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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(Some unpublished results were removed from the slides.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.)

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Acknowledgements

- Gabriel Kotliar, Kristjan Haule, Warren Pickett
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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

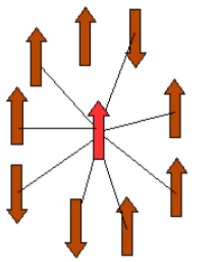
- Summary

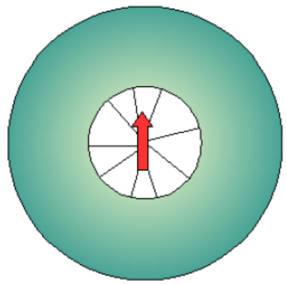
Dynamic Mean Field Theory (DMFT) in a nutshell

(G. Kotliar, S. Savrasov, K.Haule, V. Oudovenko, O. Parcollet, and C. Marianetti, RMP 78, 865-951 2006).

Weiss mean field theory for spin systems

Exact in the limit of large z





$$\sum_{ij} J_{ij}\mathbf{S}_i\mathbf{S}_j$$

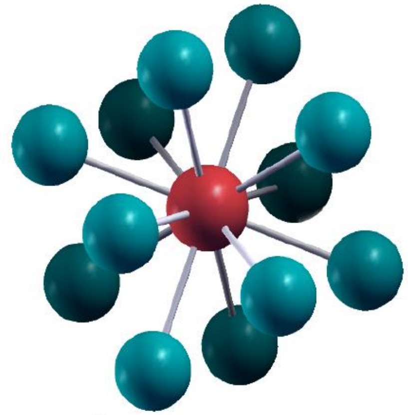
$$\sum_{i} \mathbf{B}_i\mathbf{S}_i$$

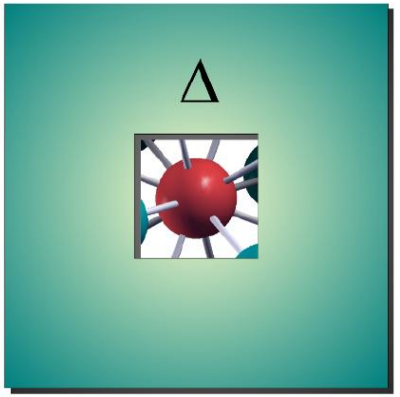
$$\mathbf{B}_i = \sum_{j\neq i} J_{ij}\langle\mathbf{S}_j\rangle$$

Classical problem of spin in a magnetic field

Dynamical mean field theory (DMFT) for the electronic problem

exact in the limit of large z





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

$$Z = \int \mathcal{D}[\psi^\dagger\psi] e^{-\sum_i S_{atom}(i) - \sum_{i}\int d\tau d\tau' \psi_i^\dagger(\tau)\hat{\Delta}(\tau,\tau')\psi_i(\tau')}$$

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

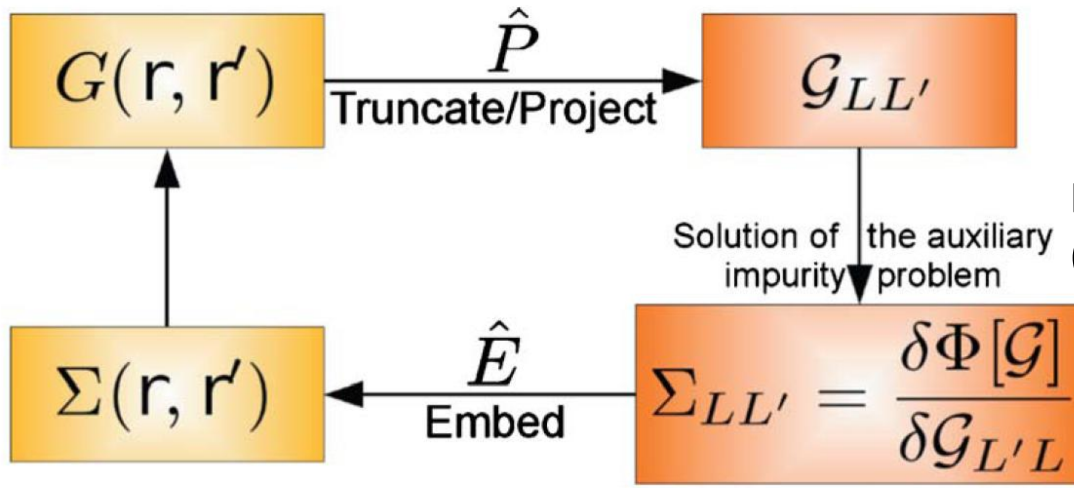
Space fluctuations are ignored, time fluctuations are treated **exactly**

DFT+DMFT

Impurity Lattice

$$\frac{1}{\omega - E_{imp} - \Sigma - \Delta} = \sum_{\mathbf{k}} P_{\mathbf{k}}[(\omega + \mu - H_{\mathbf{k}}^{\mathrm{DFT}} - E_{\mathbf{k}}\Sigma)^{-1}]$$

DFT (WIEN2k)



DMFT (CTQMC)

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

DMFT-SCC:

$$\mathcal{G} = \hat{P}\left(\omega + \mu + \nabla^2 - V_{\mathrm{KS}} - \hat{E}\frac{\delta\Phi[\mathcal{G}]}{\delta\mathcal{G}}\right)^{-1}$$

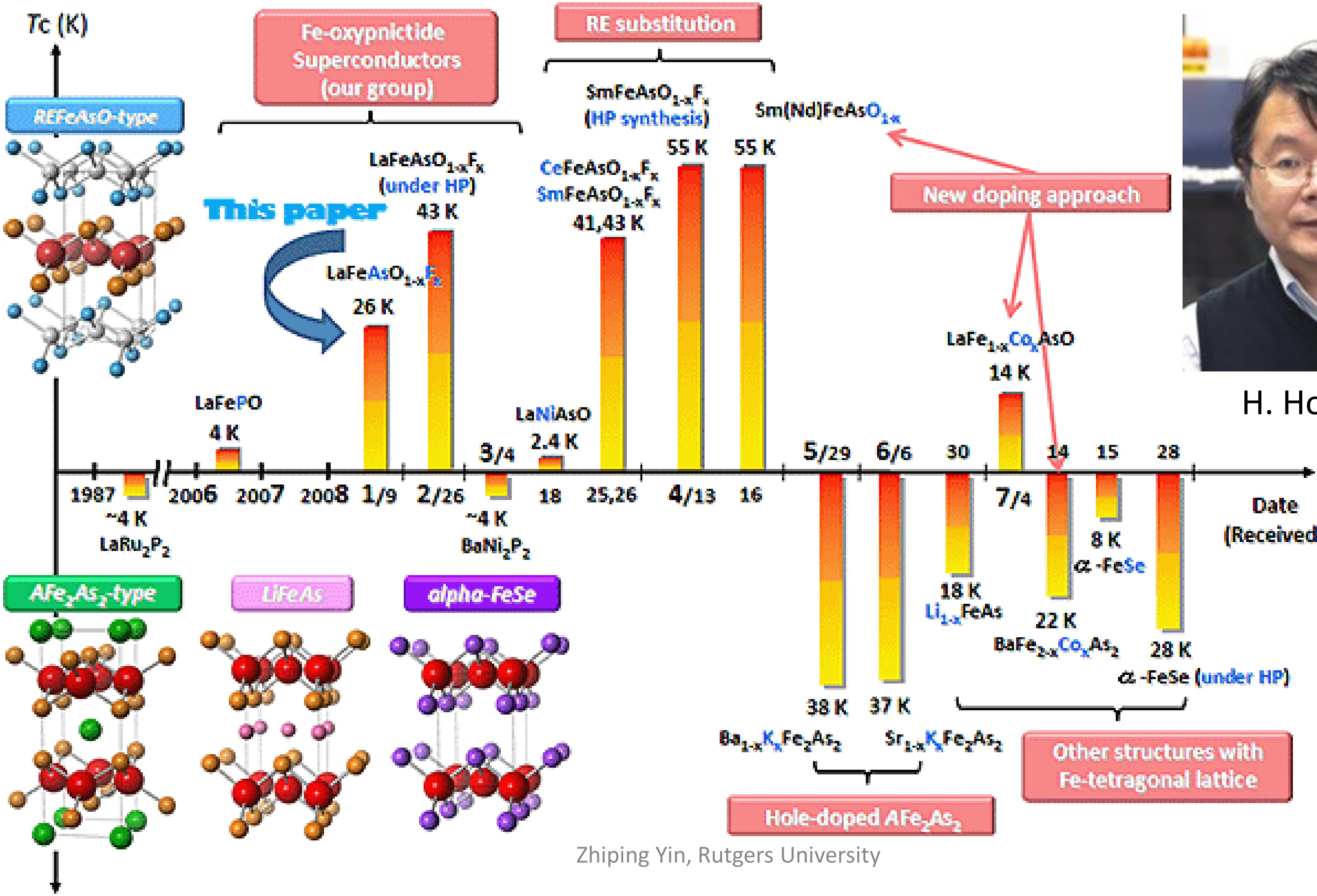
K. Haule et al, Phys. Rev. B 81, 195107 (2010).

