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CSSI Element: SI2-SSE: Collaborative Research: Software Framework for Strongly Correlated Materials: from DFT to DMFT

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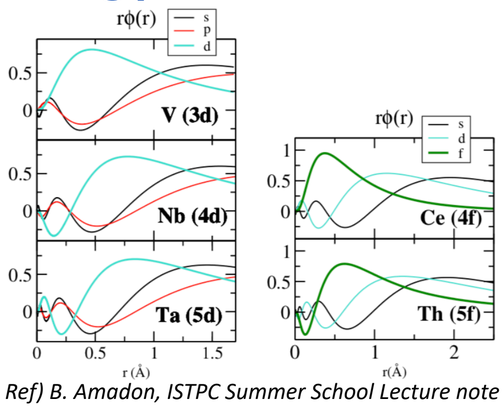
Award #: SI2-SSE 1740112

Project Goals

- **Development of an open-source and user-friendly software (DMFTwDFT) with a Python interface combining DMFT to various electronic structure packages including VASP, Siesta, and Wannier90.**
- **Application of the DMFTwDFT package to the characterization of novel electronic and structural properties of strongly correlated materials including transition metal oxides and heavy fermion systems.**

Introduction

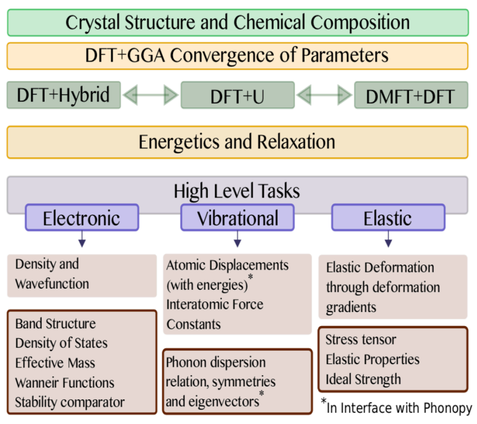
Strongly correlated materials



Ref) B. Amadon, ISTPC Summer School Lecture note

- *d* or *f* orbitals are highly localized -> Their partial occupation leads to strong electron correlations in materials.
- Conventional density functional theory (DFT) fails in strongly correlated materials.

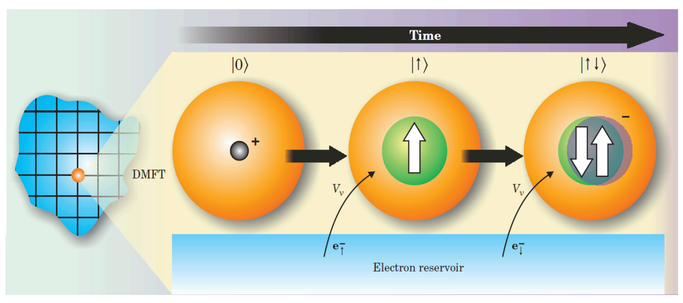
Motivation



- Needs for an efficient, high-level, open-source package beyond DFT interfaced with a Python script for users to describe strongly correlated materials

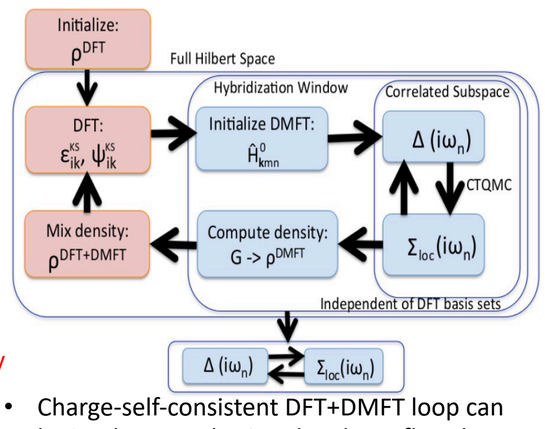
Methodology

Dynamical mean field theory



- DMFT approximates the lattice problem to a single-impurity problem hybridized to a self-consistently determined bath.
- The impurity problem can be solved using the numerically exact quantum Monte Carlo method, and it scales as O(N) where N is the number of correlated atoms.
- DMFT can define a Free energy functional in terms of the local Green's function **G(ω)** and the self-energy **Σ(ω)**

