Realistic modeling of strongly correlated materials using LDA+DMFT(QMC): Photoemission spectra of $La_{1-x}Sr_xTiO_3$

I.A. Nekrasov, K. Held, N. Blümer, A.I. Poteryaev,
V.I. Anisimov, and D. Vollhardt

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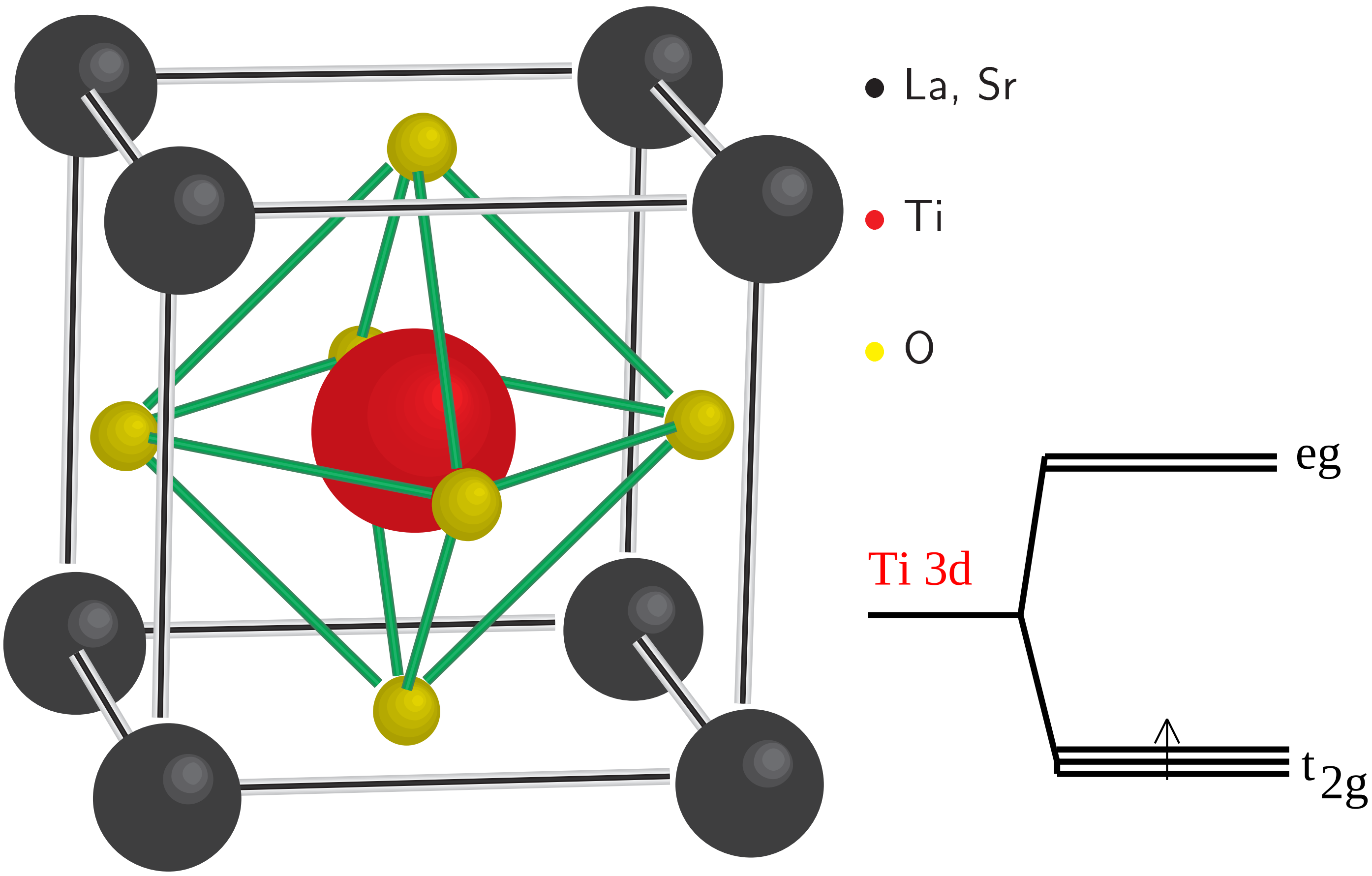
Outline

