HOW ARE CRYSTAL STRUCTURES AND SUPERCONDUCTIVITY AFFECTED BY HUNDSNESS?

Kristjan Haule

RUTGERS
THE STATE UNIVERSITY OF NEW JERSEY

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Aspen

NSF
U.S. DEPARTMENT OF ENERGY
Blavatnik Awards for Young Scientists
Alfred P. Sloan Foundation 1934
Simons Collaboration on the Many Electron Problem

Thanks to long term collaborations:

Gabi Kotliar

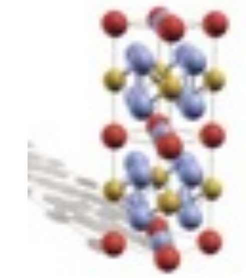
Ronald Cohen
Carnegie I.S.

Many thanks to current/former postdocs:

Subhasish Mandal
(e-ph coupling & FeSe)

Zhipin Yin
(iron superconductors)

Hunds Metals: coherence incoherence crosover



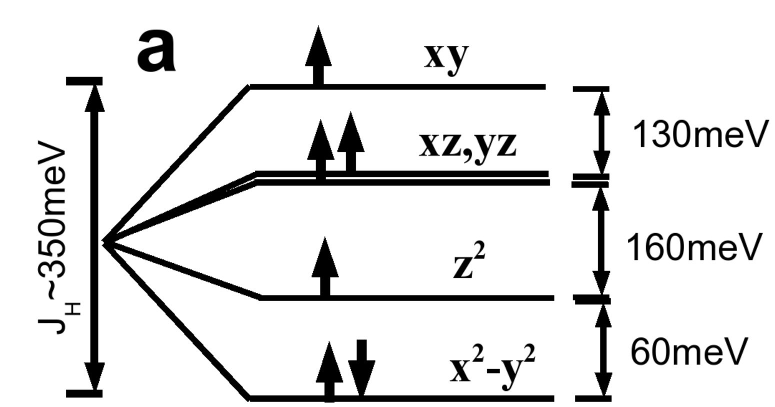
Hund's metals were found when studying the origin of mass enhancements in Fe-pnictides

Hubbard U not important

The **Hund's coupling** brings correlations!

K. Haule, G. Kotliar, arXiv:0805.0722 (2008), New Journal of Physics, 11 025021 (2009).

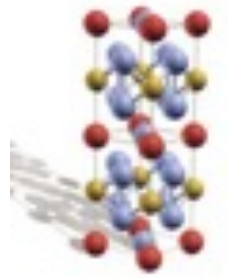
In metals when electrons are forced to be in the high-spin state (by Hund's J), the Fermi liquid state is protracted, and develops only at very low temperature. There is a "*Coherence incoherence transition*" from incoherent local-moment state at high-T to Fermi liquid state at low-T (except in the selective Mott phase).



6 electrons in 5 orbitals

Hund's coupling J ⇒high spin   Kinetic energy & Pauli ⇒ low spin

Hunds Metals: coherence incoherence crosover



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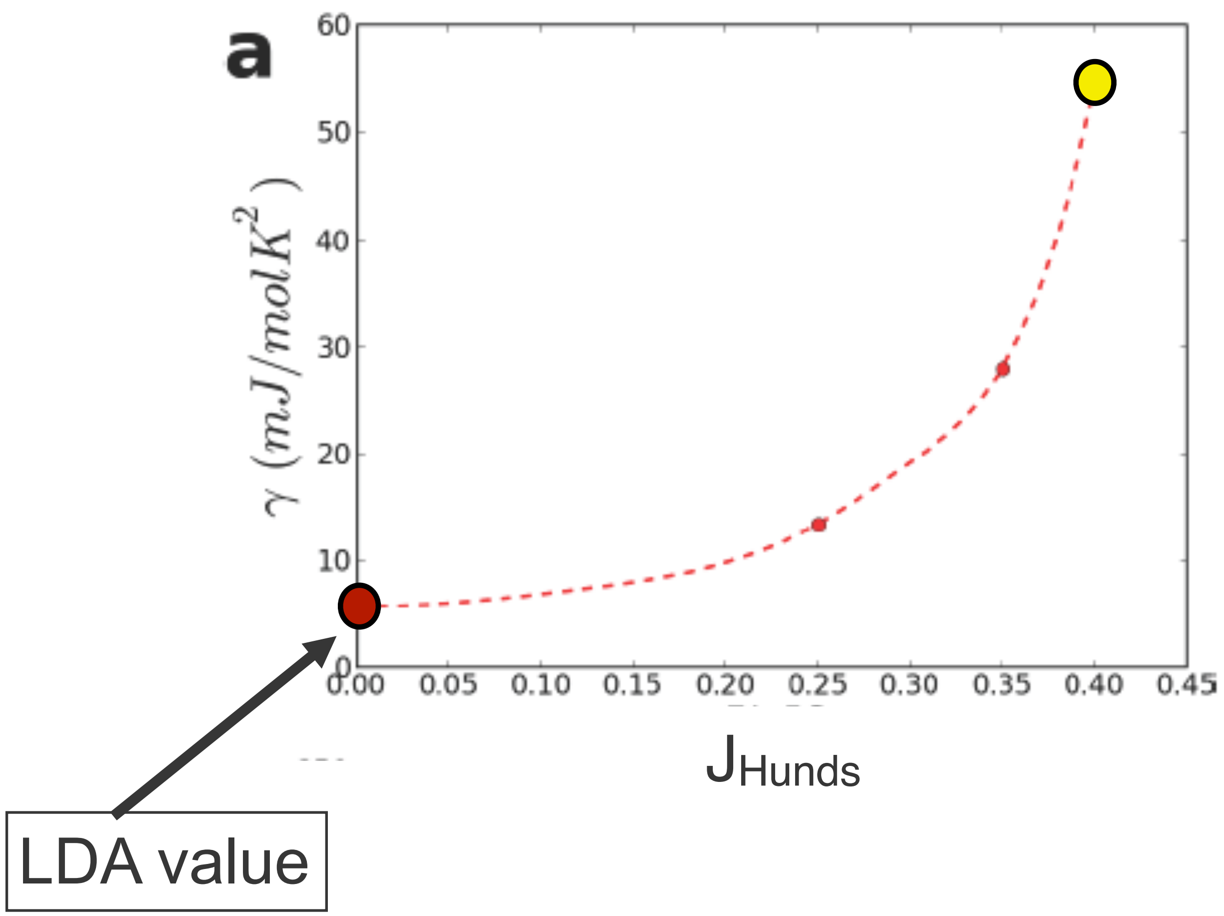
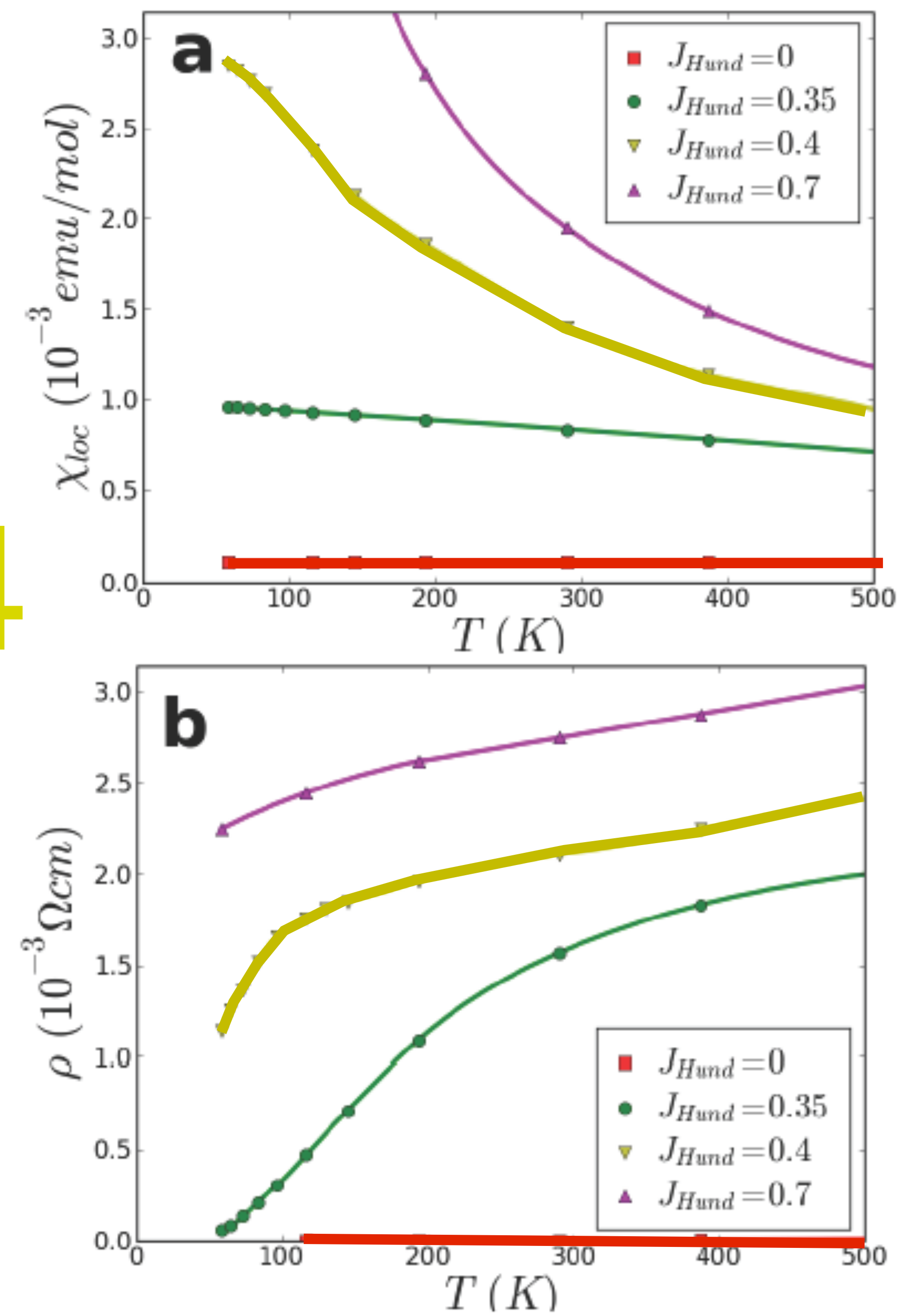


J=0.4

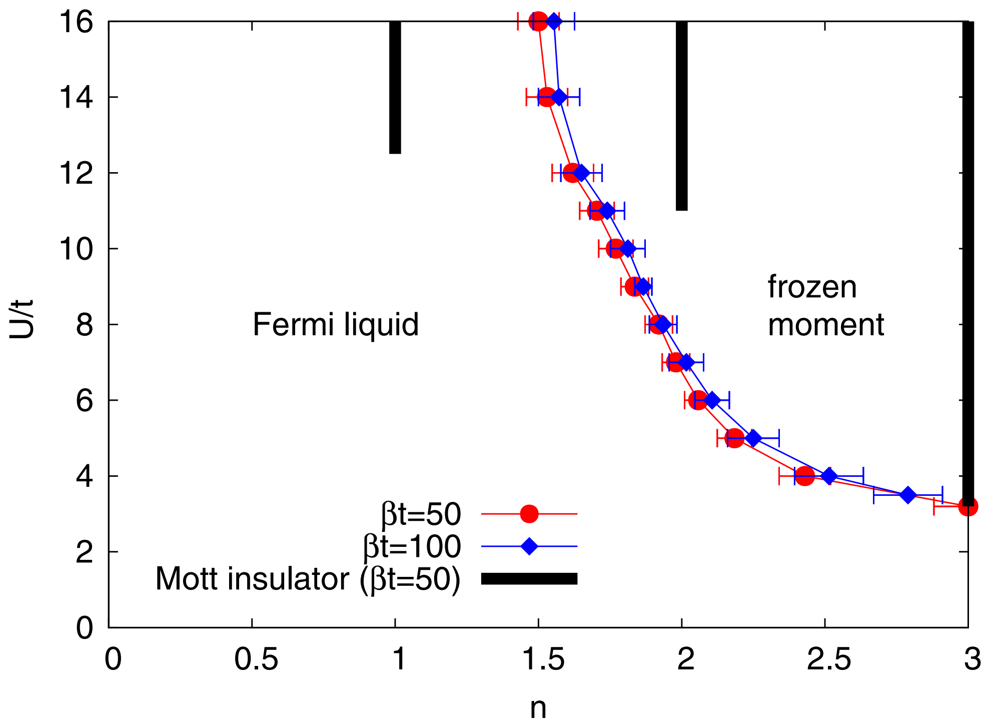


For J=0 there is negligible mass enhancement at U~W!

Spin freezing

Related early work on 3-band Hubbard model found "a quantum phase transition between paramagnetic metallic phase, and incoherent metallic phase with frozen moments." dubbed spin-freezing.

P. Werner, E. Gull, M. Troyer, and A.J. Millis



Timeline of the early works:

arXiv:0805.0722 : *Coherence-incoherence crossover in the normal state of iron-oxypnictides and importance of the Hund's rule coupling*. NJP, K.H and G.Kotliar.

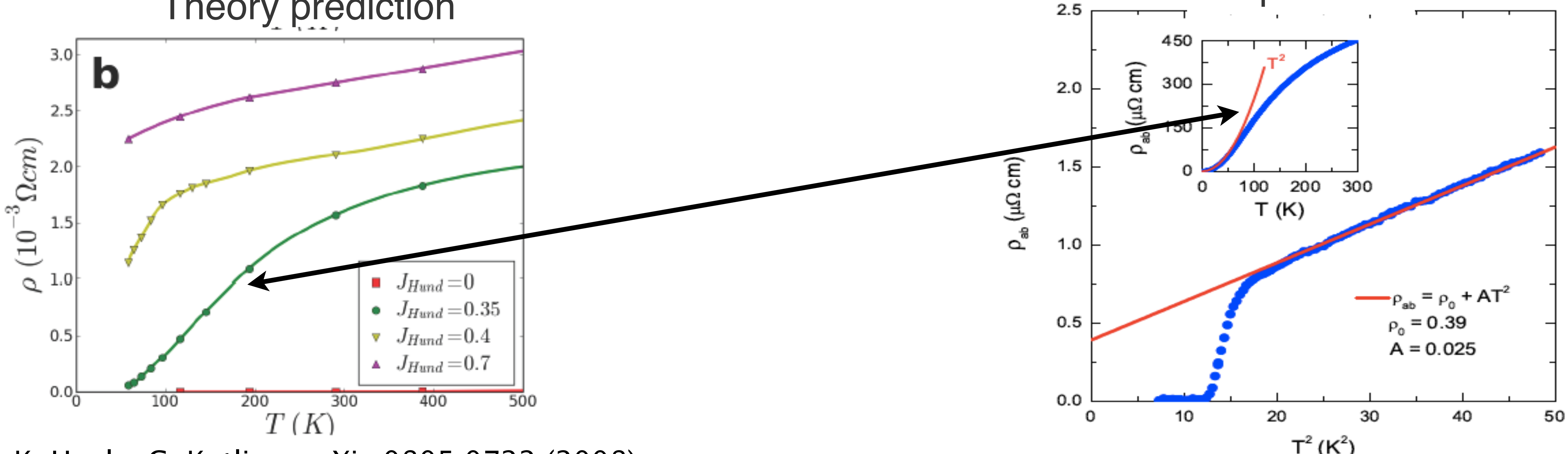
arXiv:0806.2621 : *Spin freezing transition and non-Fermi-liquid self-energy in a 3-orbital model*, PRL (2008), P. Werner et.al.

Nature Materials 10, 932-935 (2011): *The word Hund's metals was coined*, Z.P. Yin, K.H. and G. Kotliar.