Features

- Valence histogram, self-energy, Green's function
- ARPES
- Optical conductivity
- Non-colinear 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)

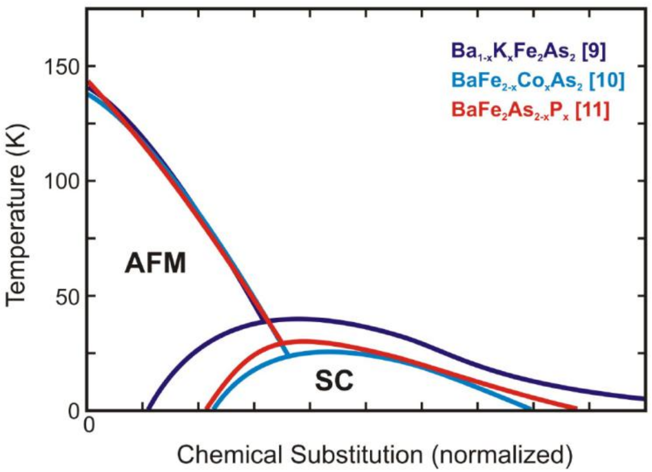
The iron-based superconductors

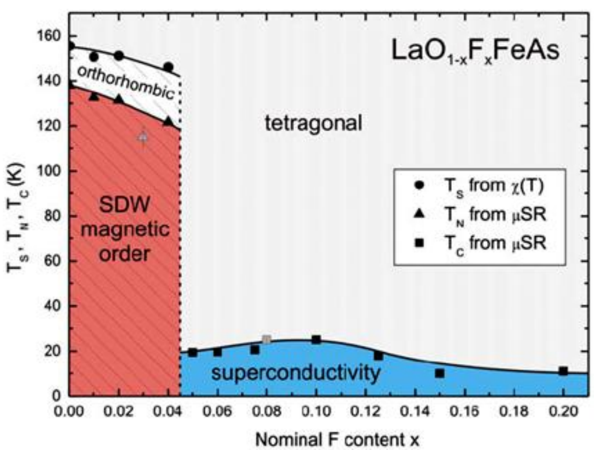
First discovery in 2008: $LaFeAsO_{1-x}F_x$, H. Hosono, JACS 130, 3296 (2/13/2008).



