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The Physics of Strongly Correlated Insulators, Metals, and Superconductors
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Hund's metals, explained

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EDUCATION SCIENCE INNOVATION

Hund's metals

LETTERS
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nature materials

Kinetic frustration and the nature of the magnetic and paramagnetic states in iron pnictides and iron chalcogenides

Z. P. Yin[1,2]★, K. Haule[1] and G. Kotliar[1]

The iron pnictide and chalcogenide compounds are a subject of intensive investigations owing to their surprisingly high temperature superconductivity[1]. They all share the same basic building blocks, but there is significant variation in their physical properties, such as magnetic ordered moments, effective masses, superconducting gaps and transition temperature (T_c). Many theoretical techniques have been applied to individual compounds but no consistent description of the microscopic origin of these variations is available[2]. Here we carry out a comparative theoretical study of a large number of iron-based compounds in both their magnetic and paramagnetic states. Taking into account correlation effects and realistic band structures, we describe well the trends in all of the physical properties such as the ordered moments, effective masses and Fermi surfaces across all families of iron compounds, and find them to be in good agreement with experiments. We trace variation in physical properties to variations in the key structural parameters, rather than changes in the screening of the Coulomb interactions. Our results also provide a natural explanation of the strongly Fermi-surface-dependent superconducting gaps observed in experiments[3].

The iron pnictides/chalcogenides are Hund's metals. In these systems the Coulomb interaction among the electrons is not strong enough to fully localize them, but it significantly slows them down, such that low-energy emerging quasiparticles have a substantially enhanced mass[4]. This enhanced mass emerges not because of the Hubbard interaction U, but because of the Hund's rule interactions that tend to align electrons with the same spin but different orbital quantum numbers when they find themselves on the same iron atom[4].

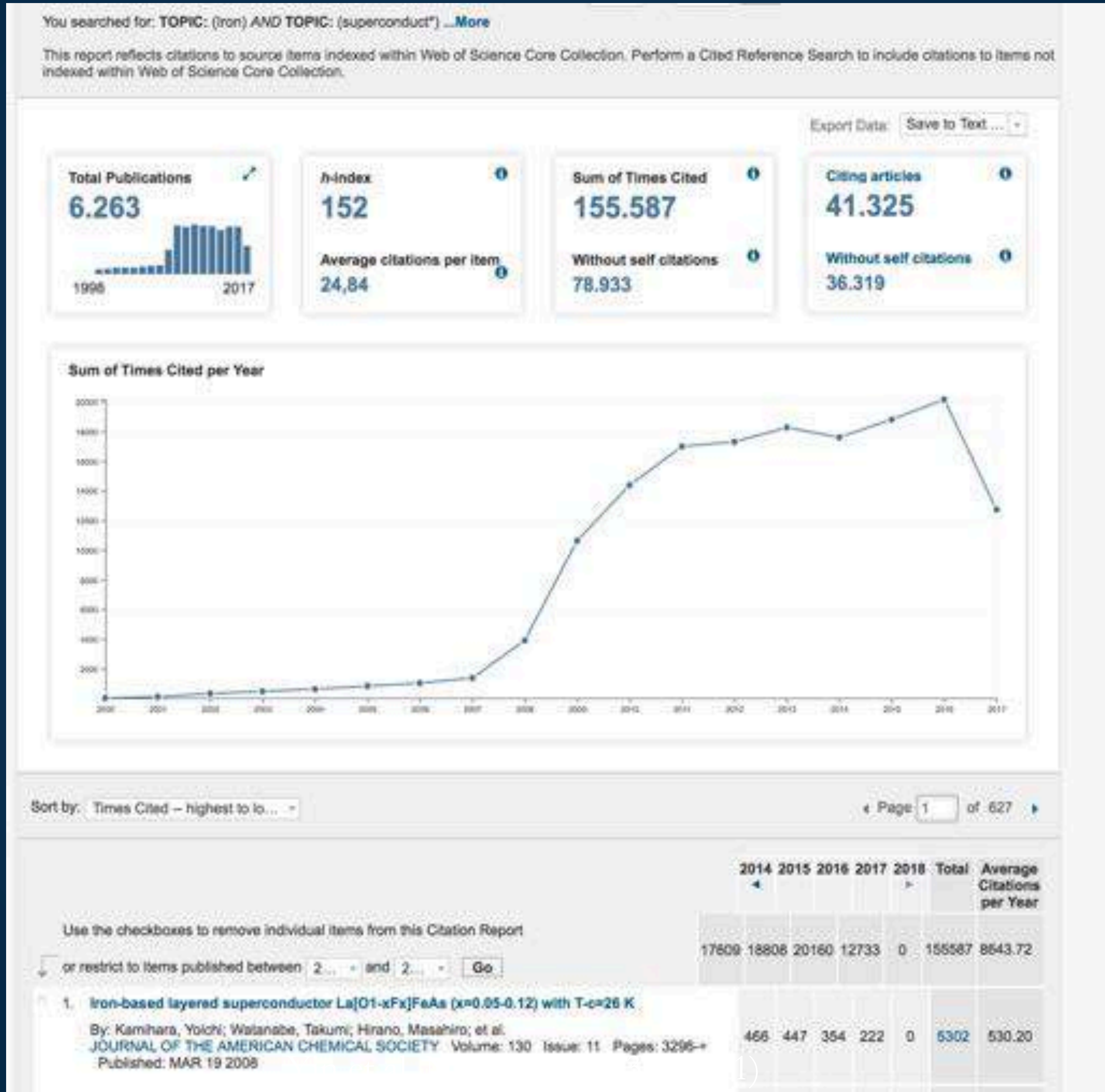
Although critical long-wavelength fluctuations certainly play a role near the phase-transition lines, we will show that the local fluctuations on the iron atom can account for the correct trend of magnetic moments and correlation strength in iron pnictide/chalcogenide layered compounds.

Using the combination of DFT and dynamical mean field theory (DFT + DMFT) (see Supplementary Information for details), we studied two different real-space orderings, the SDW ordering, characterized by wave vector $(\pi, 0, \pi)$ (this vector is written in coordinates with one iron atom per unit cell), which is experimentally found in iron arsenide compounds, and $(\pi/2, \pi/2, \pi)$ ordering, denoted by the double-stripe SDW (DSDW). The latter was found experimentally in FeTe. Figure 1a shows our theoretical results for the ordered moment in both phases together with experimentally determined values[6–13] from across all known families of iron-based superconducting compounds. There is an overall good agreement between theory and experiment; in particular, LaFePO is predicted to be non-magnetic, most 1111 and 122 compounds have an ordered moment below 1.0 μ_B (ref. 14) and FeTe orders with a DSDW moment of 2.1 μ_B.

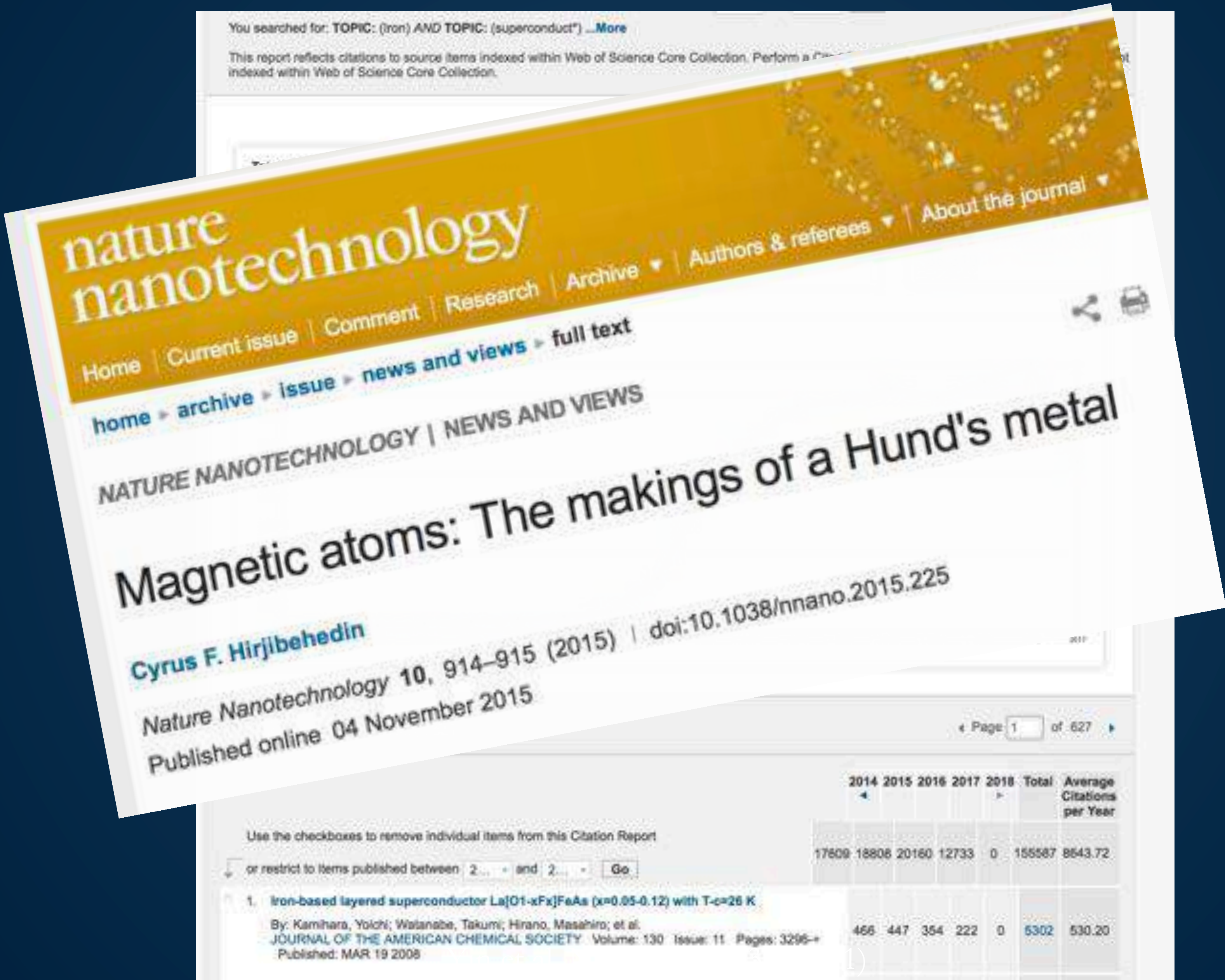
We now explain the variation of the ordered moment in terms of real-space and momentum-space concepts. The size of the fluctuating local moment, which can be extracted from neutron scattering experiments, gives an upper bound to the size of the ordered magnetic moment and is also plotted in Fig. 1a. It was computed from the atomic histogram in Fig. 2c, which shows the percentage of time the iron 3d electrons spend in various atomic configurations when the system is still in its paramagnetic state. Only high-spin states, which carry a large weight as a result of the Hund's rule coupling in iron, are shown (see also Supplementary

Yin et al, Nat Mat 10, 932 (2011)

