Fork Tensor Product States: Multi-Orbital Impurity Solver

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Motivation & Method

Published in Phys. Rev. X 7, 031013

Problem: Imaginary time impurity solvers like CTQMC rely on ill posed analytic continuation to calculate real frequency properties.

$\Rightarrow$ Real frequency solver highly desired!

MPS based methods give high resolution spectra only up to 2-orbital models.

For real material calculations at least 3-orbital solver necessary.

Anderson Impurity Models (multi orbital)

$$H = \sum_{k,m} \epsilon_{km} n_{km} + V_{km} \left(c_{0m}^\dagger c_{km} + h.c. \right) + H_{loc}$$

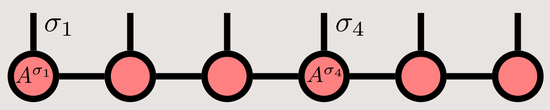
H_{loc} contains interaction terms, e.g. Kanamori Hamiltonian.

Matrix Product States (MPS)

Represent states as product of matrices (always possible):

$$|\psi\rangle = \sum_\sigma A^{\sigma_1} A^{\sigma_2} \cdots A^{\sigma_N} |\sigma_1 \sigma_2 \cdots \sigma_N\rangle$$

Understand tensor networks in general (not only MPS) by their graphical representation. For MPS:

$|\psi\rangle =$ 

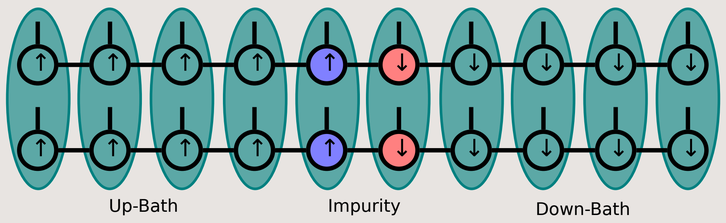
Each circle corresponds to a tensor (matrix) and each line to an index. Connected lines are summed over.

Calculation of Green's Functions

1. **Calculate ground state** $|\psi_0\rangle$
2. Apply creation/annihilation operators and **time evolve** $|\psi^<(t)\rangle = e^{iHt} c |\psi_0\rangle$
3. Calculate overlap, e.g. $G^<(t) = \langle \psi_0 | c^\dagger | \psi^<(t) \rangle$
4. Post process (i.e.: linear prediction)
5. Fourier transform to real-frequency space

Impurity Solver with MPS - the usual way

Split up- and down- spin. For multi-orbital models, combine orbitals to one larger site (for each bath site), e.g. for 2-orbital model:



Problem: For AIMs with non-interacting orbitals, the bond dimension m multiplies.
Computational effort ~ $m^{3\ n\_orbitals.}$
$\Rightarrow$ 3 band models not feasible when using a large number of bath sites.

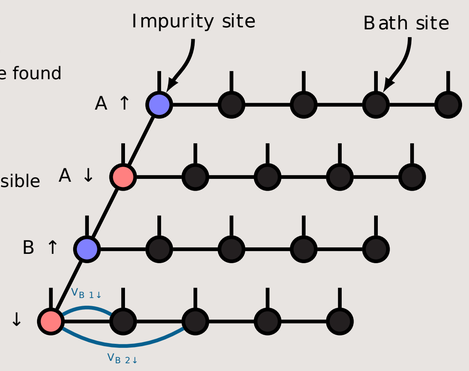
Fork Tensor Product States (FTPS) - the new way

In the AIM, the baths are not directly coupled, but interact indirectly via the impurity. Hence combining orbitals might not be the best approach.

Idea: different tensor network geometry

Cutting any bond results in two disconnected parts. Hence Schmidt decompositions can be found on any bond. Thus