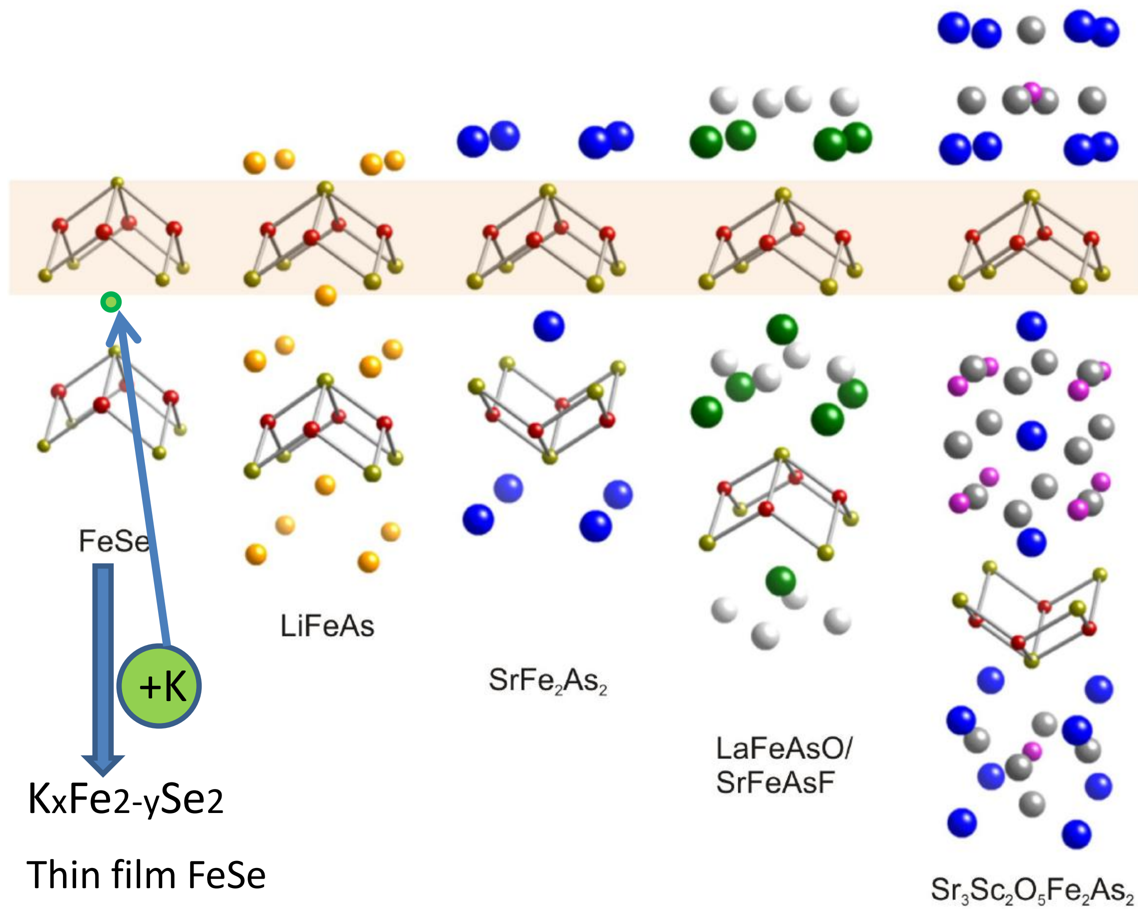
H. Hosono

The diversity of FeSC's





M. R. Norman, Physics **1**, 21 (2008).



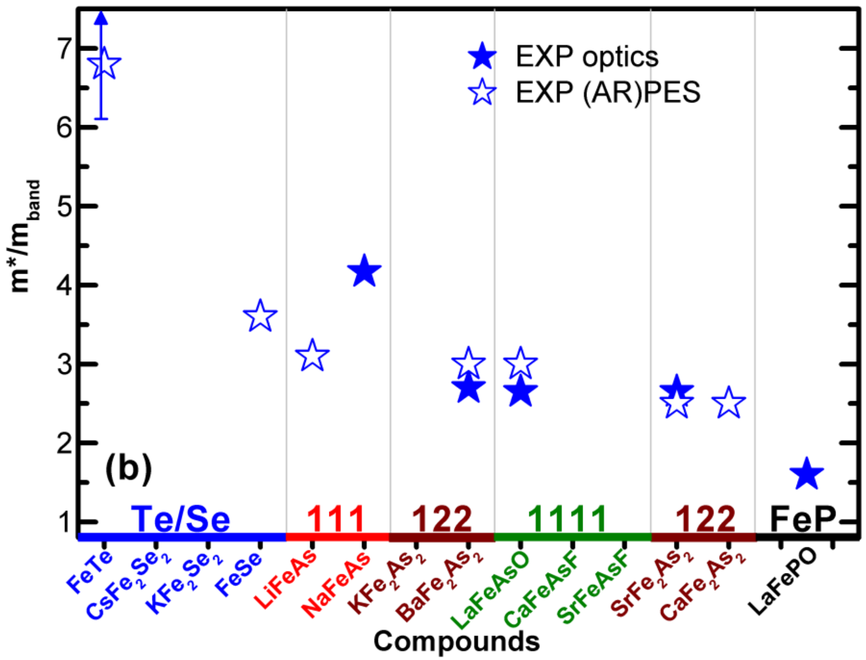
$K_xFe_{2-y}Se_2$

Thin film FeSe

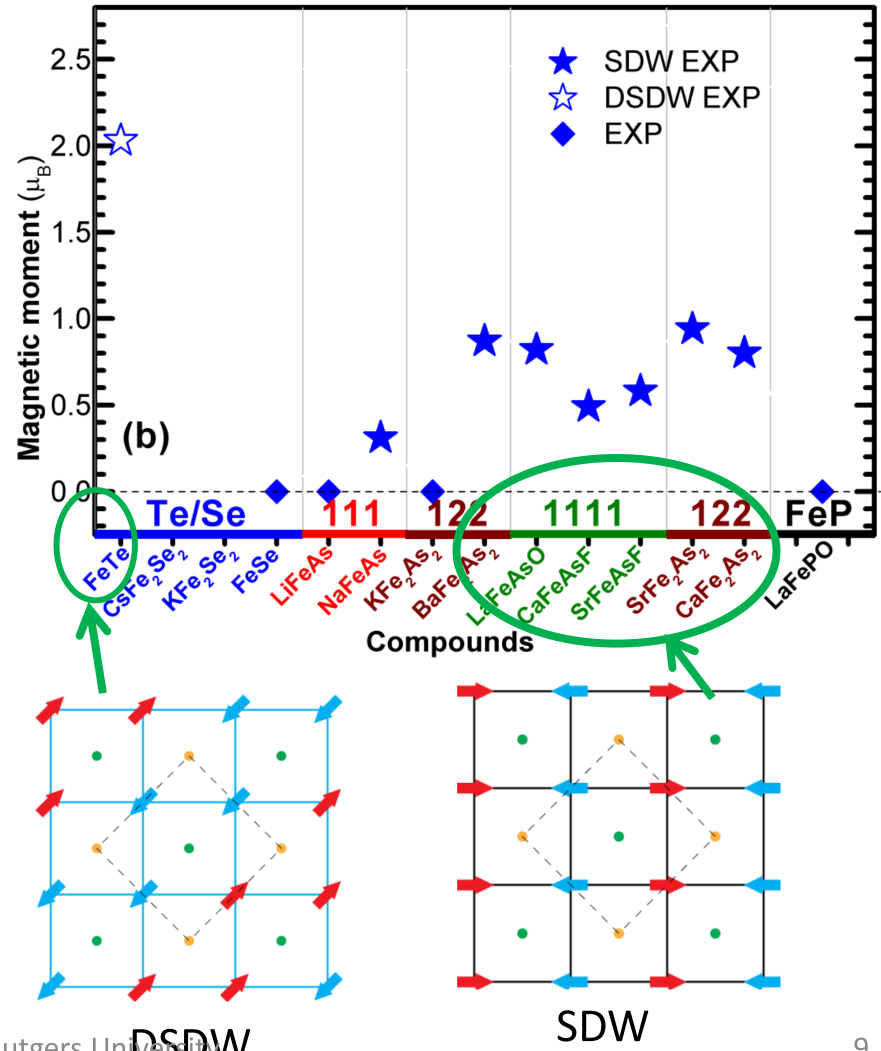
J. Paglione and R L. Greene, Nature Physics 6, 645-658 (2010).

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?

ZPY, K. Haule and G. Kotliar, Nature Materials **10**, 932 (2011).

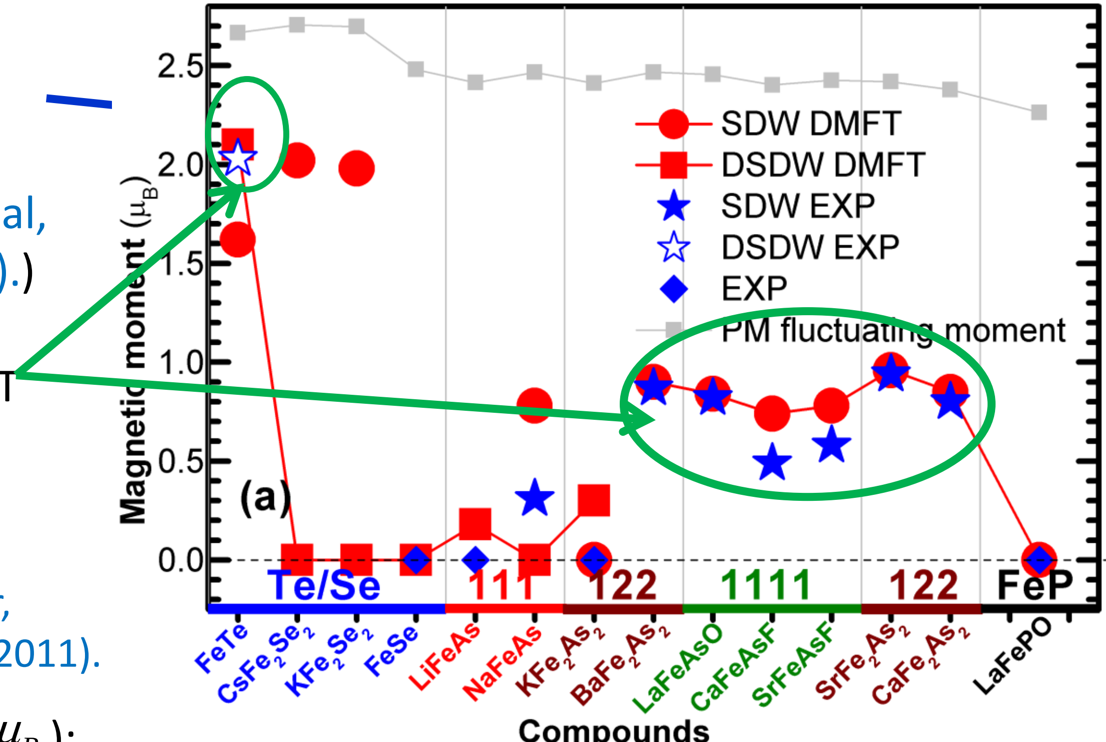
Moments and Magnetism

Moments by DFT are around 2 μ_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

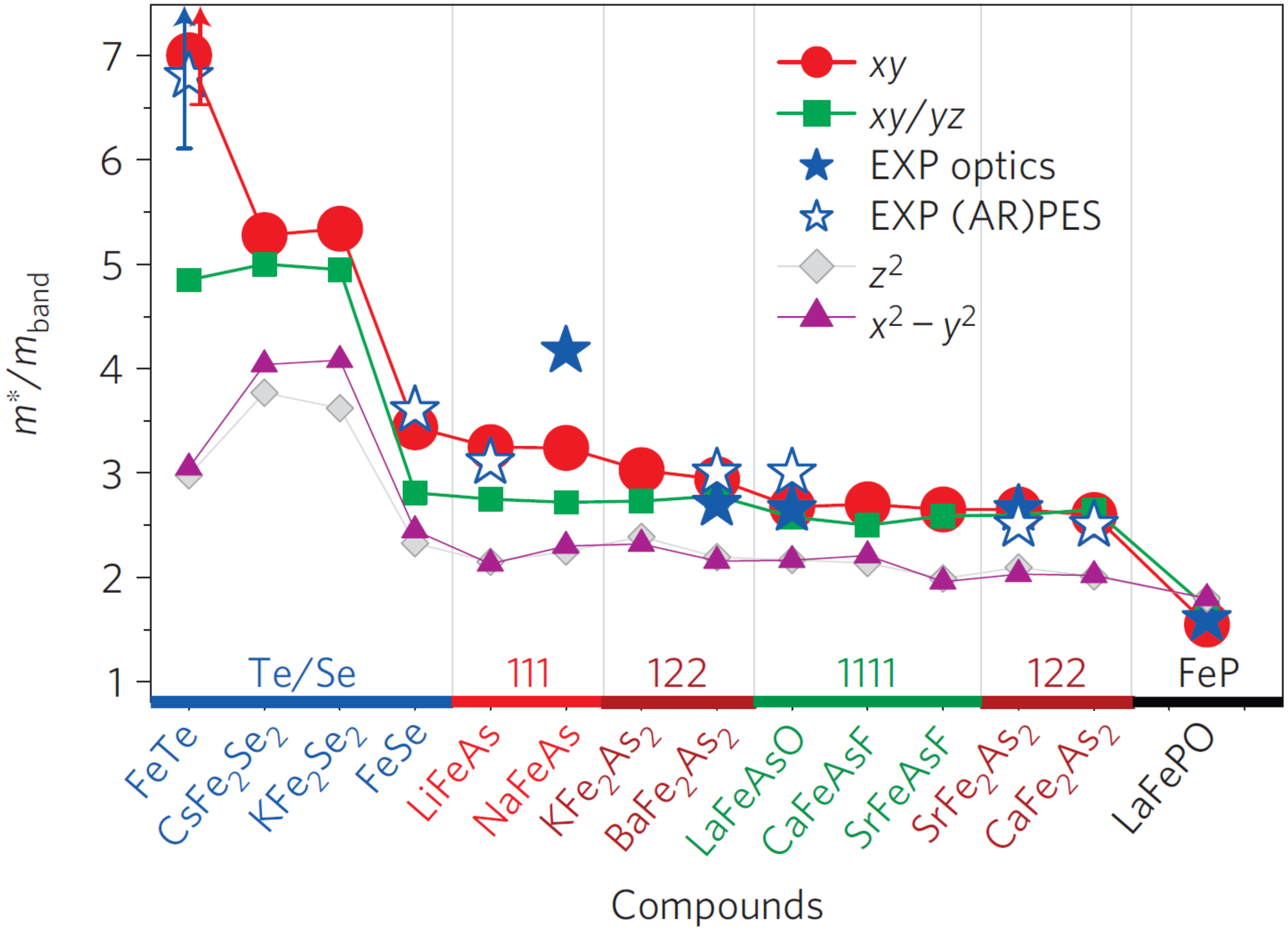
ZPY, K. Haule and G. Kotliar, Nature Materials **10**, 932 (2011).