Hund's metals



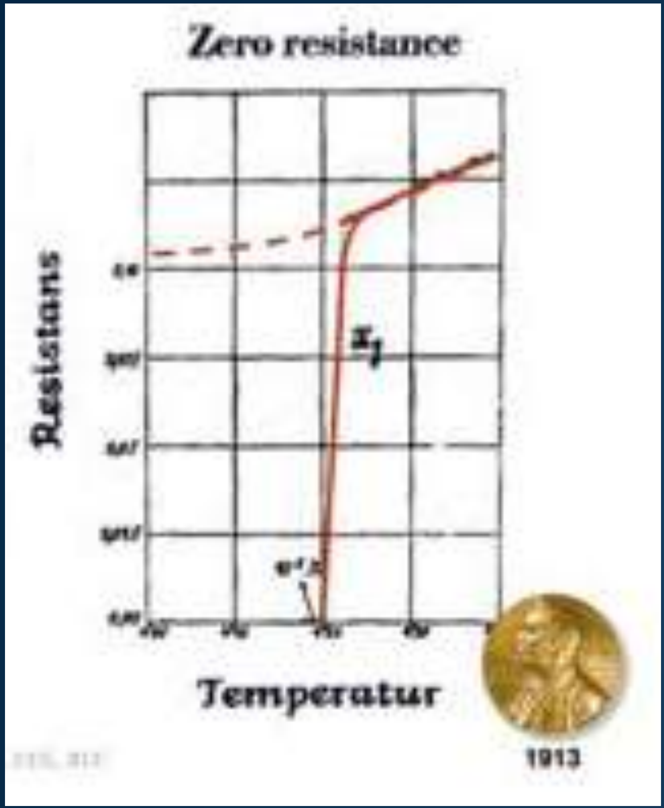
Hund's metals

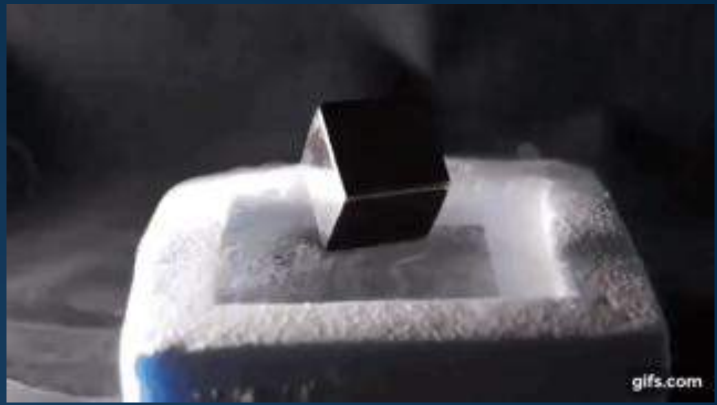


Hund’s metals

Hund’s metals

Superconducting materials



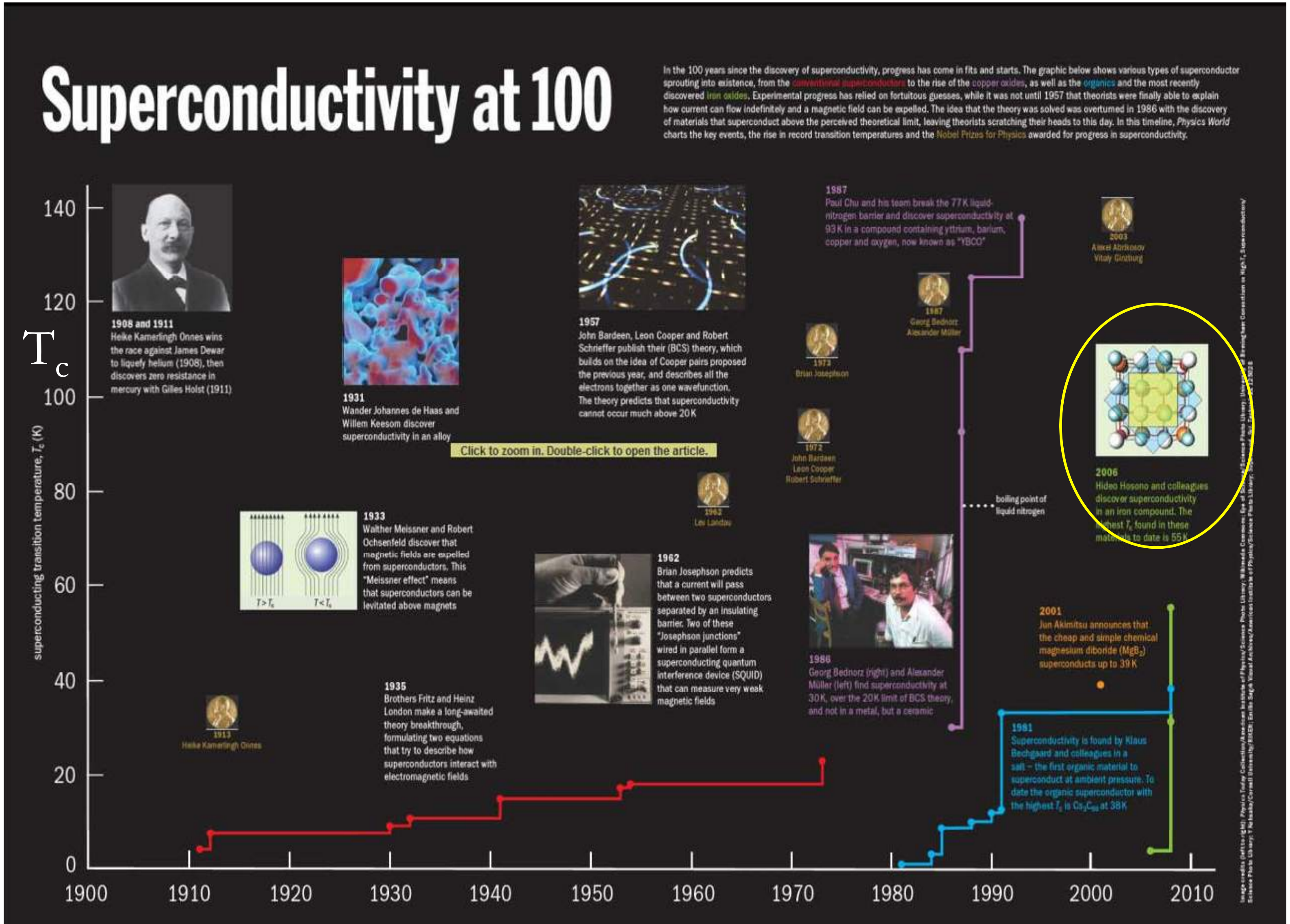


Magnetic Resonance Imaging

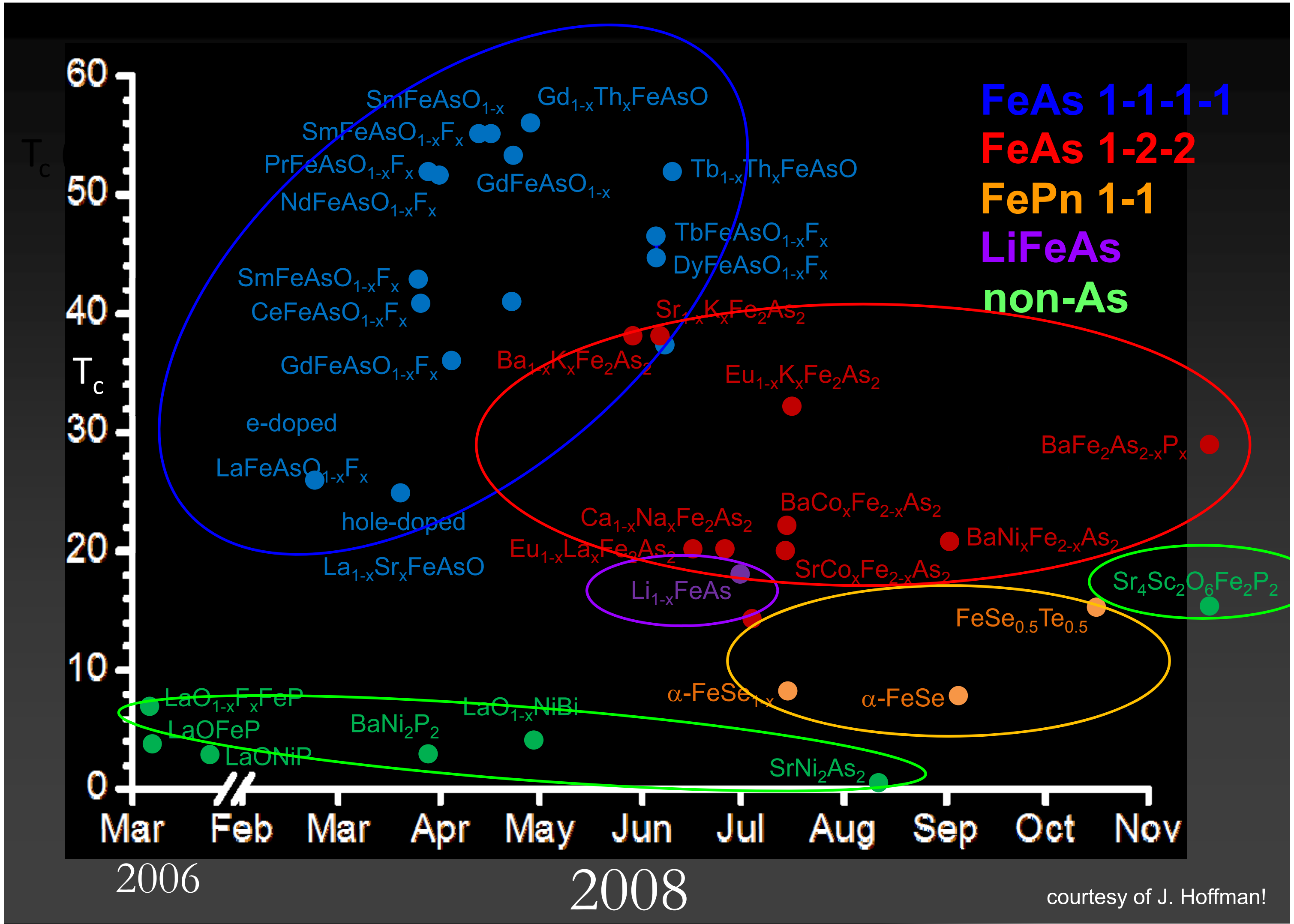
Energy transportation

MAGLEV technology

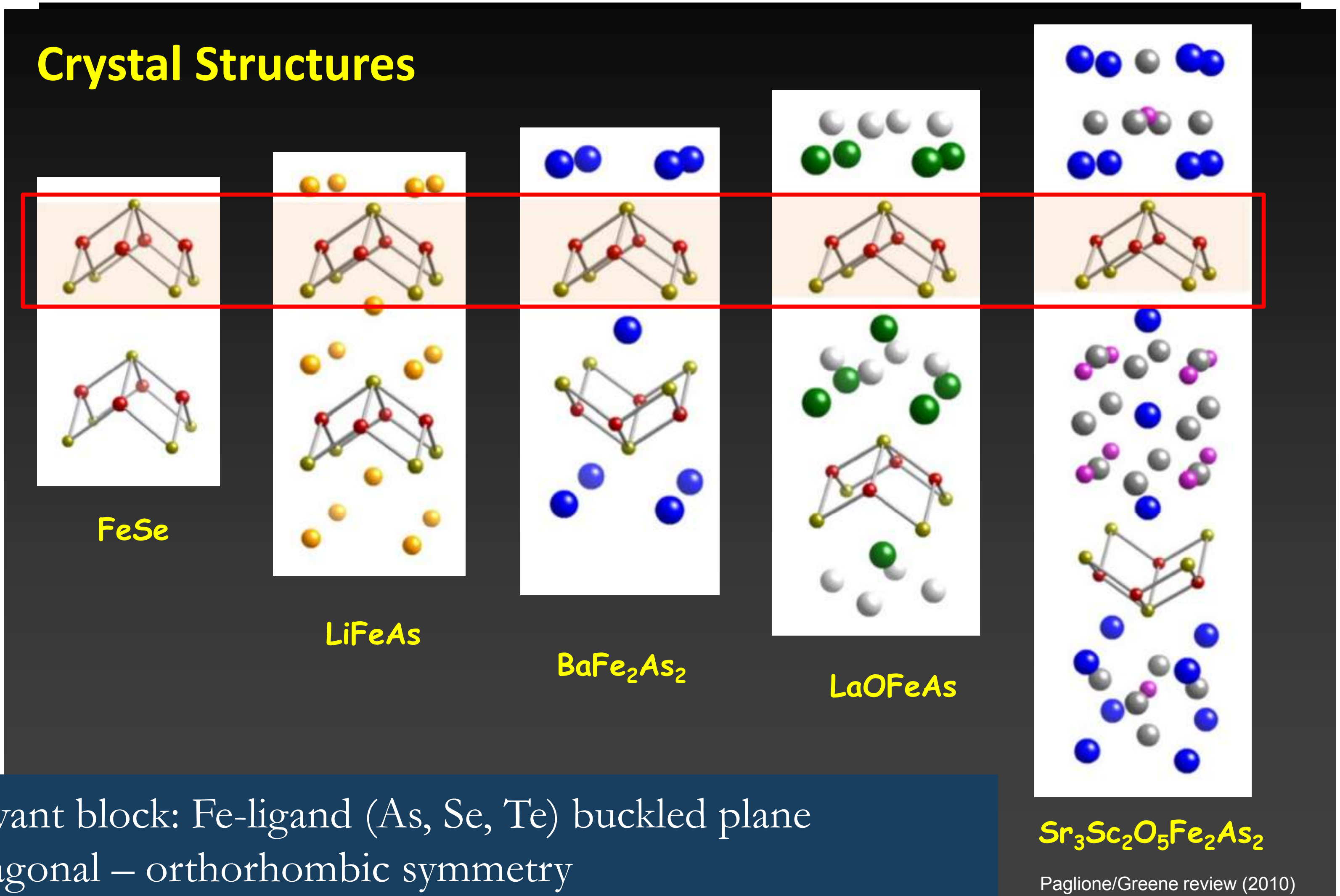
Superconducting materials



Iron-based superconductors



Iron-based superconductors



- relevant block: Fe-ligand (As, Se, Te) buckled plane
- tetragonal – orthorhombic symmetry

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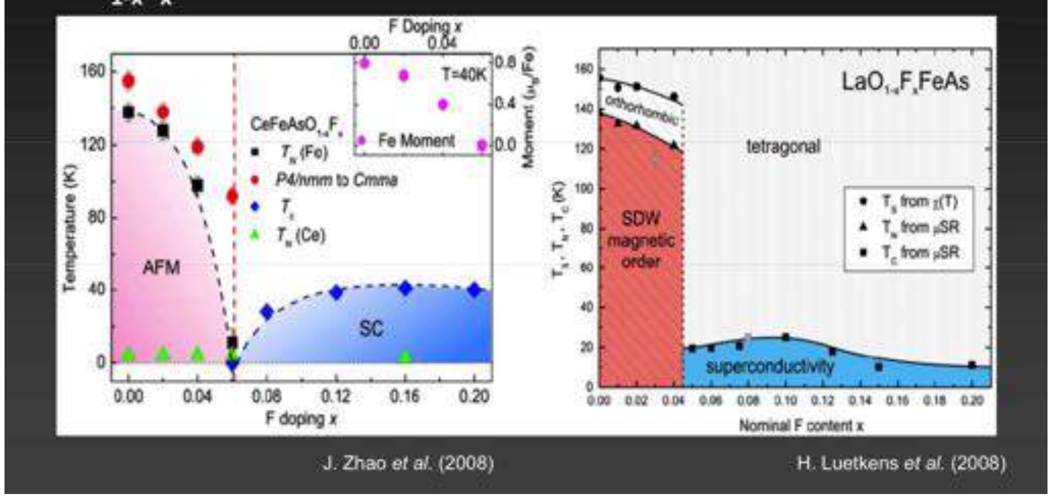
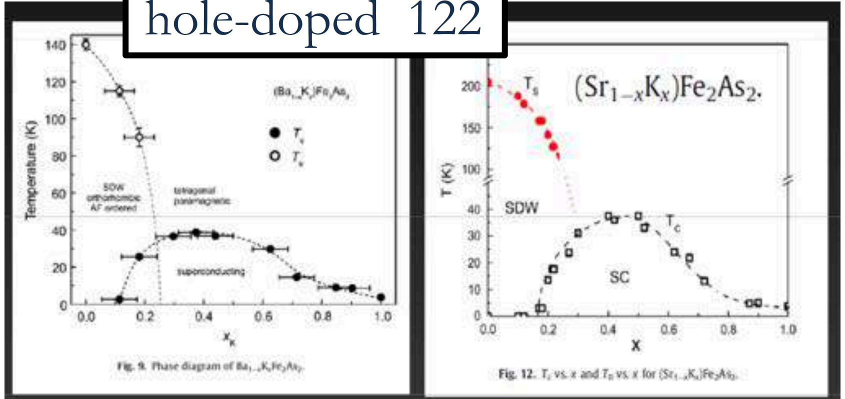
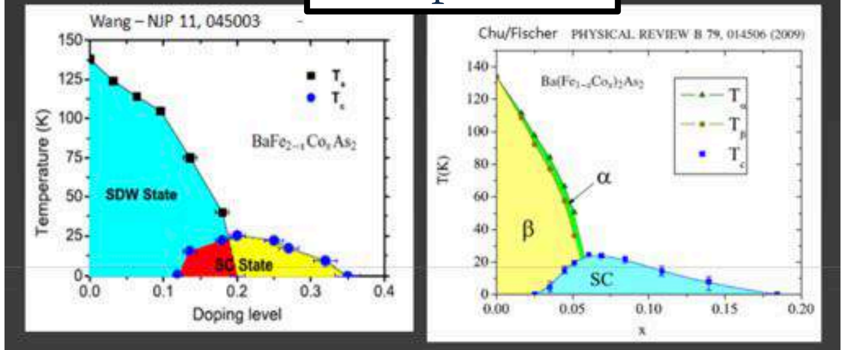
el-doped 1111