Prediction of Coherence-Incoherence crossover confirmed

Theory prediction

Experiment



K. Haule, G. Kotliar, arXiv:0805.0722 (2008)
New Journal of Physics, 11 025021 (2009).

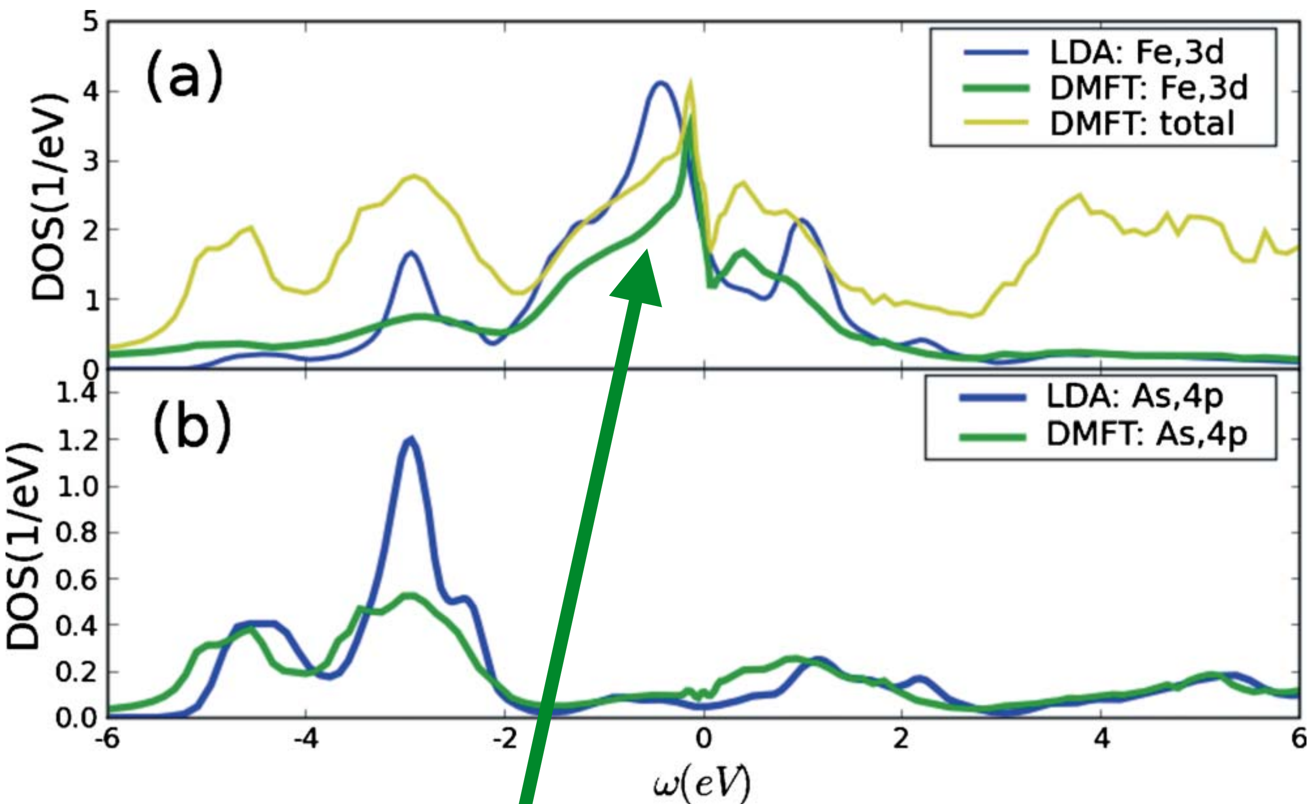
PRL 111, 027002 (2013) PHYSICAL REVIEW LETTERS week ending 12 JULY 2013

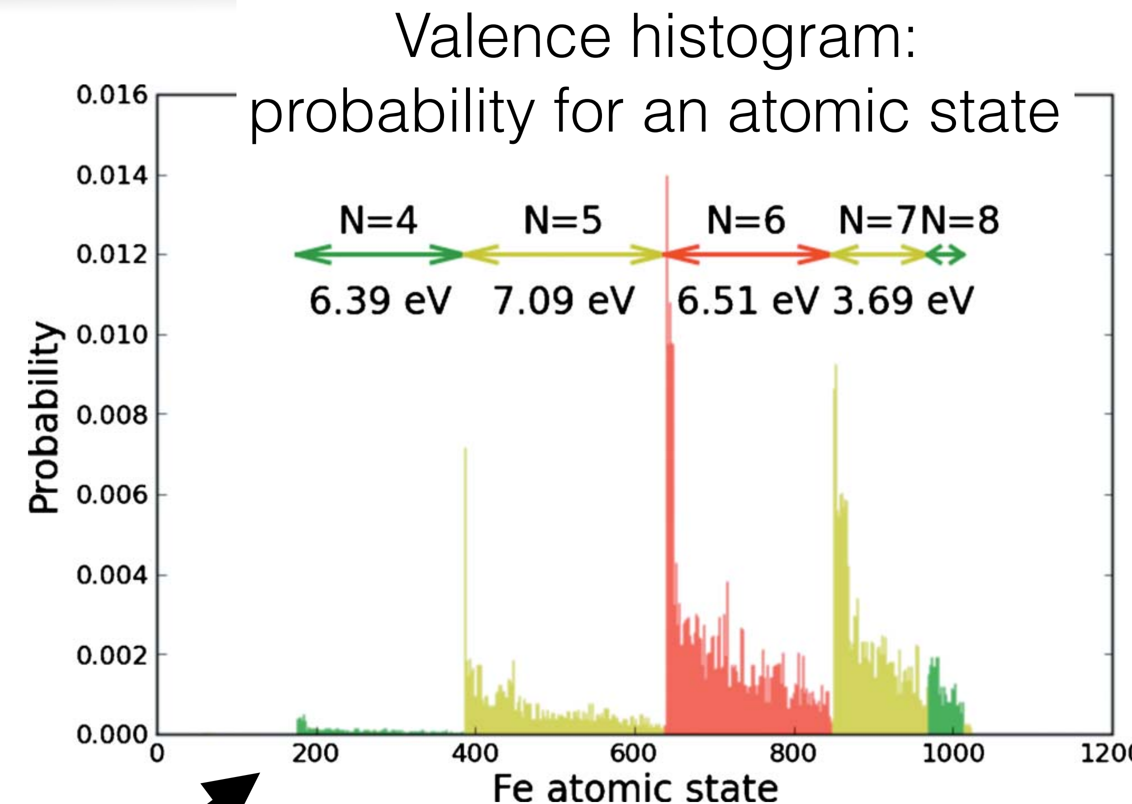
Evidence of Strong Correlations and Coherence-Incoherence Crossover in the Iron Pnictide Superconductor KFe_2As_2

F. Hardy,[1,*] A. E. Böhmer,[1] D. Aoki,[2,3] P. Burger,[1] T. Wolf,[1] P. Schweiss,[1] R. Heid,[1]
P. Adelmann,[1] Y. X. Yao,[4] G. Kotliar,[5] J. Schmalian,[6] and C. Meingast[1]

Hunds metals: correlations without Hubbard satellites, but quite localized magnetism

A. Kutepov, PRB **82**, 045105 2010.

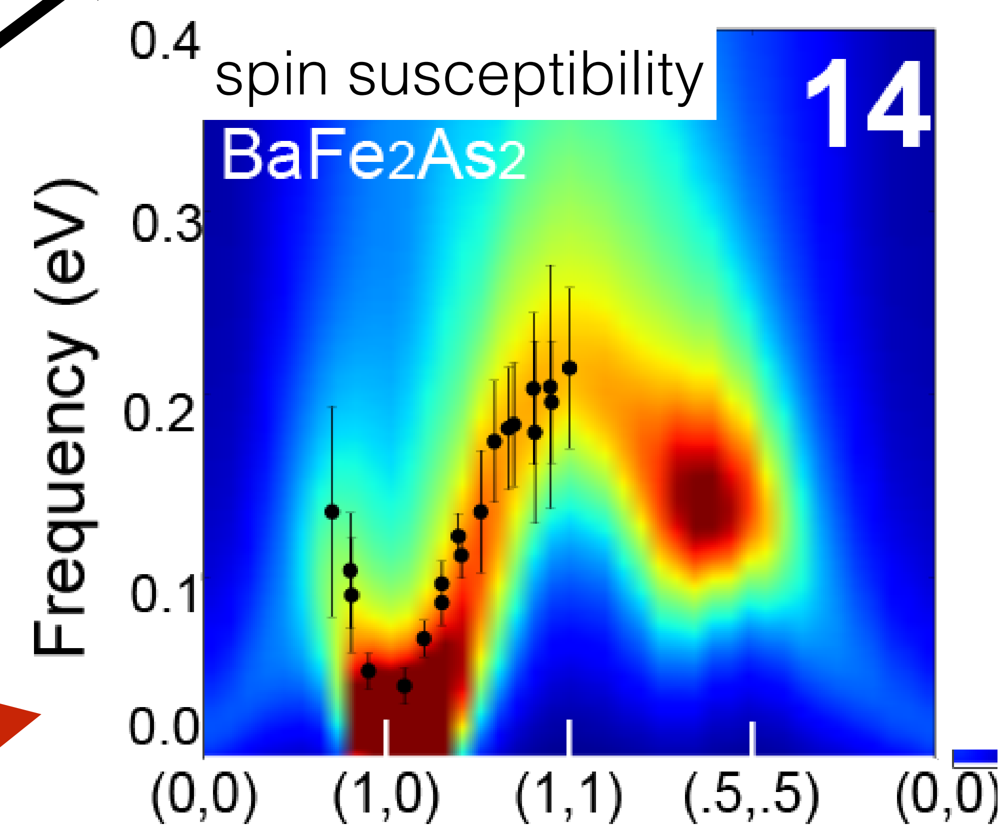




- Charge fluctuations are very fast — many atomic states are contributing to valence histogram

- Orbital fluctuations slow but faster than spin due coupling between spins and orbits.

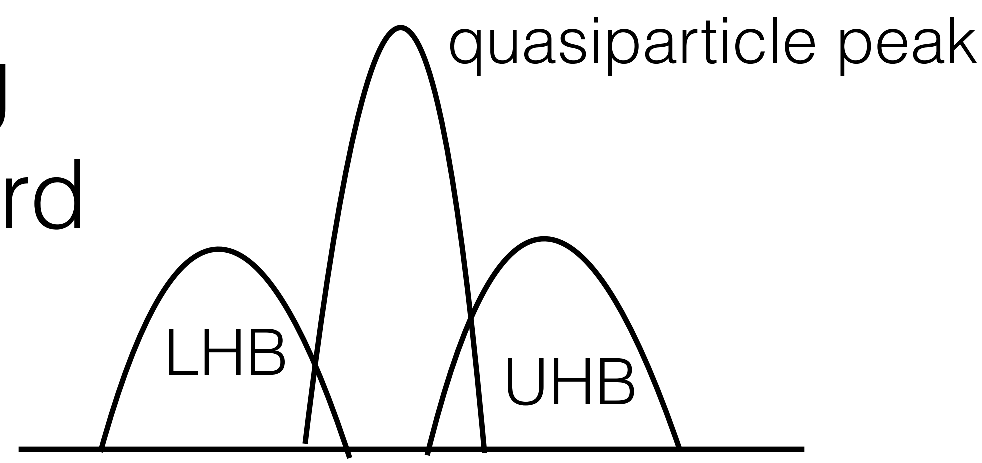
- Spin fluctuations very slow due to low energy ferromagnetic spin-spin—coupling



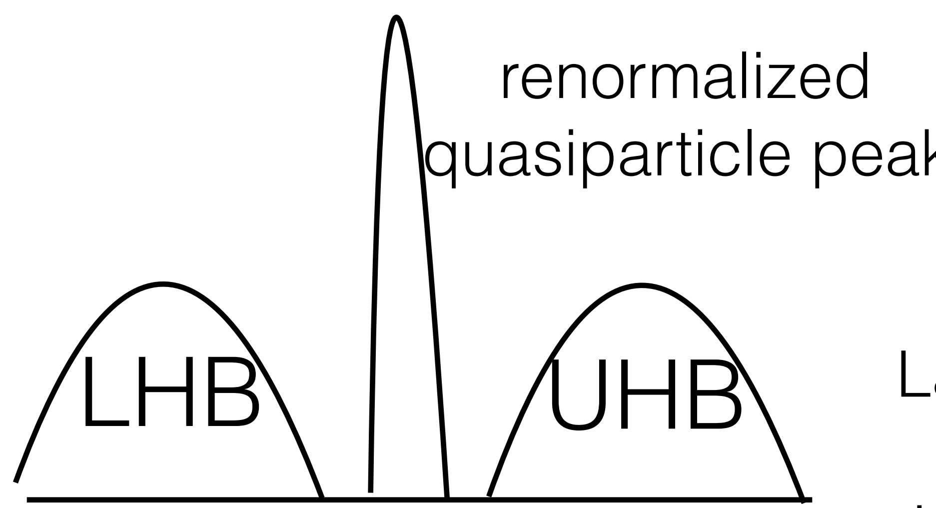
H. Park, K. Haule, G. Kotliar, PRL 107, 137007 (2011)

Qualitative idea

No Hund's coupling
just moderate Hubbard

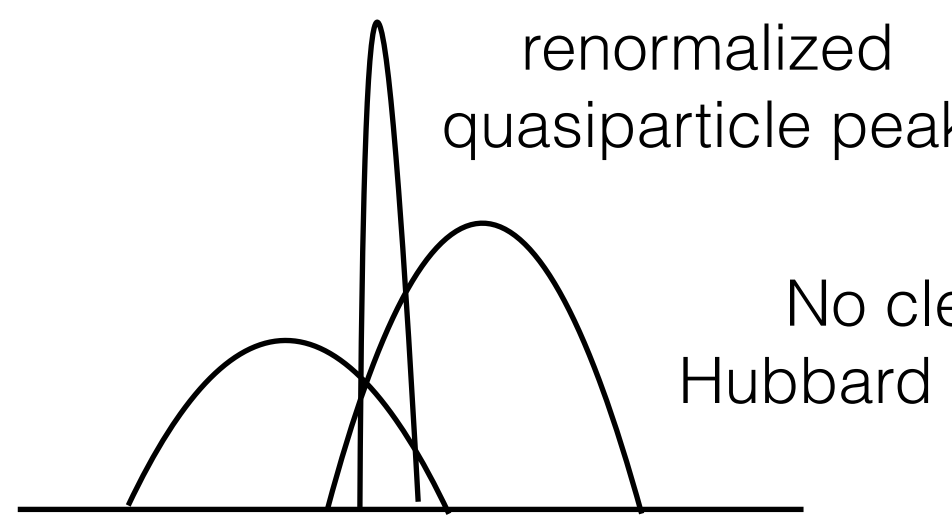


Mott physics gives:



Large separation
between
Hubbard bands

With Hund's coupling
<u>in valence state</u>
<u>away from half-filling</u>
<u>or single occupancy</u>.

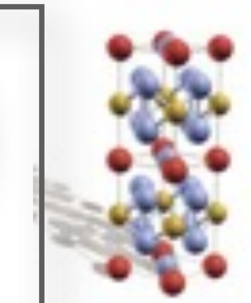


No clear
Hubbard bands

<u>Schrieffer-Wolf mathematical derivation in</u>

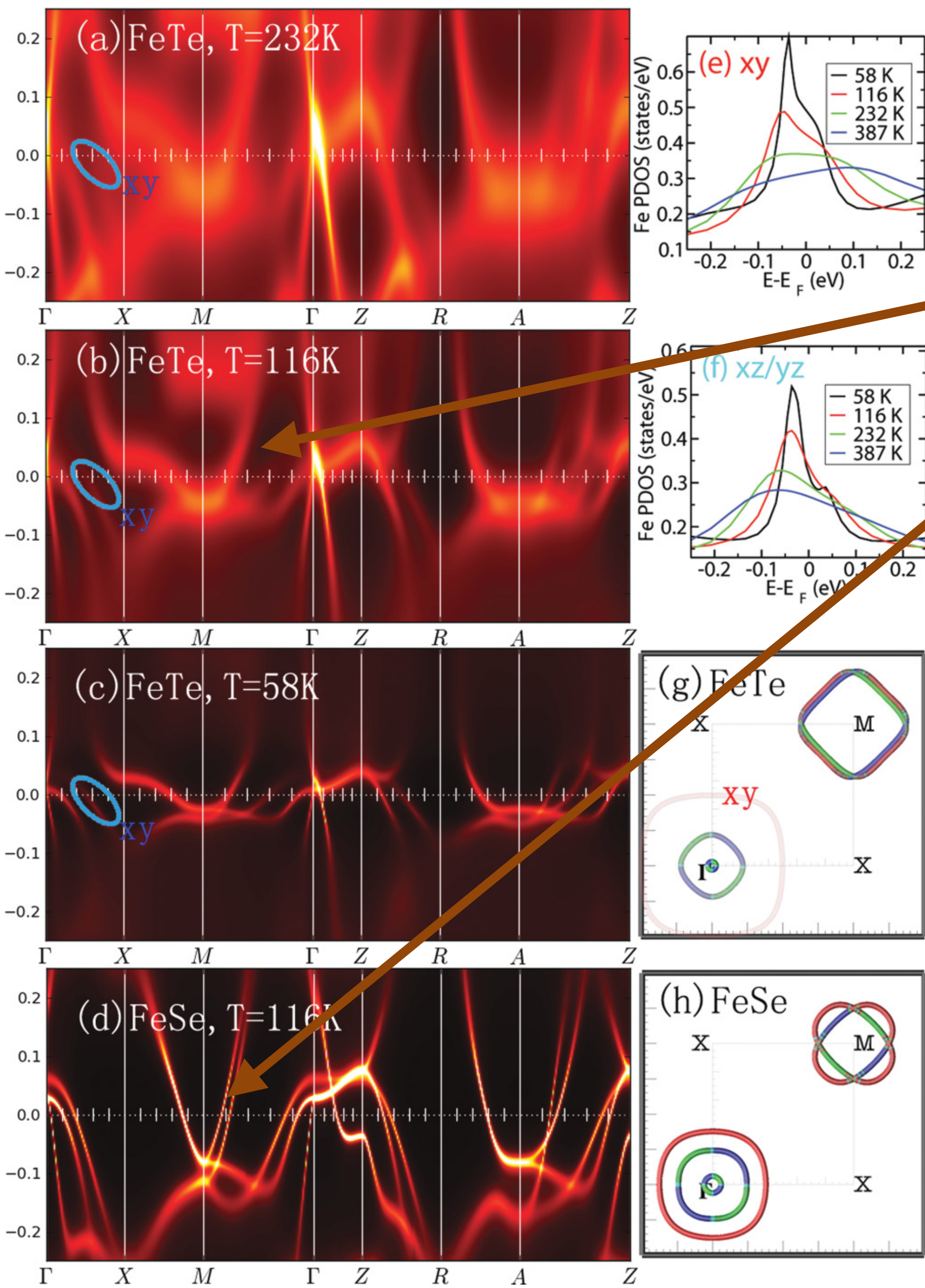
Z.Yin, KH, G. Kotliar, PRB 86, 195141 (2012).