Experimental moment (μ_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).

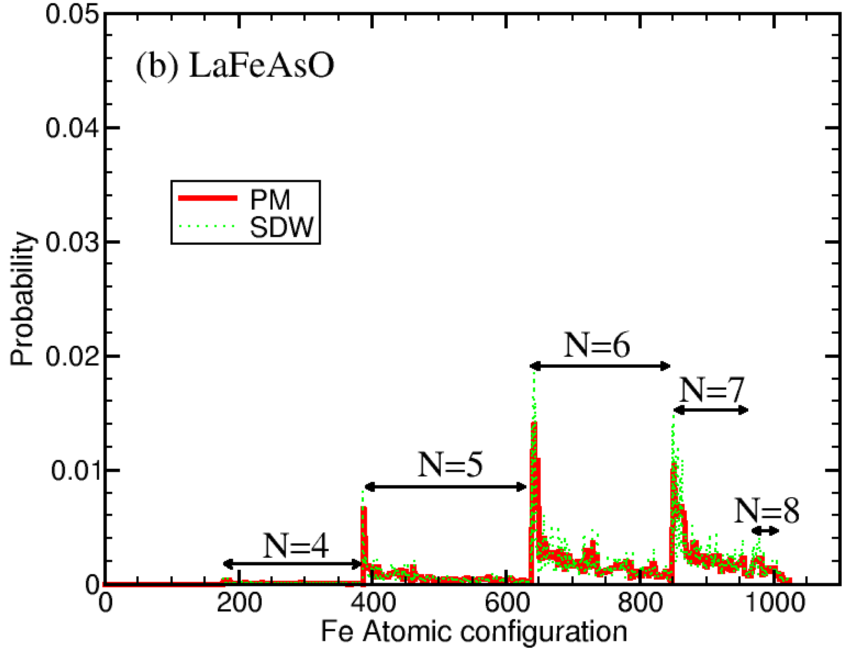
Mass enhancement

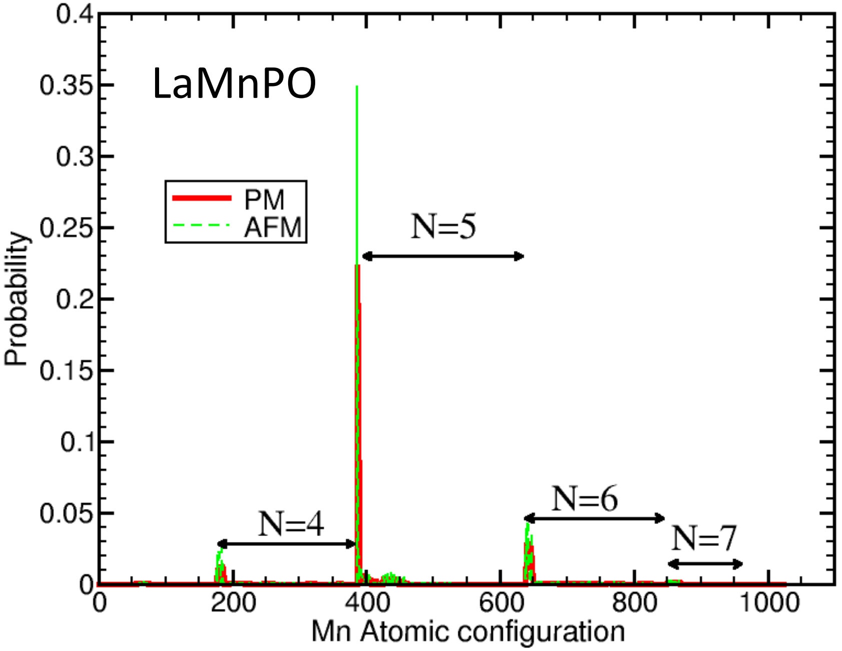


ZPY, K. Haule and G. Kotliar, Nature Materials **10**, 932 (2011).

DFT+DMFT accounts for the variations in all families without tuning U and J!

Valence Histogram



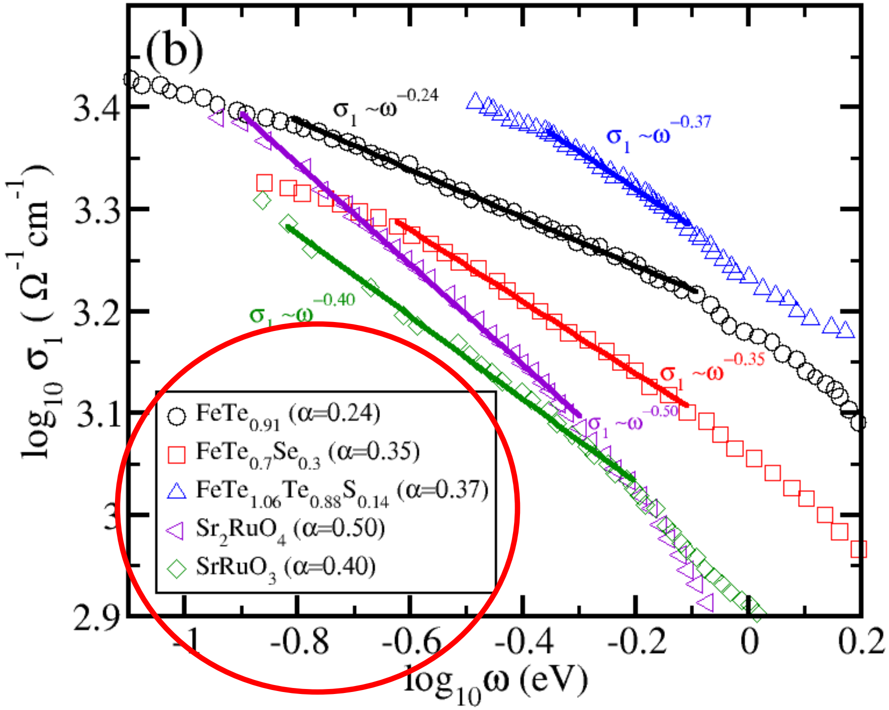


ZPY, K. Haule and G. Kotliar,
Nature Materials **10**, 932 (2011).

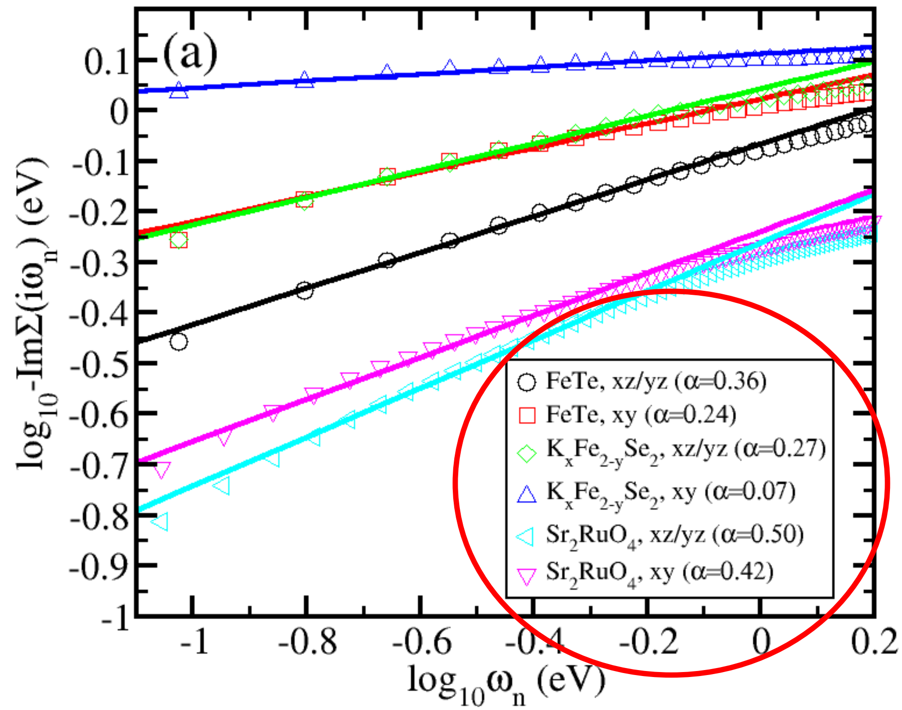
J. W. Simonson, ZPY et al., PNAS
109, E1815-E1819 (2012).