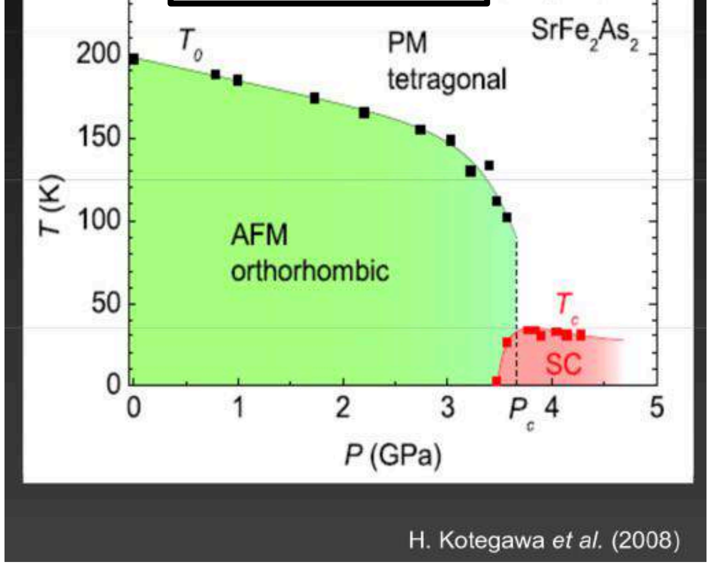
hole-doped 122

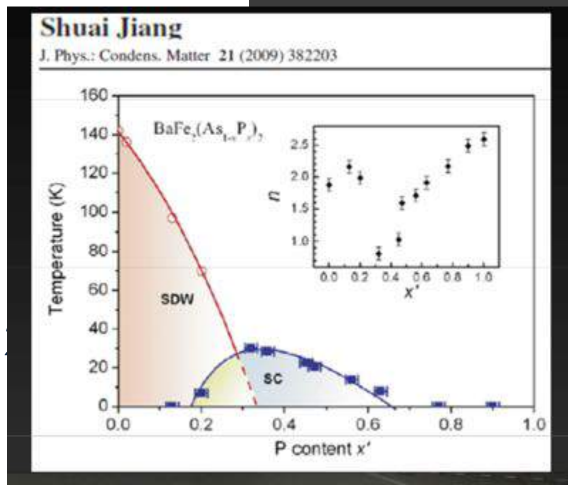


el-doped 122

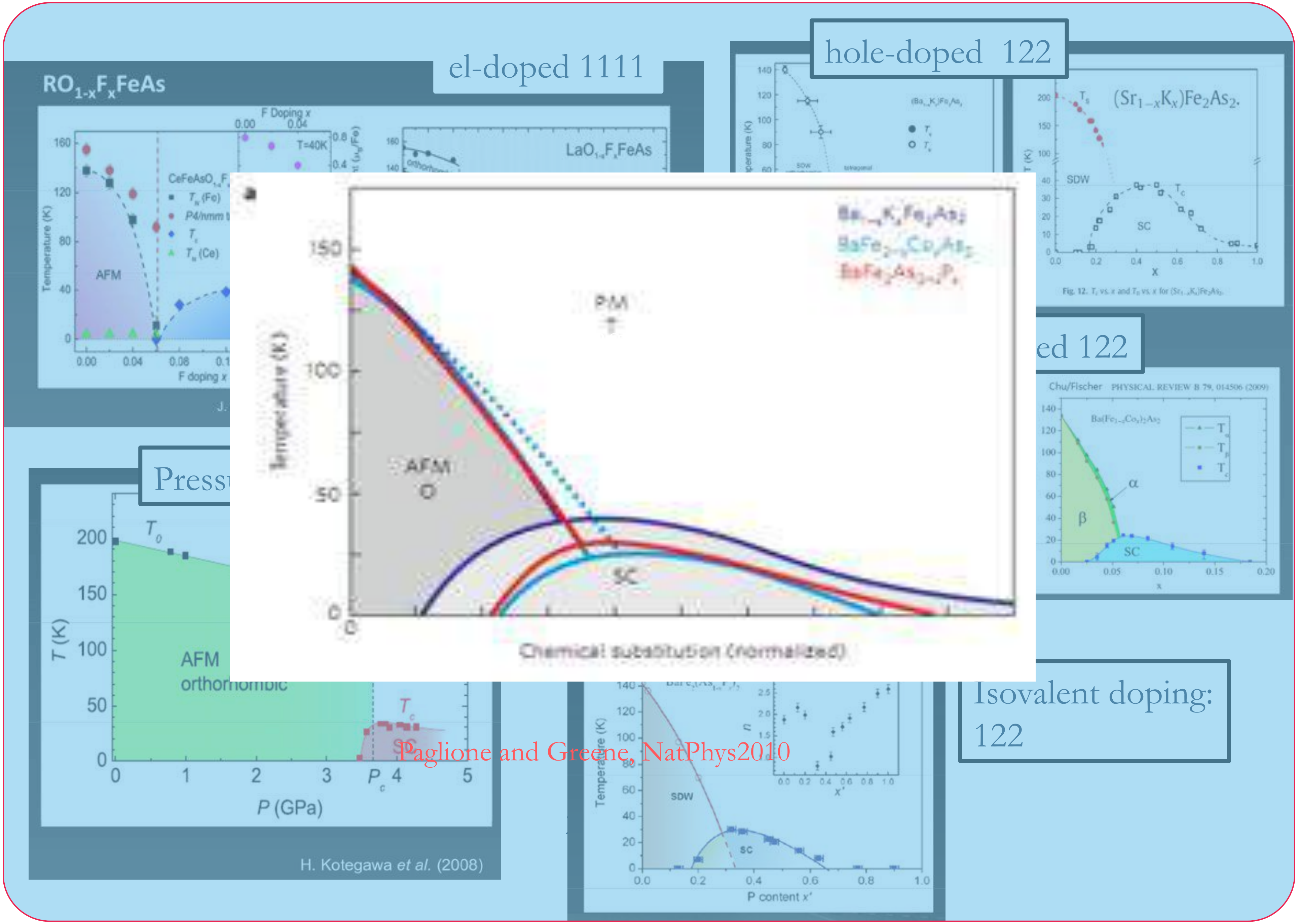


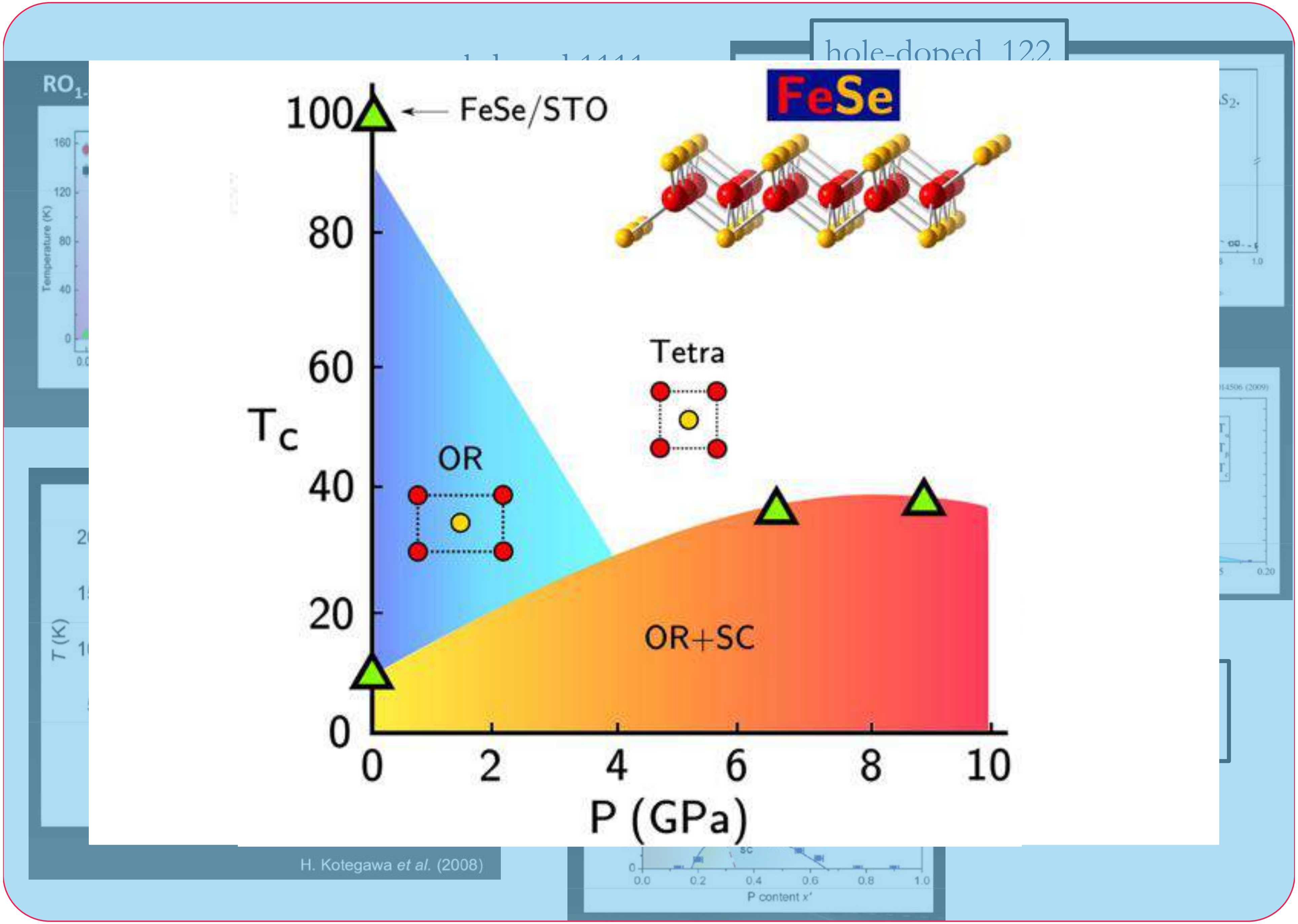
Pressure: 122





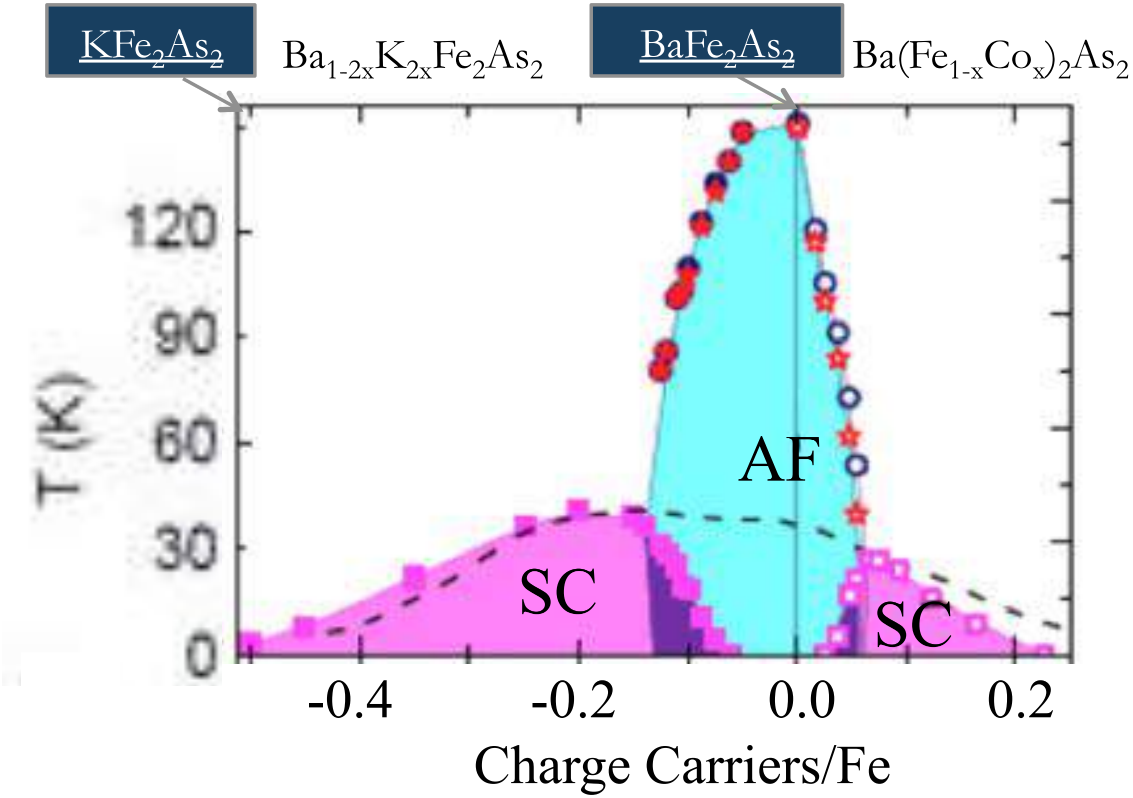
Isovalent doping: 122


el-doped 1111
$RO_{1-x}F_xFeAs$
hole-doped 122
$(Sr_{1-x}K_x)Fe_2As_2$
ed 122
Press
Isovalent doping: 122
H. Kotegawa et al. (2008)
AFM
orthorhombic
PM
T
AFM
O
SC
Temperature (K)
Chemical substitution (normalized)
$Ba_{1-x}K_xFe_2As_2$
$BaFe_{2-x}Co_xAs_2$
$BaFe_2As_{2-x}P_x$
Paglione and Greene, NatPhys2010

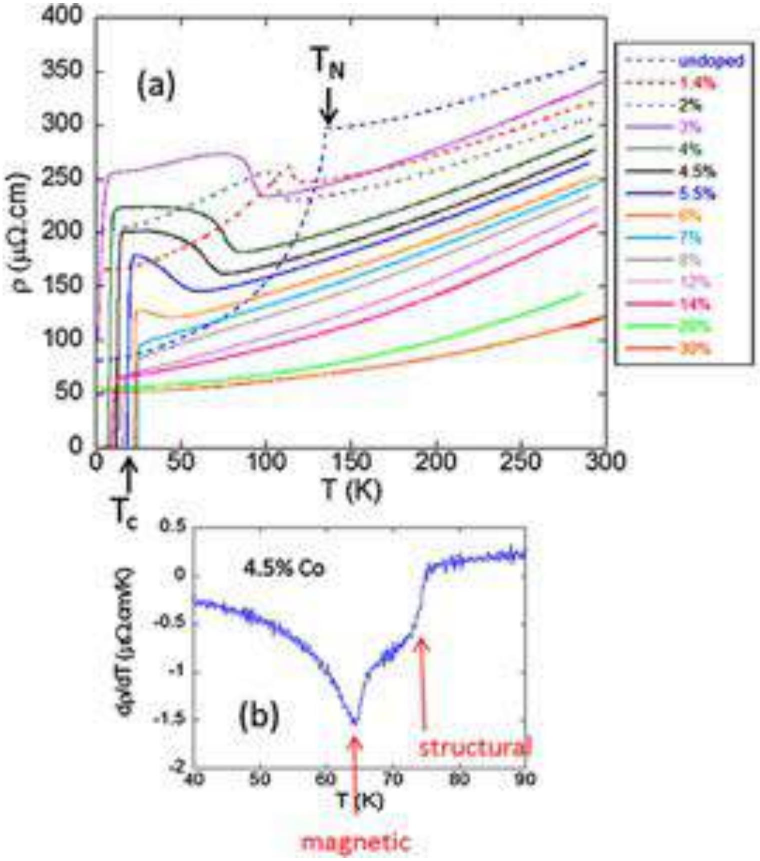

hole-doped 122
FeSe
100
80
60
40
20
0
FeSe/STO
Tc
OR
Tetra
OR+SC
0
2
4
6
8
10
P (GPa)
H. Kotegawa et al. (2008)
P content x'

Multi-orbital fermi-liquid metals

The “122” family



Avci et al. PRB 85 (2012)

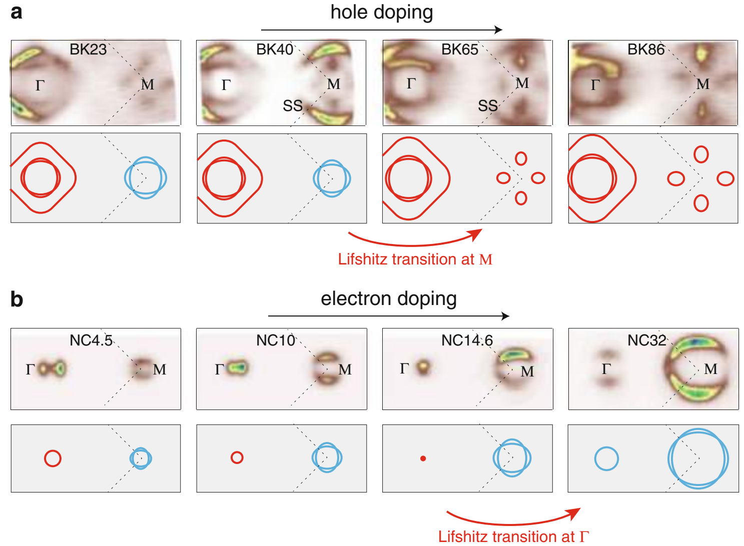


F. Rullier-Albenque, Comptes Rendus Physique 17 (2016) 164-187

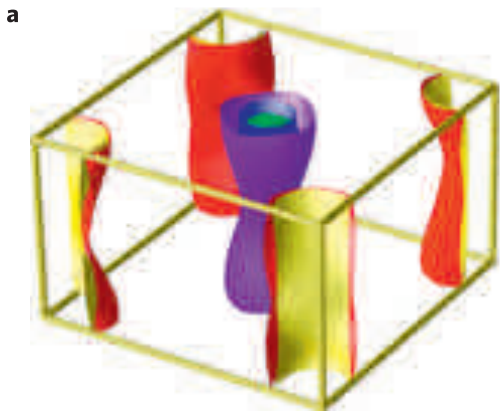
Theoretical approaches: itinerant electrons

- multi-orbital: 5 bands (Fe 3d) at the Fermi level (W~4eV)
  n=6 conduction electrons
- Partially lifted degeneracy (crystal-field splitting ~0.4eV)

Zhang et al. , Springer Book 2015

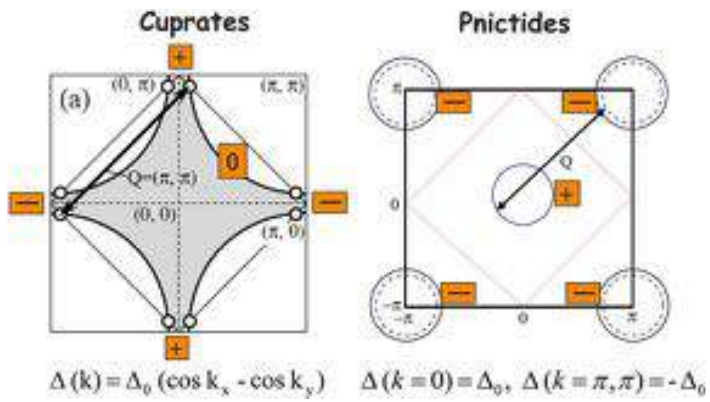


Mazin and Schmalian, Physica C 2009



Itinerant electrons:

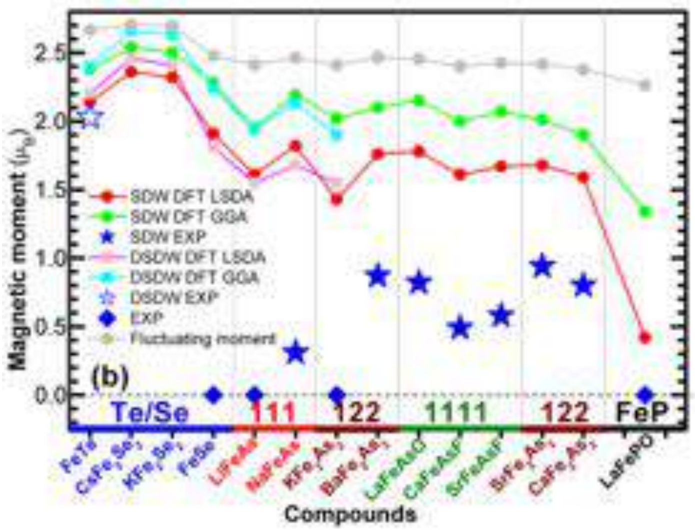
- DFT gives correct FS topology (semi-compensated metal)
- Nesting of FS pockets provides SDW and spin-fluctuations induce SC pairing ($S^{\pm}$-wave)

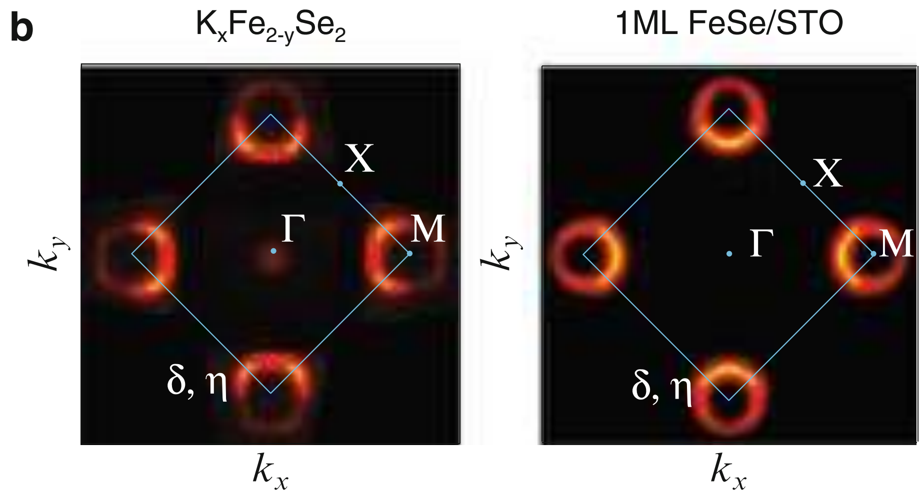


A. Chubukov, Springer Book 2015

Problems of the itinerant model

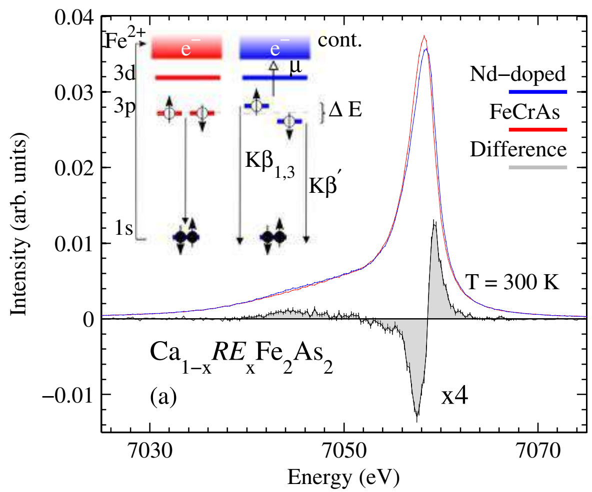
Yin et al. Nat. Mat. 2011 Zhang et al. Nat Mat 10 (2011) Tan et al. Nat Mat 12 (2014)





<u>Problems</u>

- DFT bands 2-3 times too dispersive
- LSDA unusually overestimates the ordered magnetic moment
- FeSe (Tc=8K) and LiFeAs (Tc=18K) do not have magnetic order
- $K_xFe_{2-y}Se_2$ (Tc~40K) and FeSe monolayer (Tc~100K??) only have electron pockets. KFe_2As_2 (Tc=4K) only hole pockets.
- Direct evidence of local moments in the PM phase (XES)



Electronic Correlations?

XES: Gretarsson et al. PRB 2011

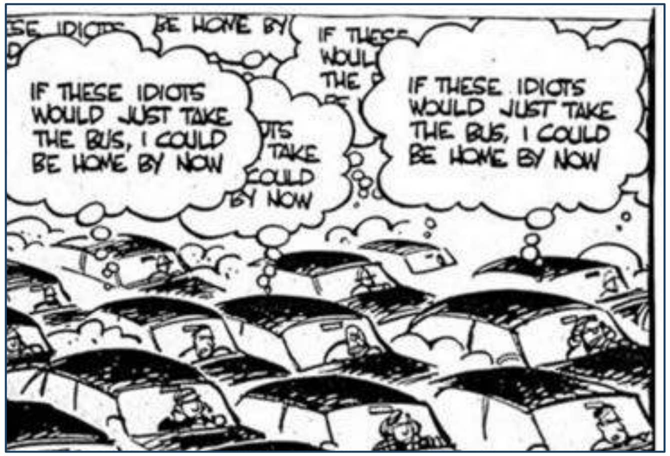
Correlated Materials

Band theory

Fermi-liquid, enhanced masses, instabilities, local magnetism,…

Mott insulator

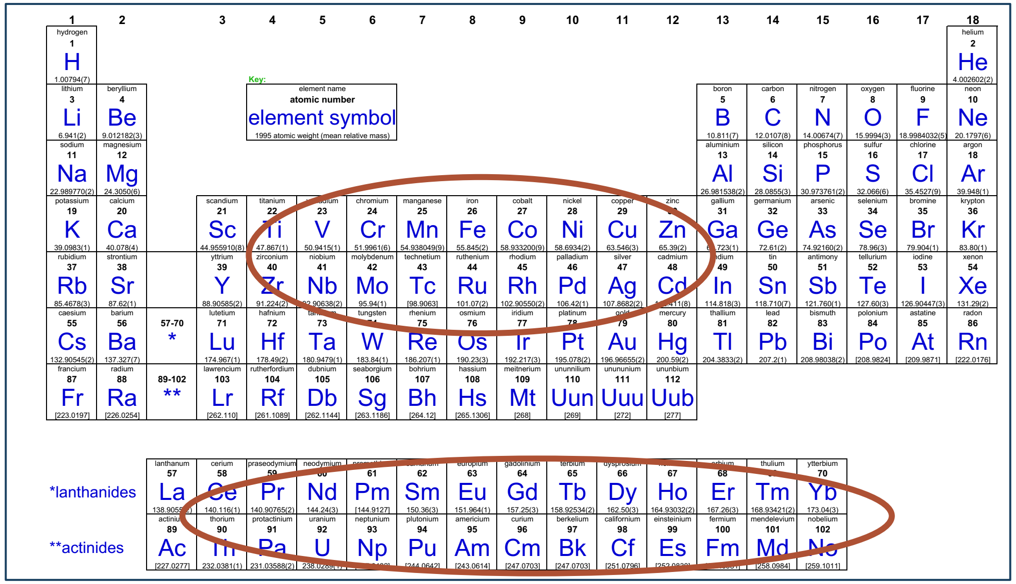
increasing electronic correlations



Unconventional phenomena:

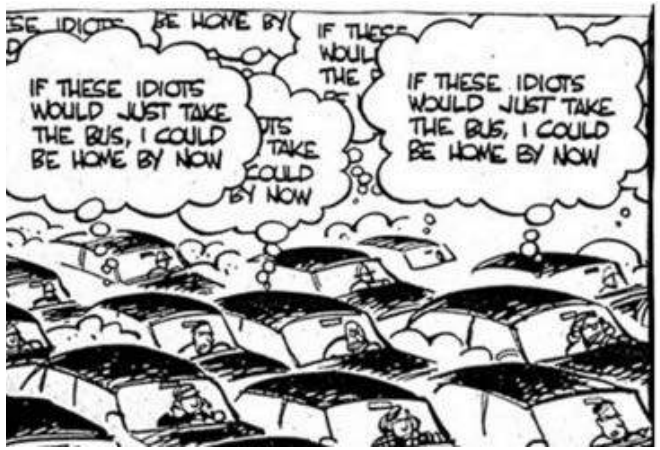
- High-Tc superconductivity
- large thermoelectric responses
- colossal magnetoresistance, …

Materials typically from 3d and 4f open shells: Cuprates, Fe-based superconductors, Manganites, heavy-fermions,…

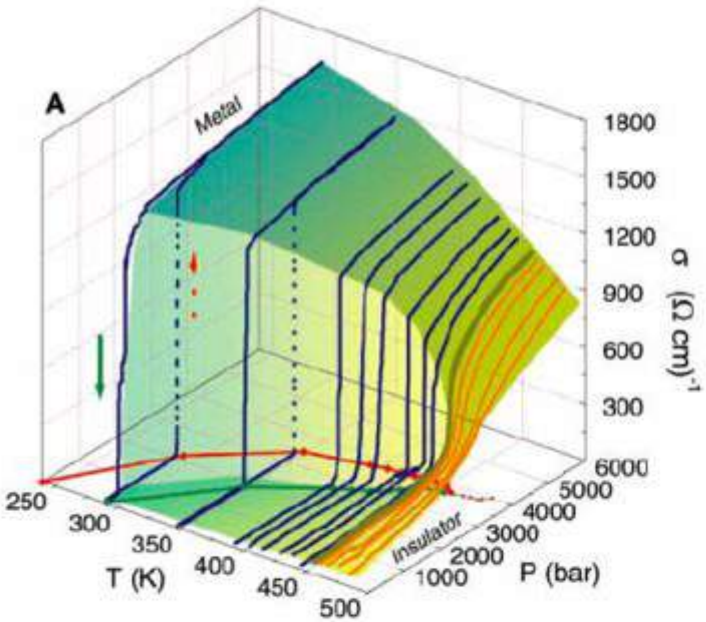


Mott Transition

<u>Localization of conduction electrons by correlations</u>



Relevant for transition metal oxides, Fullerenes, organic superconductors,…



Limelette et al. Science 302, 89 (2003)

<u>The proximity to a Mott state strongly affects the properties of a system:</u>

- reduced metallicity (Fermi-liquid: quasiparticle effective mass)
- transfer of spectral weight from low to high energy (e.g. in optical response)
- tendency towards magnetism
- …

Band Theory of electrons in a solid

$$\hat{H} = -\sum_i \frac{\hbar^2\nabla_i^2}{2m_e} - \sum_{i\alpha} \frac{Z_\alpha e^2}{|\mathbf{R}_\alpha - \mathbf{r}_i|} + \frac{1}{2}\sum_{i\neq i'} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_{i'}|}$$

Independent electron approximation

$$\hat{h}(\mathbf{r}_i) \equiv \frac{\hbar^2\nabla_i^2}{2m_e} + \boxed{v_{eff}(\mathbf{r}_i)}$$

one-particle Schrödinger equation

$$\hat{h}(\mathbf{r})\phi_a(\mathbf{r}) = \epsilon_a\phi_a(\mathbf{r})$$

In a translationally invariant system (crystal)

Bloch Functions

$$\phi_a(\mathbf{r}) \equiv \phi_{\mathbf{k}n}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}u_{n\mathbf{k}}(\mathbf{r})$$

Many-body wave function

Band theory: factorized many-body wave function

$$\psi(\mathbf{x}_1, \mathbf{x}_2) = \phi_a(\mathbf{x}_1)\phi_b(\mathbf{x}_2) \qquad E = \epsilon_a + \epsilon_b$$

Antisymmetrization

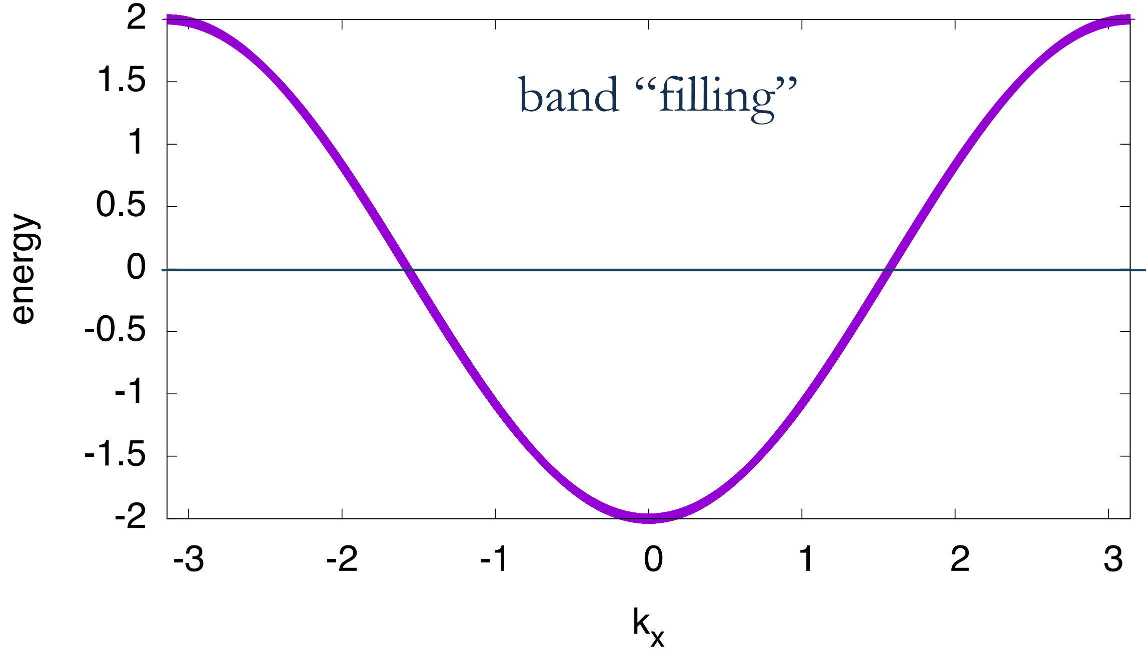
$$\psi(\mathbf{x}_1, \mathbf{x}_2) = \frac{1}{\sqrt{2}}\left[\phi_a(\mathbf{x}_1)\phi_b(\mathbf{x}_2) - \phi_b(\mathbf{x}_1)\phi_a(\mathbf{x}_2)\right]$$