DFT+DMFT

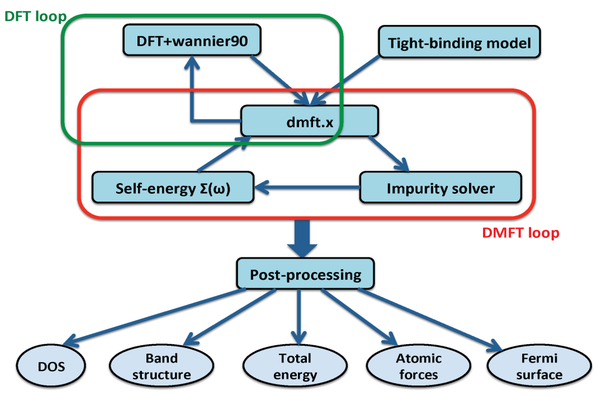


- Charge-self-consistent DFT+DMFT loop can be implemented using the above flowchart.
- Our code adopts the Wannier orbitals (obtained from wannier90 code) for the construction of hybridization and correlation subspaces.

Features of the DMFTwDFT package

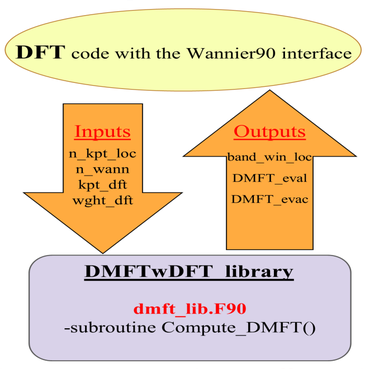
DMFTwDFT website: https://github.com/DMFTwDFT-project/DMFTwDFT

Overall structure



- We provide various post-processing tools

Library mode



- Our Library mode provides an efficient link of our package to an arbitrary DFT code for the charge-self-consistent DFT+DMFT implementation without much modifications of the DFT code

Interface to different DFT

W90 VASP siesta

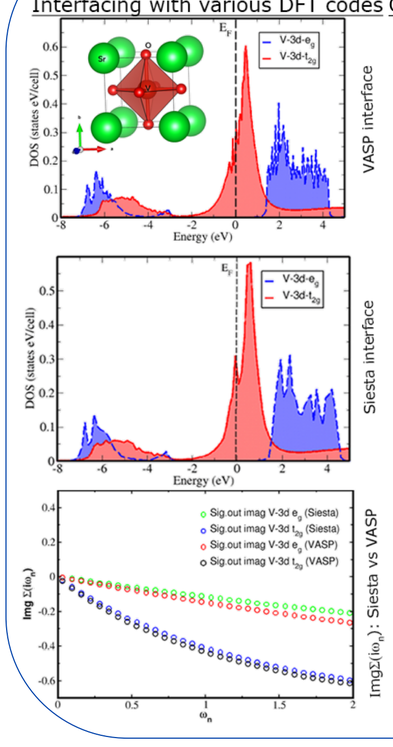
Parallelization

- We provide the parallelization of the k-point mesh for DMFT calculations using MPI.

Main results of DMFTwDFT (arXiv:2002.00068)