Introduction:	LDA versus model-Hamiltonian approaches the doped Mott insulator $La_{1-x}Sr_xTiO_3$
Theory:	DFT(LDA); extensions for strong correlations dynamical mean-field theory (DMFT) quantum Monte Carlo (QMC)
Results:	(partial) densities of states photoemission spectra
Conclusion/Outlook	

Introduction

	DFT/LDA		Model Hamiltonians
+	no empirical parameters: "ab initio"	+	systematic many-body investigation possible
+	well-tested code packages		
+	computationally cheap		
−	fails for strong correlations	−	choice of input parameters?
−	self interaction	−	computations highly complex
−	no systematic improvement		

$La_{1-x}Sr_xTiO_3$

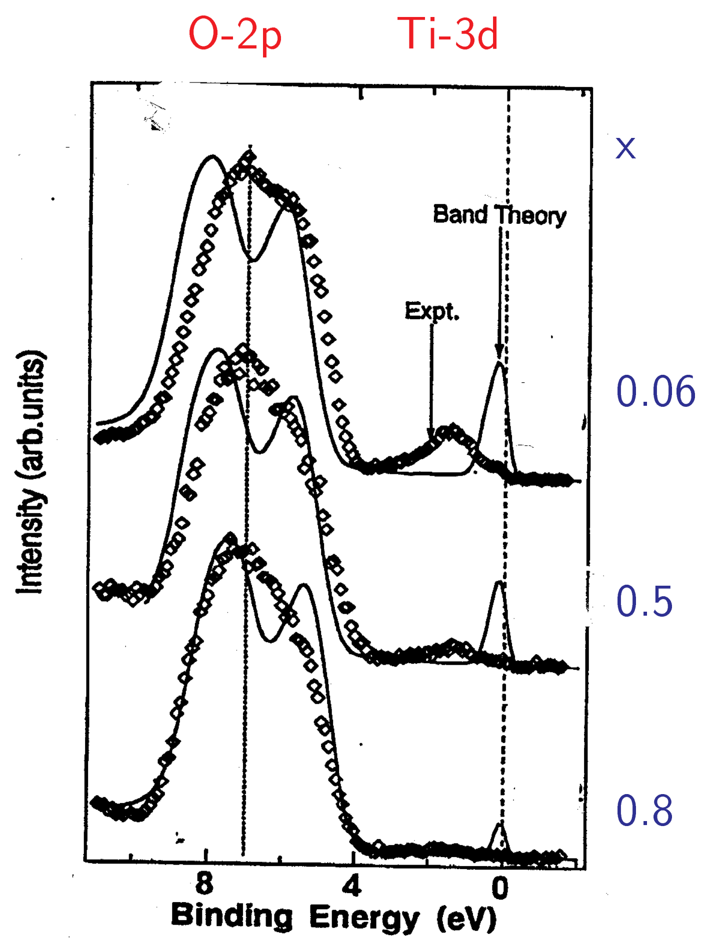


Cubic Perowskite, slight orthorombic distortion

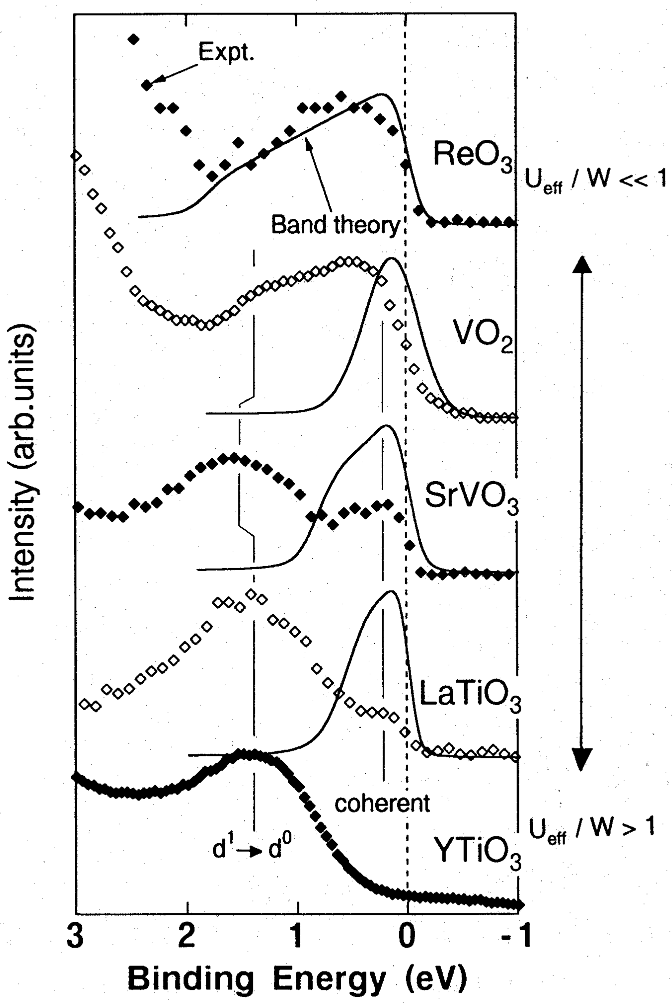
$x = 0$	$LaTiO_3$: AF insulator for $T < T_N = 125K$
$x > 0.05$	strongly correlated paramagnetic metal
$x = 1$	$SrTiO_3$: band insulator