- DMRG possible
- Real time evolution possible
- Easy to generalize

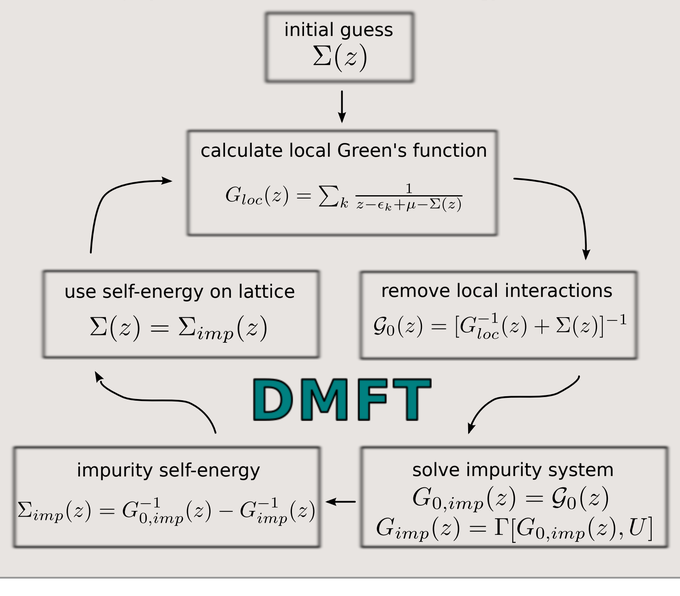


Tradeoff: Small bath tensors and a few 4-legged tensors on the impurity (expensive) instead of many big 3-legged bath tensors (MPS)
Spoiler: FTPS much better

DFT + Dynamical Mean Field Theory (DMFT)

Starting from a density functional theory (DFT) band structure we identify the correlated subspace and calculate a multi-orbital Hubbard model by e.g.: Wannier projection.

Then we employ DMFT to calculate the local self energy.



Results: $SrVO_3$

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Properties and Hamiltonian

$SrVO_3$ has a cubic crystal structure and is a weakly correlated metal with one electron in the V-d shell.

Due to the crystal field, the 5 degenerate d-states are split into two e_g and three t_{2g} states. The latter form the correlated subspace.

As interaction we choose the Kanamori Hamiltonian:

$$U \sum_m n_{m\uparrow} n_{m\downarrow} + (U-2J) \sum_{m'>m,\sigma} n_{m\sigma} n_{m'\bar{\sigma}} + (U-3J) \sum_{m'>m,\sigma} n_{m\sigma} n_{m'\sigma} + H_{SF-PH}$$

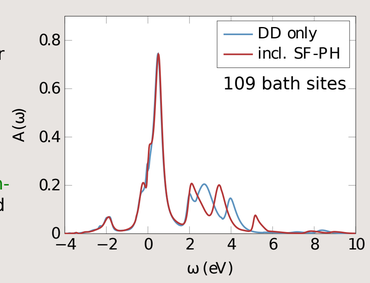
$H_{SF\text{-}PH}$ are the spin flip and pair hopping terms.

Impurity Spectral function, U=4 eV, J=0.6 eV

Typical structure: lower Hubbard band, quasi particle peak and upper Hubbard band.

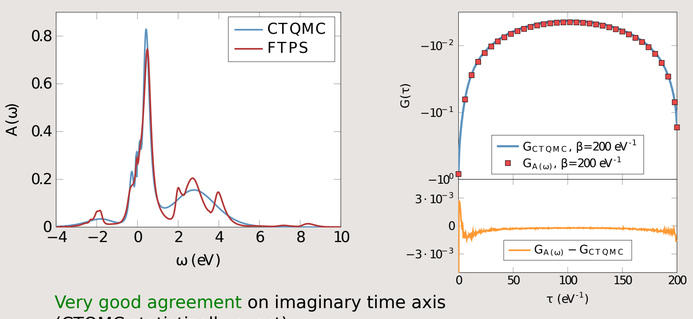
The upper Hubbard band shows a distinct 3-peak structure.

Peak position different whether spin-flip and pair-hopping terms included (incl. SF-PH) or not.



In the following we discuss results using only DD-interactions. Similar conclusions will hold for full rotational symmetry.

Comparison to CTQMC

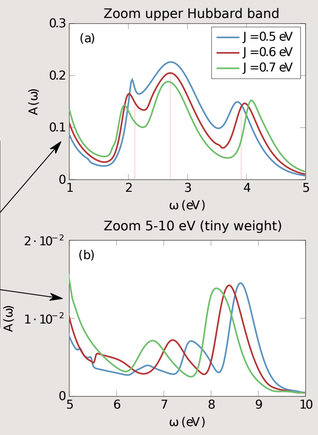


Very good agreement on imaginary time axis (CTQMC statistically exact).
3-peak structure not visible with CTQMC + MaxEnt.