Coherence incoherence crossover predicted for FeTe->FeSe, verified by ARPES



Z.P. Yin, K. Haule, G. Kotliar, PRB 86, 195141 (2012)

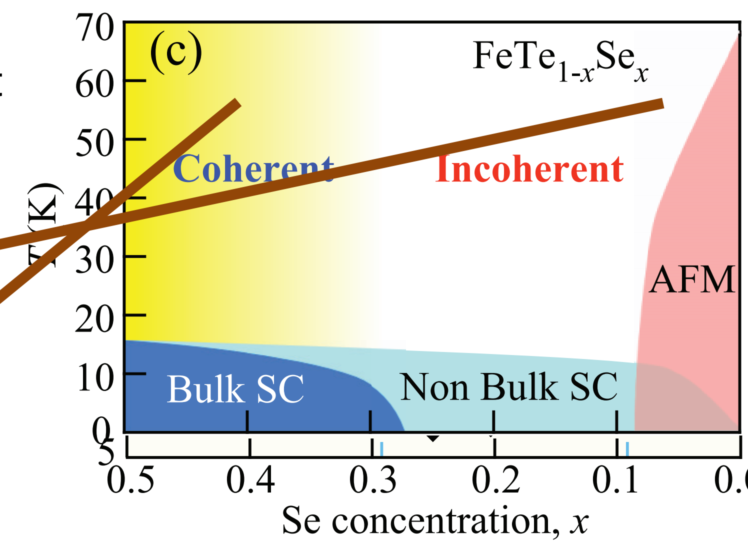
Theory prediction

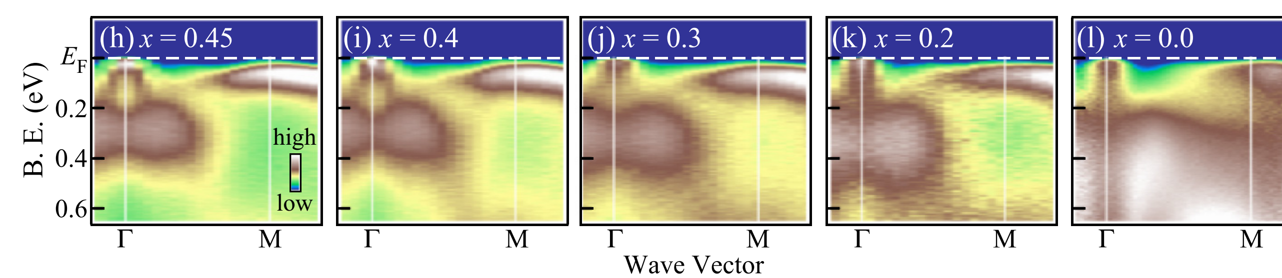


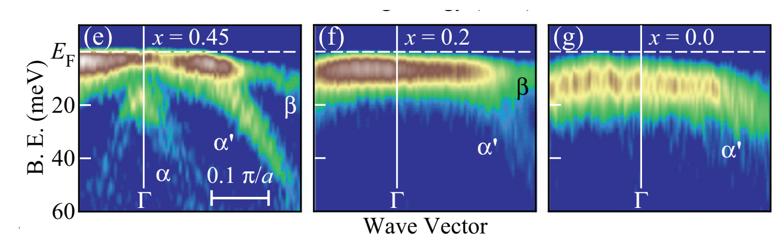
E. Ieki, & H. Ding et.al, PRB 89, 140506(R)(2014)

ARPES measurements

very incoherent



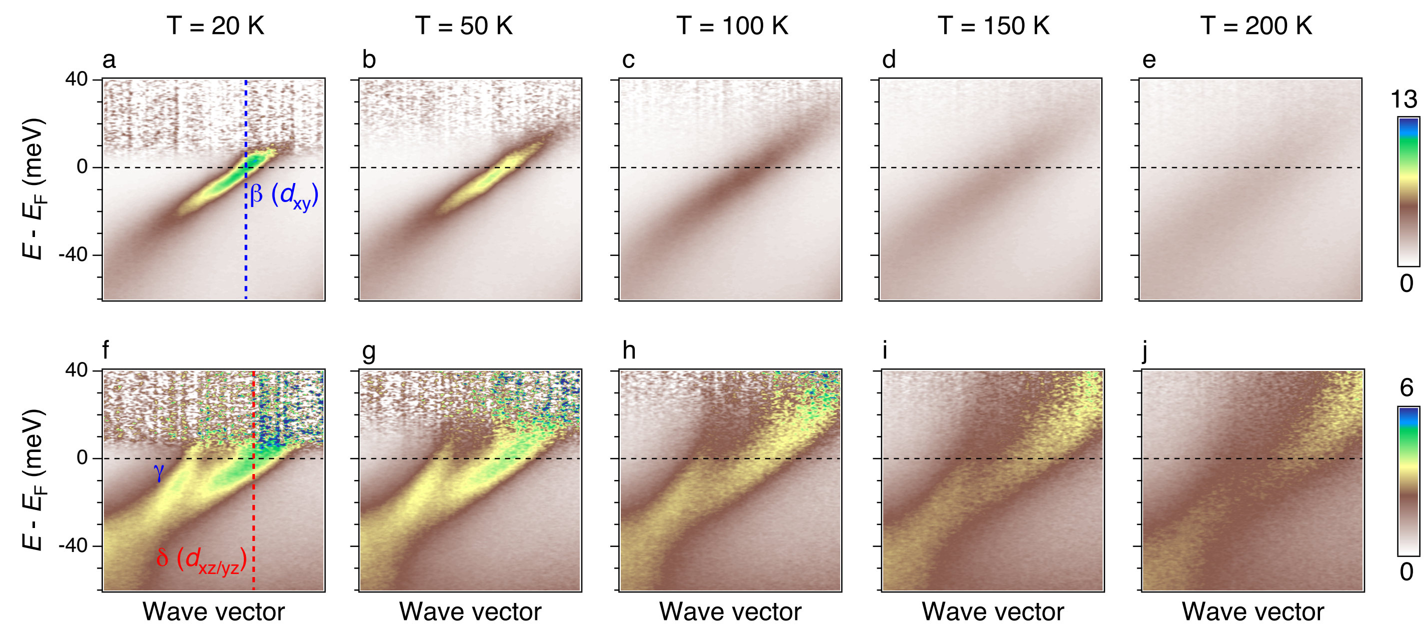




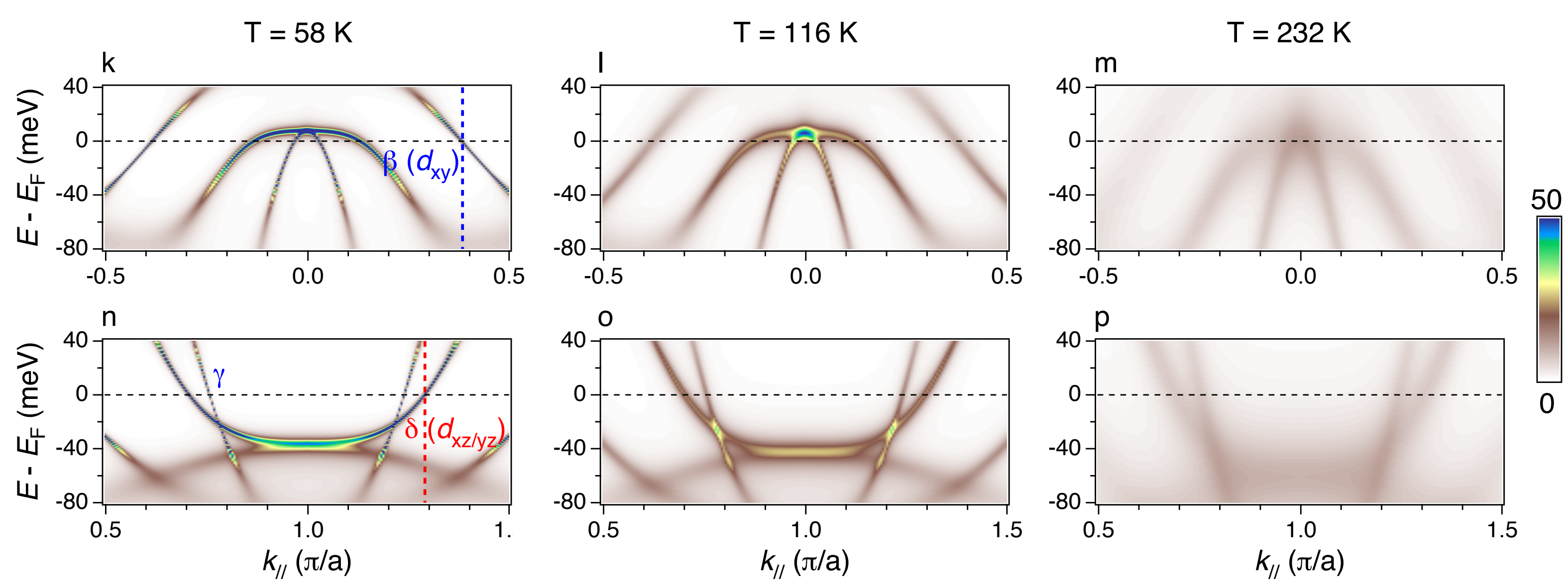
BANDS IN FLUCTUATING MOMENT/HUNDS SYSTEMS

Momentum resolved spectra of LiFeAs

ARPES



DFT-DMFT for LiFeAs:



m*/m~3

H Miao,…Haule, Kotliar, H. Ding, Phys. Rev. B 94, 201109(R) (2016).

Bands are sharp only very near the fermi level and only at low temperature (Fermi liquid).

Above the coherence temperature, electrons are better described as fluctuating moments, rather than plane waves.

DMFT predicted coherence incoherence crossover in Fe-SC (Hund's metals)

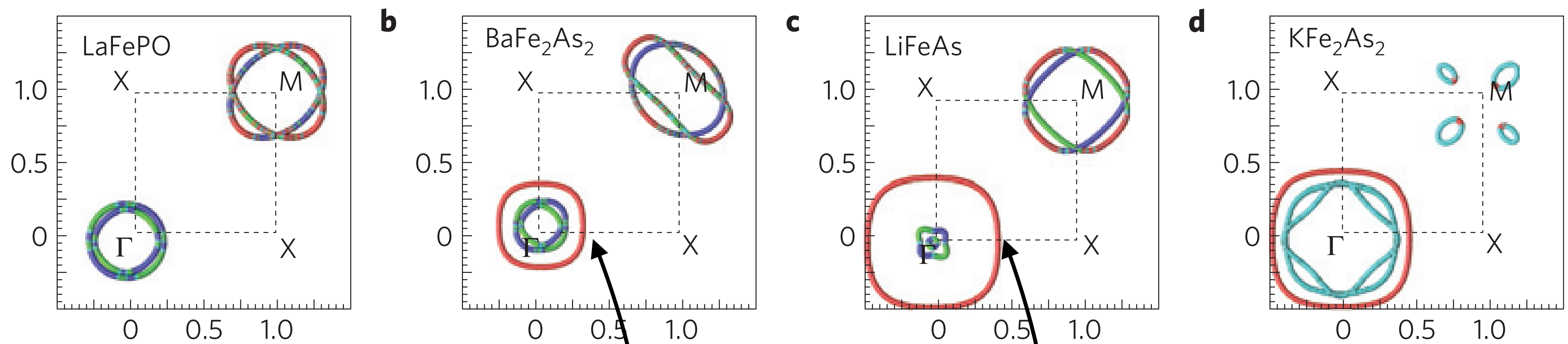
(Haule & Kotliar NJP 11, 25021 (2009))

Fermi surface can be strongly affected by correlations due to Hund’s

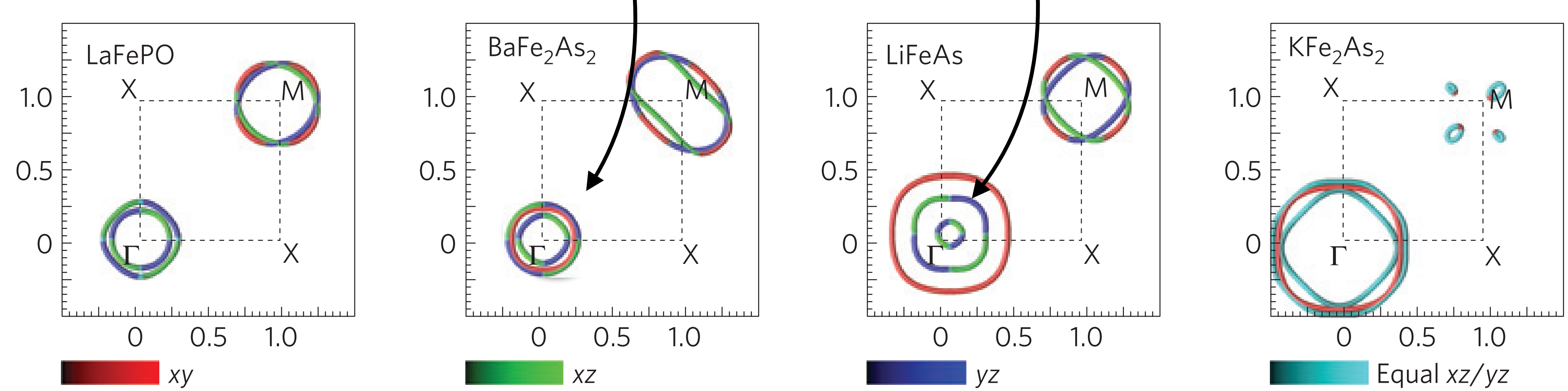
Z.P. Yin, K. Haule and G. Kotliar, Nature Materials **10**, 932 (2011).

