DFT+DMFT to Correlated Electronic Structures: Recent Developments and Applications to Iron-based Superconductors

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(Some unpublished results were removed from the slides. Interested readers can contact the author via email yinzping at physics.rutgers.edu for details.)

Electronic Structures 2013, Williamsburg, VA, June 11-14, 2013
Acknowledgements

• Gabriel Kotliar, Kristjan Haule, Warren Pickett
• Hyowon Park, Jan Tomczak, and other group members (Rutgers)
• Girsh Blumberg (Rutgers)
• Pengcheng Dai (UTK)
• Meigan Aronson (Stony Brook, BNL)
• NSF and AFOSR for support
Outline

• DMFT and DFT+DMFT in a nutshell

• Features of our implementation of DFT+DMFT

• Applications of DFT+DMFT to FeSCs
  Spectroscopy
  Susceptibility
  Superconductivity
  Resonant Raman, Thermoelectric

• Summary
Dynamic Mean Field Theory (DMFT) in a nutshell

Weiss mean field theory for spin systems
Exact in the limit of large $z$

$$\sum_{ij} J_{ij} S_i S_j$$

Dynamical mean field theory (DMFT) for the electronic problem
exact in the limit of large $z$

$$Z = \int \mathcal{D}[\psi^\dagger \psi] e^{-\sum_i S_{atom}(i) - \sum_{ij} \int d\tau \psi_i^\dagger(\tau) \hat{H}_{ij} \psi_j(\tau)}$$

Classical problem of spin in a magnetic field

Problem of a quantum impurity (atom in a fermionic band)

Space fluctuations are ignored, time fluctuations are treated **exactly**

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DFT+DMFT

Impurity \[ \frac{1}{\omega - E_{imp} - \Sigma - \Delta} = \sum_k P_k[(\omega + \mu - H_k^{\text{DFT}} - E_k \Sigma)^{-1}] \]

Lattice

Charge self-consistent implementation, avoids construction of low energy models

Features

- Valence histogram, self-energy, Green’s function
- ARPES
- Optical conductivity
- Non-collinear magnetism with spin-orbit coupling (non-perturbative)
- Thermoelectric power coefficient using proper dipole transition matrix element (unlike Boltztrap and BoltzWann: Peierls appr., no phase factors)
- Local and momentum dependent spin and charge susceptibility including two-particle vertex corrections
- Superconducting gap symmetry and coupling strength including vertex corrections (available soon)
- Resonant Raman spectra (in progress)
The iron-based superconductors

First discovery in 2008: LaFeAsO$_{1-x}$F$_x$, H. Hosono, JACS 130, 3296 (2/13/2008).
The diversity of FeSC’s


All FeSCs share the same FePn layer, but there are large variations among them.

Mass enhancement in the PM phase.

Magnetic moment in the ordered phases.

Can DFT+DMFT account for these variations without tuning U and J?

Moments by DFT are around 2 $\mu_B$, overestimated by a factor of two (ZPY et al, PRL 101, 047001 (2008)).

Moments by DFT+DMFT are in good agreement with experiments.

Theory

Experimental moment ($\mu_B$):
- FeTe: 2.03, W. Bao et al., PRL 102, 247001 (2009).
- NaFeAs: 0.31, L. Ma et al., PRB 83, 132501 (2011).
- Ba122: 0.87, Q. Huang et al., PRL 101, 257003 (2008).
- LaFeAsO: 0.82, H.-F. Li et al., PRB 82, 064409 (2010).
- CaFeAsF: 0.49, Y. Xiao et al., PRB 79, 060504(R) (2009).
- SrFeAsF: 0.58, Y. Xiao et al., PRB 81, 094523 (2010).
- Sr122: 0.94, J. Zhao et al., PRB 78, 140504(R) (2008).
- Ca122: 0.80, A. I. Goldman et al., PRB 78, 100506(R) (2008).
DFT+DMFT accounts for the variations in all families without tuning U and J!

Self-energy: Fractional power-law behavior in some FeSC’s

\[ \sigma_1(\omega) \sim \omega^{-\alpha} \]

\[ \Sigma''(\omega) \propto -\omega^\alpha \]

\( \alpha \) is orbital and material dependent, not necessarily 1/2.

ZPY et al., PRB 86, 195141 (2012).
T-dependence: Coherence-incoherence crossover

Very low coherence temperature

Theory: ZPY et al., PRB 86, 195141 (2012).

