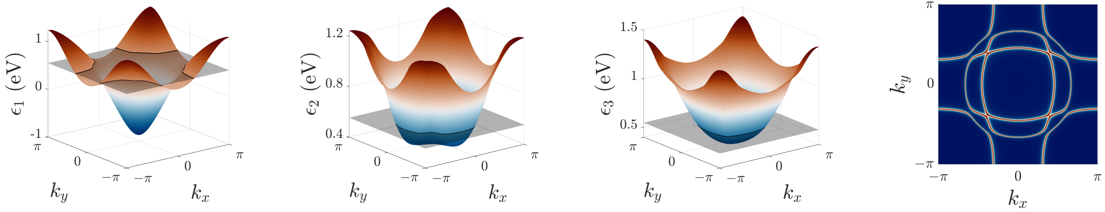
Automatic, high-order, adaptive Brillouin zone integration

FLATIRON INSTITUTE
Center for Computational Quantum Physics

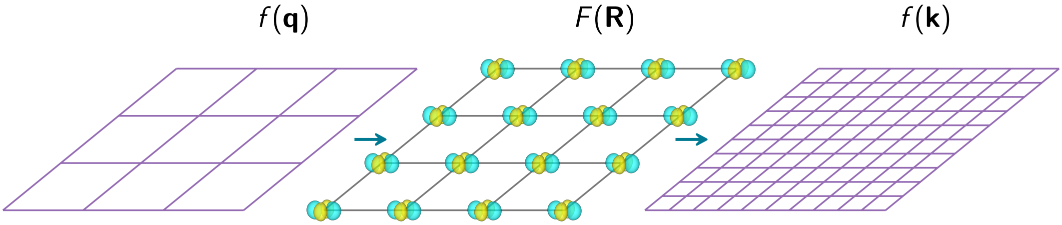
Task: compute local single-particle Green's function (i.e. DOS)

$$G(\omega) = \int_{\mathsf{BZ}} \mathrm{d}^3\mathbf{k}\,\mathrm{Tr}\left[\left(\omega - H(\mathbf{k}) - \Sigma(\mathbf{k},\omega)\right)^{-1}\right]$$

- **Applications**: self-consistency loops in DMFT and post-processing
- **Setting**: $H(\mathbf{k})$ obtained from a Wannier Hamiltonian $H(\mathbf{R})$, $\Sigma(\mathbf{k},\omega) = \mathrm{i}\eta$
- **Goal**: fully automatic, high-order and adaptive algorithm



Wannier interpolation



$$\mathcal{O}_{nm}(\mathbf{q}) = \langle u_{n\mathbf{q}} | \hat{\mathcal{O}}(\mathbf{q}) | u_{m\mathbf{q}} \rangle$$

$$\mathcal{O}^{(W)}_{nm}(\mathbf{R}) = \frac{1}{N_0} \sum_{\mathbf{q}} e^{-i\mathbf{q}\cdot\mathbf{R}} \mathcal{O}^{(W)}_{nm}(\mathbf{q})$$

$$\mathcal{O}^{(W)}_{nm}(\mathbf{k}) = \sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}} \mathcal{O}^{(W)}_{nm}(\mathbf{R})$$

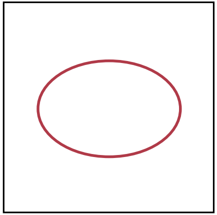
J. R. Yates, X. Wang, D. Vanderbilt, and I. Souza, Phys. Rev. B 75, 195121 (2007)

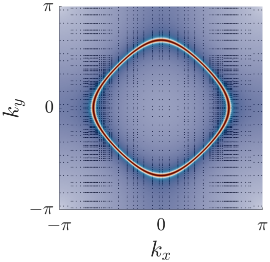
Integration schemes applied to BZ integrals and their scalings

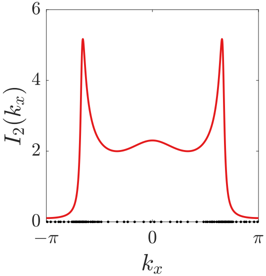
FLATIRON INSTITUTE
Center for Computational Quantum Physics

- periodic trapezoidal rule (PTR): $\mathcal{O}(\eta^{-3})$
- iterated adaptive integration (IAI)[1]: $\mathcal{O}(\log^3(\eta^{-1}))$

$$\int\int \mathrm{d}k_x\, \mathrm{d}k_y\, f(k_x,k_y) = \int \mathrm{d}k_x\, I_2(k_x), \qquad I_2(k_x) = \int \mathrm{d}k_y\, f(k_x,k_y)$$