Peak Structure - Atomic Multiplets

Eigen energies (difference to the ground state) and example eigen states of the DD-only Kanamori Hamiltonian.

particle sector	atomic eigen energies	state
0	ε_0	$\|0,0,0\rangle$
1	0	$\|\uparrow,0,0\rangle$
2	U - 3J+ε_0	$\|\uparrow,\uparrow,0\rangle$
	U - 2J+ε_0	$\|\uparrow,\downarrow,0\rangle$
	U +ε_0	$\|\uparrow\downarrow,0,0\rangle$
3	3U - 9J + 2ε_0	$\|\uparrow,\uparrow,\uparrow\rangle$
	3U - 7J + 2ε_0	$\|\uparrow,\uparrow,\downarrow\rangle$
	3U - 5J + 2ε_0	$\|\uparrow\downarrow,\uparrow,0\rangle$



Peak structure resolved by FTPS !

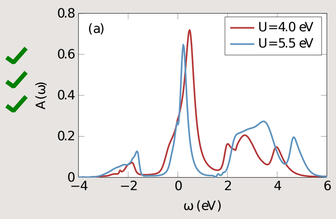
Here, it can be understood using atomic physics with effective parameters (e.g: U = 4.0 eV, U_{eff} = 6 eV and J = 0.6 eV, J_{eff} = 0.66 eV)

Coupling of bath: ground state has contributions from the N=2 particle sector of impurity.

$\Rightarrow$ even excitations into the N=3 sector visible

Atomic Physics - is this everything?

When increasing U we expect:
Quasi-particle peak gets slimmer. ✓
Lower and upper Hubbard band gain weight. ✓
Upper Hubbard band shifts (lower not). ✓

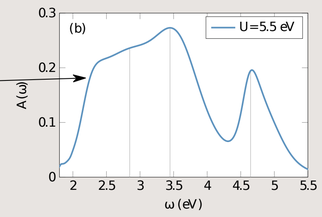


BUT: Atomic multiplet gone?

No, still there, accompanied by a feature at the inside of the Hubbard band!

$\Rightarrow$ Maybe Hubbard-side peak?

See:
Karski et al. PRB 72, 113110 (2005)
Lee et al. arXiv:1705.03910 (2017)



CPU-time for one Iteration:
FTPS: 80h
CTQMC : 32h

Results: $SrMnO_3$

Properties & Band Structure

Difference to $SrVO_3$: nominally half filled t_{2g} shell (3 electrons).
$\Rightarrow$ Mott Insulator
We present results for the paramagnetic, cubic phase.

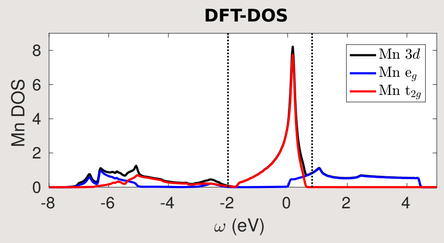
Here we are interested in the influence of the Wannier function construction on the result:

1. Oxygen hybridization: "oxygen bands" have t_{2g} / e_g weight:

$\Rightarrow$ Small energy window or large projective energy window?

2. e_g bands directly at Fermi energy:

$\Rightarrow$ Include e_g and use 5-bands?

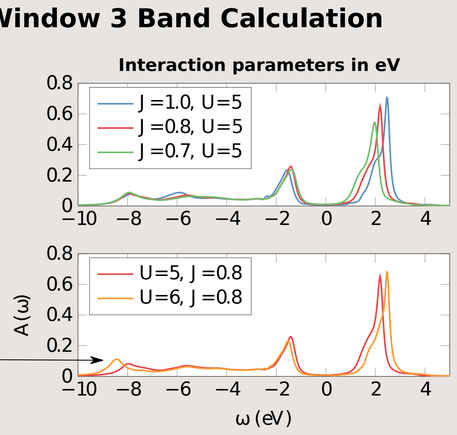


Large Energy Window 3 Band Calculation

Lower Hubbard band at -2.5 eV.
Interaction with t_{2g} weight from oxygen bands
$\Rightarrow$ "d-dp model"

Broad structure with several peaks in the occupied spectrum.