Photoemission spectra

$La_{1-x}Sr_xTiO_3$



[Fujimori et al., PRB **46**, 9841 (1992)]



[Fujimori et al., PRL **69**, 1796 (1992)]

- LDA fails when correlations become strong
- $La_{1-x}Sr_xTiO_3$ ($x \ll 1$) is typical strongly correlated transition metal oxide

Theory

Density functional theory (DFT):

- mapping onto effective single-particle problem
- ground state method

Kohn-Sham equations:

$$\left[-\nabla^2 + v_{ext}(\mathbf{r}) + 2\int d\mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + v_{xc}(\mathbf{r},\{\rho\})\right]\varphi_i(\mathbf{r}) = \varepsilon_i \varphi_i(\mathbf{r})$$

Local (spin) density approximation, L(S)DA:
$v_{xc}(\mathbf{r},\{\rho\}) = v_{xc}(\rho_\uparrow(\mathbf{r}), \rho_\downarrow(\mathbf{r}))$,
obtained from QMC solution of homogeneous electron gas.

\+ good for simple metals
− only Stoner-type long range order

- gradient corrections
- self-interaction correction

$\Big\}$ no general improvement

LDA+U: introduces Hubbard U for localized orbitals, treatment in static mean-field theory [Anisimov, Zaanen, Andersen, PRB. **44**, 943 (1991)]

$$E = E_{LDA} + \frac{1}{2}U \sum_{m\sigma \neq m'\sigma'} n_m^\sigma n_{m'}^{\sigma'} - \frac{1}{2}UN(N-1); \quad N = \sum_{m\sigma} n_m^\sigma$$

\+ insulating, long-range ordered phases (AF, OO) can be described
\+ U can be extracted from constrained supercell LDA
− no Kondo effect, no paramagnetic insulator
$\longrightarrow$ dynamical theory needed

LDA + multi-band Hubbard Hamiltonian

1) LDA-LMTO bandstructure calculation $\rightarrow$ tight-binding model

$$H^{\mathrm{LMTO}} = \sum_{ilm,jl'm',\sigma} \left(t_{ilm,jl'm'} \hat{c}^{\dagger}_{ilm,\sigma} \hat{c}_{jl'm',\sigma} + \varepsilon_{ilm} \hat{n}_{ilm,\sigma} \delta_{ilm,jl'm'} \right)$$

$\longrightarrow E^{\mathrm{LMTO}}$: total LDA energy

2) Add local Coulomb correlations

$$H_{\mathrm{corr}} = \frac{1}{2} \sum_{ilml'm'\sigma\sigma'}' U_{il,mm'} \hat{n}_{ilm,\sigma} \hat{n}_{ilm',\sigma'} \quad + \text{ off-diagonal parts}$$

$\longrightarrow E^{\mathrm{LMTO}}_{\mathrm{corr}}(\{n_{d,f}\})$: average correlation energy

only single d-orbital: $E^{\mathrm{LMTO}}_{\mathrm{corr}} = \frac{1}{2} U n_d (n_d - 1)$