- Orbital differentiation gives different mass and different shifts to different orbitals:
  - xy is orbital more correlated, its volume can be different than in DFT
  - xz/yz orbitals compensate for the volume change in yx orbital (Luttinger theorem)

DMFT Fermi surfaces:



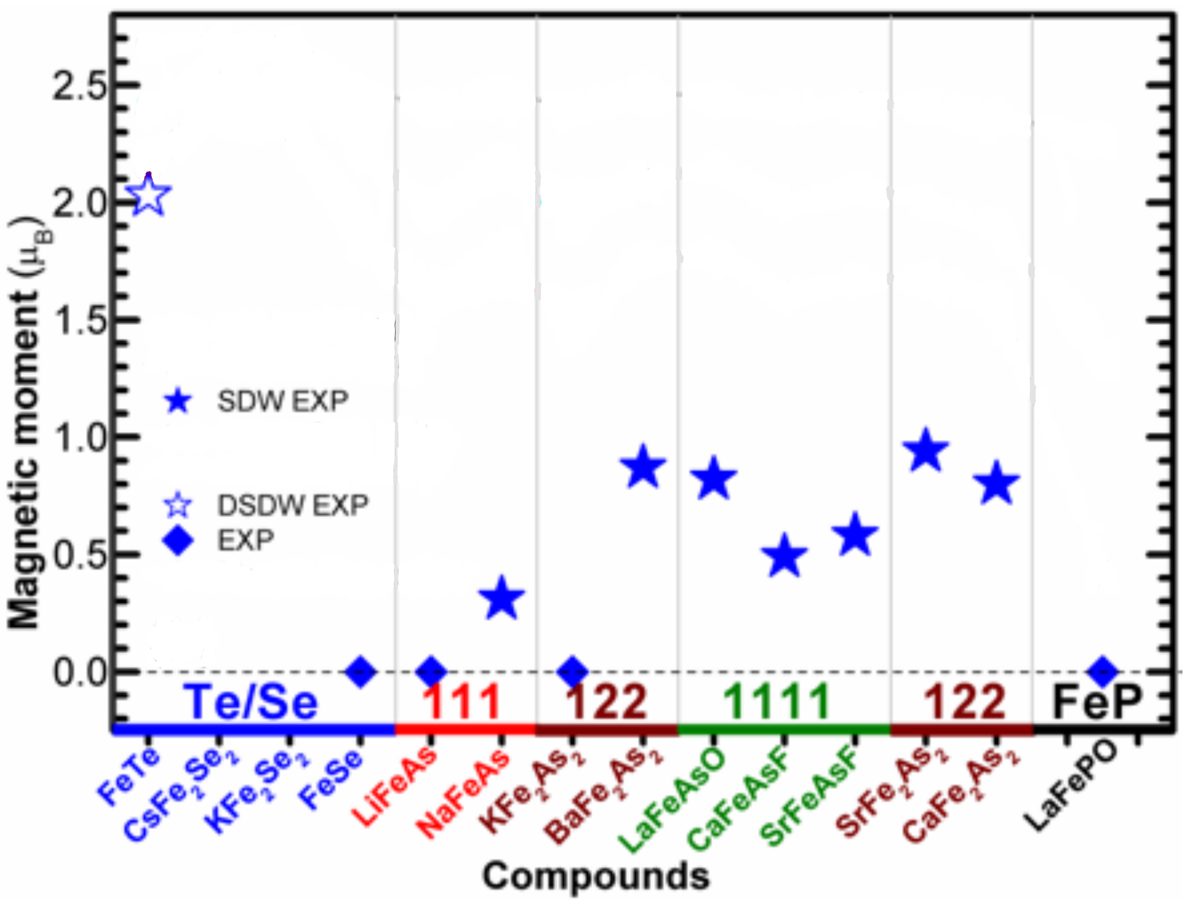
DFT Fermi surfaces:

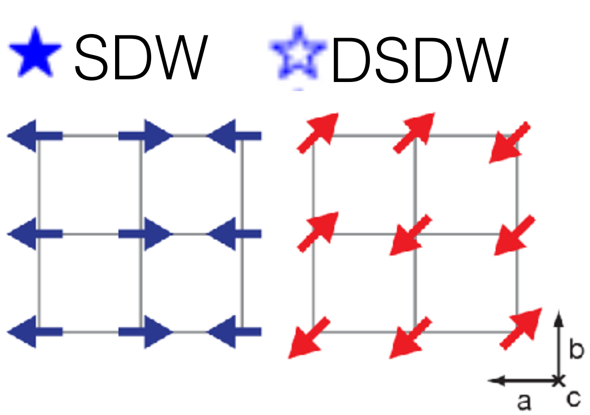


Puzzle: Parent compounds of Fe SC have very different ordered magnetic moments

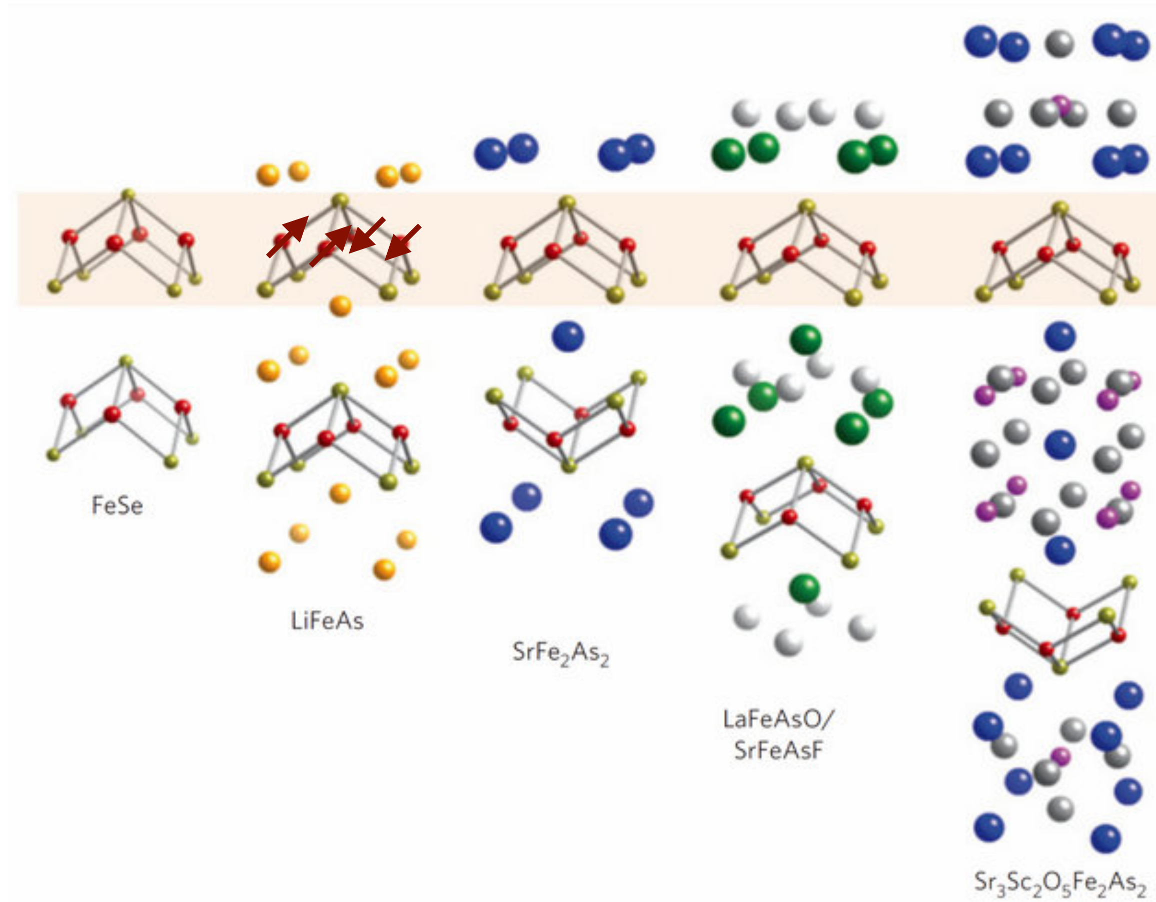
Experiment: Ordered magnetic moments across many Fe superconductors

no understanding what determines the size of the ordered moments





many families of Fe superconductors share the same Fe-As tetrahedral unit

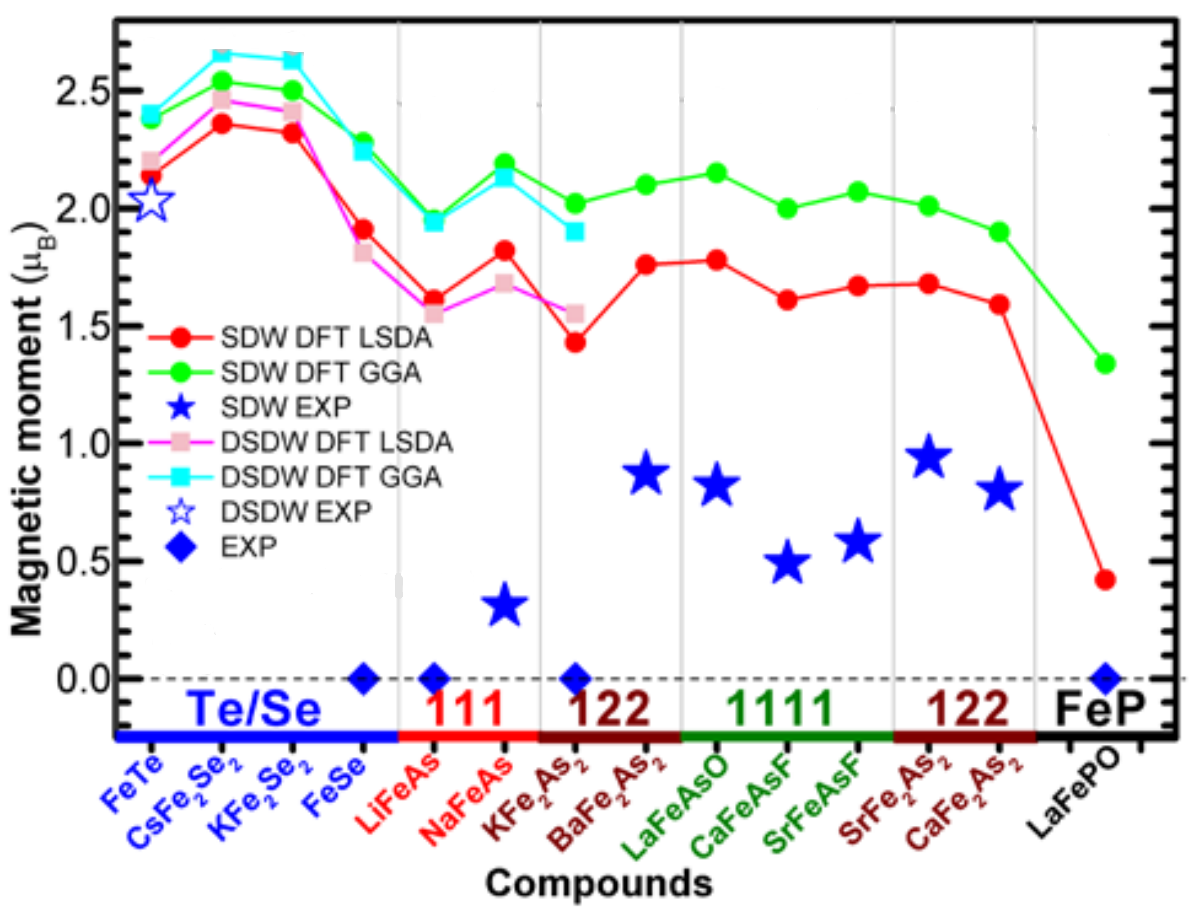


Contrast with cuprates, where the moments correspond to spin s=1/2.

What governs magnetism in Fe compounds?

DFT does not predict well the moments

blue symbols : experiment
green line: GGA, red line: LSDA



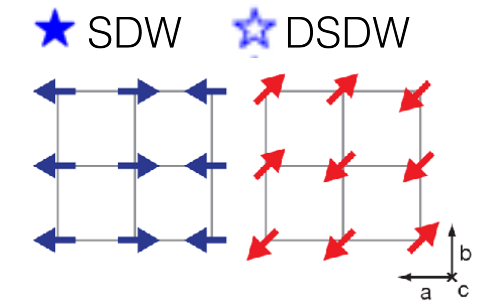
- DFT overestimates the ordered moment by roughly a factor of 2 (very uncommon)
- Many compounds appear magnetic in theory and non-magnetic in exp.

Very unusual because in

- bcc-Fe (weak itinerant magnet) LDA/GGA predicts moment very accurately ~2.1μ_B
- strong magnets like MnO LDA/GGA also very accurate

LDA/GGA accurate when very little fluctuating moment left in the ordered state. The entire magnetic moment has to statically order.

DMFT predicted magnetic moments in Fe-superconductors

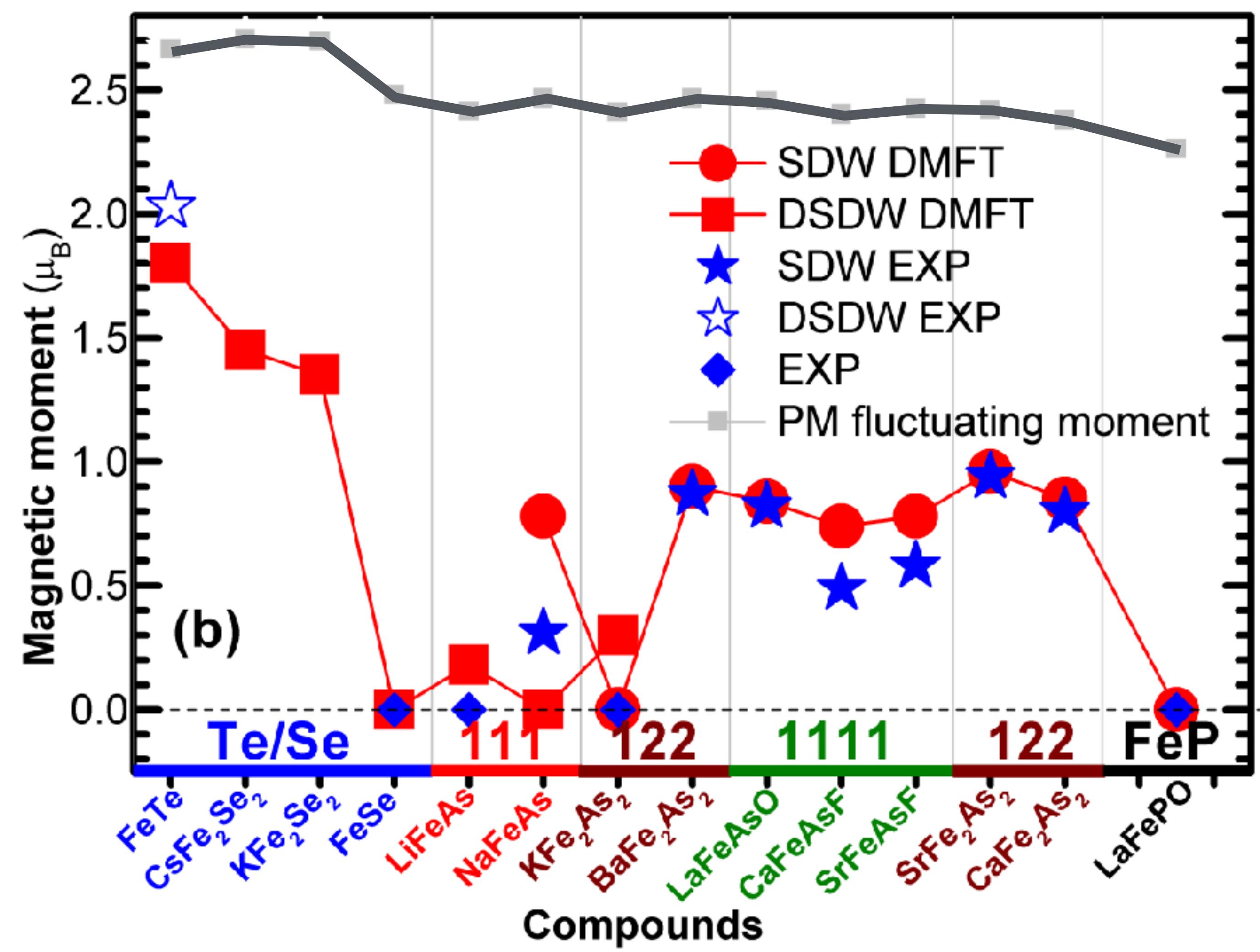


blue symbols : experiment

red symbols : DFT-eDMFT

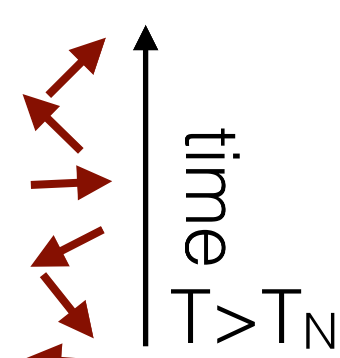
Can distinguish between the fluctuating moment <S²>, and the ordered moment <S>². In para <S>=0 on each site.

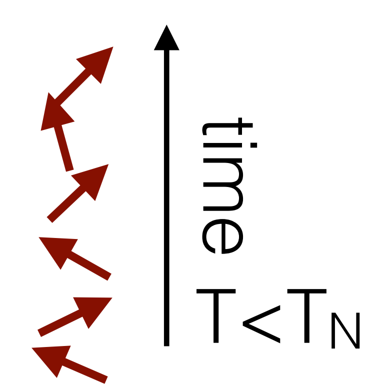
Substantial fluctuating moment remains in the ordered state



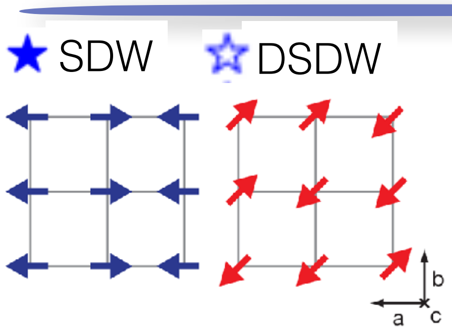
Yin, Haule & Kotliar, Nature Physics 7, 294-297 (2011).

Prediction: large fluctuating moment (2-2.5μ_B) in para state
Only a part of the fluctuating moments orders.