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

Experiments



Theory (DFT+DMFT)



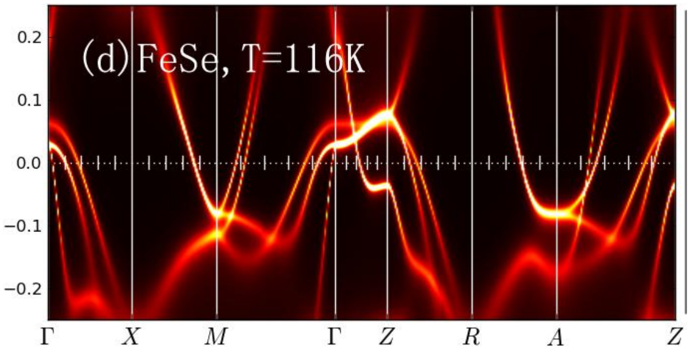
$$\sigma_1(\omega) \sim \omega^{-\alpha} \quad \longleftrightarrow \quad \Sigma''(\omega) \propto -\omega^{\alpha}$$

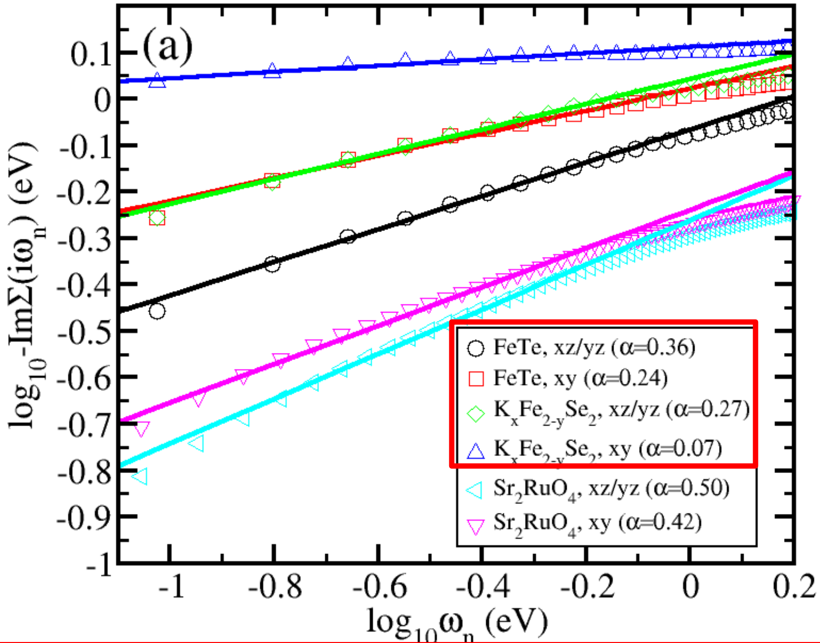
$$\sigma_1(\omega) \propto \mathrm{Re}\left[\frac{1}{\omega + i\Sigma''(\omega) + \Sigma'(\omega) - \Sigma'(\omega=0)}\right]$$

α is orbital and material dependent, not necessarily 1/2.

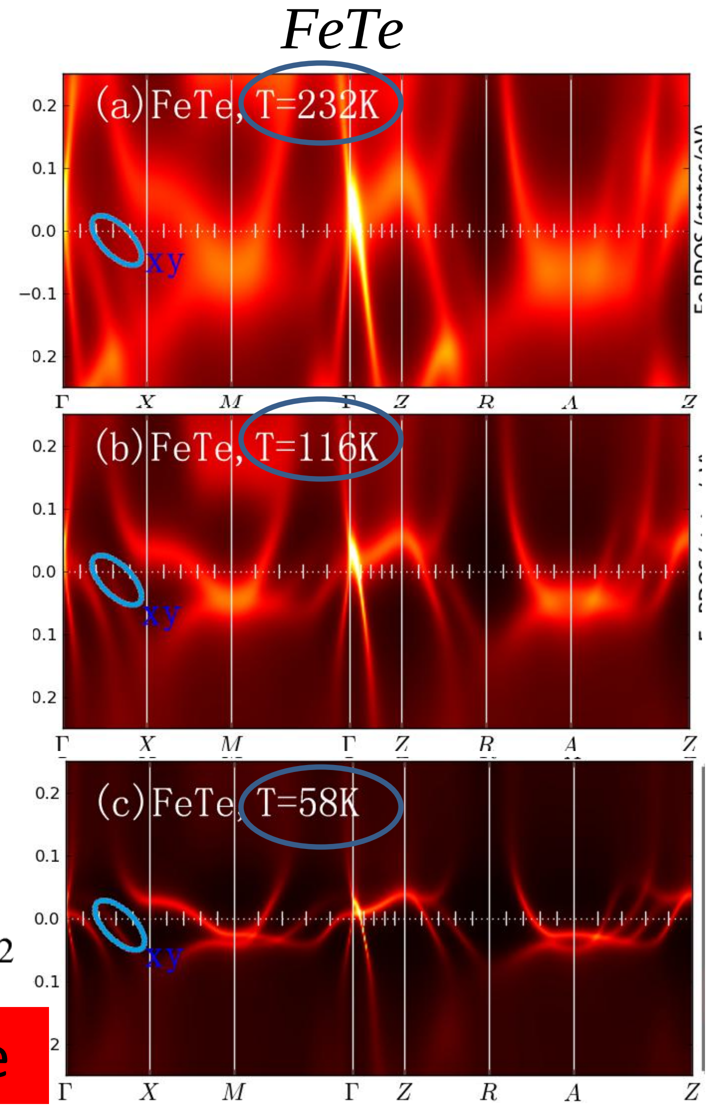
ZPY *et al.*, PRB **86**, 195141 (2012).

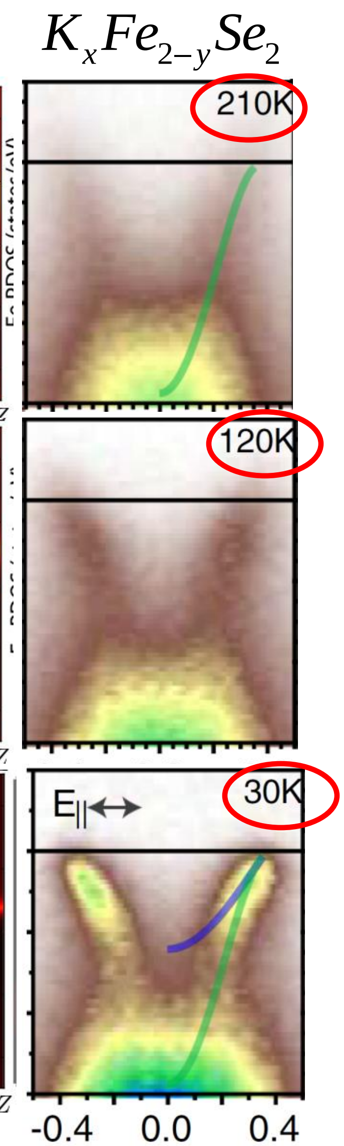
T-dependence: Coherence-incoherence crossover





Very low coherence temperature





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

Experiments: M. Yi *et al.*, PRL **101**, 067003 (2013).