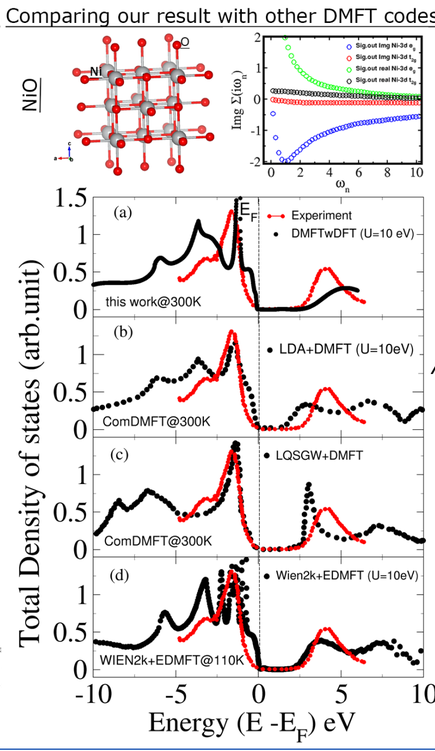
$SrVO_3$

Interfacing with various DFT codes



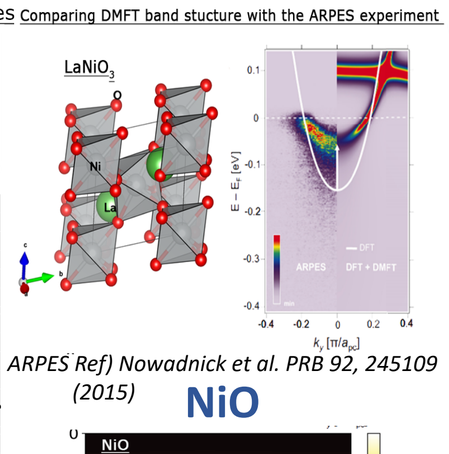
NiO

Comparing our result with other DMFT codes



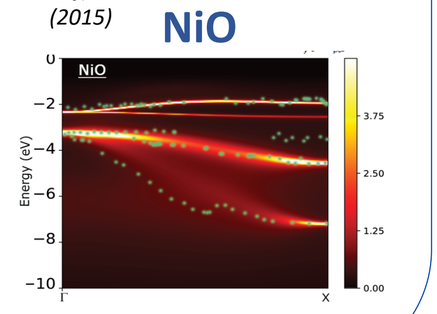
$LaNiO_3$

Comparing DMFT band stucture with the ARPES experiment



ARPES Ref) Nowadnick et al. PRB 92, 245109 (2015)

NiO



ARPES Ref) Shen et al. PRB 44 3604 (1991)

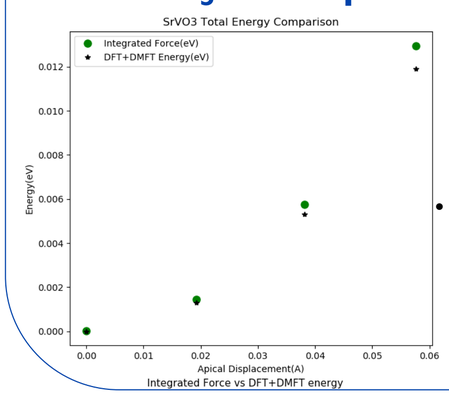
Atomic Force calculations

Force formula in DFT+DMFT

$$\mathbf{F} = -\frac{\delta E}{\delta \mathbf{R}}$$

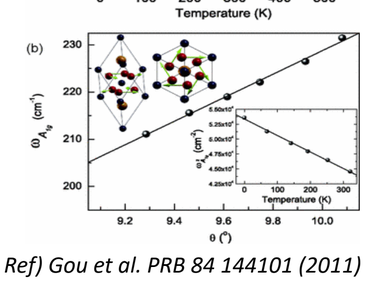
$$\begin{aligned}\delta E &= Tr[(\delta\epsilon_{i\mathbf{k}} + \delta\Sigma_{ij}(i\omega_n))G_{ji}(i\omega_n)] - Tr[\delta V^{Hxc}\rho] - Tr[\delta\Sigma G] \\ &= \mathbf{F}^{DFT} - Tr[\delta\epsilon_{i\mathbf{k}} f_i] + Tr[\delta\epsilon_{i\mathbf{k}} G_{ji}] \\ &+ Tr[\delta(\langle\psi_i|\phi_m\rangle)\Sigma_{mn}\langle\phi_n|\psi_j\rangle G_{ji}] + Tr[\langle\psi_i|\phi_m\rangle\Sigma_{mn}\delta(\langle\phi_n|\psi_j\rangle)G_{ji}]\end{aligned}$$

$SrVO_3$ example



- Our $SrVO_3$ run example shows the consistent trend between integrated forces and energies (The discrepancy can be due to the change of Wannier orbitals in the above formula).
- We will use our DMFTwDFT package for the study of $LaNiO_3$ phonon modes and the structural phase transition at finite temperatures.