DMFT predicted moments much closer to experiment

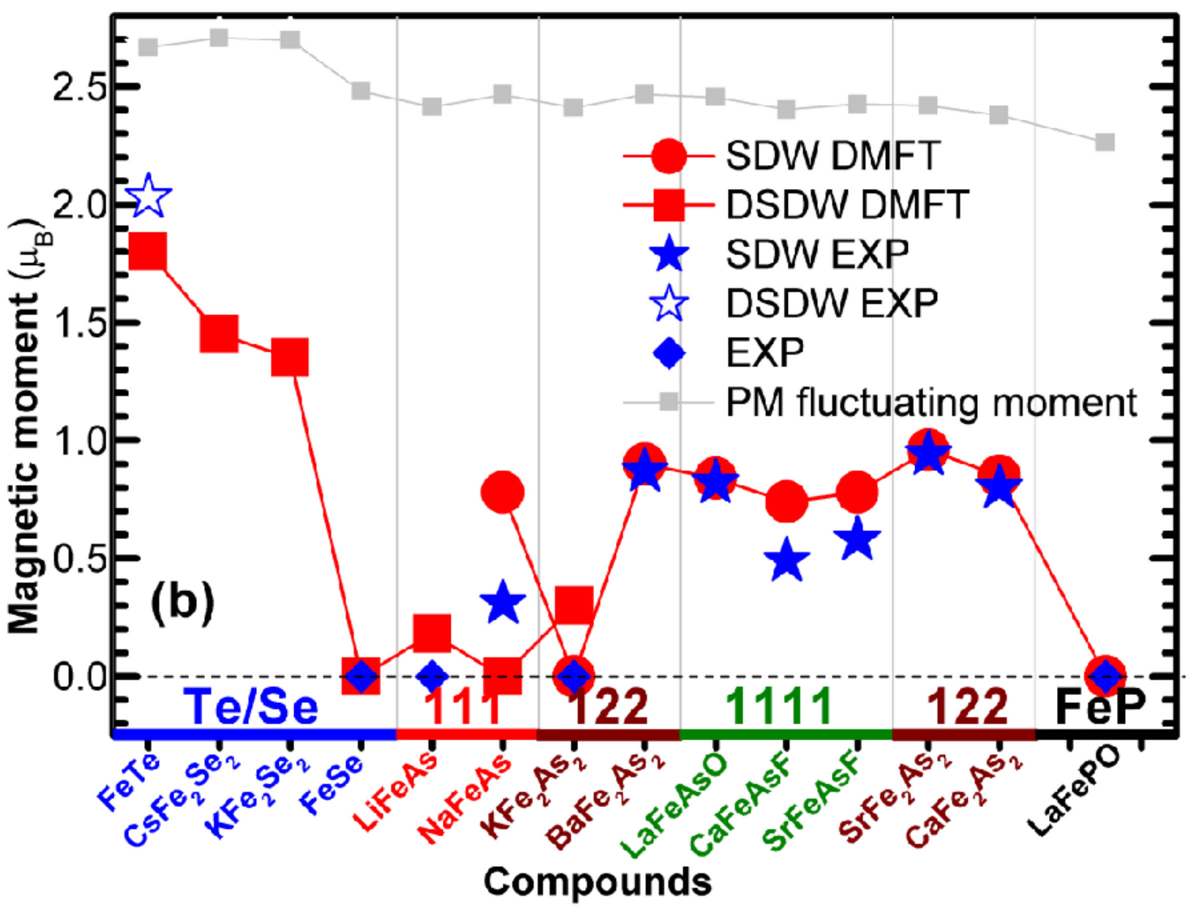


several competing effects:

- correlation strength (m*) (left versus right)
- competing ordered states (1,0),(0,1) with (1/2,1/2)
- substantial fluctuating moment remains in the ordered state

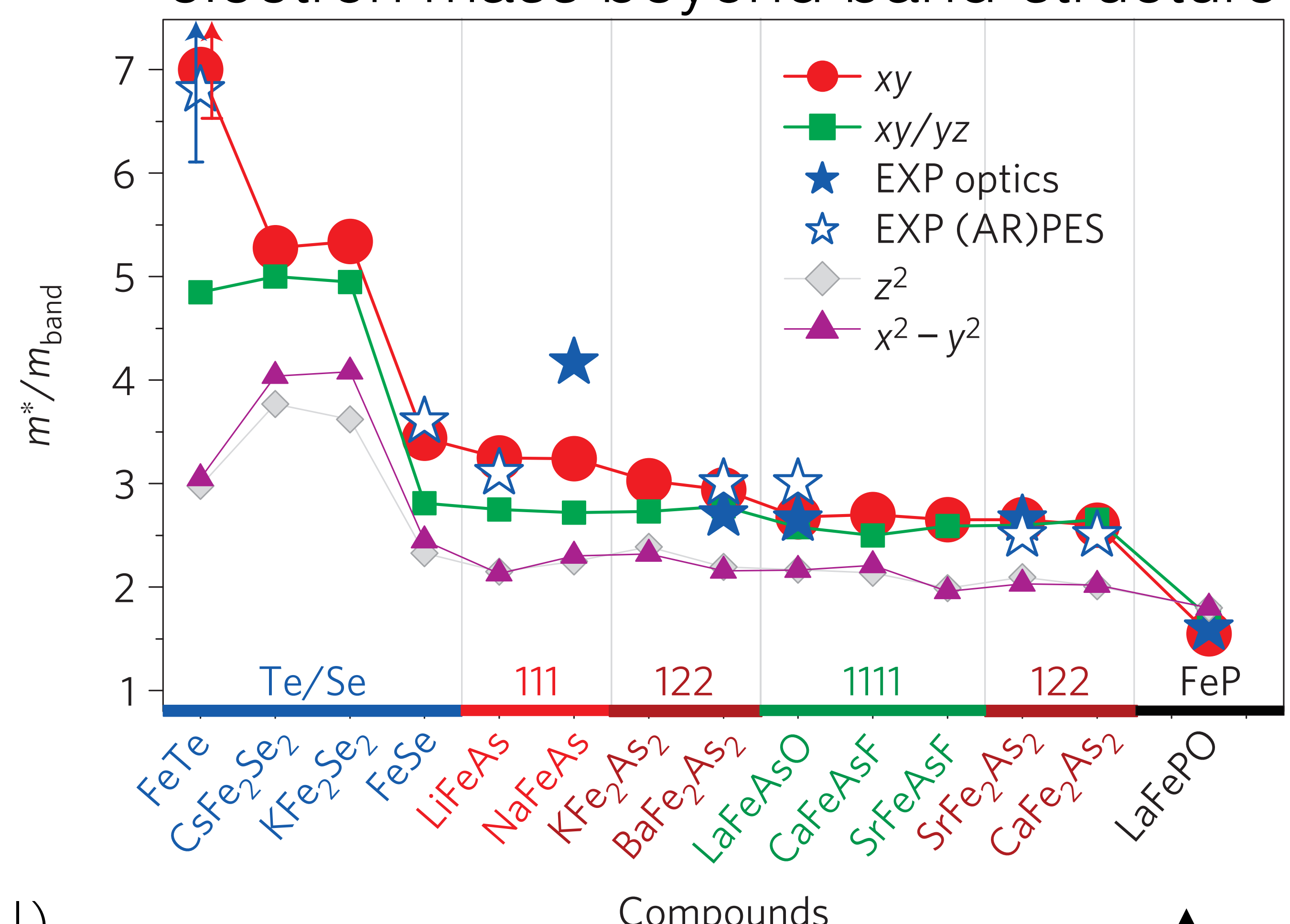
blue symbols : experiment

red symbols : DFT-eDMFT



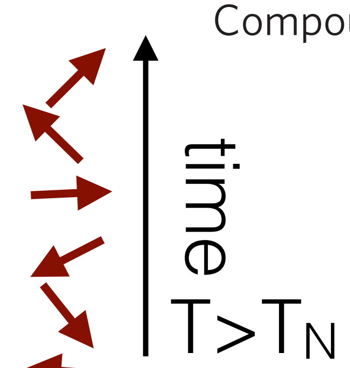
Yin, Haule & Kotliar, Nature Physics 7, 294-297 (2011).

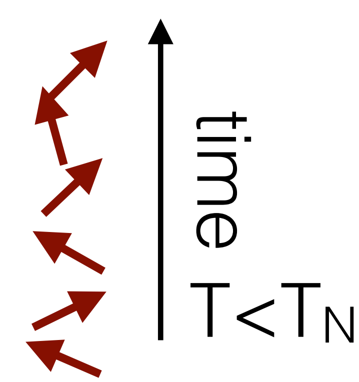
electron mass beyond band-structure



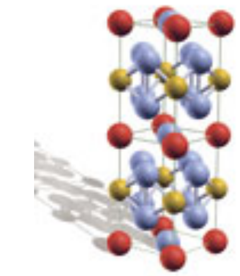
Prediction: large fluctuating moment (2-2.5μ_B) in para state

Only a part of the fluctuating moments orders.



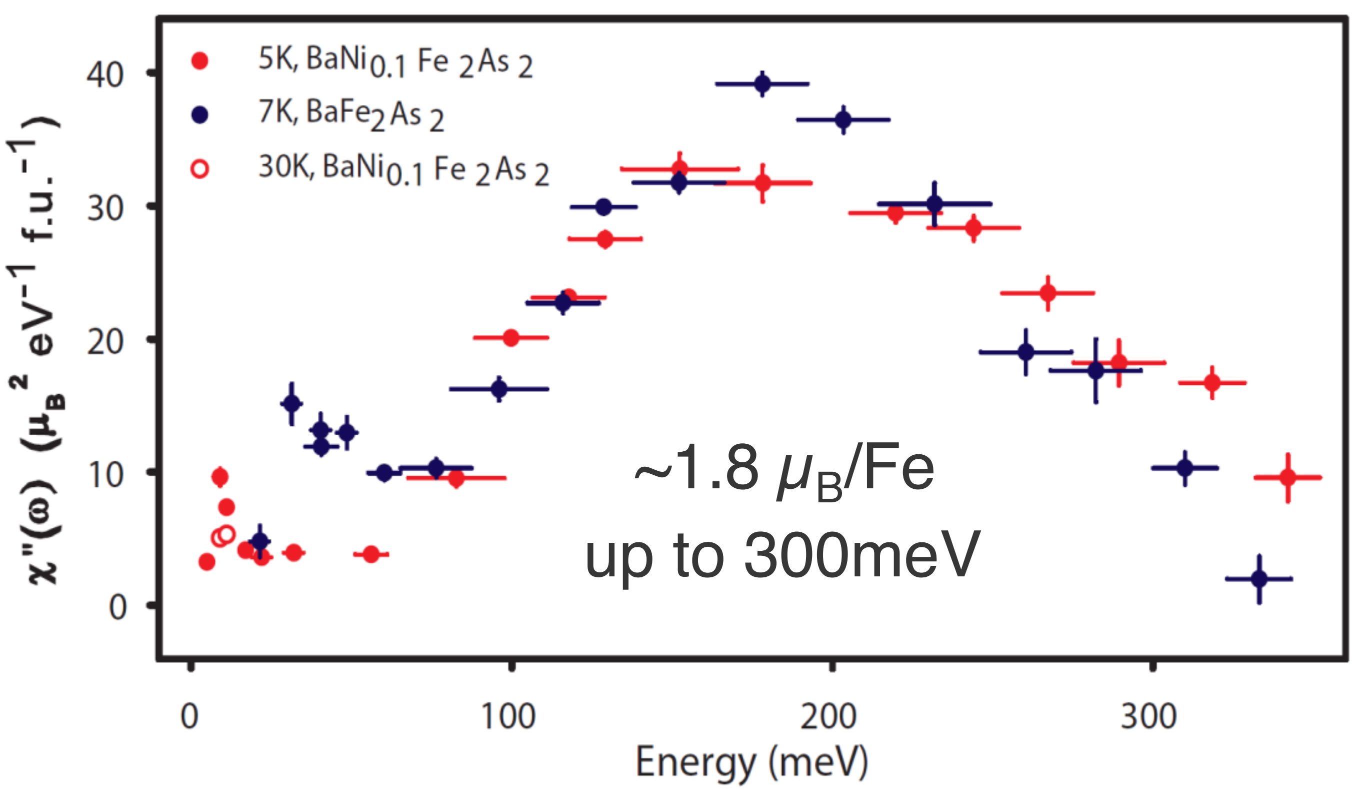


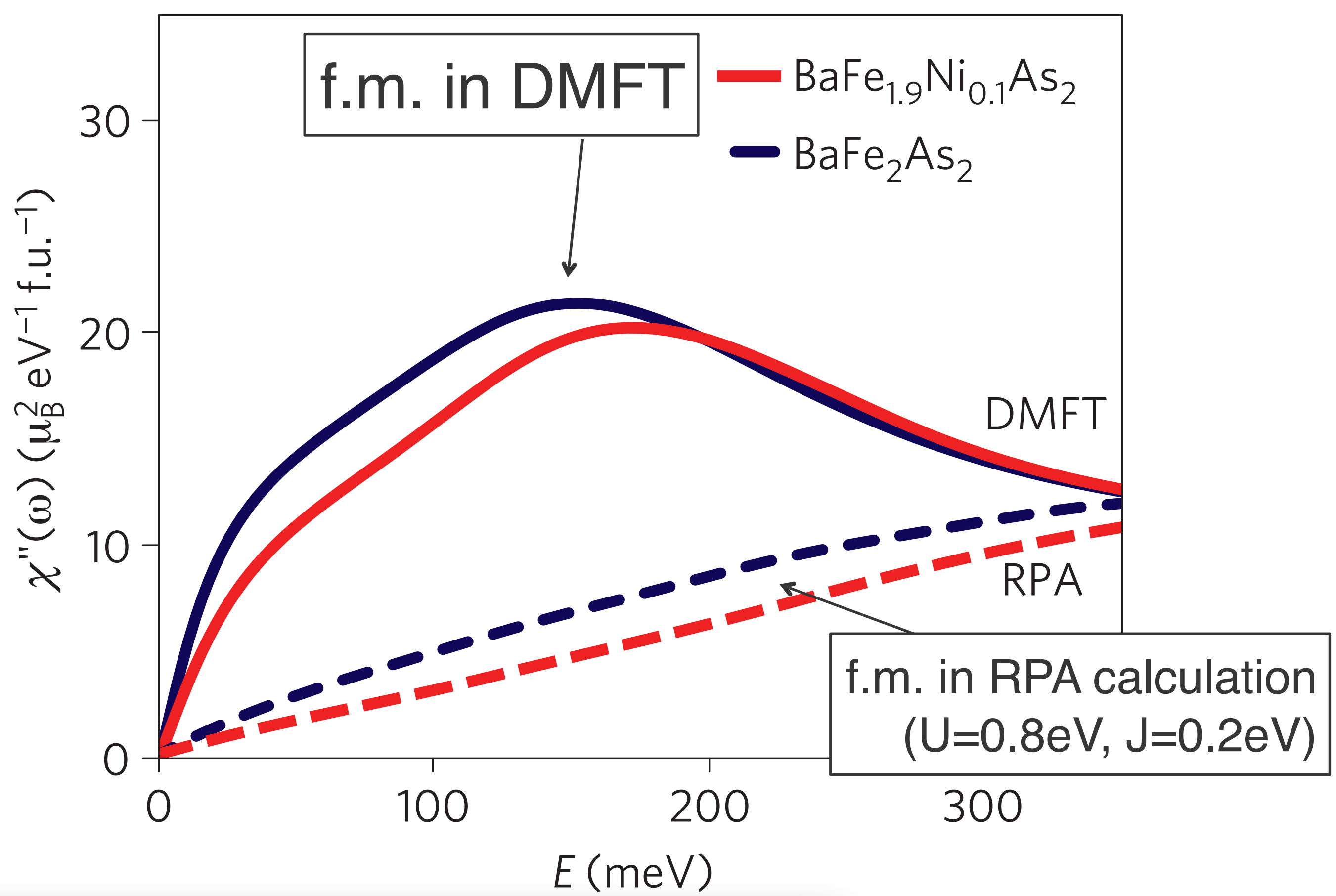
Large fluctuating moment confirmed



Fluctuating moment measured by neutrons - integral of local susceptibility:

$$\langle \mu^2 \rangle = \int \frac{d\omega}{\pi} n(\omega) \chi''(\omega)$$





Large fluctuating moment can not be explained by a purely itinerant model - property of Hundsness!

The DMFT account for a dual nature of electrons in Hund's metals: itinerant and localized nature.

M. Liu,… K. Haule, Kotliar, P.Dai, et.al., Nature Physics 8, 376-381 (2012)

Electron-phonon coupling in Hund's metals

In Fe-SC electron-phonons coupling is too weak to explain high Tc's.