3) Subtract local Coulomb correlations included in LDA

shift localized levels

$$\varepsilon_{ilm} \longrightarrow \varepsilon^0_{ilm} = \begin{cases} \varepsilon_{ilm}, & l \neq l_d \\ \varepsilon_{ilm} - U(n_d - \frac{1}{2}), & l = l_d \end{cases}$$

$$H^{\mathrm{LMTO}}_0 := H^{\mathrm{LMTO}}\{t_{ilm,jl'm'}, \varepsilon^0_{ilm}\} \quad \text{“non-interacting part”}$$

$$H = H^{\mathrm{LMTO}}_0 + H_{\mathrm{corr}} \quad \text{“ab initio Hamiltonian”}$$

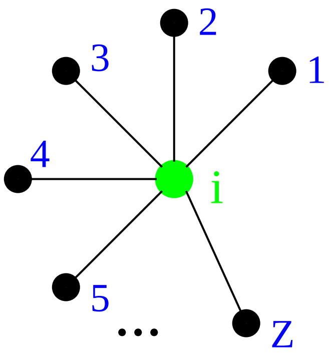
Approximation needed for treatment of H_{corr}:

DMFT: Anisimov, Poteryaev, Korotin, Anokhin, Kotliar, J. Phys. **9**, 7359 (1997)

FLEX: Lichtenstein, Katsnelson, PRB **57**, 6884 (1998)(also Hubbard-I, DMFT)

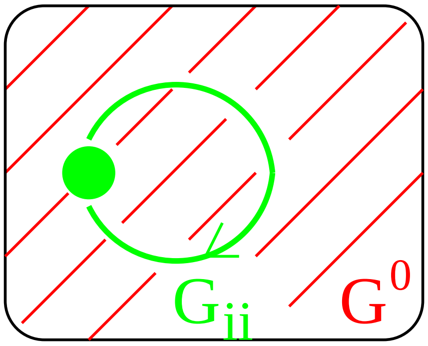
Drchal, Janiš, Kudrnovský, PRB **60**, 15664 (1999)

LDA + DMFT



$\xrightarrow{\text{DMFT}}$

exact for $Z \to \infty$



lattice problem

single impurity Anderson model
+ self consistency condition

Metzner, Vollhardt, PRL **62**, 324 (1989); Georges et al., RMP **44**, 187 (1995)

(i) self-consistency condition ($\mathbf{k}$-integrated Dyson equation)

$$G^{\sigma}_{qlm,q'l'm'}(z) = \frac{1}{v_{\mathrm{BZ}}} \int d\mathbf{k} \left[z - H^{\mathrm{LMTO}}_{0,qlm,q'l'm'}(\mathbf{k}) - \Sigma^{\sigma}_{qlm,q'l'm'}(z)\,\delta_{ql,q_d l_d} \right]^{-1}$$

q, q': atomic index in unit cell ↑ local selfenergy

For weak hybridization of d-orbital, orbital/spin-degenerate case:

$G \equiv G|_d$, Σ diagonal $\longrightarrow G(z), \Sigma(z)$ for $l = l_d$, $q = q_d$

$$G(z) = G^0(z - \Sigma(z)) = \int d\varepsilon \frac{\rho^0(\varepsilon)}{z - \Sigma(z) - \varepsilon}$$

(ii) multi-band SIAM

$$G_{\nu m\sigma} = -\langle \psi_{\nu m\sigma} \psi^*_{\nu m\sigma} \rangle_{\mathcal{A}\{\psi,\psi^*; G^{-1}+\Sigma\}}; \quad z \to \omega_\nu = \frac{(2\nu+1)\pi}{\beta}$$

Numerical solution of LDA+DMFT(X)

IPT	Anisimov, Poteryaev, Korotin, Anokhin, Kotliar, J. Phys. **9**, 7359 (1997)
NCA	Zölfl, Pruschke, Keller, Poteryaev, Nekrasov, Anisimov, PRB **61**, 12810 (2000)
QMC	Katsnelson, Lichtenstein, PRB **61**, 8906 (2000)
	Liebsch, Lichtenstein, PRL **84**, 1591 (2000)

QMC solution of impurity problem (M interacting orbitals):

- discretization $\Delta\tau$ of imaginary time $\beta = \Lambda\Delta\tau$
- Trotter decomposition
- discrete Hubbard-Stratonovich transformation
  $\rightarrow$ Λ-component Ising field for each pair of interacting orbitals/spins
  $\hat{=} 2^{M(2M-1)\Lambda}$ different non-interacting systems
- MC importance sampling over auxiliary Ising fields
- total computational cost $\propto M(2M-1)\beta^3$