Optical conductivity: $BaFe_2As_2$

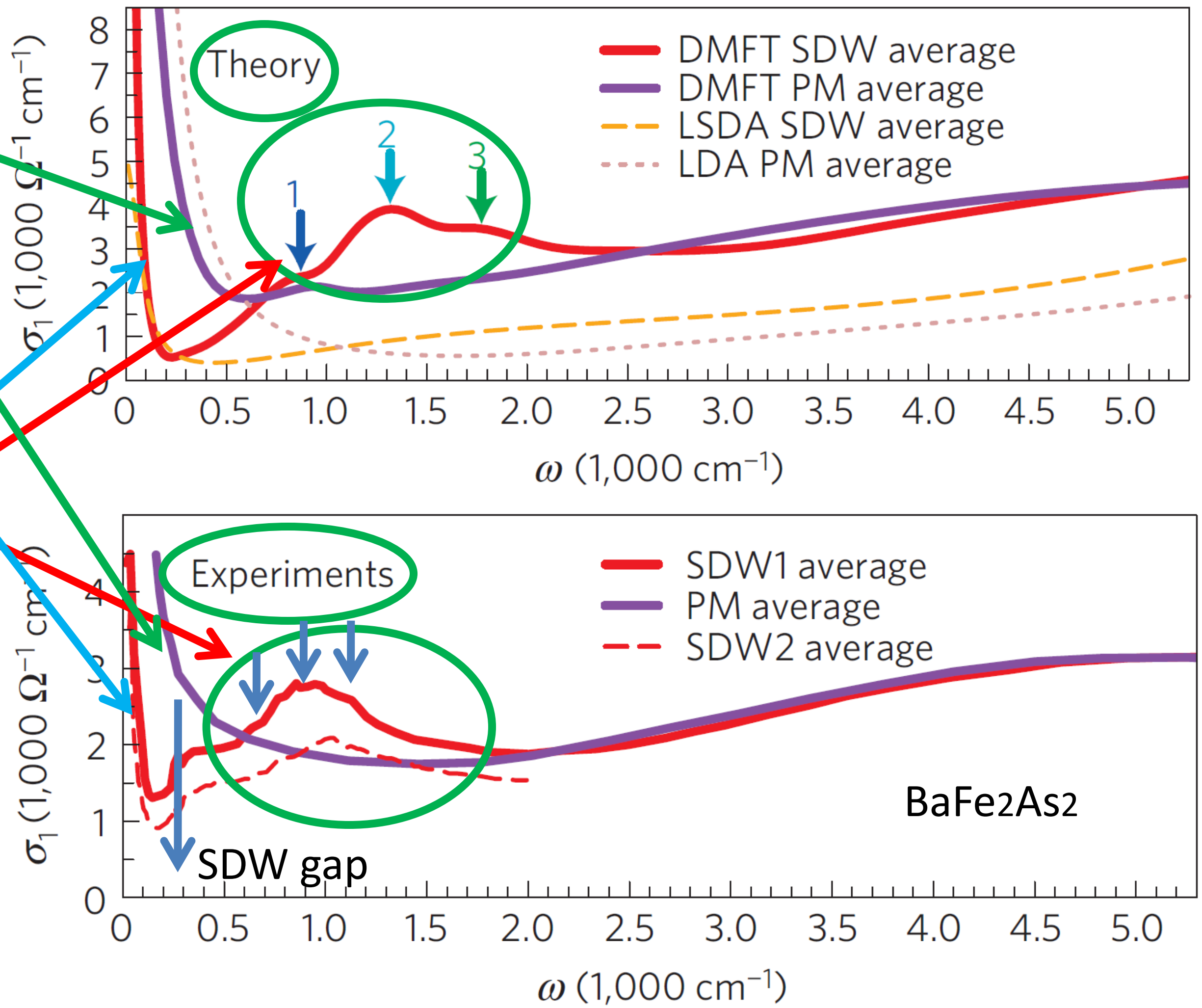
ZPY, K. Haule, G. Kotliar, Nature Physics **7**, 294 (2011).

PM state→broader Drude peak
Correct plasma ω_{p}:
DMFT ~ 1.6eV
Exp ~ 1.6eV
LDA ~ 2.6eV

SDW state more coherent→ 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



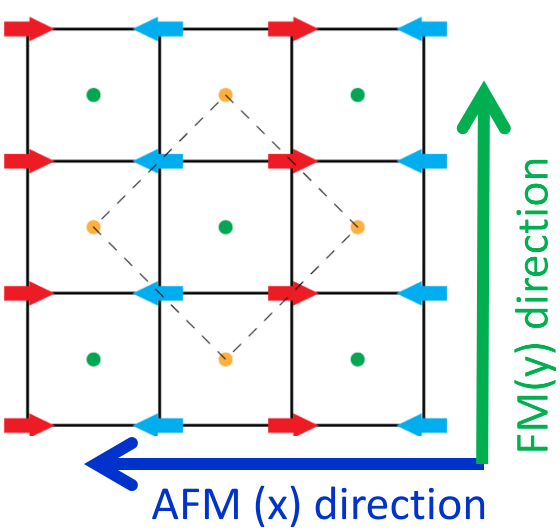
Experiments: W. Z. Hu, et al, PRL 101, 257005 (2008).
Nakajima, M. et al. Phys. Rev. B 81, 104528 (2010).

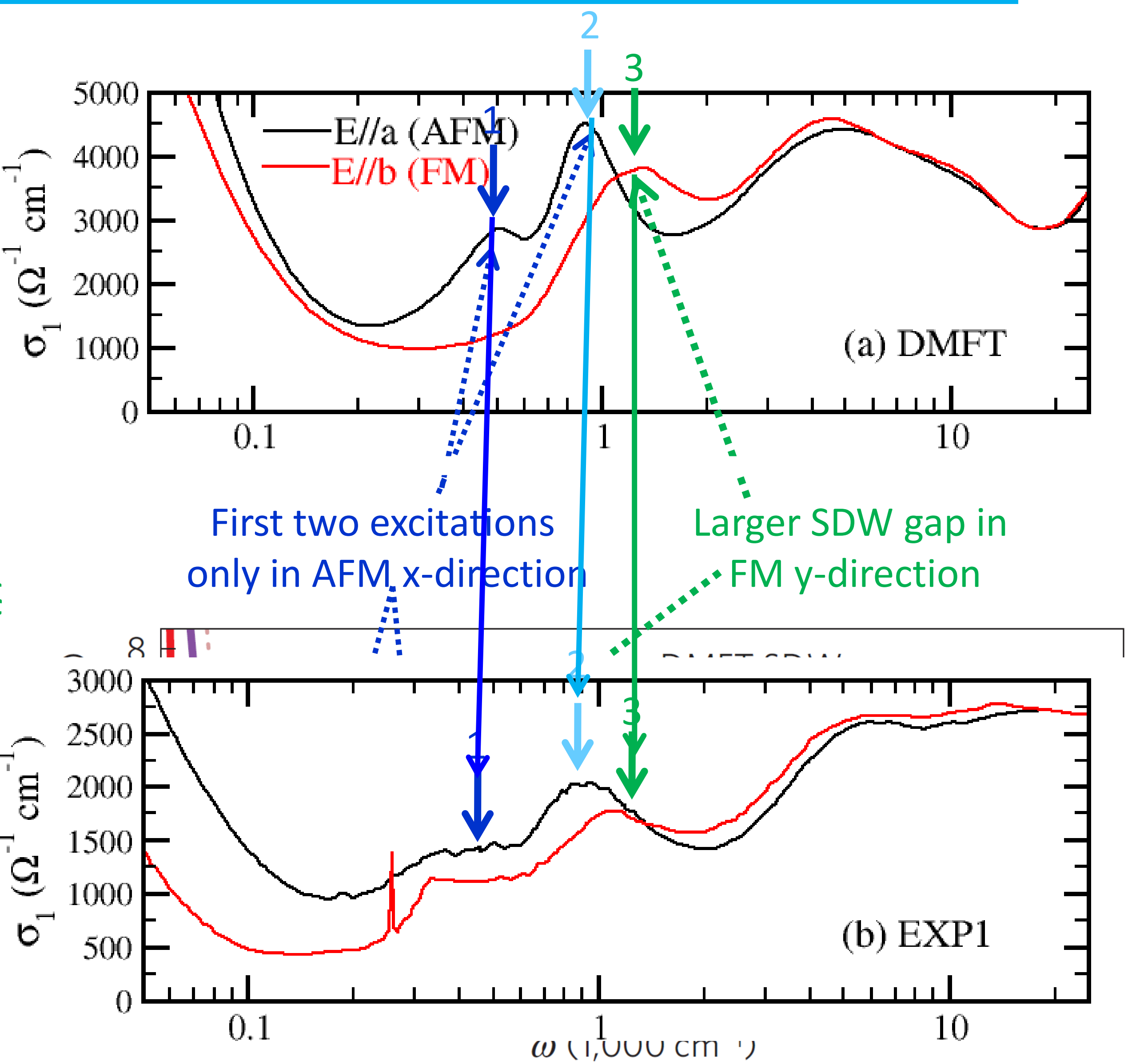
Optical in-plane anisotropy predicted by DFT+DMFT

$BaFe_2As_2$ in the SDW phase

DMFT prediction:

Nature Physics 7, 294 (04/2011), arXiv:1007.2867 (07/2010).