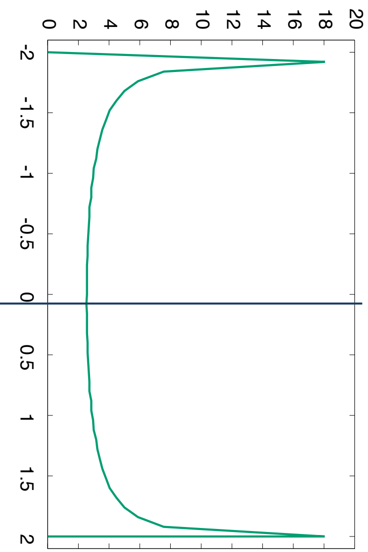
Slater determinant

$$\psi(\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_N) = \frac{1}{\sqrt{N!}}\begin{vmatrix} \phi_{a_1}(\mathbf{x}_1) & \phi_{a_1}(\mathbf{x}_2) & \ldots & \phi_{a_1}(\mathbf{x}_N) \\ \phi_{a_2}(\mathbf{x}_1) & \phi_{a_2}(\mathbf{x}_2) & \ldots & \phi_{a_2}(\mathbf{x}_N) \\ \vdots & \vdots & \vdots & \vdots \\ \phi_{a_N}(\mathbf{x}_1) & \phi_{a_N}(\mathbf{x}_2) & \ldots & \phi_{a_3}(\mathbf{x}_N) \end{vmatrix}$$

$$E = \sum_i \epsilon_{a_i}$$

Band “filling”, excitations, DOS





density of states (DOS)

$$N(\epsilon)$$

compressibility

$$\chi = N(\epsilon_F)$$

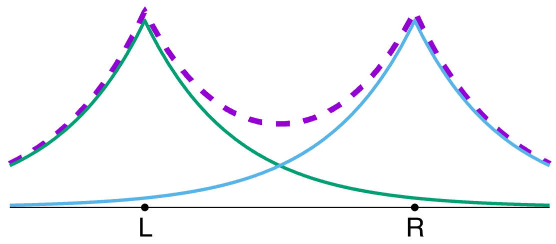
specific heat “Sommerfeld” coefficient

$$C_V \sim \gamma T$$

$$\gamma = \frac{\pi^2 k_B^2}{3} N(\epsilon_F)$$

Factorized wave function: a two-site example

$$\phi_a(\mathbf{x}) = c_{aL}\phi_L(\mathbf{x}) + c_{aR}\phi_R(\mathbf{x})$$



$$\phi_b(\mathbf{x}) = c_{bL}\phi_L(\mathbf{x}) + c_{bR}\phi_R(\mathbf{x})$$

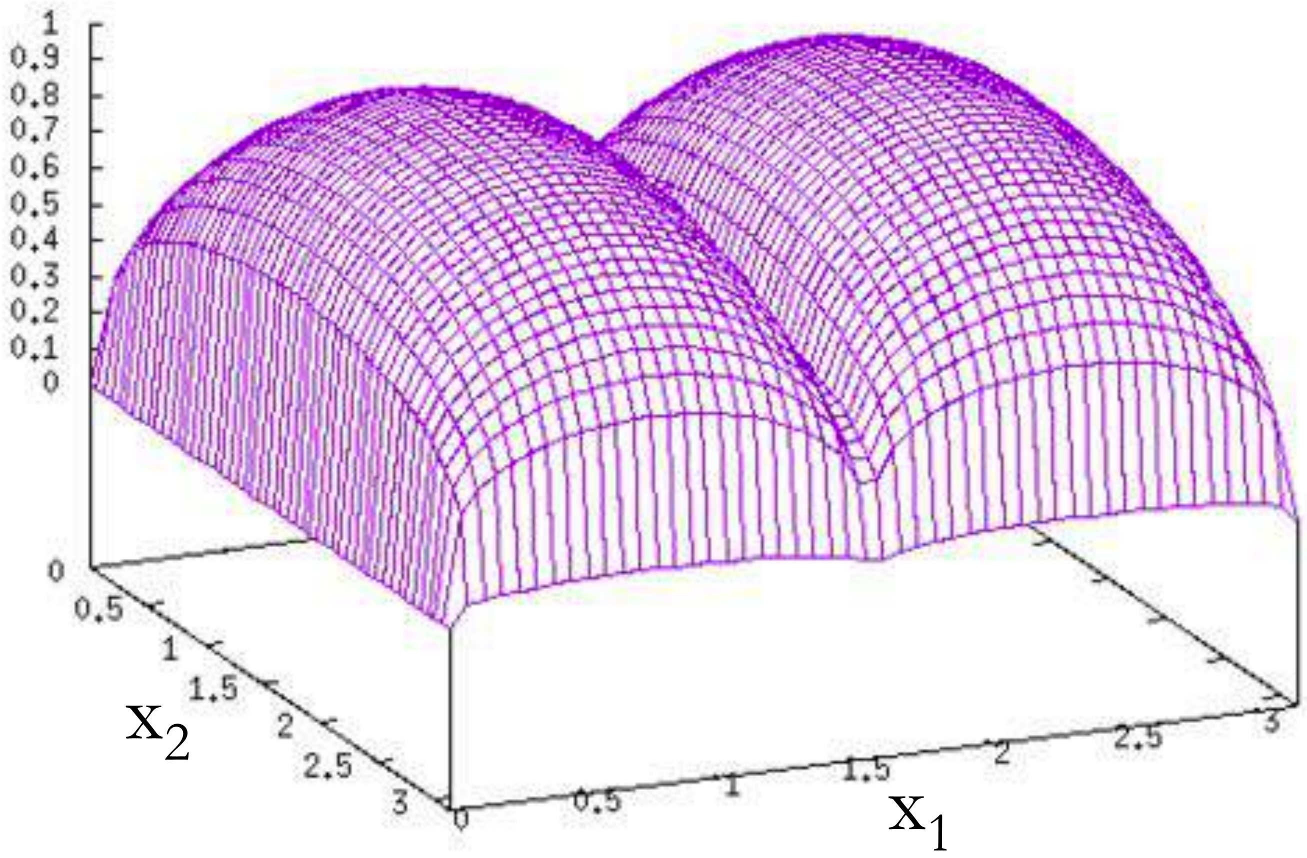
There is no way, by changing the one-electron wave functions (i.e. by changing c_{aL}, c_{aR}, c_{bL} and c_{bR}) to:

disfavour these contributions

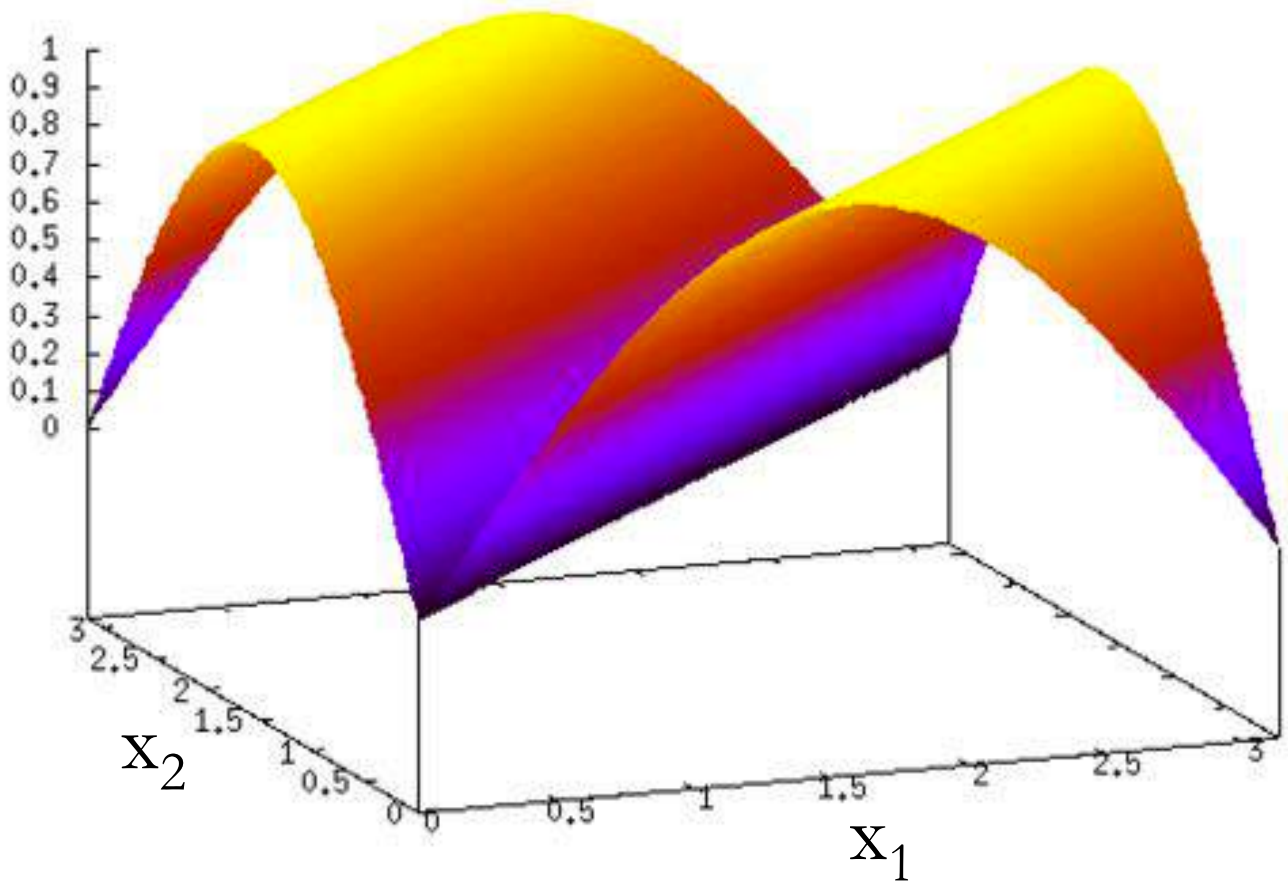
$$\phi_a(\mathbf{x}_1)\phi_b(\mathbf{x}_2) = c_{aL}c_{bL}\phi_L(\mathbf{x}_1)\phi_L(\mathbf{x}_2) + c_{aR}c_{bR}\phi_R(\mathbf{x}_1)\phi_R(\mathbf{x}_2)$$
$$+c_{aL}c_{bR}\phi_L(\mathbf{x}_1)\phi_R(\mathbf{x}_2) + c_{aR}c_{bL}\phi_R(\mathbf{x}_1)\phi_L(\mathbf{x}_2))$$

favour these contributions

a factorized wave function



non-factorized wave function



orbital

single-orbital case

$$\hat{H} = \sum_{ij\sigma} t_{ij} d^{\dagger}_{i\sigma} d_{j\sigma} + U \sum_{i} \tilde{n}_{i\uparrow} \tilde{n}_{i\downarrow}$$

$$n_{im\sigma} = d^{\dagger}_{im\sigma} d_{im\sigma}$$

multi-orbital case

$$\tilde{n}_{m\sigma} \equiv n_{m\sigma} - 1/2$$

$$\hat{H} = \sum_{ijmm'\sigma} t^{mm'}_{ij} d^{\dagger}_{im\sigma} d_{jm'\sigma} + H_{int}$$

$$H_{int} = U \sum_{m} \tilde{n}_{m\uparrow} \tilde{n}_{m\downarrow} + (U - 2J) \sum_{m \neq m'} \tilde{n}_{m\uparrow} \tilde{n}_{m'\downarrow} + (U - 3J) \sum_{m < m', \sigma} \tilde{n}_{m\sigma} \tilde{n}_{m'\sigma}$$

Intra-orbital interaction

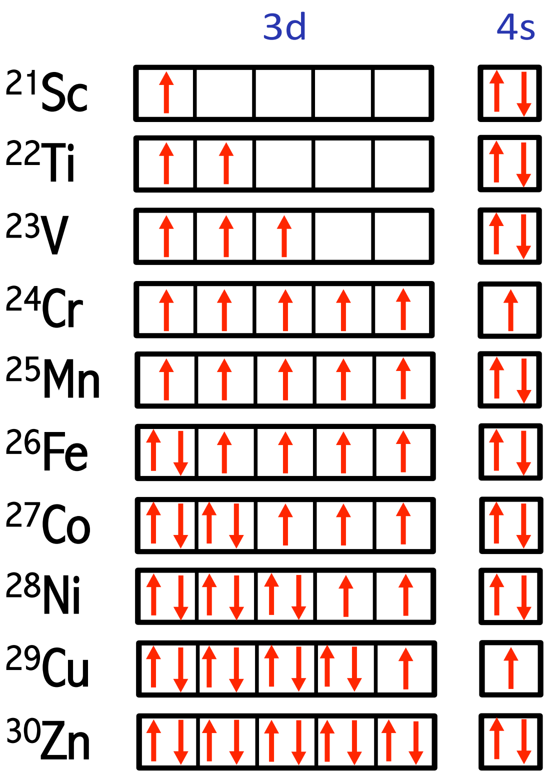
Inter-orbital interaction
U'<U
for opposite spins

Inter-orbital interaction
U'-J<U'<U
for parallel spins

J: "Hund's coupling" In correlated solids typically J=0.1÷0.2U

Hund's rules

Aufbau



<u>Hund's Rules</u>

In open shells:

1. Maximize total spin S
2. Maximize total angular momentum T

(3. Dependence on J=T+S, Spin-orbit effects)

Slave-spin mean field

Slave variables mean fields (general)

Recipe:

- Enlarge the local Hilbert space (new variables + constraint)
- Decouple the pseudo-fermions from the slave variables (renormalized non-interacting fermionic model)
- Treat the slave variables in a local mean-field
- Treat the constraint on average

Examples:

- Slave Bosons (Coleman, PRB 29, 3035 (1984) - Kotliar and Ruckenstein, PRL 57, 1362 (1987))
- Slave Rotors (Florens and Georges, PRB70, 035114 (2004))

Slave-spins

de' Medici et al. PRB 72, 205124 (2005)
S. R. Hassan and LdM, PRB 81, 35106 (2010)

Hilbert Space mapping:

$$|n^d_{i\sigma} = 1\rangle \Longleftrightarrow |n^f_{i\sigma} = 1,\ S^z_{i\sigma} = +1/2\rangle$$

$$|n^d_{i\sigma} = 0\rangle \Longleftrightarrow |n^f_{i\sigma} = 0,\ S^z_{i\sigma} = -1/2\rangle$$

$$f^\dagger_{i\sigma} f_{i\sigma} = S^z_{i\sigma} + \frac{1}{2}$$

constraint,
to exclude unphysical states

Slave-spin mean field

The density-density interactions are easily re-expressed through slave-spin operators

$$n^d_{i\sigma} \Longleftrightarrow n^f_{i\sigma}$$

$$n^d_{i\sigma} \Longleftrightarrow S^z_{i\sigma} + \frac{1}{2}$$

$$(\tilde{n}_{m\sigma} \equiv n_{m\sigma} - 1/2)$$

$$H_{int} = U \sum_m \tilde{n}_{m\uparrow}\tilde{n}_{m\downarrow} + (U-2J) \sum_{m \neq m'} \tilde{n}_{m\uparrow}\tilde{n}_{m'\downarrow} + (U-3J) \sum_{m<m',\sigma} \tilde{n}_{m\sigma}\tilde{n}_{m'\sigma}$$

$$H_{int}[S] = U \sum_m S^z_{m\uparrow} S^z_{m\downarrow} + (U-2J) \sum_{m \neq m'} S^z_{m\uparrow} \tilde{S}^z_{m'\downarrow} + (U-3J) \sum_{m<m',\sigma} S^z_{m\sigma} S^z_{m'\sigma}$$

Hopping terms

$$d_{i\sigma} \rightarrow f_{i\sigma} O_{i\sigma}$$

$$d^\dagger_{i\sigma} \rightarrow f^\dagger_{i\sigma} O^\dagger_{i\sigma}$$

$$O_{m\sigma} = S^-_{m\sigma} + c_{m\sigma} S^+_{m\sigma}, \qquad c_m = \frac{1}{\sqrt{\langle n^f_{m\sigma} \rangle (1 - \langle n^f_{m\sigma} \rangle)}} - 1$$

in the constrained space

$$H - \mu \hat{N} = \sum_{i \neq j \sigma} t^{mm'}_{ij} O^\dagger_{im\sigma} O_{jm'\sigma} f^\dagger_{im\sigma} f_{jm'\sigma} + \sum_{im\sigma} (\epsilon_m - \mu) n^f_{im\sigma} + H_{int}[S]$$

Slave-spin mean field

Finally: mean-field equations (constraint treated on average: lagrange multipliers λ_m)

$$H_f = \sum_{i\neq j, mm'\sigma} t_{ij}^{mm'} \sqrt{Z_m Z_{m'}} f^\dagger_{im\sigma} f_{jm'\sigma} + \sum_{im\sigma} (\epsilon_m - \lambda_m - \mu) n^f_{im\sigma},$$

$$H_s = \sum_{m,\sigma} \left[(h_m O^\dagger_{m\sigma} + H.c.) + \lambda_m (S^z_{m\sigma} + \frac{1}{2}) \right] + \hat{\tilde{H}}_{int}[S],$$

$$h_{m\sigma} = \sum_{m'} \langle O_{m'\sigma} \rangle_s \sum_{j(\neq i)} t_{ij}^{mm'} \langle f^\dagger_{im\sigma} f_{jm'\sigma} \rangle_f$$

$$Z_m = |\langle O_{m\sigma} \rangle|^2 \qquad \langle S^z_{i\sigma} \rangle + \frac{1}{2} = \langle f^\dagger_{i\sigma} f_{i\sigma} \rangle$$

Analogous to multi-orbital Kotliar-Ruckenstein slave-bosons and Gutzwiller approximation (F. Gebhard lecture)
Similarly, the Slave-spin mean-field (SSMF) describes a Fermi-liquid

Pedagogical introduction:
"Modeling Many-Body Physics with Slave-Spin Mean-Field : Mott and Hund's Physics in Fe-Superconductors", L. de' Medici and M. Capone in The Iron Pnictide Superconductors, Springer Series in Solid-State Sciences, Vol. 186, pp. 115-185, Mancini, F., Citro, R. (Eds) 2017.