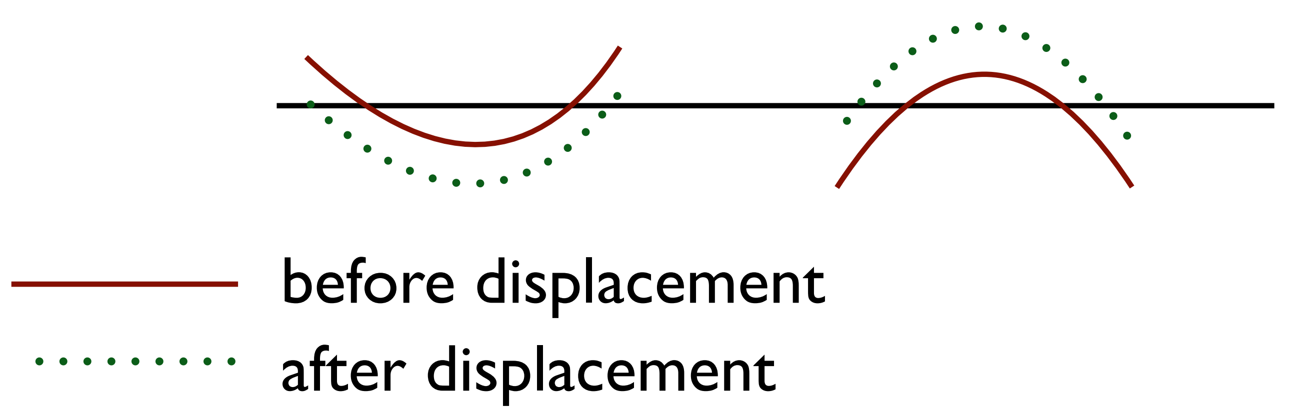
K. Haule, J. H. Shim, G. Kotliar, Phys. Rev. Lett. 100, 226402 (2008)

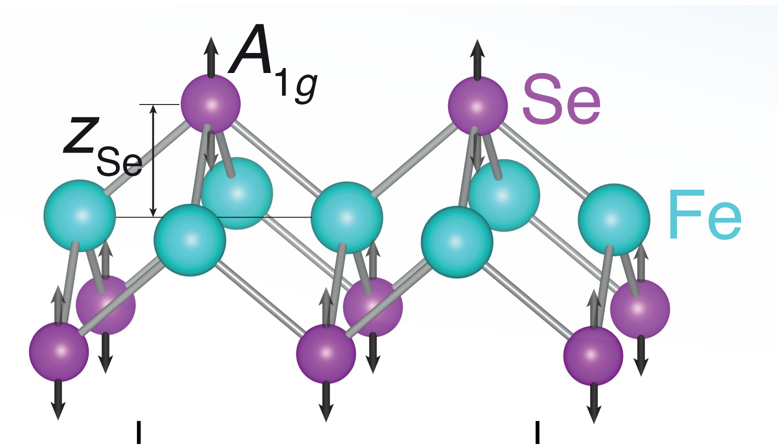
L. Boeri, O.V. Dolgov, and A. A. Golubov, PRL 101, 026403 (2008).

Nevertheless, phonons can boost Tc when cooperating with unconventional spin-mediated (correlation-driven) superconducting mechanisms.

The phonon enhancement of Tc is determined by electron-phonon coupling:

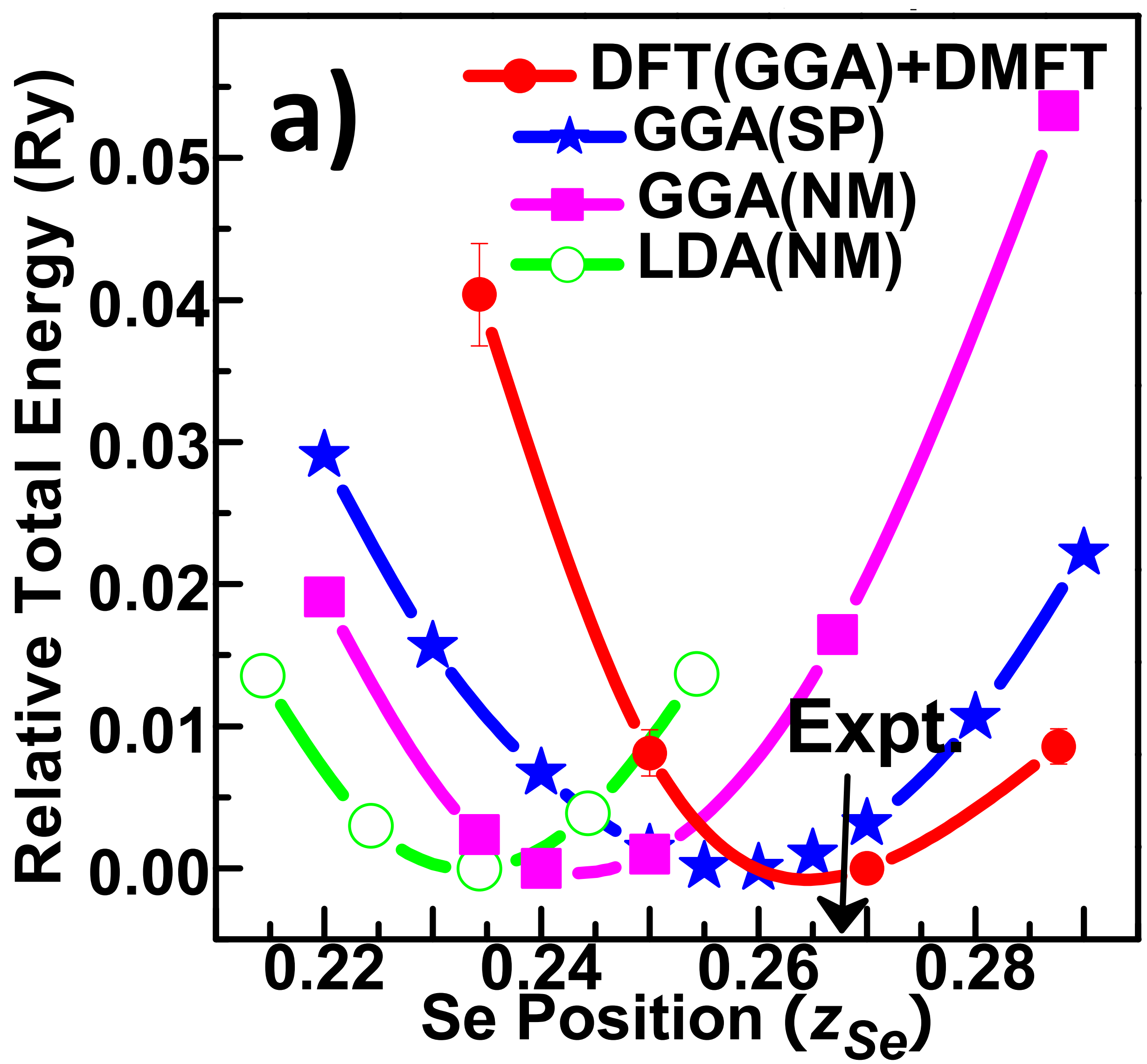
Change of the band structure due to displacement of the ions in the direction of a phonon mode.





Ion positions in fluctuating moment systems

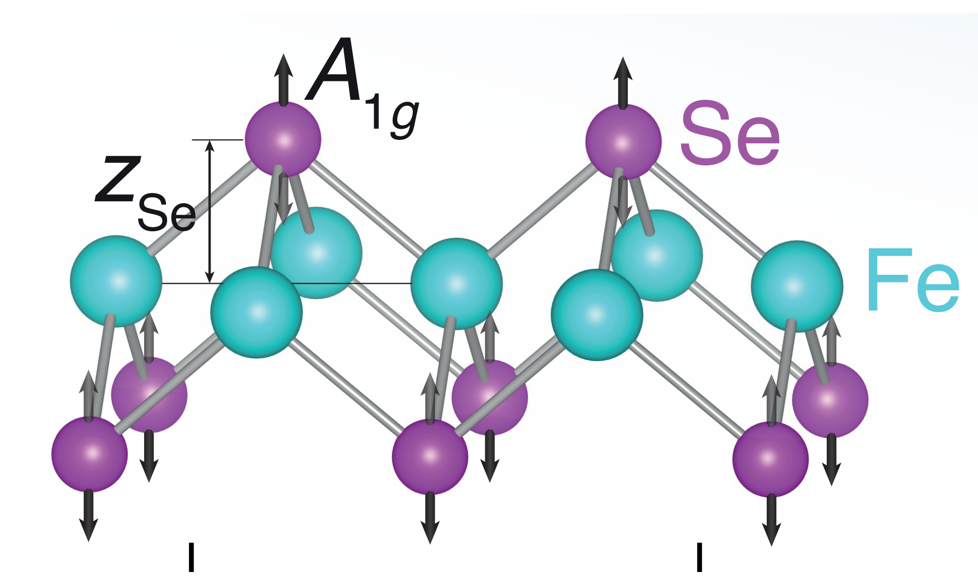
The vertical distance between Fe and Se atoms by different theories



DFT+DMFT Prediction: **Subhasish Mandal**, Cohen, & Haule, PRB **89**, 220502(R) (2014).

Correlation on Fe ion push away other ions (Se).

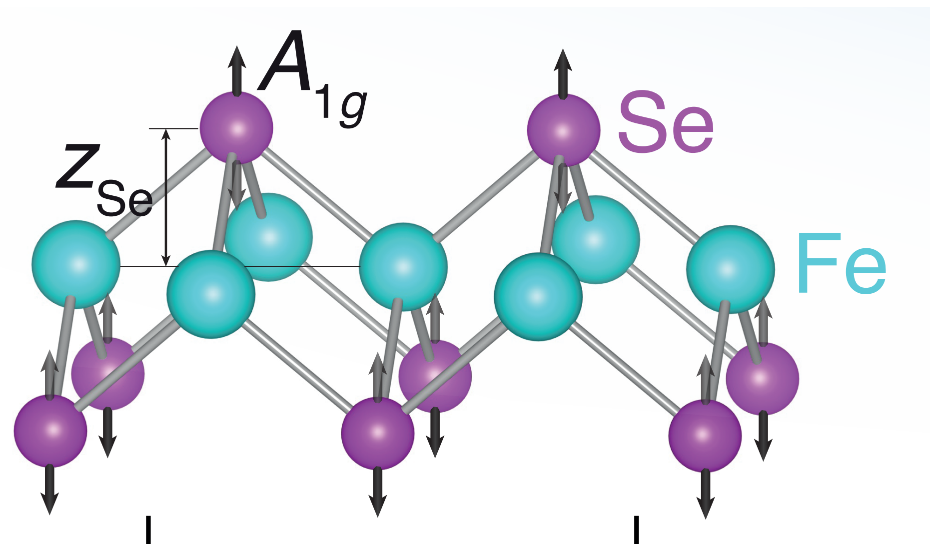
electrons in correlated orbitals need more space

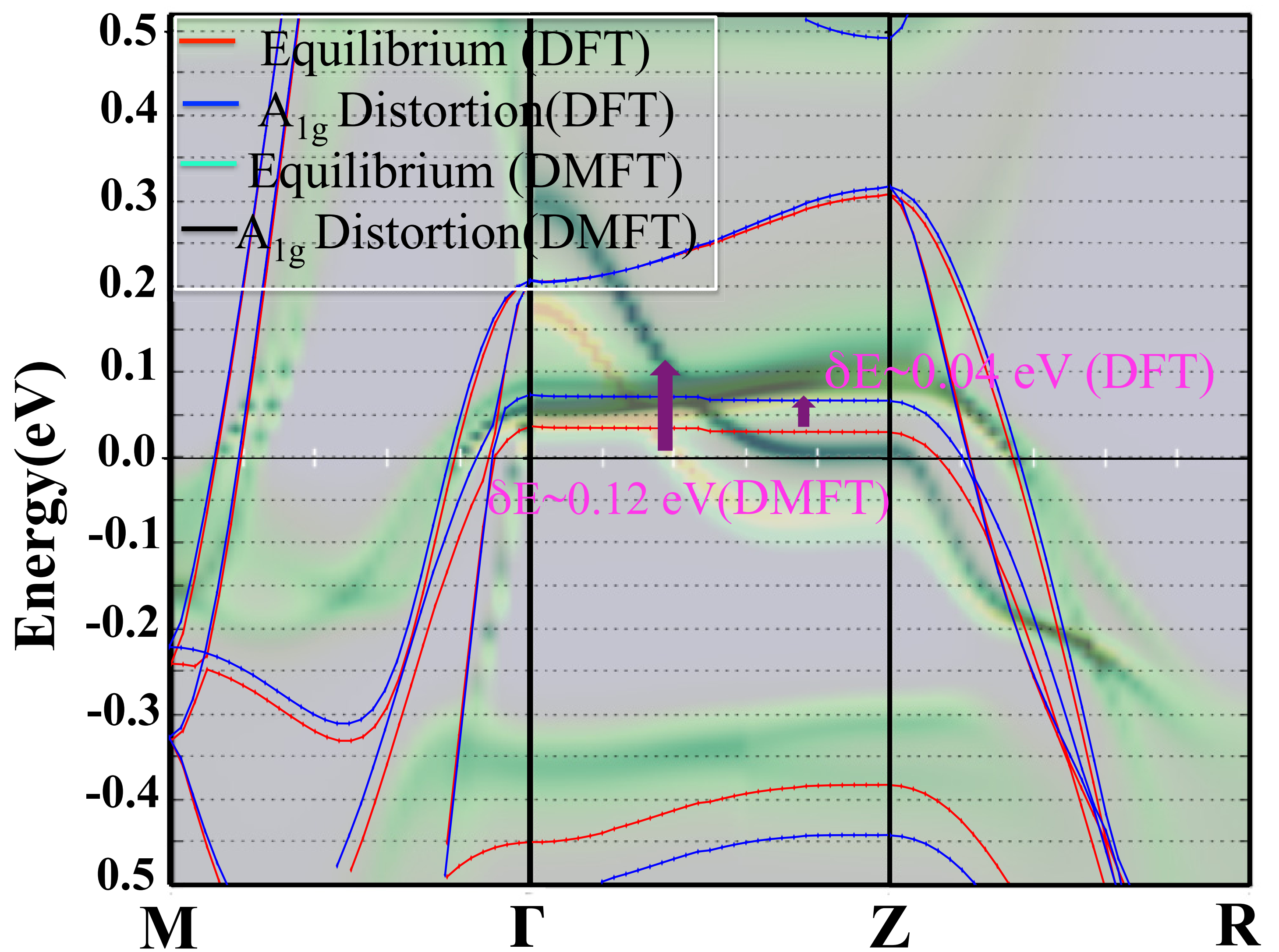


Electron-phonon coupling in fluctuating moment systems

The DMFT electronic states are much more sensitive to Se displacement than predicted by DFT.

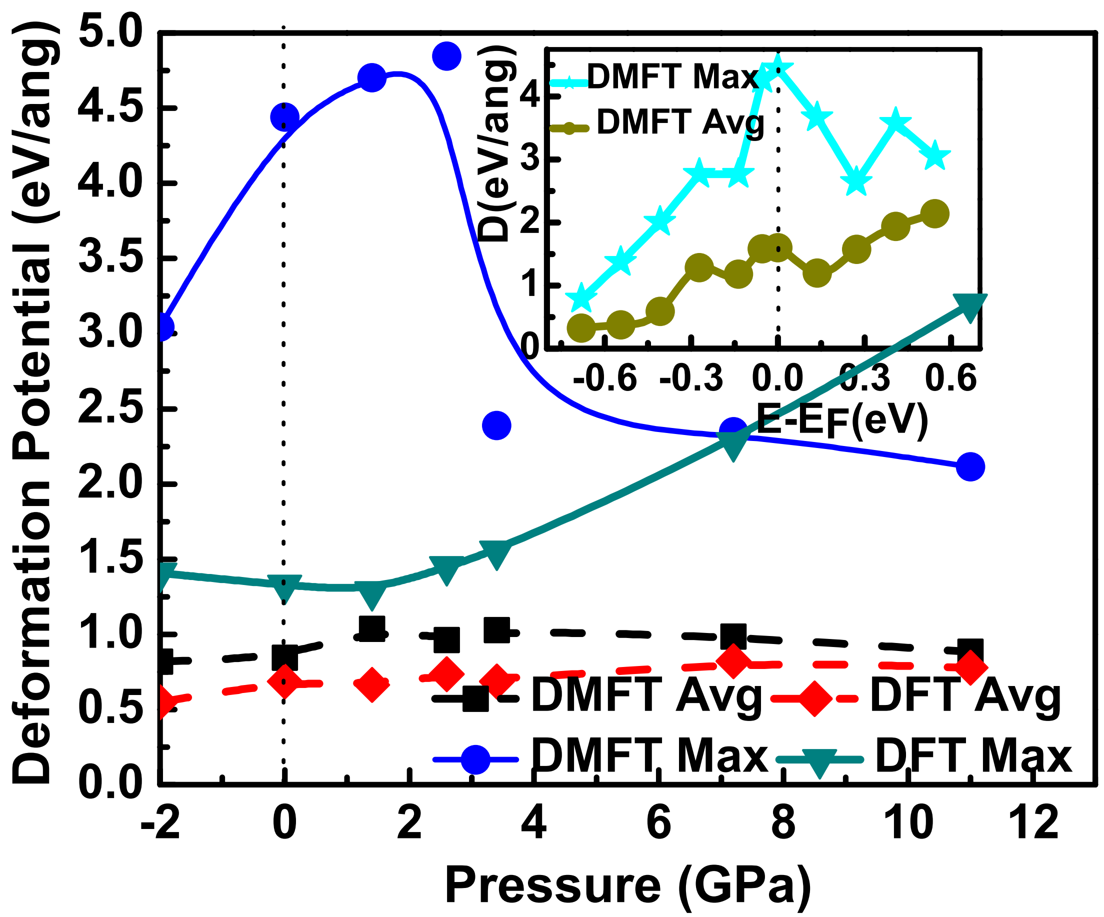
Can be understood as a feedback effect of correlations on electrons through structure.
Correlations push Se away from Fe, which reduces hybridization strength of Fe with Se, which increases correlations, and push Se further away (Kondo coupling exponentially sensitive to hybridization)





Mandal, Cohen, & Haule, PRB 89, 220502(R) (2014).