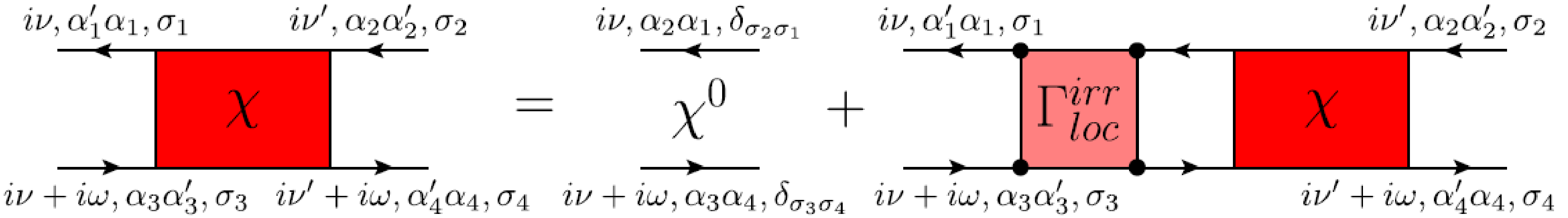


Experiment:

M. Nakajima, …,S Uchida, **PNAS 108**, 12238 (07/2011), arXiv: 1106.4967 (06/2011).

Susceptibility with vertex correction

Bethe-Salpeter equation:



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

One particle Green's function

Sampled by ctqmc

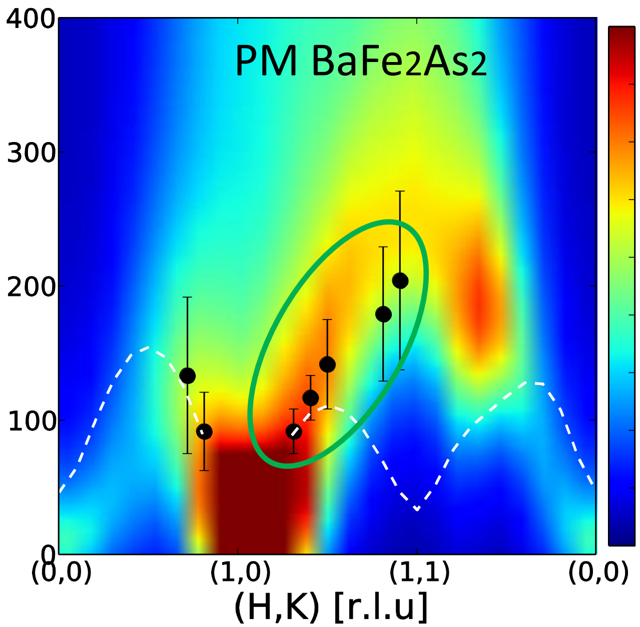
$$\chi^{\alpha_1\sigma_1,\alpha_2\sigma_2}_{\alpha_3\sigma_3,\alpha_4\sigma_4}(i\nu, i\nu')_{\mathbf{q},i\omega} = [(\chi^0)^{-1}_{\mathbf{q},i\omega} - T\Gamma^{\text{irr}}_{\text{loc}}]^{-1}.$$

$$\chi(\mathbf{q}, i\omega) = T\sum_{i\nu,i\nu'}\sum_{\substack{\alpha_1\alpha_2\\ \alpha_3\alpha_4}}\sum_{\substack{\sigma_1\sigma_2\\ \sigma_3\sigma_4}} \mu^z_{\substack{\alpha_1\sigma_1\\ \alpha_3\sigma_3}}\mu^z_{\substack{\alpha_2\sigma_2\\ \alpha_4\sigma_4}}\chi^{\alpha_1\sigma_1,\alpha_2\sigma_2}_{\alpha_3\sigma_3,\alpha_4\sigma_4}(i\nu, i\nu')_{\mathbf{q},i\omega}.$$

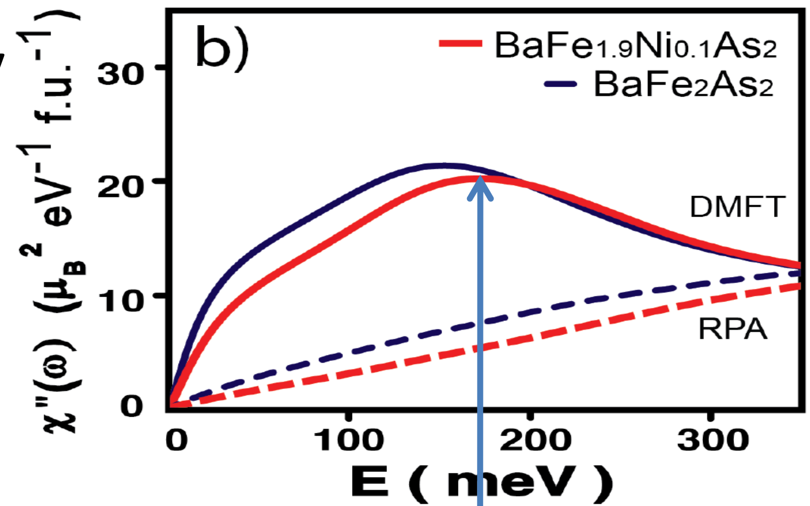
Easily replaced with charge or orbital to obtain charge or orbital susceptibility