Fermi Liquid Theory

quasiparticles instead of particles

$$\frac{1}{\tau} = a(\epsilon - \epsilon_F)^2 + bT^2$$

infinite lifetime at T=0 at the Fermi level

$$\epsilon_{n\mathbf{k}} \longrightarrow \epsilon^*_{n\mathbf{k}}$$

renormalized dispersion

$$N(\epsilon_F) \longrightarrow N^*(\epsilon_F)$$

renormalized DOS

$$\mathbf{v}^*_{n\mathbf{k}} = \frac{1}{\hbar}\nabla_{\mathbf{k}}\epsilon^*_{n\mathbf{k}} \qquad |\mathbf{v}^*_{n\mathbf{k}}| = \frac{\hbar k}{m^*}$$

effective mass

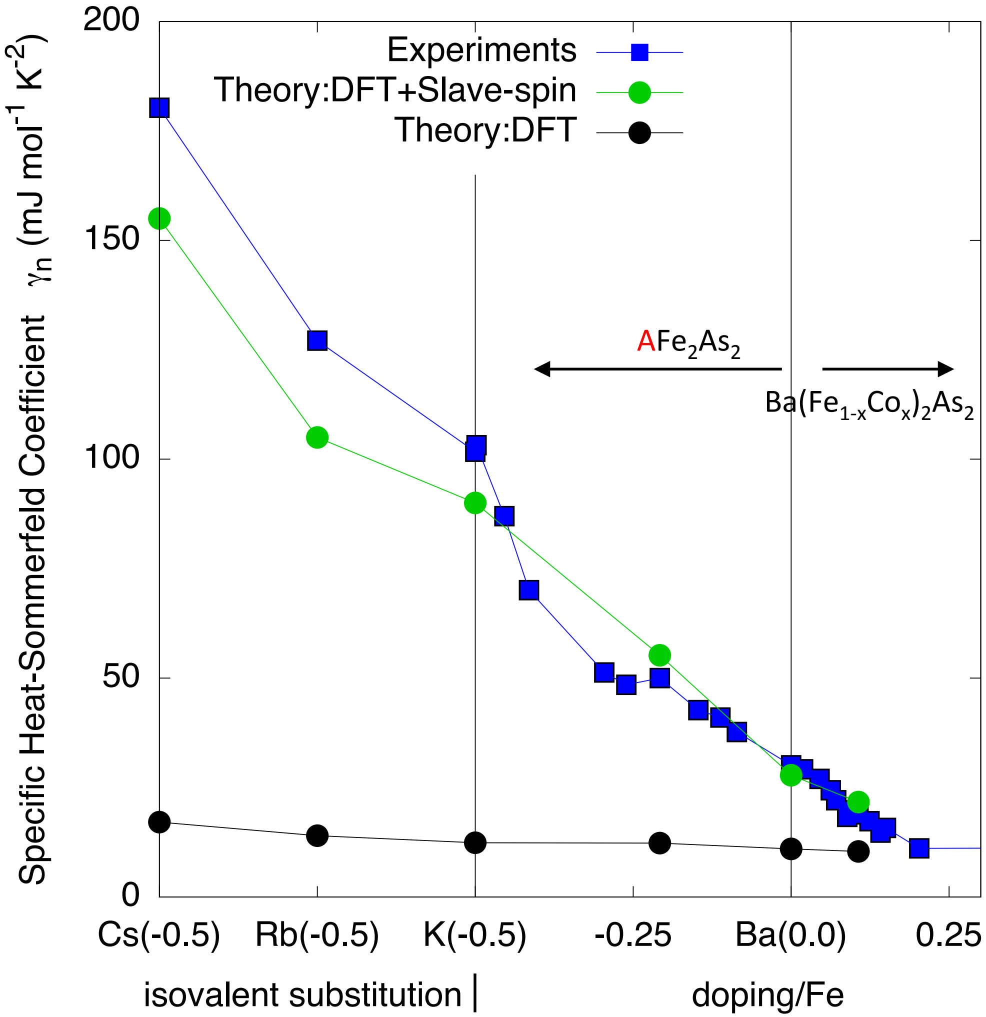
$$\chi = \frac{N^*(\epsilon_F)}{1 + F_0^s}$$

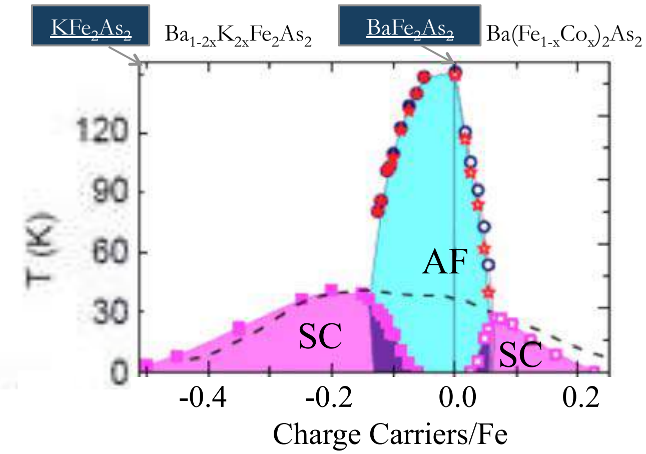
compressibility

$$\gamma = \frac{\pi^2 k_B^2}{3} N^*(\epsilon_F)$$

Sommerfeld coefficient

Fe-superconductors: specific heat



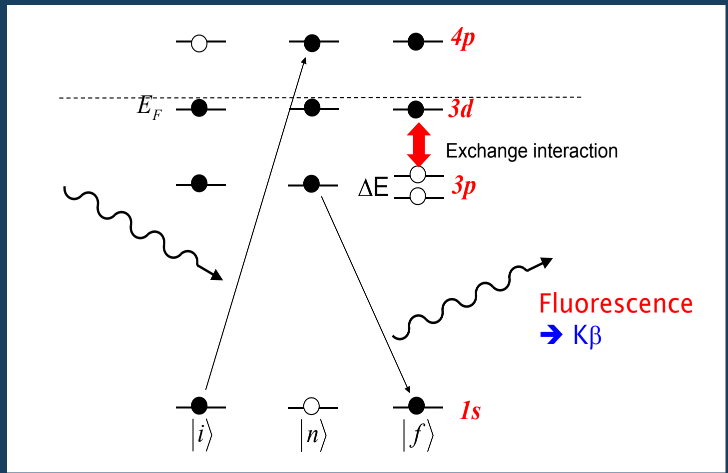


Theory (DFT+Slave-spin):
- the same interaction parameters (U=2.7eV, J/U=0.25) capture the whole material trend
- DFT results are completely off: strong correlations

Experiments: C. Meingast's group in Karlsruhe. F. Hardy,…, LdM et al. PRB 94, 205113 (2016)

Fe-superconductors: local moments

X-ray Emission Spectroscopy
(K-β/β' line shift)



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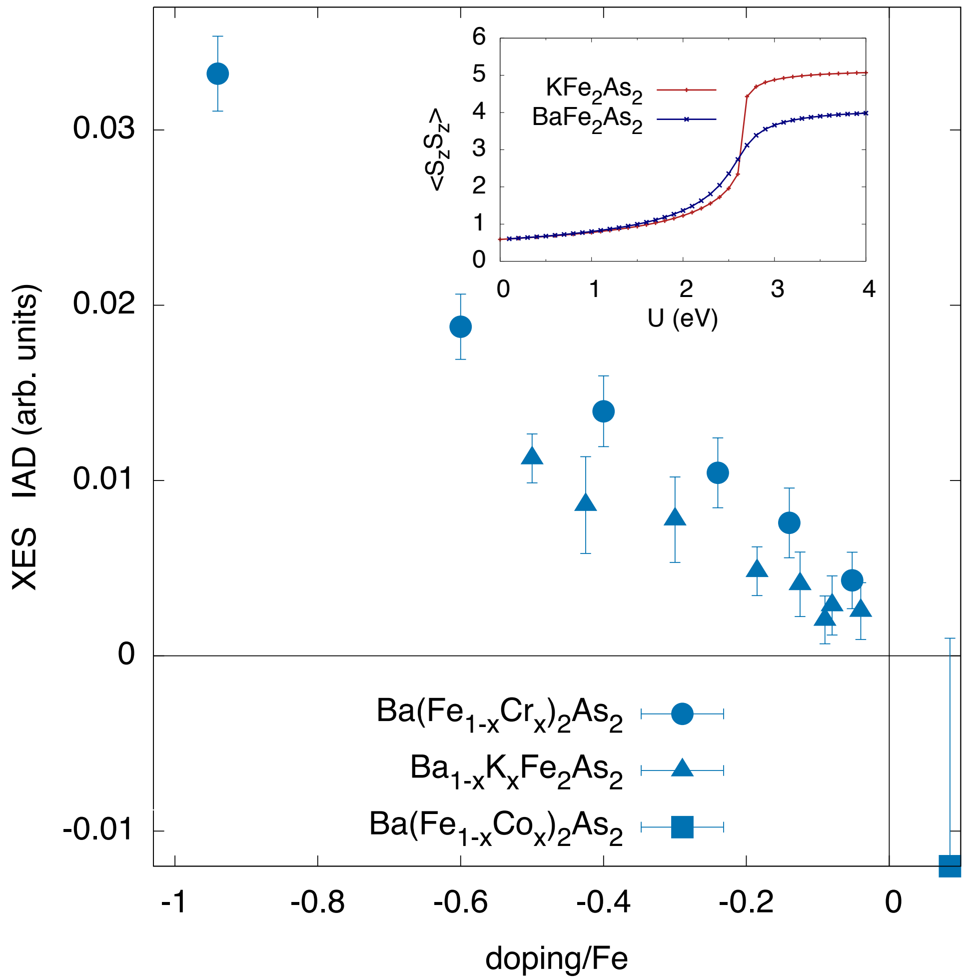


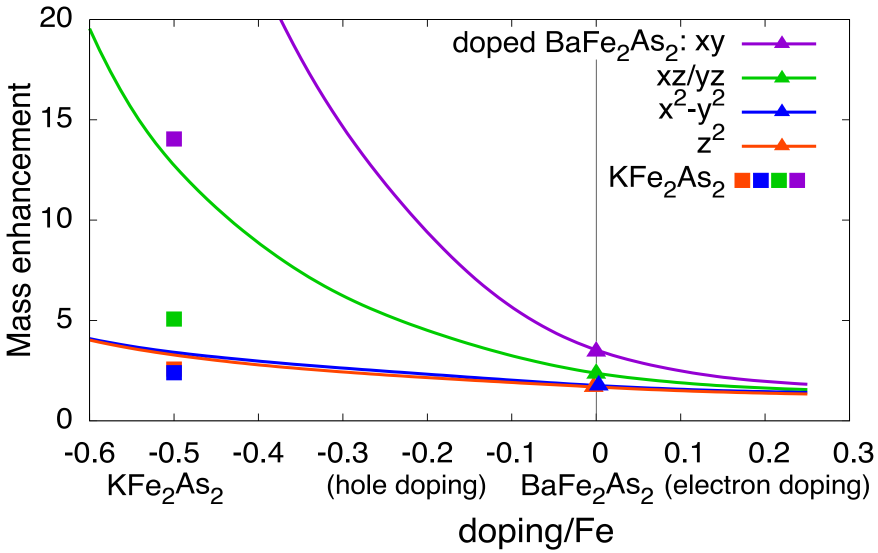
P. Glatzel group @ ESRF (ID26)

S. Lafuerza, …, LdM et al., PRB 96, 045133 (2017)

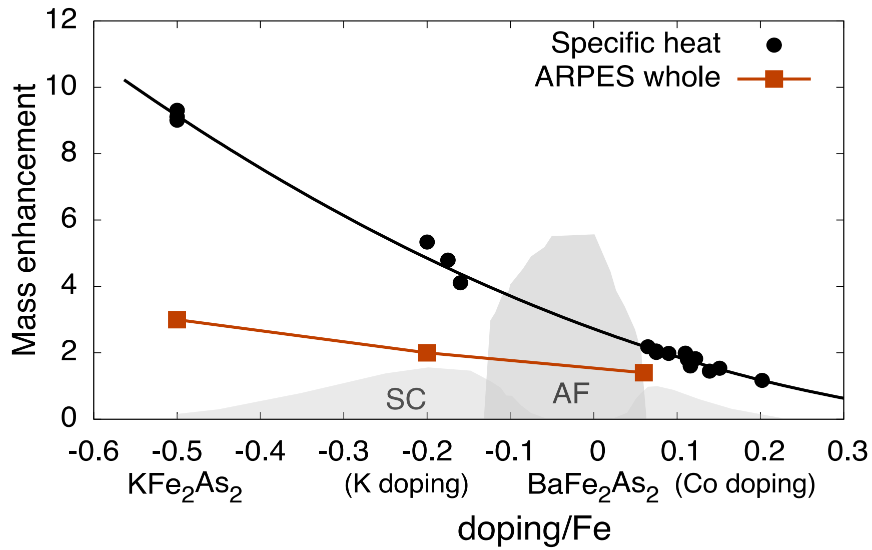
see also Pelliciari et al. Sci. Rep. 7, 8003 (2017)

Fe-superconductors: orbital-selectivity

Theory (LDA+Slave-spins)



Experimental data
(high-T tetragonal phase)



LdM, Giovannetti, Capone, PRL 2014 "Selective Mott Physics as a Key to Iron Superconductors"

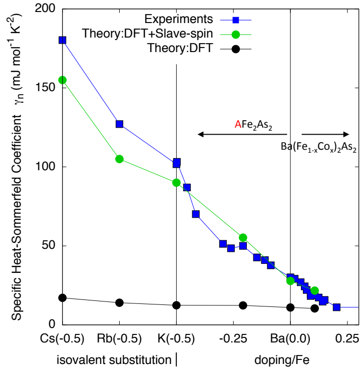
Selective correlation strength:
strongly *and* weakly correlated electrons coexisting

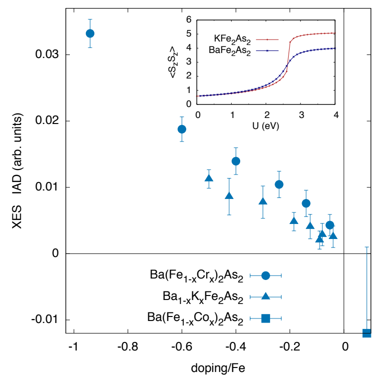
LdM, *Weak AND strong correlations in Fe-SC*, in "Iron-based Superconductivity", Springer book 2015