Hirsch and Fye, PRL **56**, 2521 (1986)

M. Jarrell, PRL **69**, 168 (1992); George et al., RMP **44**, 187 (1995)

Rozenberg, PRB **55**, R4855 (1997); Held, Vollhardt, EPJ B **5**, 473 (1998)

Analytic continuation onto real axis using Maximum Entropy method

- ill-conditioned inversion of $G(\tau) = \int d\omega \rho(\omega) e^{-\tau\omega}/(1 + e^{-\beta\omega})$
- minimize χ^2 and entropy function
- can be biased by choice of default model

Jarrell, Gubernatis, Physics Reports **269**, 133 (1996)

Results

LDA-LMTO calculation for $LaTiO_3$

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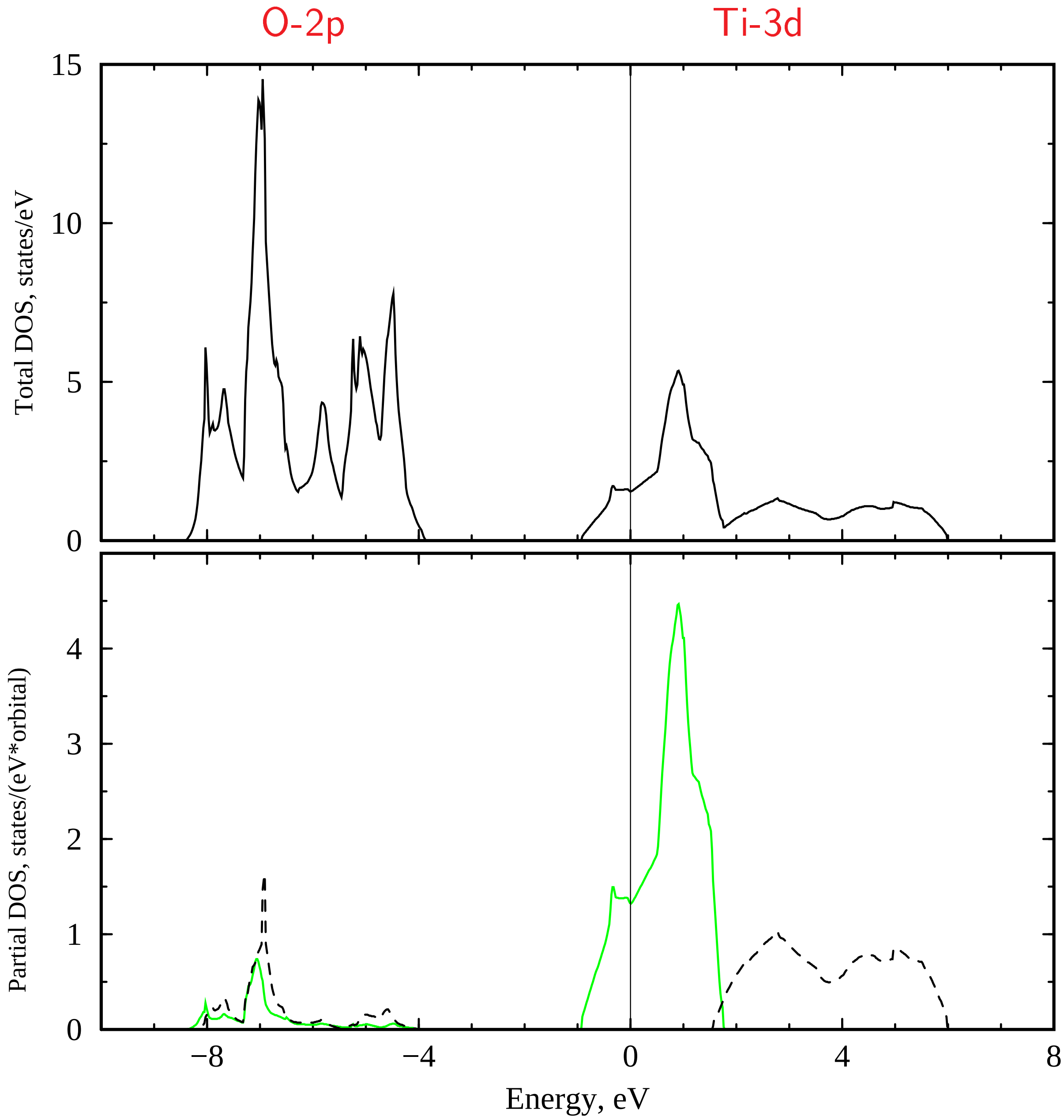

Upper figure: total density of states;
lower figure: partial t_{2g} (—) and e_g (- -) densities of states.