shift with U but not with J

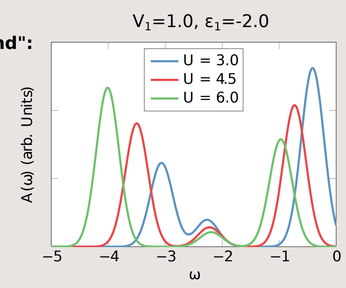


What we see here:
1. Increasing interactions opens the gap (both U and J)
2. Peak at -8.0 eV is affected by U and grows with U
3. Peak at -6.0 eV is affected by J

How we understand the "lower Hubbard band":
Toy model: Single orbital AIM with one bath site

Already this simple model shows most features present in the spectrum:

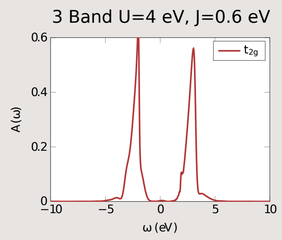
$\Rightarrow$ First peak becomes smaller with U
$\Rightarrow$ Last peak becomes larger with U
$\Rightarrow$ Last peak shifts more than first peak



CPU-time for one Iteration:
3-Band d-dp: FTPS: 240h CTQMC : 38h
5-Band d-dp: FTPS: 720h CTQMC : 500-700h

Small Energy Window Calculations

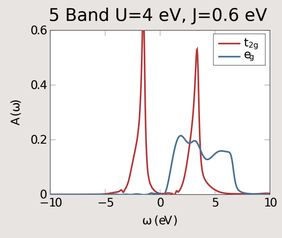
3 band calculation:
typical Mott-insulator
Lower and upper Hubbard band without multiplets
$\Rightarrow$ ground state mainly $|\uparrow,\uparrow,\uparrow\rangle + |\downarrow,\downarrow,\downarrow\rangle$
(Hund's rule and large U)



5 band calculation:
e_g orbitals nearly empty:
$\Rightarrow$ t_{2g} spectrum does not change (see 3-Band)
$\Rightarrow$ eg does (compared to DFT)
add particle into e_g: high spin or low spin:

$|\uparrow,0\rangle \otimes |\uparrow,\uparrow,\uparrow\rangle$ $|\downarrow,0\rangle \otimes |\uparrow,\uparrow,\uparrow\rangle$

$\Rightarrow$ spectrum broadens!



Large Energy Window 5 Band Calculation

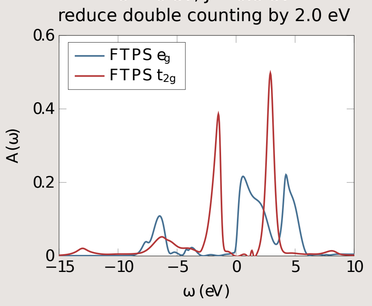
e_g bands not empty - they also posses weight in the oxygen bands.
$\Rightarrow$ Include them - perform full 5 band calculation

U = 6 eV, J = 0.8 eV
reduce double counting by 2.0 eV

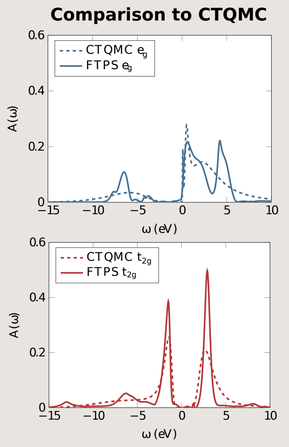
Gap determined by e_g - not t_{2g}-t_{2g} gap!

Not insulating (CTQMC and FTPS)

Majority- and minority spin splitting of e_g orbitals enhanced

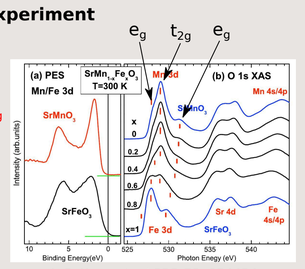


Comparison to CTQMC



Comparison to Experiment

Experimental data also shows alternating e_g-t_{2g}-e_g peak structure.



J.-S. Kang et. al PRB 78, 054434

Conclusion:
FTPS is a new, real-frequency impurity solver for material calculations that gives high resolution results for 3-band and even 5-band calculations.

ACKNOWLEDGMENTS
Austrian Science Fund (FWF):
SFB ViCoM F41 (P04 and P03)
Project P26220
START program Y746
NAWI-Graz
National Science Foundation
Grant No. NSF PHY-1125915
Vienna Sci-entific Cluster (VSC)
ITensor library (http://itensor.org/)