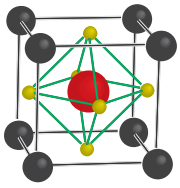
Calculation of photoemission spectra of the doped Mott insulator $La_{1-x}Sr_xTiO_3$ using LDA+DMFT(QMC)

N. Blümer[1,2], I.A. Nekrasov[3], K. Held[2,4], A.I. Poteryaev[3], V.I. Anisimov[3], and D. Vollhardt[2]

[1]Institut für Physik, KOMET 337, Universität Mainz; [2] Theoretische Physik III, Universität Augsburg; [3] Institute of Metal Physics, Russian Academy of Sciences, Yekaterinburg; [4] Physics Departement, Princeton University

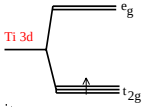
Introduction

Structure



$La_{1-x}Sr_xTiO_3$

- La, Sr
- Ti
- O

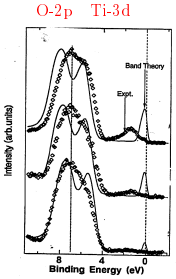


Cubic Perowskite
slight orthorombic distortion

Effect of doping

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

photoemission spectra (PES)



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

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

Theory

Conventional approaches

DFT/LDA:	+	material specific: "ab initio"
	−	fails for strong correlations
	+	fast code packages
model Hamiltonians:	−	input parameters unknown
	+	systematic many-body approach
	−	computationally expensive

Density functional theory (Hohenberg-Kohn): $E_0 = E_0[\rho]$
Levy: $E[\rho] = \inf_\varphi \left\{ \langle\varphi|\hat{H}|\varphi\rangle \;\middle|\; \rho(\varphi,\mathbf{r}) = \rho(\mathbf{r}) \right\}$
$E[\rho] = E_{\rm kin}[\rho] + E_{\rm ion}[\rho] + E_{\rm Hartree}[\rho] + E_{\rm xc}[\rho]$
local density approximation: $E_{xc}[\rho] \to \int d^3r\, E_{xc}^{\rm jellium}(\rho(\mathbf{r}))$

Extensions of LDA for correlated systems

use Kohn-Sham Lagrange parameters as single-particle energies and localized basis (linear muffin tin orbitals, LMTO)
$\longrightarrow$ tight-binding Hamiltonian

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

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

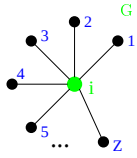
here: many-body approach

$$\hat{H} = \hat{H}^{\rm LMTO} + \frac{1}{2} \sum_{m\sigma \neq m'\sigma'} U_{mm'}\, \hat{n}^\sigma_m \hat{n}^{\sigma'}_{m'} - \frac{1}{2} U n_d (n_d - 1)$$

Approximation needed for treatment of $\hat{H}_{\rm corr}$:

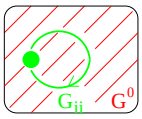
DMFT:	Anisimov et. al., J. Phys. **9**, 7359 (1997)
FLEX:	Lichtenstein, Katsnelson, PRB **57**, 6884 (1998)
	Drchal, Janiš, Kudrnovský, PRB **60**, 15664 (1999)

Dynamical mean-field theory (DMFT)



DMFT $\longrightarrow$ 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)

LDA+DMFT

(i) self-consistency condition (**k**-integrated Dyson equation)

$$G^\sigma_{qlm,q'l'm'}(z) = \frac{1}{v_{\rm BZ}} \int d\mathbf{k} \Big[z - H^{\rm LMTO}_{0,qlm,q'l'm'}(\mathbf{k}) - \delta_{ql,q_dl_d} \Sigma^\sigma_{qlm,q'l'm'}(z) \Big]^{-1}$$

q, q': atomic index in unit cell $\quad\uparrow$ local selfenergy

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

$$G \equiv G|_d,\ \Sigma \text{ diagonal} \longrightarrow G(z), \Sigma(z) \quad \text{for } l = l_d,\ q = q_d$$

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

(ii) multi-band SIAM

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

Numerical solution of LDA+DMFT(X)