Orthorombic structure approximated by cubic structure with same volume
$\longrightarrow$ slight overestimation of effective bandwidth

DMFT calculation for $La_{1-x}Sr_xTiO_3$

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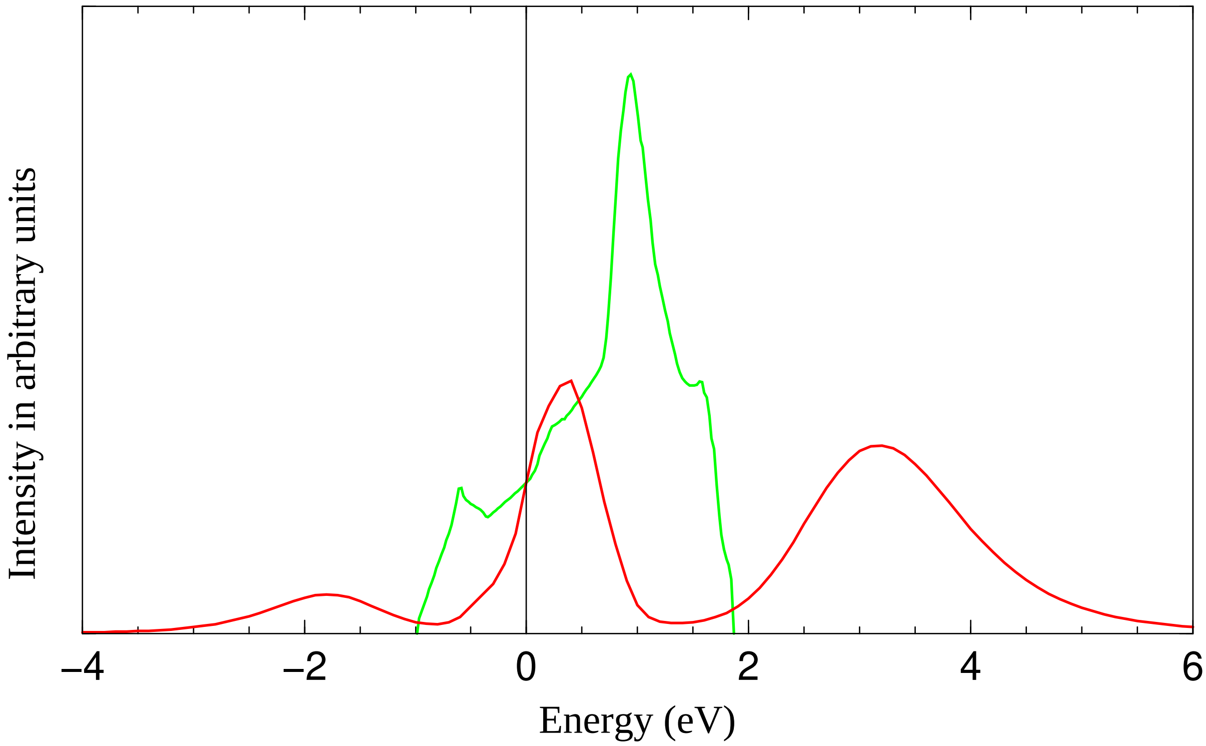
Partial t_{2g} densities of states



LDA (—) LDA+DMFT(QMC) (—)

Parameters for DMFT:
$x = 0.06$, $U = 4.0$ eV, $T = 0.1$ eV~ 1000K, 3 degenerate bands