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Optical conductivity: BaFe$_2$As$_2$

- PM state $\rightarrow$ broader Drude peak
- Correct plasma $\omega_p$:
  - DMFT $\sim$ 1.6eV
  - Exp $\sim$ 1.6eV
  - LDA $\sim$ 2.6eV

- SDW state more coherent $\rightarrow$ sharper Drude peak
- Above SDW gap: 3 peaks

- Magnetic moment:
  - Exp. : $0.87 \mu_B$
  - DMFT: $0.9 \mu_B$
  - LSDA: $2.0 \mu_B$

- Good agreement at low energy in both paramagnetic and magnetic phases

Optical in-plane anisotropy predicted by DFT+DMFT

BaFe$_2$As$_2$ in the SDW phase

DMFT prediction:

Experiment:
Susceptibility with vertex correction

Bethe-Salpeter equation:

\[
\Gamma_{\text{loc}}^{\text{irr}}_{\alpha_1 \sigma_1, \alpha_2 \sigma_2, \alpha_3 \sigma_3, \alpha_4 \sigma_4}(i \nu, i \nu')_{i \omega} = \frac{1}{T} \left[ \left( \chi_{\text{loc}}^0 \right)^{-1}_{i \omega} - \chi_{\text{loc}}^{-1} \right].
\]

One particle Green’s function

\[
\chi^{\alpha_1 \sigma_1, \alpha_2 \sigma_2, \alpha_3 \sigma_3, \alpha_4 \sigma_4}_{\alpha_3 \sigma_3, \alpha_4 \sigma_4}(i \nu, i \nu')_{q, i \omega} = \left[ \left( \chi_{\text{loc}}^0 \right)^{-1}_{q, i \omega} - T \Gamma_{\text{loc}}^{\text{irr}} \right]^{-1}.
\]

\[
\chi(q, i \omega) = T \sum_{i \nu, i \nu'} \sum_{\alpha_1 \sigma_1, \alpha_2 \sigma_2, \alpha_3 \sigma_3, \alpha_4 \sigma_4} \mu_{\alpha_1 \sigma_1} \mu_{\alpha_2 \sigma_2} \chi^{\alpha_1 \sigma_1, \alpha_2 \sigma_2, \alpha_3 \sigma_3, \alpha_4 \sigma_4}_{\alpha_3 \sigma_3, \alpha_4 \sigma_4}(i \nu, i \nu')_{q, i \omega}.
\]

H. Park et al., PRL 107, 137007 (2011).

Easily replaced with charge or orbital to obtain charge or orbital susceptibility

Sampled by ctqmc
Spin susceptibility in iron pnictides


Experiments: L. W. Harriger et al, PRB 84, 054544 (2011)

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Superconductivity

Superconductivity

$$\Gamma^{\text{irr},p-p,(s)}_{\alpha_2,\alpha_4} (k, iv, k', iv') = \Gamma^{f-\text{irr},(s)}_{\alpha_2,\alpha_4} (iv, iv') + \left[ \frac{3}{2} \tilde{\Gamma}^{p-h,(m)}_{\alpha_1,\alpha_3} - \frac{1}{2} \tilde{\Gamma}^{p-h,(d)}_{\alpha_2,\alpha_4} (iv, -iv')_{k'=-k, iv'=-iv} \right] + \frac{1}{2} \left[ \frac{3}{2} \tilde{\Gamma}^{p-h,(m)}_{\alpha_1,\alpha_3} - \frac{1}{2} \tilde{\Gamma}^{p-h,(d)}_{\alpha_1,\alpha_2} (iv, iv')_{k'=-k, iv'=-iv} \right]$$

(4.47)

$$\chi^{p-p} = \chi^{0,p-p} \cdot \left[ 1 + \Gamma^{\text{irr},p-p,(s)} \cdot \chi^{0,p-p} \right]^{-1}$$

$$-\frac{T}{N_k} \sum_{k', iv'} \sum_{\alpha_2,\alpha_4,\alpha_5,\alpha_6} \Gamma^{\text{irr},p-p,(s)}_{\alpha_2,\alpha_4} (k, iv, k', iv') \cdot \chi^{0,p-p}_{\alpha_5,\alpha_6} (k', iv') \cdot \phi^\lambda_{\alpha_5,\alpha_6} (k', iv') = \lambda \cdot \phi^\lambda_{\alpha_1,\alpha_3} (k, iv)$$

Leading eigenvalue approaches 1 gives $T_c$

The corresponding eigenfunction gives the pairing symmetry

Superconductivity: pairing symmetry

Orbital space

Ground State

First Excited State

Band space: on FS

S+-

d-wave
Raman susceptibility

Experimental data from V. K. Thorsmolle and G. Blumberg (Rutgers University)
Thermoelectric Power: FeSi

Summary

• DFT+DMFT is shown to capture quantitatively many experimental observables in the correlated iron-based superconductors.

• DFT+DMFT is a promising tool to study correlated materials and can be used, in collaborations with experiments, to rationally design novel correlated functional materials with desirable properties such as high temperature superconductivity and large thermoelectric power.