Hund's metals

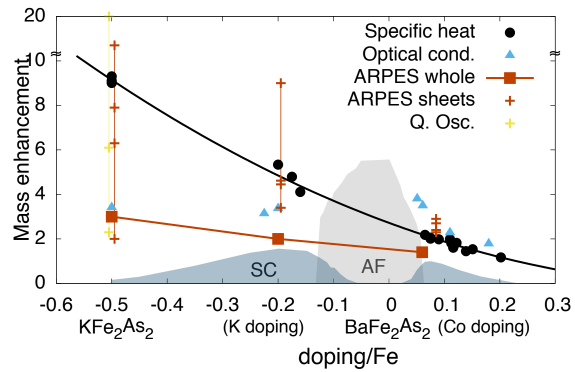
3 main features:

- enhanced electron correlations and masses
- high local spin configurations dominating the paramagnetic fluctuations
- orbital-selectivity of the electron correlation strength





which are due to the proximity to a Hund's favored Mott insulating state for half-filled conduction bands (1 hole/Fe doping)

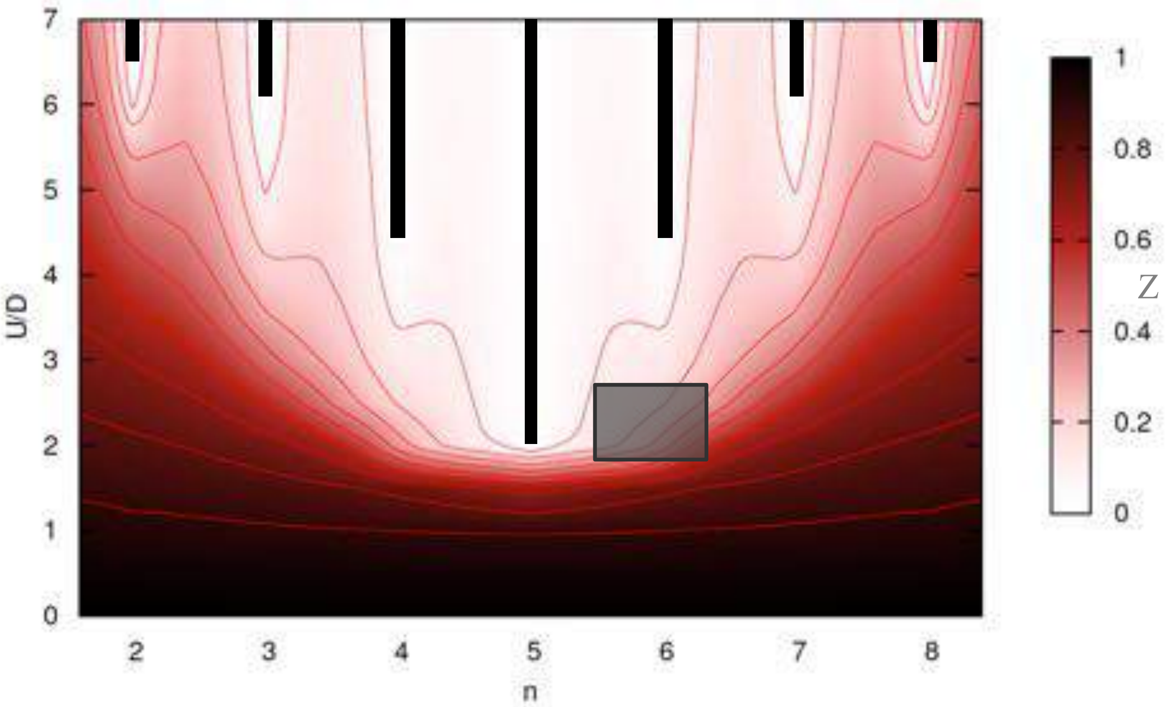


5-orbital Hubbard model

J/U=0.15

J/U=0.20

J/U=0.25



Semicircular DOS of width W=2D
(Bethe lattice with hopping t, D=2t)

$$D(\epsilon) = \frac{2}{\pi D}\sqrt{1 - \frac{\epsilon^2}{D^2}}$$

LdM, *Weak AND strong correlations in Fe Superconductors*, in “Iron-based Superconductivity”, Springer Series in Material Sciences, Vol 211, pp 409-441 (2015) - ArXiv: 1506.01678

5-orbital Hubbard model

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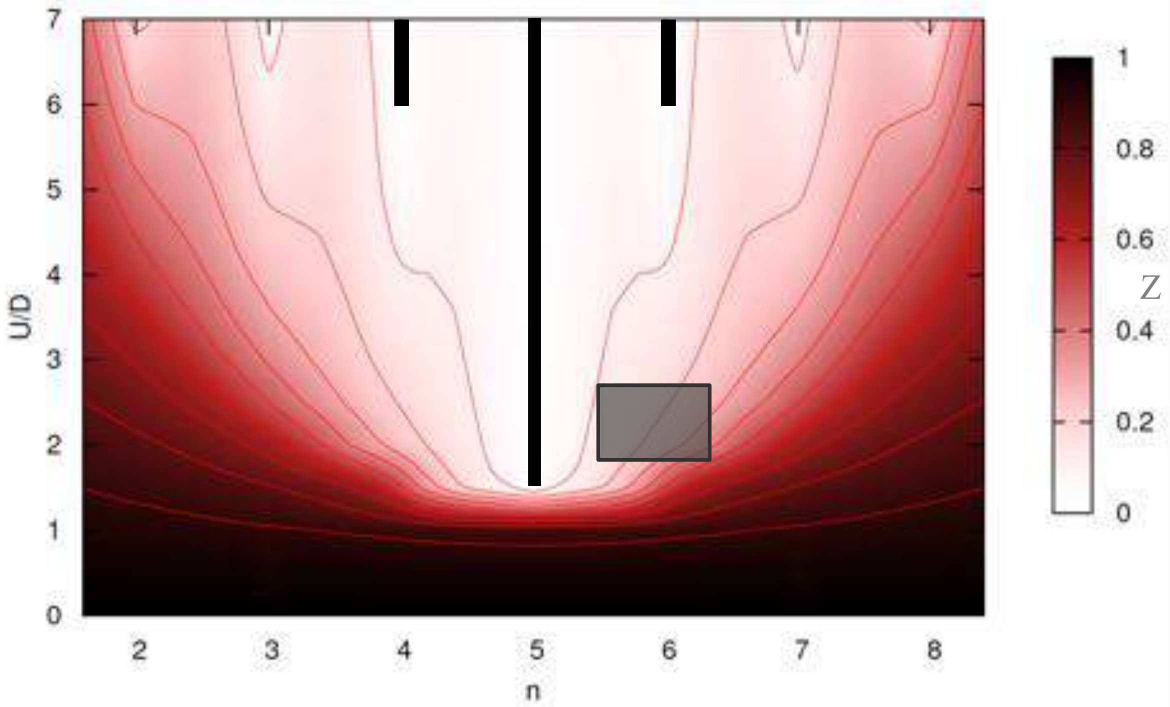

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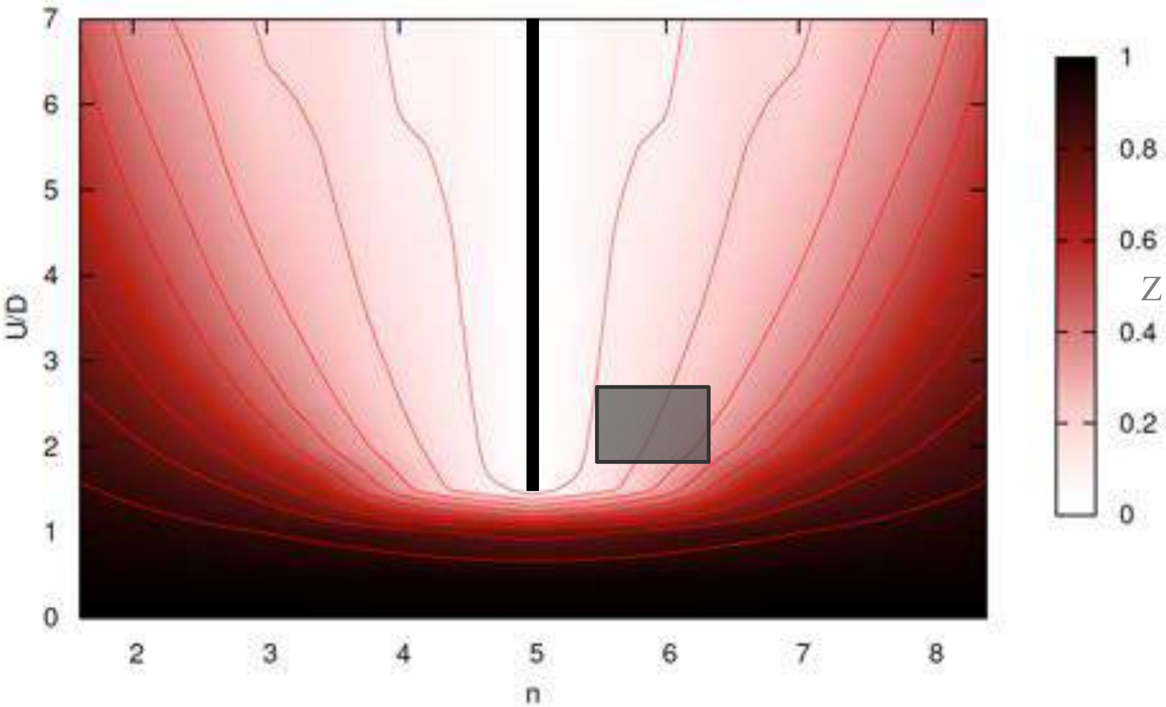

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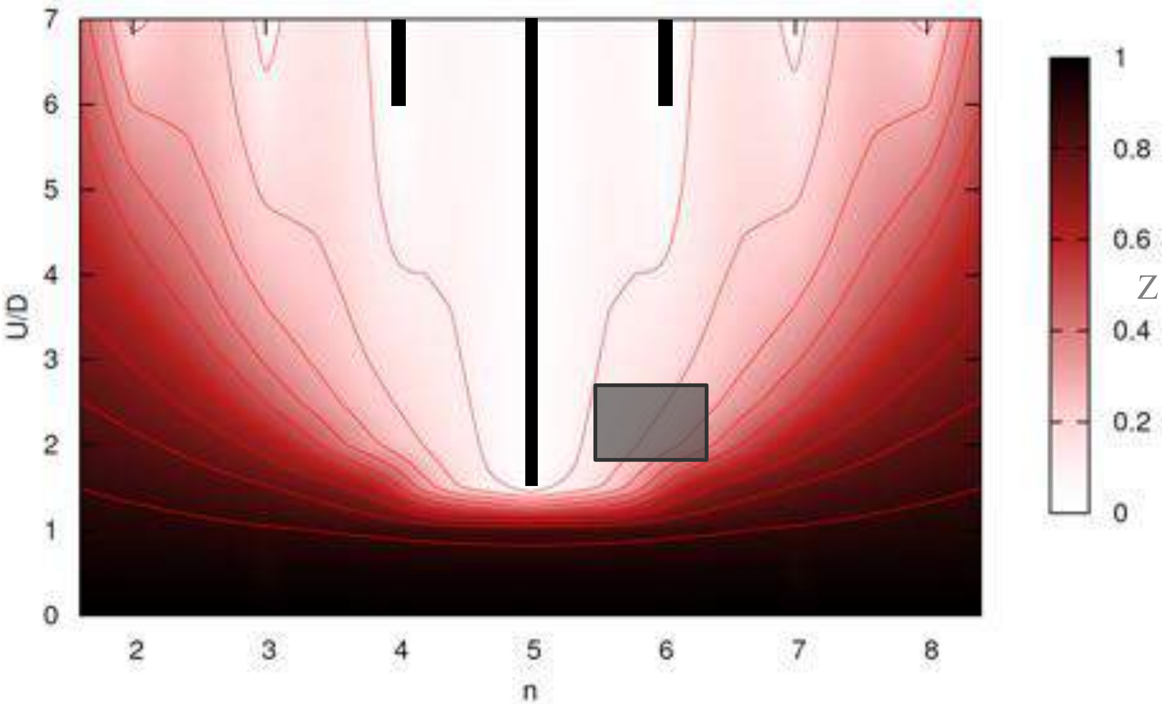
5-orbital Hubbard model

J/U=0.15

$\boxed{\text{J/U=0.20}}$

J/U=0.25



Semicircular DOS of width W=2D
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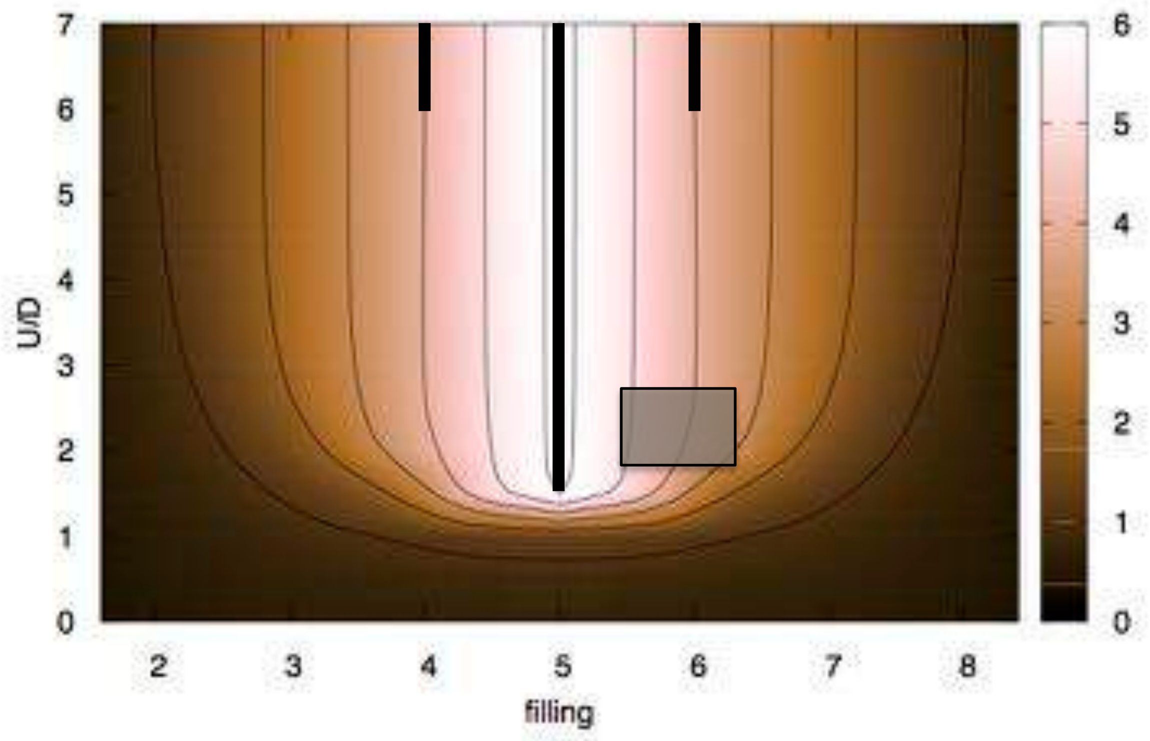
$$D(\epsilon) = \frac{2}{\pi D}\sqrt{1 - \frac{\epsilon^2}{D^2}}$$

LdM, *Weak AND strong correlations in Fe Superconductors*, in "Iron-based Superconductivity", Springer Series in Material Sciences, Vol 211, pp 409-441 (2015) - ArXiv: 1506.01678