Comparison of different DMFT methods

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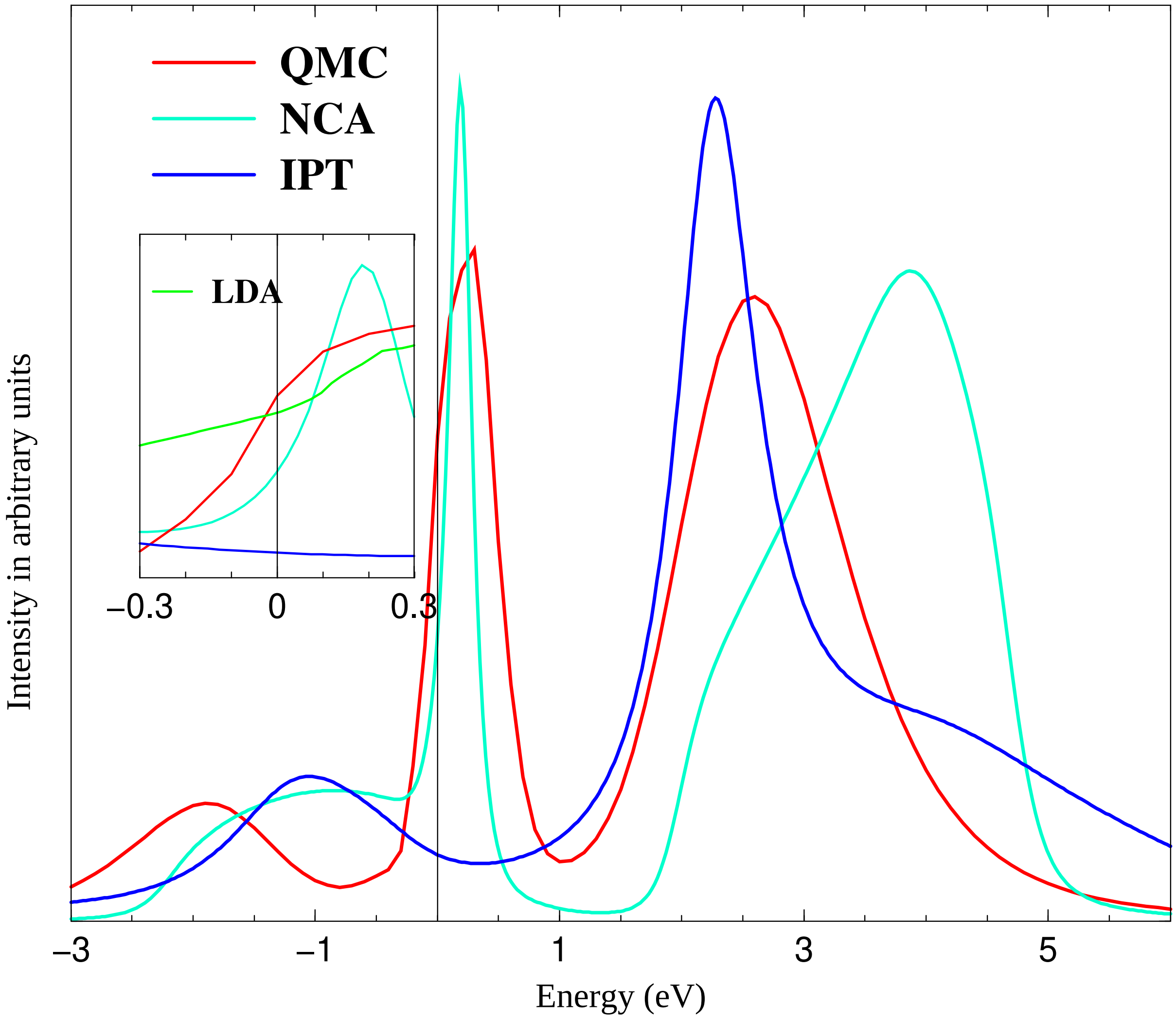
Spectral densities of $La_{1-x}Sr_xTiO_3$ as calculated by LDA+DMFT



QMC and approximations IPT and NCA at $x = 0.06$, $T = 0.1$ eV.
Inset: Behavior at Fermi level including LDA DOS ($x = 0$, $T = 0$).