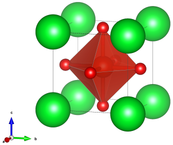


[1] J. Kaye, SB, A. Barnett, L. Van Muñoz, and O. Parcollet, arxiv:2211.12959 (2022)

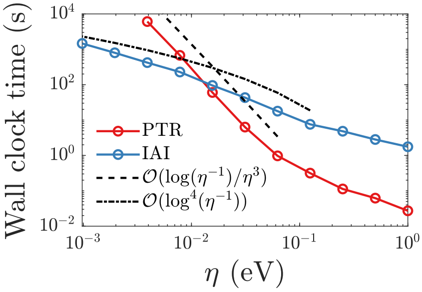
Example: density of states

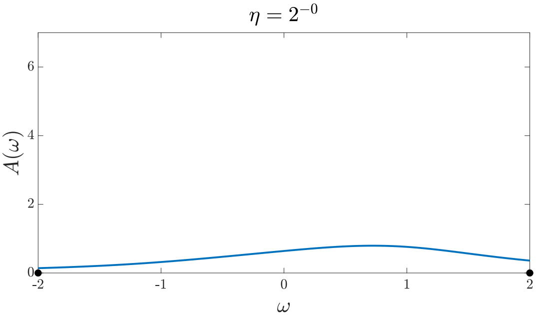
FLATIRON INSTITUTE
Center for Computational Quantum Physics

DOS of $SrVO_3$, three t_{2g} orbitals:



$$A(\omega) = -\frac{1}{\pi}\mathrm{Im}G(\omega) = -\frac{1}{\pi}\mathrm{Im}\int_{\mathrm{BZ}} \mathrm{d}^3\mathbf{k}\,\mathrm{Tr}\left[(\omega - H(\mathbf{k}) - i\eta)^{-1}\right]$$

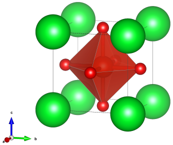


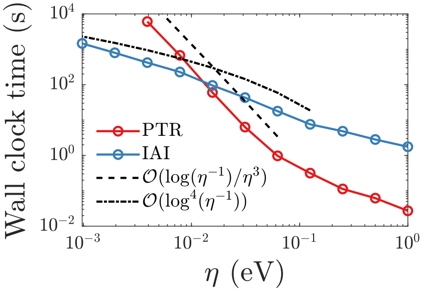


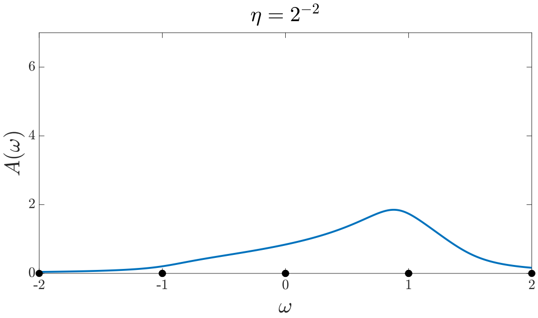
Example: density of states

DOS of $SrVO_3$, three t_{2g} orbitals:

$$A(\omega) = -\frac{1}{\pi}\mathrm{Im}G(\omega) = -\frac{1}{\pi}\mathrm{Im}\int_{\mathrm{BZ}} \mathrm{d}^3\mathbf{k}\,\mathrm{Tr}\left[\left(\omega - H(\mathbf{k}) - i\eta\right)^{-1}\right]$$



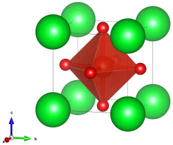


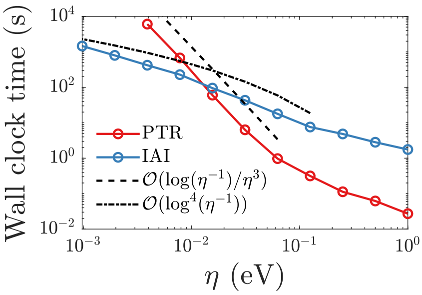


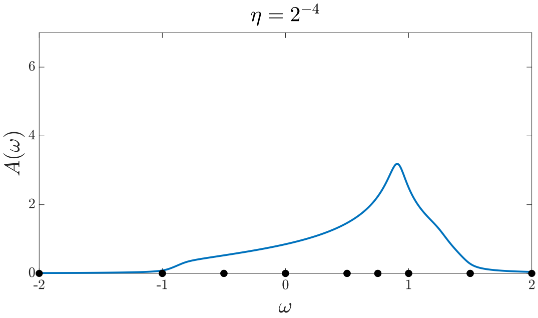
Example: density of states

DOS of $SrVO_3$, three t_{2g} orbitals:

$$A(\omega) = -\frac{1}{\pi}\mathrm{Im}G(\omega) = -\frac{1}{\pi}\mathrm{Im}\int_{\mathrm{BZ}} \mathrm{d}^3\mathbf{k}\,\mathrm{Tr}\left[(\omega - H(\mathbf{k}) - i\eta)^{-1}\right]$$



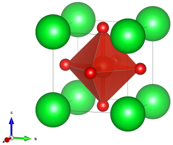


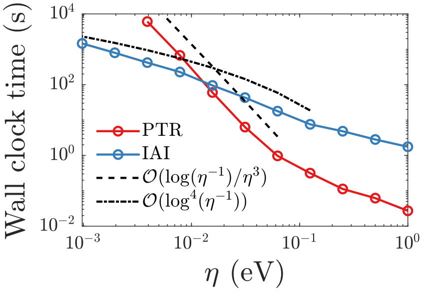


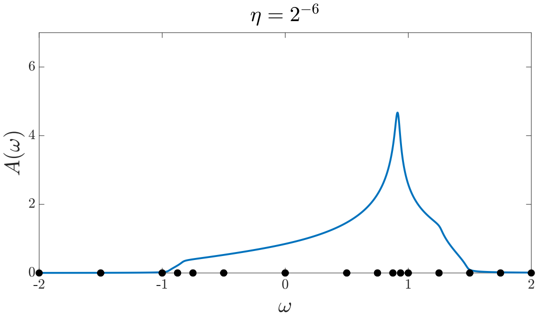
Example: density of states

DOS of $SrVO_3$, three t_{2g} orbitals:

$$A(\omega) = -\frac{1}{\pi}\mathrm{Im}G(\omega) = -\frac{1}{\pi}\mathrm{Im}\int_{\mathrm{BZ}} \mathrm{d}^3\mathbf{k}\,\mathrm{Tr}\left[(\omega - H(\mathbf{k}) - i\eta)^{-1}\right]$$



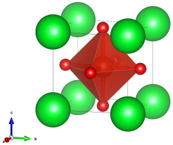


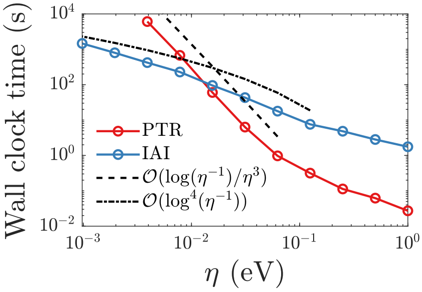


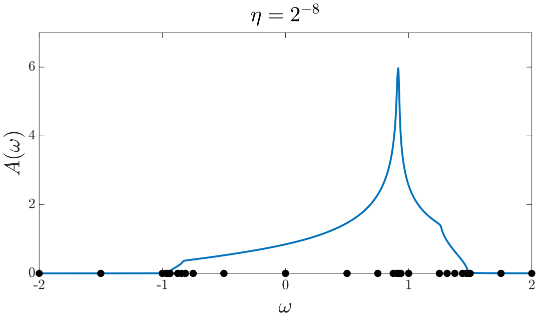
Example: density of states

DOS of $SrVO_3$, three t_{2g} orbitals:

$$A(\omega) = -\frac{1}{\pi}\mathrm{Im}G(\omega) = -\frac{1}{\pi}\mathrm{Im}\int_{\mathrm{BZ}} \mathrm{d}^3\mathbf{k}\,\mathrm{Tr}\left[(\omega - H(\mathbf{k}) - i\eta)^{-1}\right]$$