Electron-phonon coupling in FeSe



- *Pressure dependence tracks Tc of FeSe*
- *Phonons boost SC in FeSe*
- *eDMFT suggests up to one order of magnitude stronger e-ph coupling than DFT (A1g mode)*

DFT+eDMFT Prediction:
Mandal, Cohen, & Haule, PRB **89**, 220502(R) (2014).

		DFT	DFT+eDMFT (2014)	Experiment
position of Se	z_{Se}(r.l.u)	0.2456	0.27	0.2653
A1g frequency	f_{A1g}(THz)	6.5±0.3	5.7	
e-ph coupling	$\Delta E_{xz/yz}/\Delta z_{Se}$(meV/pm)	-1.6±0.2	-10.3 to -13.4	

Stanford pioneering exp: direct measurement of e-ph c.

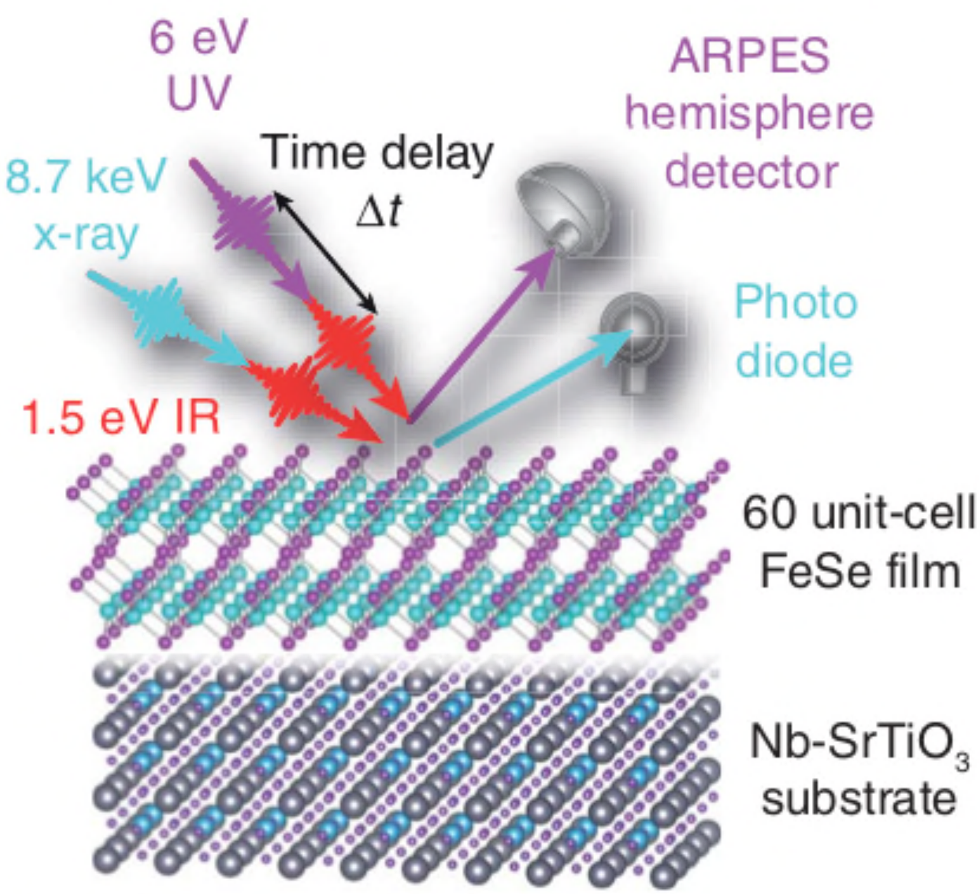
send IR pump pulse to excite A1g phonon

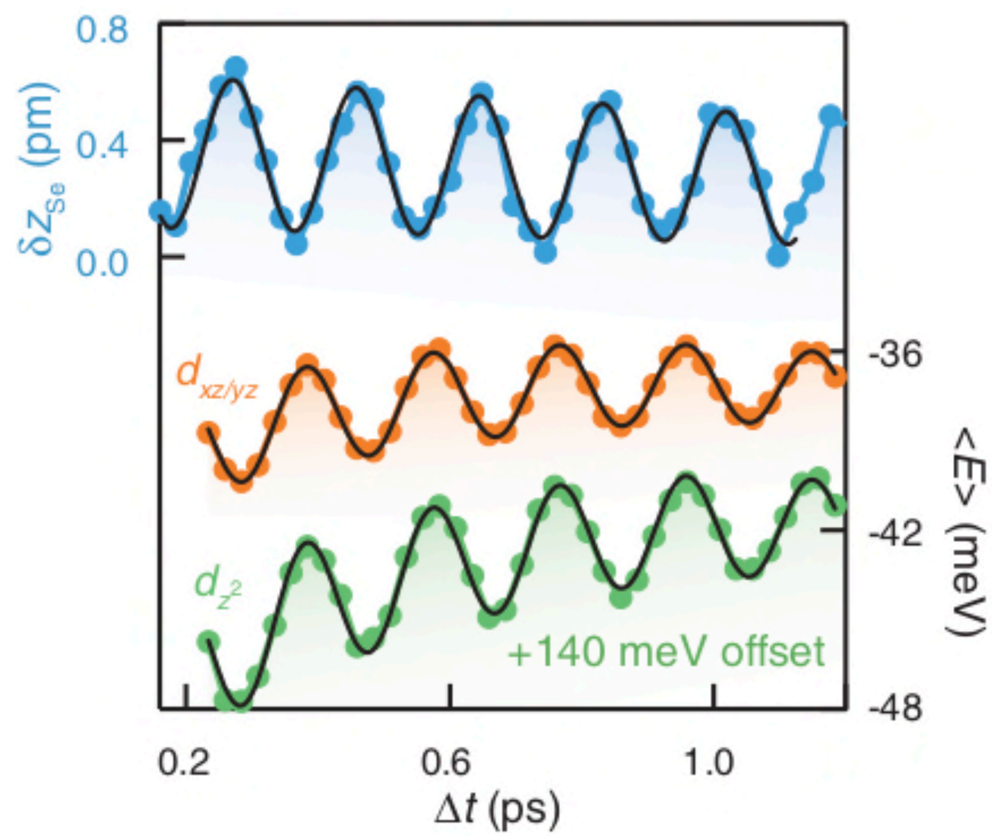
measure time resolved X-ray

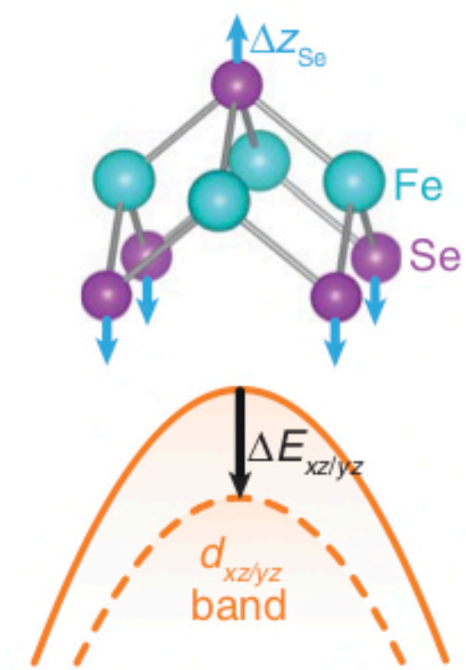
measure time resolved ARPES

Brag peak position is oscillating with A1g phonon frequency

Bands are oscillating with the same frequency.





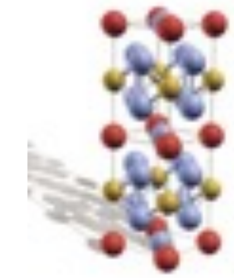


DFT+eDMFT Prediction:
Mandal, Cohen, & Haule, PRB **89**, 220502(R) (2014).

Experiment:
S. Gerber, …,Z.X. Shen et.al., Science 357, 71 (2017).

		DFT	DFT+eDMFT (2014)	Experiment (2017)
position of Se	z_{Se}(r.l.u)	0.2456	0.27	0.2653
A1g frequency	f_{A1g}(THz)	6.5±0.3	5.7	5.30±0.05
e-ph coupling	$\Delta E_{xz/yz}/\Delta z_{Se}$(meV/pm)	-1.6±0.2	-10.3 to -13.4	-13.0±2.5

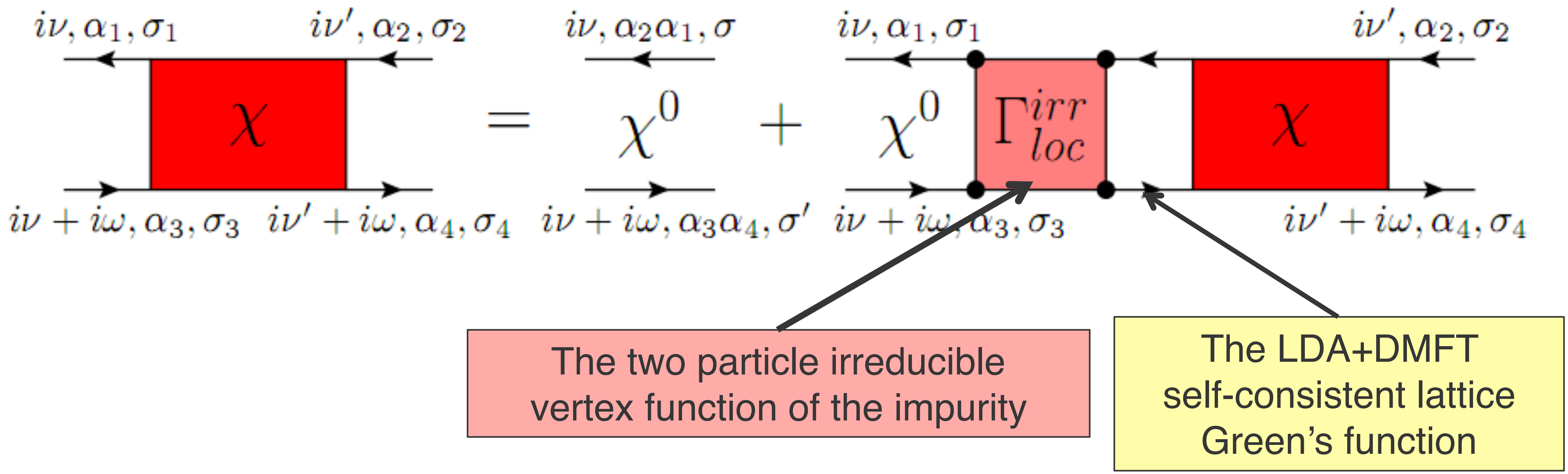
Direct confirmation of DFT+eDMFT prediction



Two particle response of Hund's metals: Dynamical structure factor

$$S(\mathbf{q},\omega) = \frac{\chi''(\mathbf{q},\omega)}{1 - e^{-\hbar\omega/k_B T}}$$

Computed from the two particle response functions
using the fact that the irreducible vertex is local.

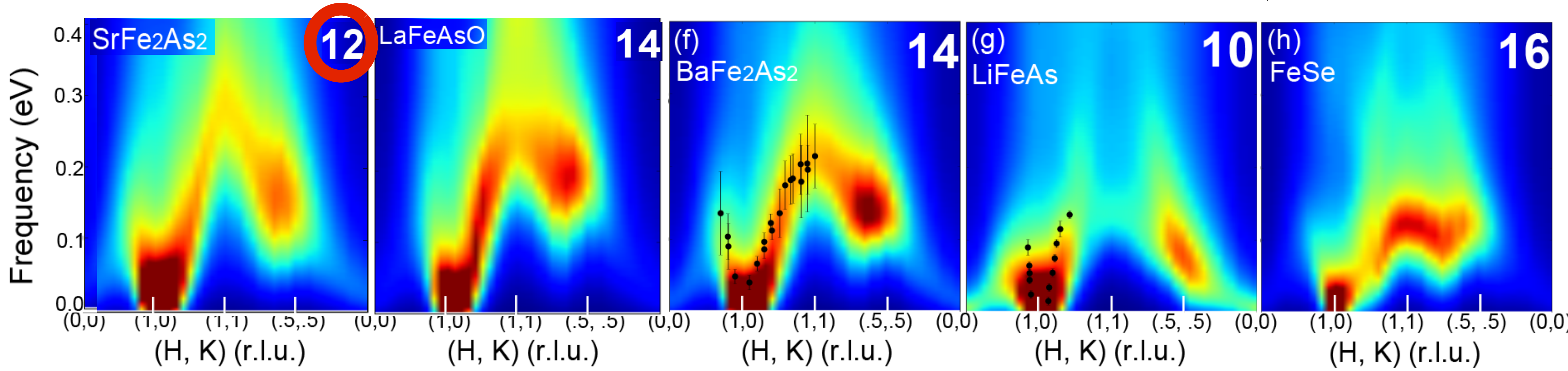


H. Park, K. Haule, G. Kotliar, PRL 107, 137007 (2011)

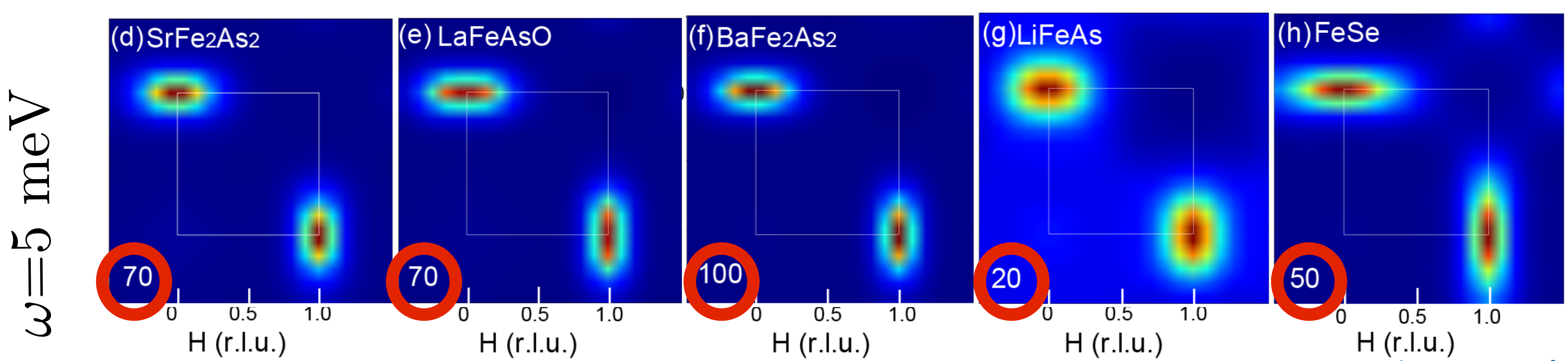
Dynamical structure factor of Fe superconductors

High-Tc compounds

$T_C^{max} = 37K$ $T_C^{max} = 43K$ $T_C^{max} = 39K$ $T_C = 18K$ $T_C^{max} = 37K$



high Tc: **Strong commensurate low energy excitations**, strong high energy dispersive exc.