Comparison with experiment

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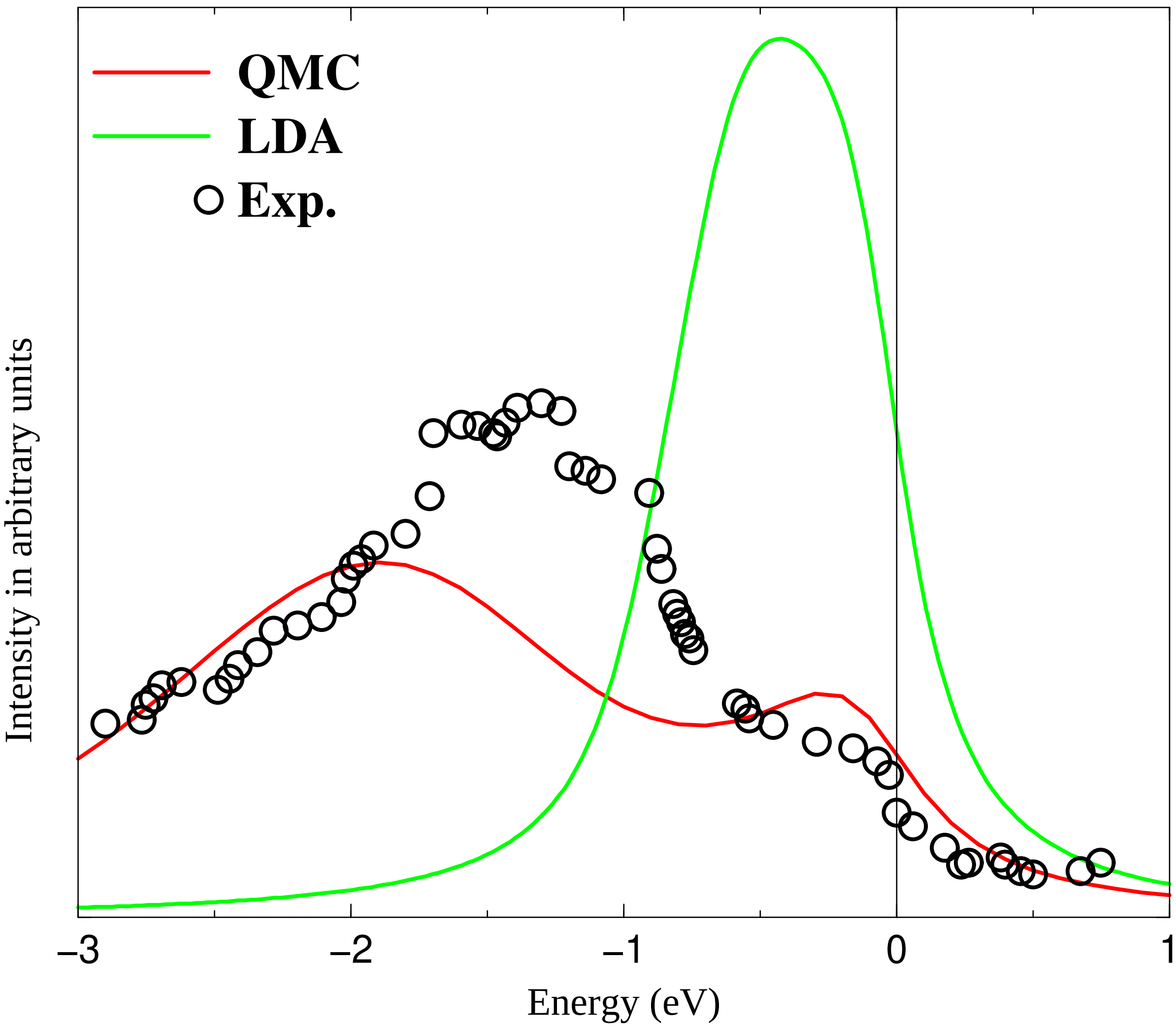

Comparison of the experimental photoemission spectrum ($T = 80$ K, resolution $\Delta\varepsilon = 0.3$ eV) [Fujimori et al., PRL **69**, 1796 (1992)], the LDA result ($T = 0$), and the LDA+DMFT(QMC) calculation ($T = 1000$ K) for $LaTiO_3$ with 6% hole doping.

Conclusion

LDA + DMFT(QMC) reproduces

- qualitative picture of spectral weight transfer from qp-band to lower (and upper) Hubbard band
- position of lower Hubbard band
- relative intensities between qp-band and lower Hubbard band
- narrowing of qp-band

big improvement over LDA

substantial deviations from NCA, IPT

Outlook

- check influence of Hund's rule couplings
- use $\mathbf{k}$-dependent self-consistency equation
- other materials
- full self-consistency: QMC output $\longrightarrow$ LDA

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