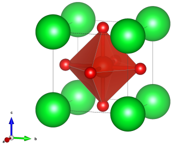




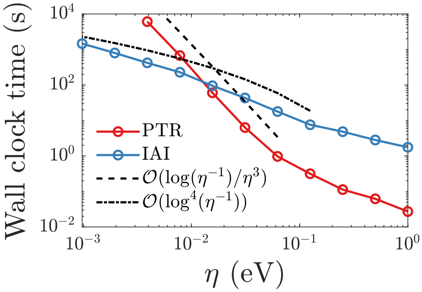


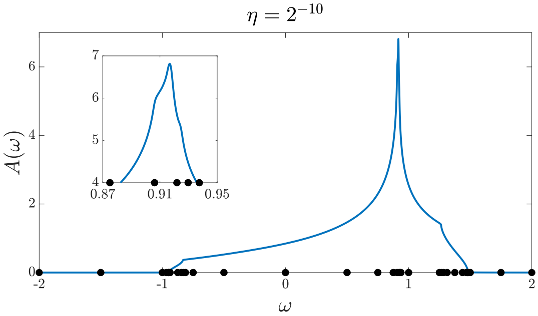
Example: density of states

DOS of $SrVO_3$, three t_{2g} orbitals:

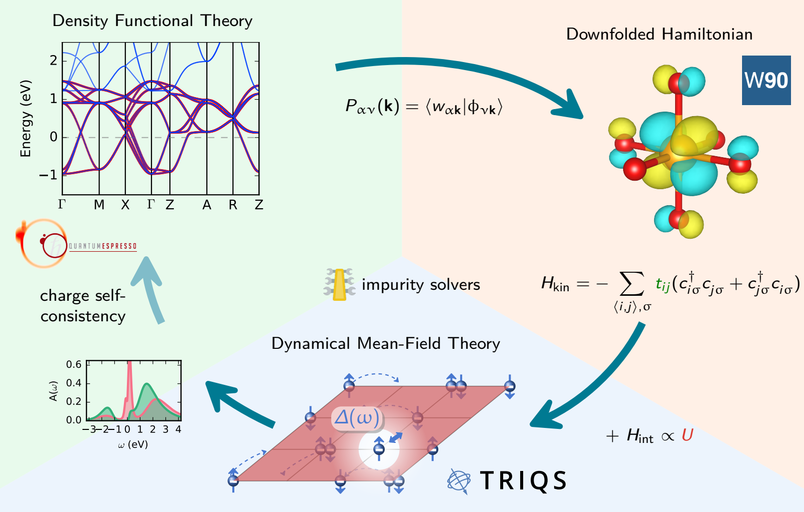


$$A(\omega) = -\frac{1}{\pi}\mathrm{Im}G(\omega) = -\frac{1}{\pi}\mathrm{Im}\int_{\mathrm{BZ}} \mathrm{d}^3\mathbf{k}\,\mathrm{Tr}\left[(\omega - H(\mathbf{k}) - i\eta)^{-1}\right]$$

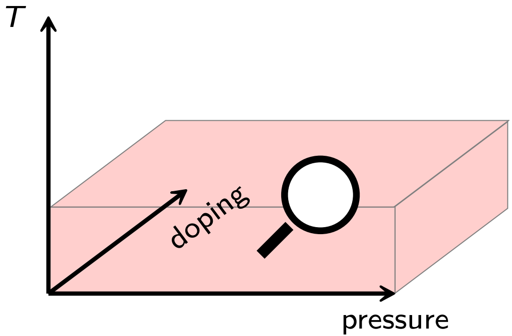




Summary


Density Functional Theory
Energy (eV)
Γ M X Γ Z A R Z
QUANTUMESPRESSO
charge self-consistency
$P_{\alpha\nu}(\mathbf{k}) = \langle w_{\alpha\mathbf{k}} | \phi_{\nu\mathbf{k}} \rangle$
Downfolded Hamiltonian
W90
$H_{\mathrm{kin}} = -\sum_{\langle i,j \rangle,\sigma} t_{ij}(c^\dagger_{i\sigma} c_{j\sigma} + c^\dagger_{j\sigma} c_{i\sigma})$
$+ H_{\mathrm{int}} \propto U$
impurity solvers
Dynamical Mean-Field Theory
$\Delta(\omega)$
TRIQS
A(ω)
ω (eV)

More problems - more literature



- double counting
- more orbitals, more complex systems
- screening
- (real-frequency) impurity solvers and analytic continuation
- superconductivity
- out of equilibrium
- low-T, exotic states

Jülich, Autumn School on Correlated Electrons www.cond-mat.de/events/correl.html