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

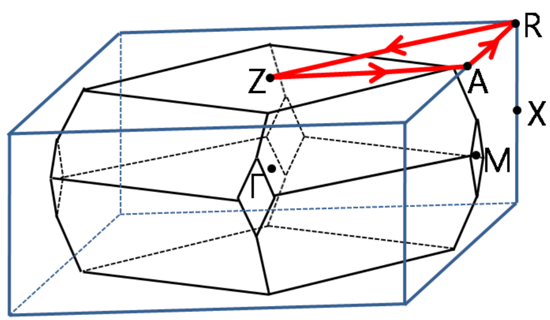
Spin susceptibility in iron pnictides

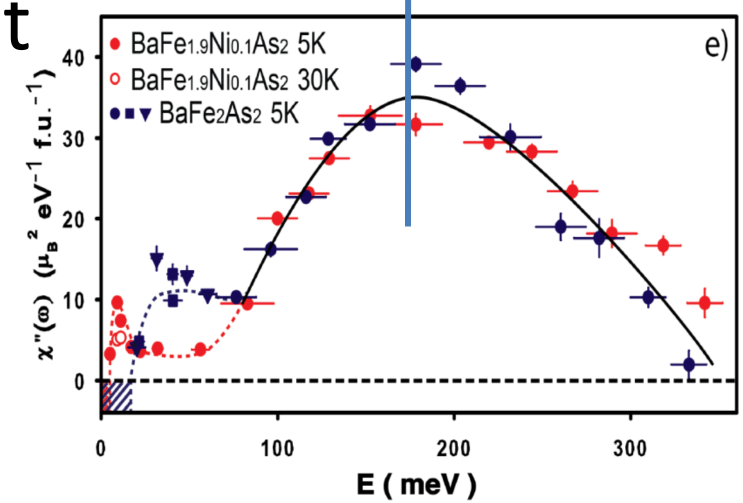


Theory



Experiment

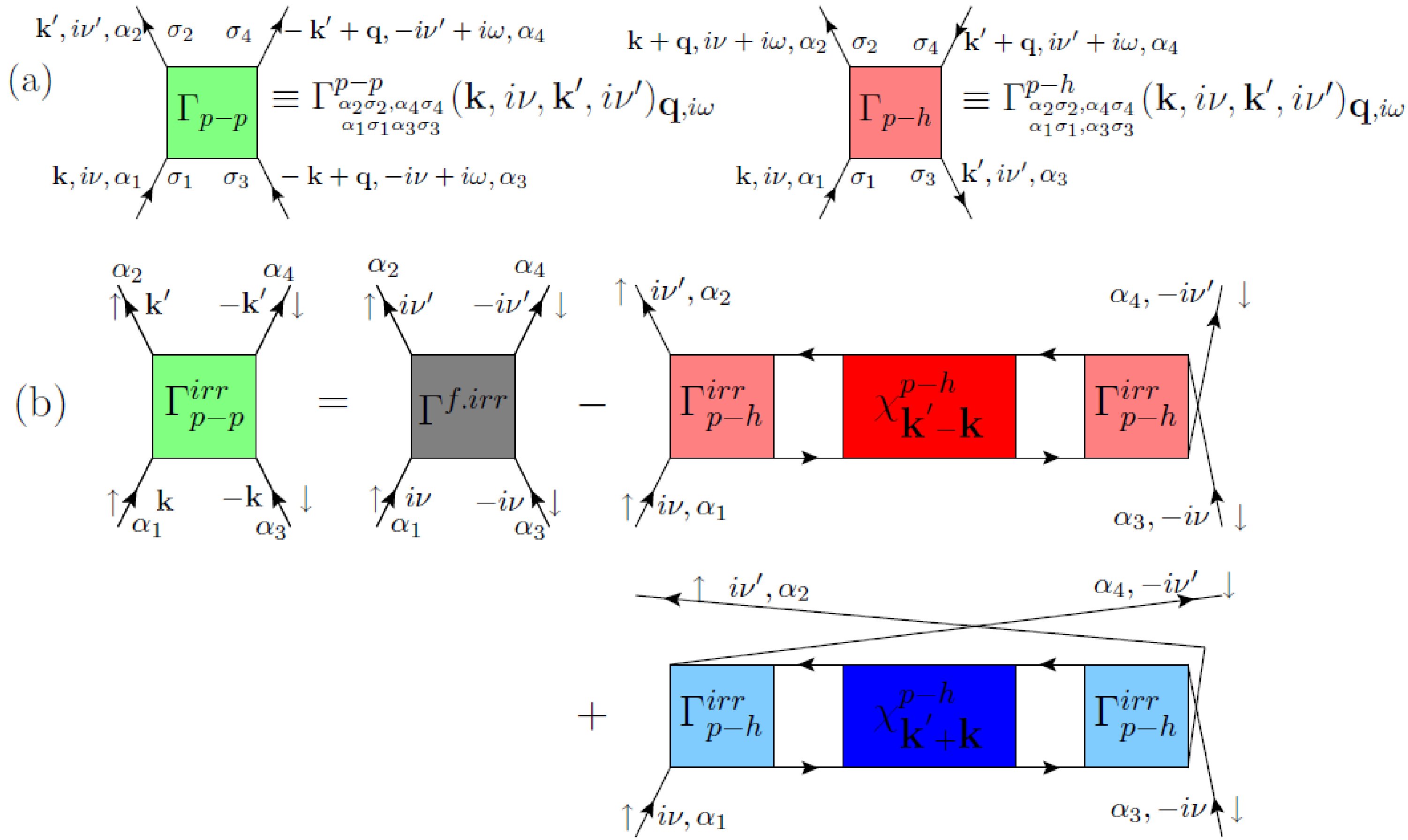




Theory: H. Park et al., PRL 107, 137007 (2011).
Experiments: L. W. Harriger et al, PRB 84, 054544 (2011)

Theory and Experiments: M. Liu et al., Nature Physics 8, 376-381 (2012), M. Wang et al, arXiv:1303.7339

Superconductivity



H. Park, Ph.D thesis, Rutgers University (2011).

Superconductivity

$$\Gamma^{irr,p-p,(s)}_{\substack{\alpha_2,\alpha_4\\ \alpha_1,\alpha_3}}(\mathbf{k},i\nu,\mathbf{k}',i\nu') = \Gamma^{f-irr,(s)}_{\substack{\alpha_2,\alpha_4\\ \alpha_1,\alpha_3}}(i\nu,i\nu') + [\frac{3}{2}\widetilde{\Gamma}^{p-h,(m)} - \frac{1}{2}\widetilde{\Gamma}^{p-h,(d)}]_{\substack{\alpha_2,\alpha_3\\ \alpha_1,\alpha_4}}(i\nu,-i\nu')_{\mathbf{k}'-\mathbf{k},i\nu'-i\nu} + \frac{1}{2}[\frac{3}{2}\widetilde{\Gamma}^{p-h,(m)} - \frac{1}{2}\widetilde{\Gamma}^{p-h,(d)}]_{\substack{\alpha_4,\alpha_3\\ \alpha_1,\alpha_2}}(i\nu,i\nu')_{-\mathbf{k}'-\mathbf{k},-i\nu'-i\nu} \quad (4.47)$$

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

$= 0$

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

Leading eigenvalue approaches 1 gives Tc

The corresponding eigenfunction gives the pairing symmetry

H. Park, Ph.D thesis, Rutgers University (2011).

Superconductivity: pairing symmetry

Orbital space

Band space: on FS

Ground State

S+-

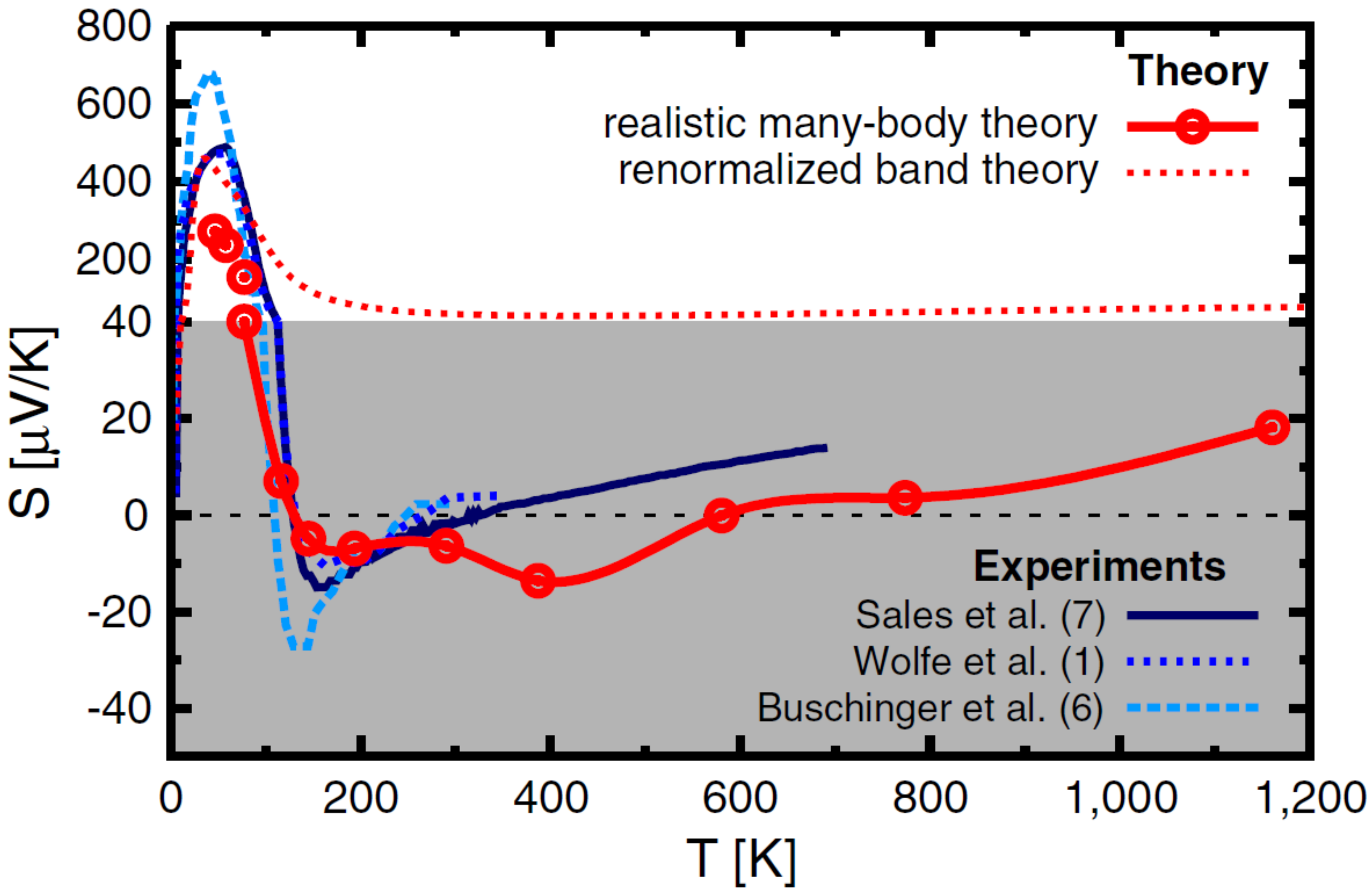
First Excited State

d-wave

Raman susceptibility

Experimental data from V. K. Thorsmolle and G. Blumberg (Rutgers University)

Thermoelectric Power: FeSi



J. M. Tomczak, K. Haule and G. Kotliar, PNAS 109 (9), 3243-3246 (2012)

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.