IPT	Anisimov et. al. J. Phys. **9**, 7359 (1997)
NCA	Zölfl et. al. PRB **61**, 12810 (2000)
QMC	Katsnelson, Lichtenstein, PRB **61**, 8906 (2000)

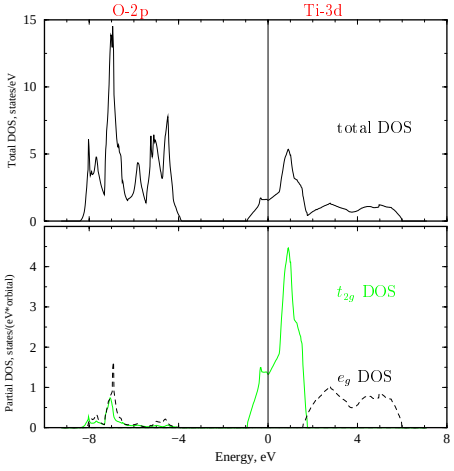
QMC solution of impurity problem (M interacting orbitals):

- discretization $\Delta\tau$ of imaginary time $\beta = \Lambda\Delta\tau$ ($\beta = 1/T$)
- discrete Hubbard-Stratonovich transformation $\to 2^{M(2M-1)\Lambda}$ different non-interacting systems
- MC importance sampling; comp. cost $\propto M(2M-1)\beta^3$

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

Results: LDA

LDA-LMTO(ASA) calculation for $LaTiO_3$

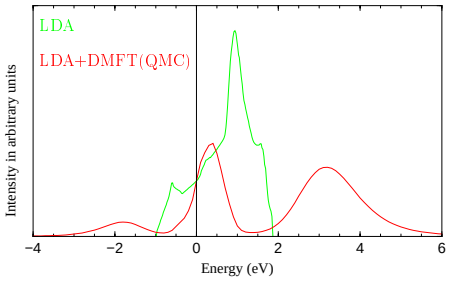


Orthorhombic structure approximated by cubic structure with same volume
$\longrightarrow$ slight overestimation of effective bandwidth

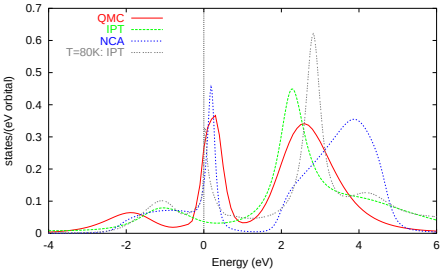
constrained LDA $\longrightarrow U_d \approx 3 - 5$ eV

Results: LDA+DMFT(QMC)

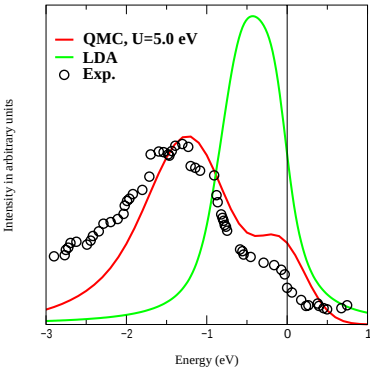
DMFT calculation for $La_{1-x}Sr_xTiO_3$: partial t_{2g} DOS
$x = 0.06$, $U = 4.0$ eV, $T = 0.1$ eV ~ 1000K, 3 degenerate bands



Comparison to approximate DMFT methods



Comparison with experiment: photoemission spectra (PES)



Experiment: Fujimori et al., PRL **69**, 1796 (1992)
Theory: DOS multiplied with Fermi step function; broadening 0.3 eV

Conclusion / Outlook

- LDA + DMFT(QMC) yields qualitatively correct picture
- significant improvement over LDA
- QMC solution of DMFT superior to NCA, IPT
- other experiments: XAS, optical conductivity, magnetic susceptibility, heat capacity
- include more orbitals, Hund's rule couplings
- other materials: V_2O_3 (see Held et al., cond-mat/0011518)

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I.A. Nekrasov, K. Held, N. Blümer, A.I. Poteryaev, V.I. Anisimov, and D. Vollhardt, Eur. Phys. J. B **18**, 55 (2000)