Maximum intensity

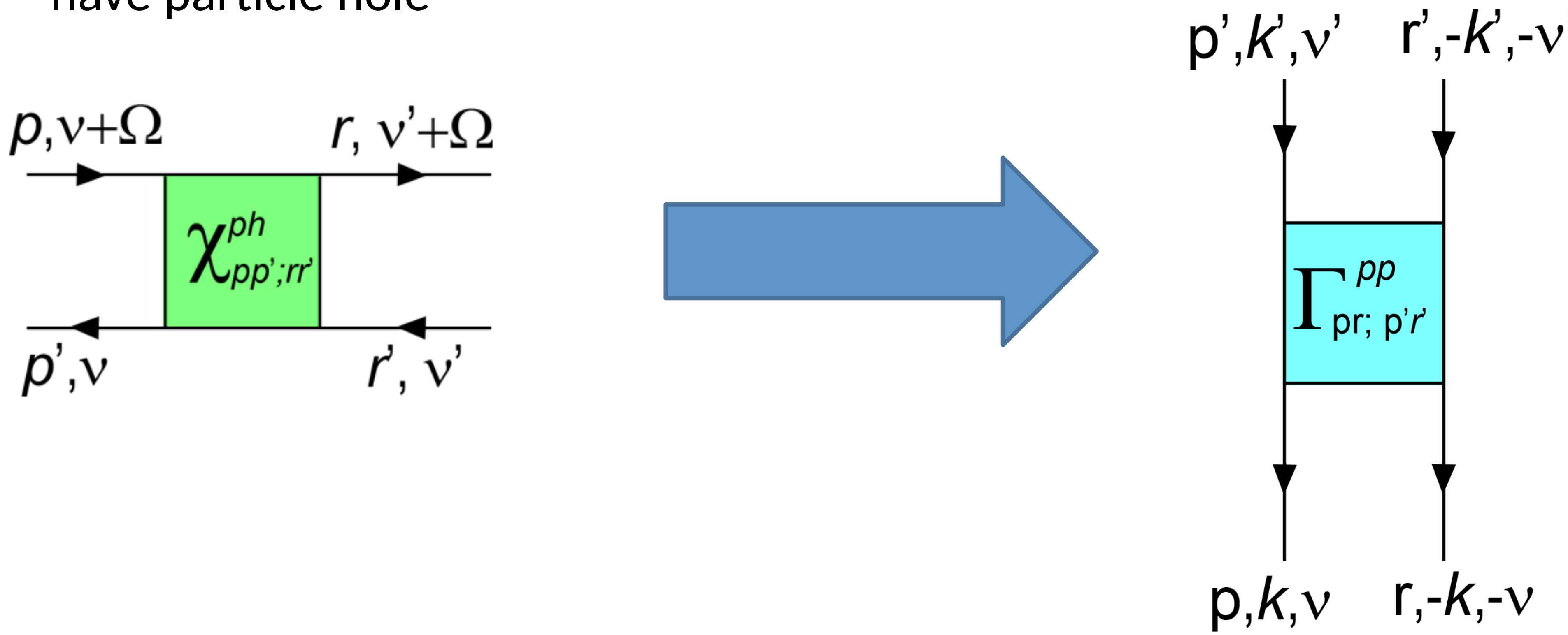
Z.P. Yin, K. Haule, G. Kotliar, Nature Physics 10, 845 (2014)

Particle-particle irreducible vertex

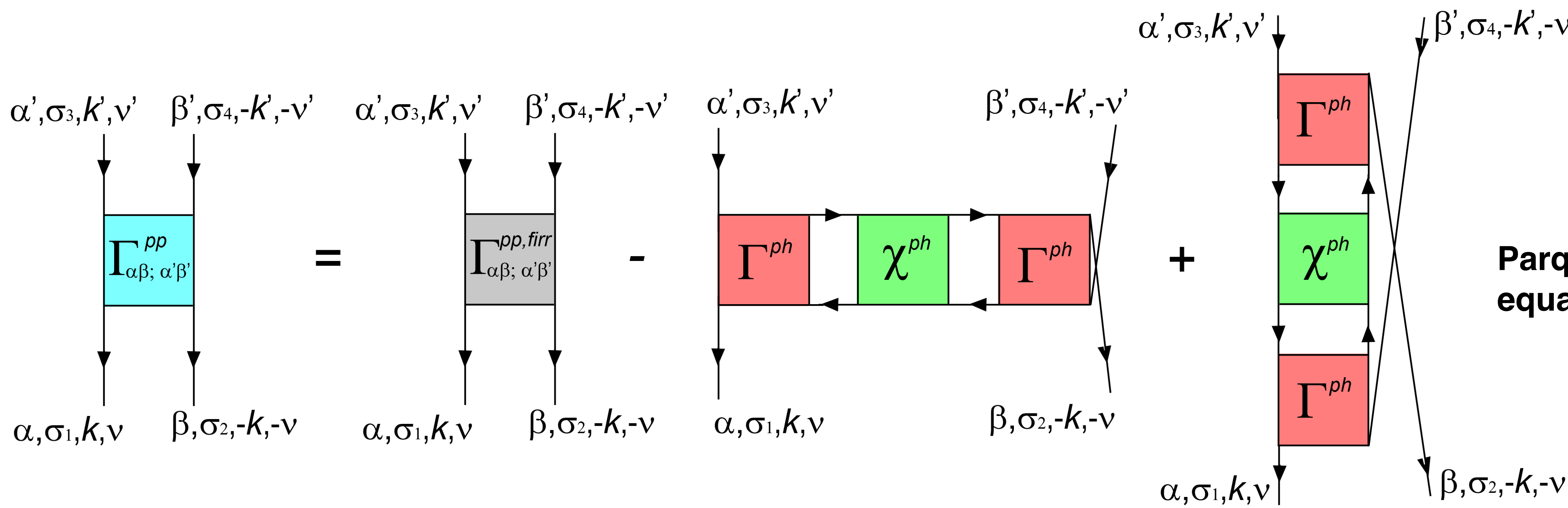
Z.P. Yin, K. Haule, G. Kotliar, Nature Physics 10, 845 (2014)

have particle hole

want



We can compute particle-particle vertex using parquet equations:

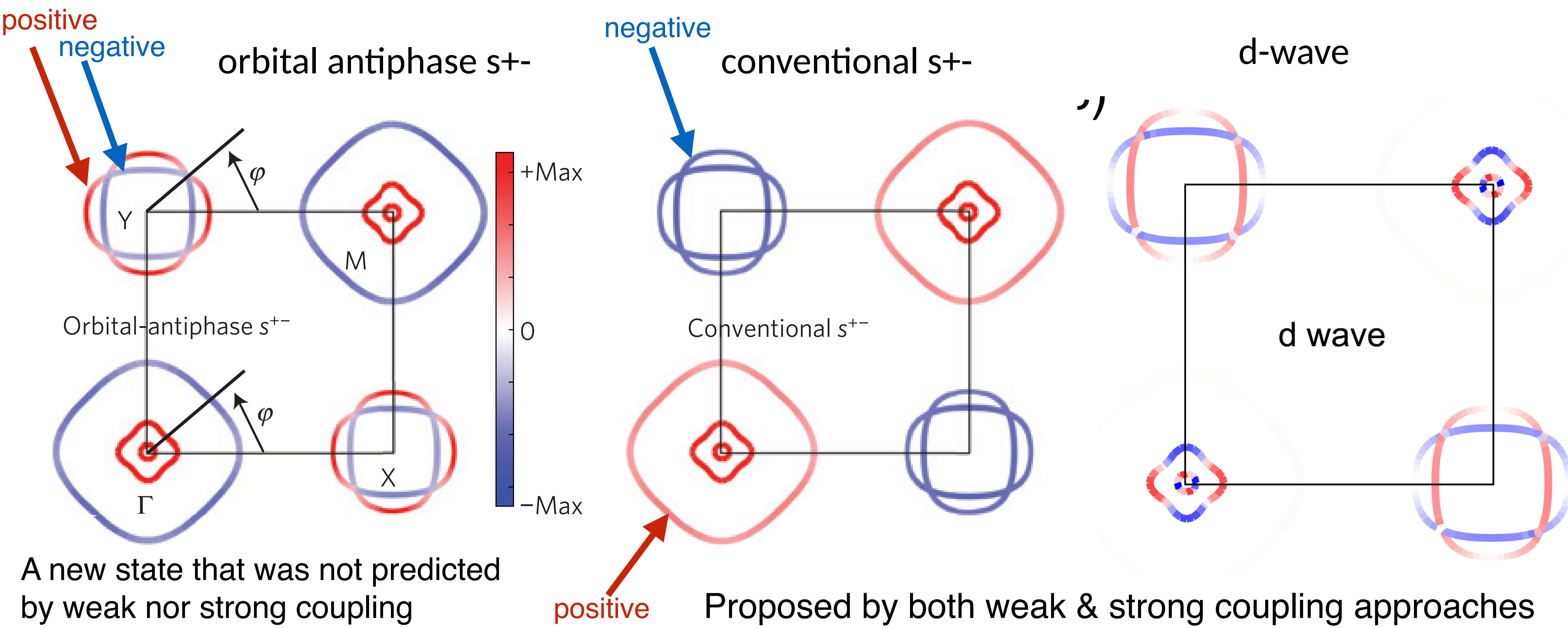


Parquet equations

Superconducting and pairing symmetry

Elliashberg equastions solved in orbital/momentum space with ab-initio two particle vertex

$$-k_BT\sum_{k'\nu'\alpha'\beta'\gamma\delta}\Gamma^{pp,s}(\alpha\beta k\nu;\alpha'\beta'k'\nu')\chi^{0,pp}_{\alpha'\beta'\gamma\delta}(k'\nu')\Delta_{\gamma\delta}(k'\nu')=\lambda\Delta_{\alpha\beta}(k\nu)$$



A new state that was not predicted by weak nor strong coupling theories.

Proposed by both weak & strong coupling approaches

Closely competing states:
Depending on the details, system can change from one to another

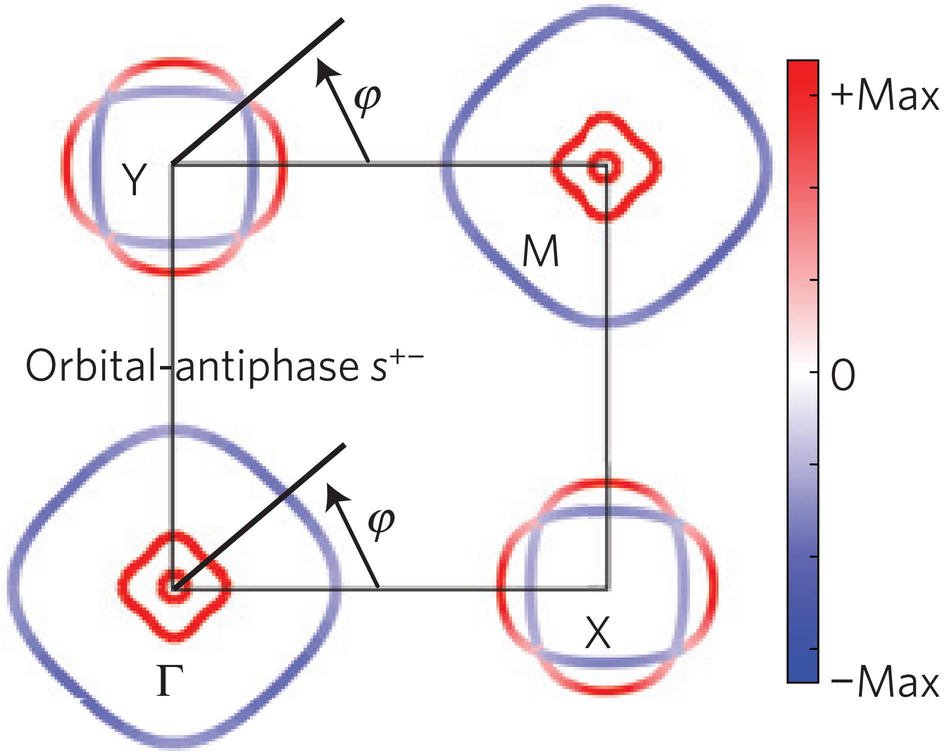
Z.P. Yin, K. Haule, G. Kotliar, Nature Physics 10, 845 (2014)

Example: LiFeAs, orbital antiphase

P. Zhang, ... H. Ding et.al., PRX 4, 031001 (2014)

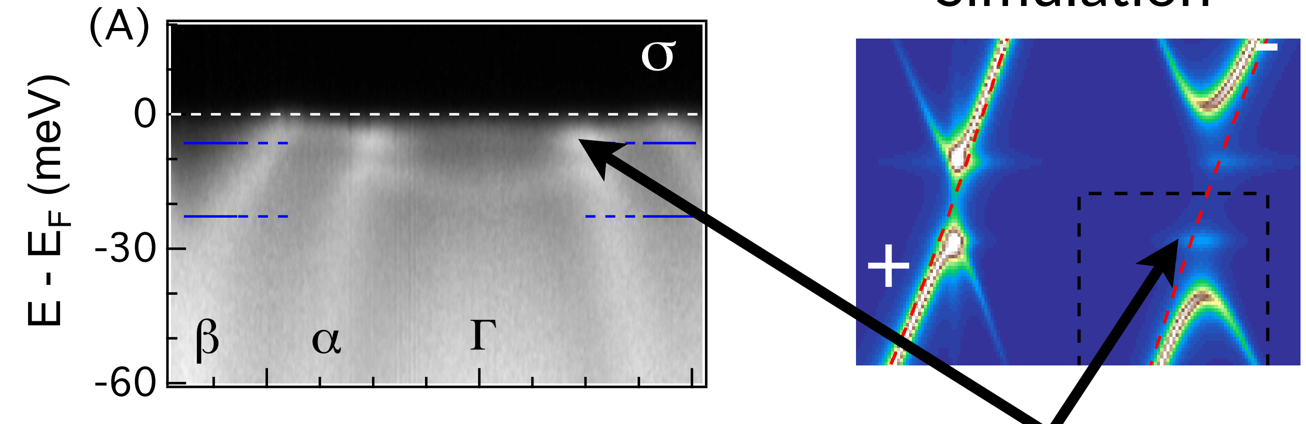
P.Richard,…H Ding et.al., J. Phys.: Cond. Matt. 27, 293203 (2015)

weak scatterer and in-gap state:Observation of momentum-confined in-gap impurity state in $Ba_{0.6}K_{0.4}Fe_2As_2$: evidence for anti-phase s± pairing.

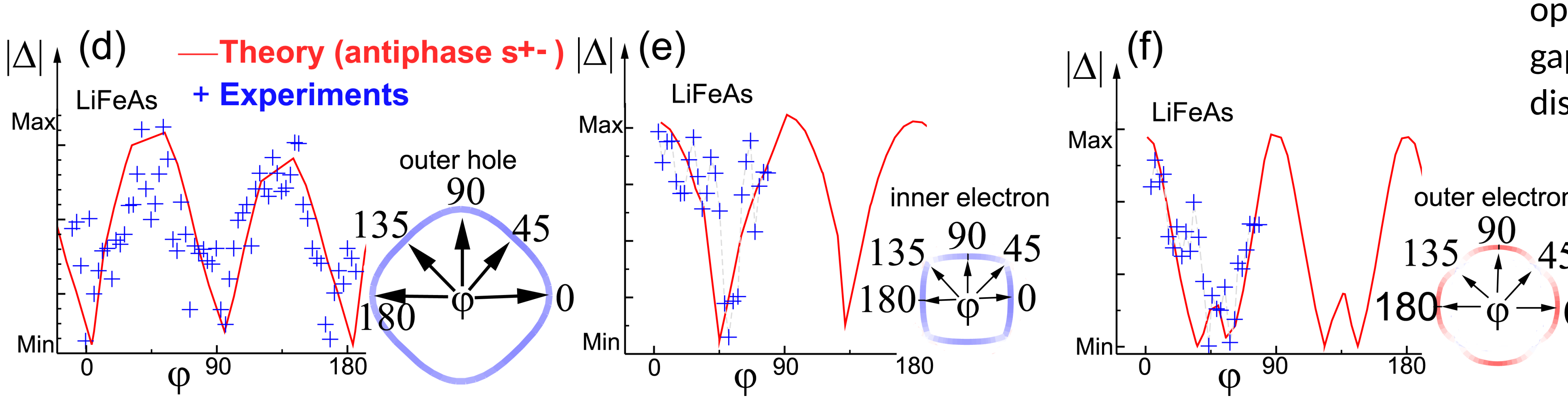


ARPES

Simulation



Two nearby bands of opposite gap sign - in gap state with peculiar dispersion



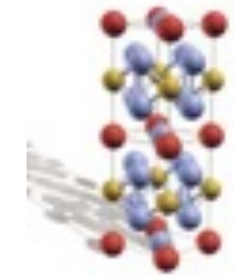
Orbital-antiphase matches best with ARPES
conventional s+- does not match

Z.P. Yin, K. Haule, G. Kotliar, Nature Physics 10, 845 (2014)

Thank you for your attention!

Origin of fluctuating local moments

Low Energy (Schrieffer-Wolff)



Z.Yin, KH, G. Kotliar, PRB 86, 195141 (2012).

Analyzing the quantum impurity model in the d^6 configuration:

low energy effective model has these terms:

spin-spin: $$J_1 \vec{S} \sum_{a,\sigma\sigma'} c^\dagger_{a\sigma} \vec{\sigma}_{ss'} c_{a\sigma'}$$

orbital-orbital: $$J_2 \vec{\Lambda} \sum_{ab,\sigma} c^\dagger_{a\sigma} \vec{\lambda}_{ab} c_{a\sigma}$$

cross term: orbital+spin-orbital+spin $$J_3 \left(\vec{\Lambda} \otimes \vec{S}\right) \sum_{ab,\sigma\sigma'} c^\dagger_{a\sigma} \left(\vec{\lambda}_{ab} \otimes \vec{\sigma}_{ss'}\right) c_{b\sigma'}$$

Gell-Mann matrices for SU(3)

$$\lambda_1 = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad \lambda_2 = \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad \lambda_3 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

$$\lambda_4 = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix} \quad \lambda_5 = \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix}$$

$$\lambda_6 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix} \quad \lambda_7 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix} \quad \lambda_8 = \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}$$

Effective Kondo coupling for J_H=0 are AFM:

$$J_1 = 1/3 J_0$$ well screened spins

$$J_2 = 1/4 J_0$$

$$J_3 = 1/2 J_0$$

well screened orbital fluctuations

Effective Kondo coupling for J_H=∞

$$\boxed{J_1 = -1/9 J_0}$$ poorly screened spins -> local moments

$$J_2 = 1/3 J_0$$

$$J_3 = 1/3 J_0$$

couple to orbital and make also orbital fluctuations slower