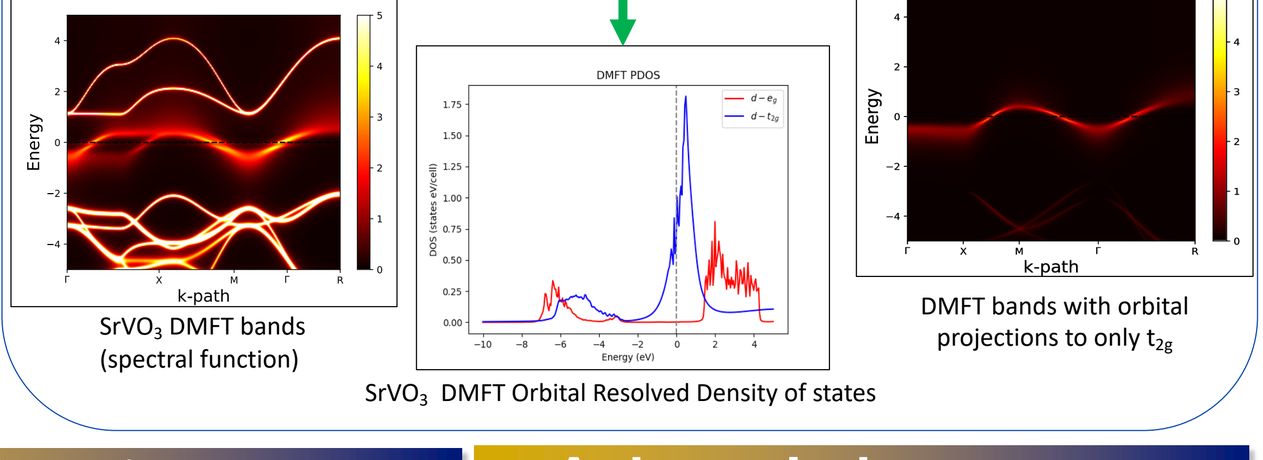
Phonon calculation in $LaNiO_3$



Ref) Gou et al. PRB 84 144101 (2011)

- Phonon modes in $LaNiO_3$ has been computed by Gou *et al* using DFT zero-temperature energetics.

Automated post-processing tools



$SrVO_3$ DMFT bands (spectral function)

$SrVO_3$ DMFT Orbital Resolved Density of states

DMFT bands with orbital projections to only t_{2g}

Conclusions

- An open-source and Python-interfaced computational package, DMFTwDFT, combining DMFT to various DFT codes including VASP and Siesta has been developed.
- Our package provides various features including the interface with the wannier90 code, a library mode for an efficient link of our package to an arbitrary DFT code, and various post-processing tools for data analysis of band structures, density of states, and total energies.
- Electronic structures of $SrVO_3$ obtained from, both VASP+DMFT and SIESTA+DMFT interfaces, is in good agreement, showing the moderate mass enhancement of Sr t_{2g} orbitals. Our NiO calculation shows that Ni e_g orbital develops a Mott gap, and band structures below the Fermi energy are consistent with ARPES measurements.

Future Directions

- We will apply our DMFTwDFT package to the study of electronic and structural properties of various strongly correlated materials including the rare-earth materials with strongly correlated *f* orbitals hybridized with other bands and transition metal oxides with vacancies and disorder.
- We will implement both the atomic force and the Fermi surface calculations within our DMFTwDFT package for the next release of the package.
- We will implement the calculation of U and J values within our package based on ab initio linear-response theory from the constrained DFT method.

Acknowledgements

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