High fluctuating magnetic moment

J/U=0.20



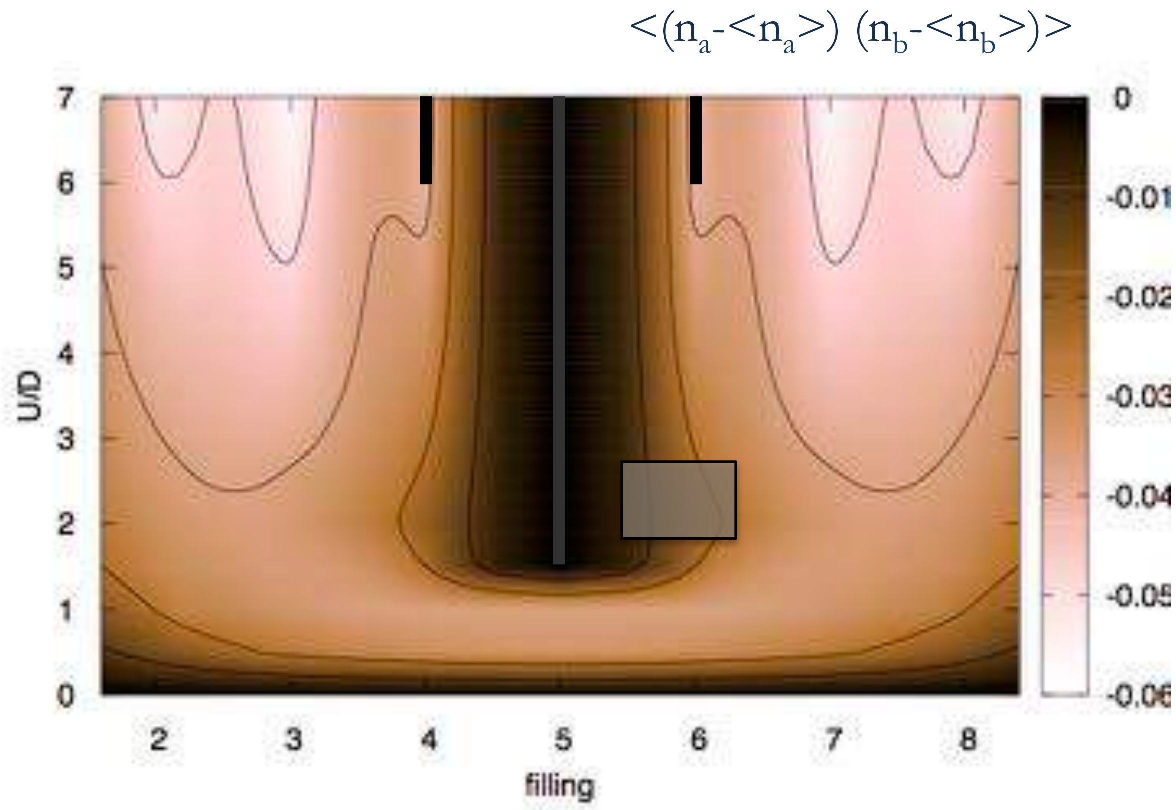
A metal in which high-spin configurations prevail

"Hund's metal"

LdM, *Weak AND strong correlations in Fe-SC*, in "Iron-based Superconductivity", Springer book 2015

inter-orbital charge correlations

J/U=0.20



charge fluctuations in different orbitals become uncorrelated near the half-filled Mott insulator

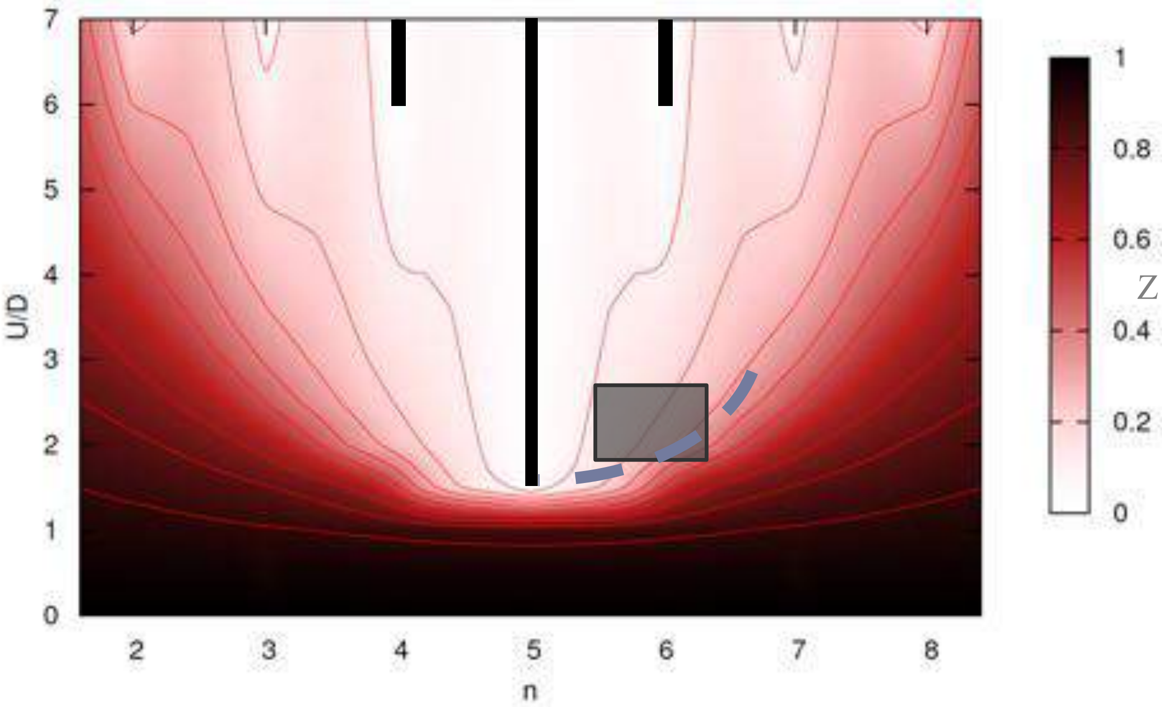
LdM, *Weak AND strong correlations in Fe-SC*, in "Iron-based Superconductivity", Springer book 2015

Hund's metal and half-filled Mott insulator

J/U=0.15

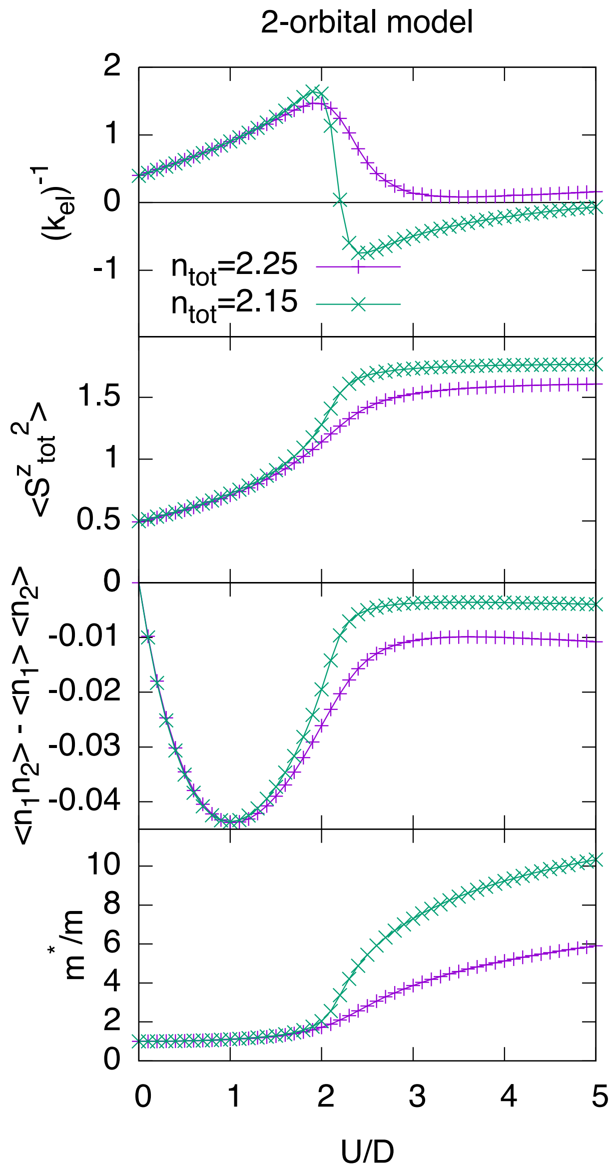
J/U=0.20

J/U=0.25

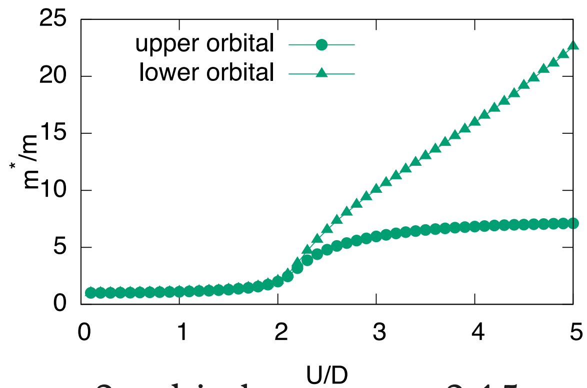


The cross-over departs from the Mott transition at half-filling

Hund's metal frontier: 2-orbital Hubbard model



J/U=0.25



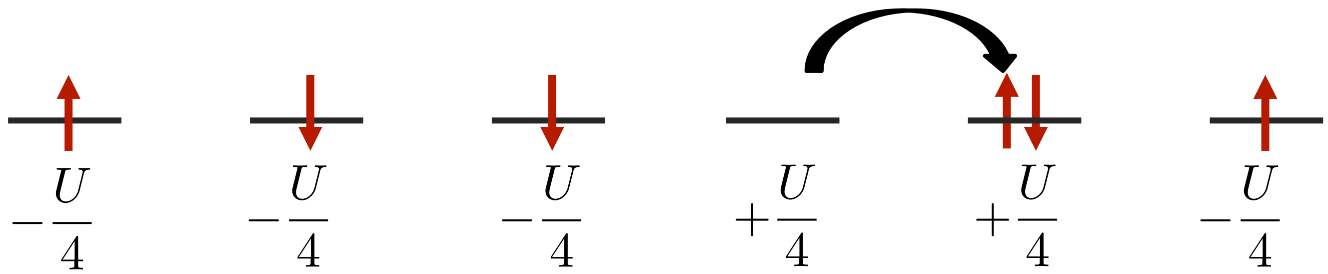
2-orbital case n_{tot}=2.15, with a small crystal-field splitting ε_1-ε_2=0.05D

Hund's phenomenology analogous to the 5-orbital (and 3-orbital) case and to the realistic simulations for the Fe-superconductors: **generic**

Mott transition: the atomic Mott gap

Single band Hubbard model
in the atomic limit

$$\sum_i U(n_{i\uparrow} - \frac{1}{2})(n_{i\downarrow} - \frac{1}{2})$$



Potential energy barrier to overcome to establish conduction
(ionization energy - electron affinity)

$$\begin{aligned}\Delta_{at} &= E_{at}(n+1) - E_{at}(n) + E_{at}(n-1) - E_{at}(n) \\ &= E_{at}(n+1) + E_{at}(n-1) - 2E_{at}(n)\end{aligned}$$

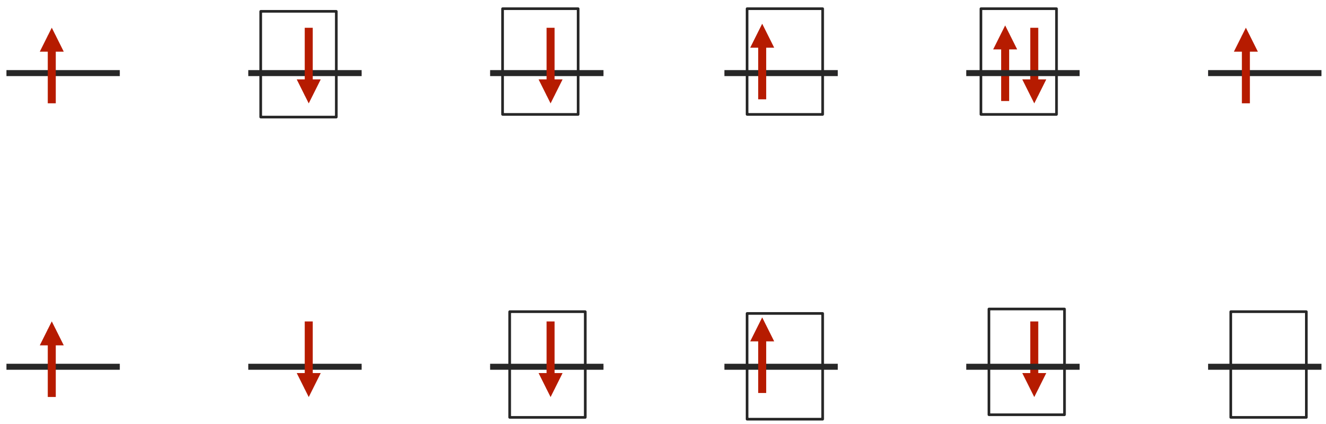
$$\boxed{\Delta_{at} = U}$$

Mott transition: Hubbard bands

However these excited states are mobile
(high degeneracy removed by hopping → energy dispersion ~W)

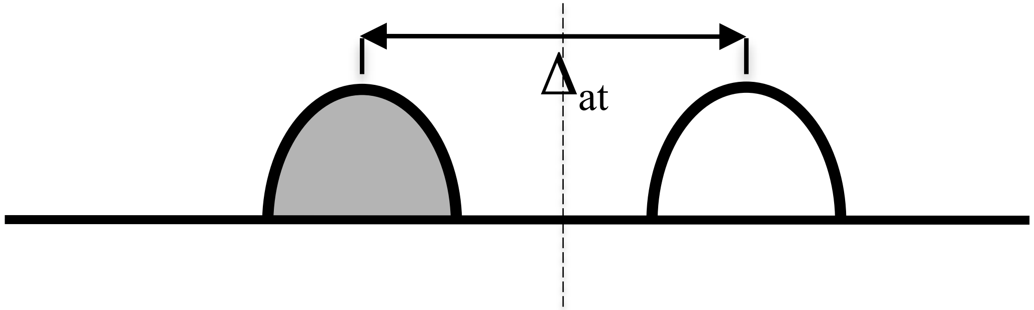
particle excitation

hole excitation

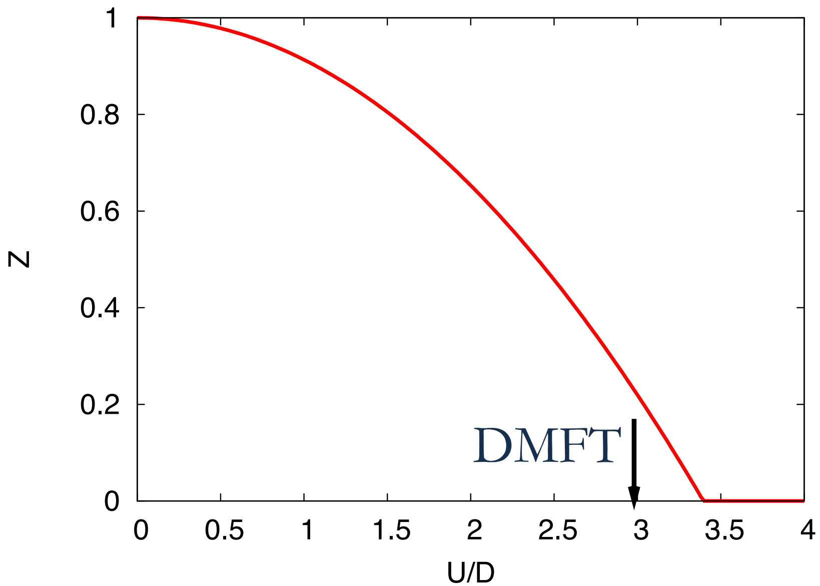


Formation of two "Hubbard bands" in the spectrum of excitations

Mott transition: theHubbard criterion



$$\Delta = U - W \quad \Longrightarrow \quad \boxed{U_c = W}$$



Slave-spin mean-field
half-filled 1-band Hubbard model
Semi-circular Density of states
of width W=2D

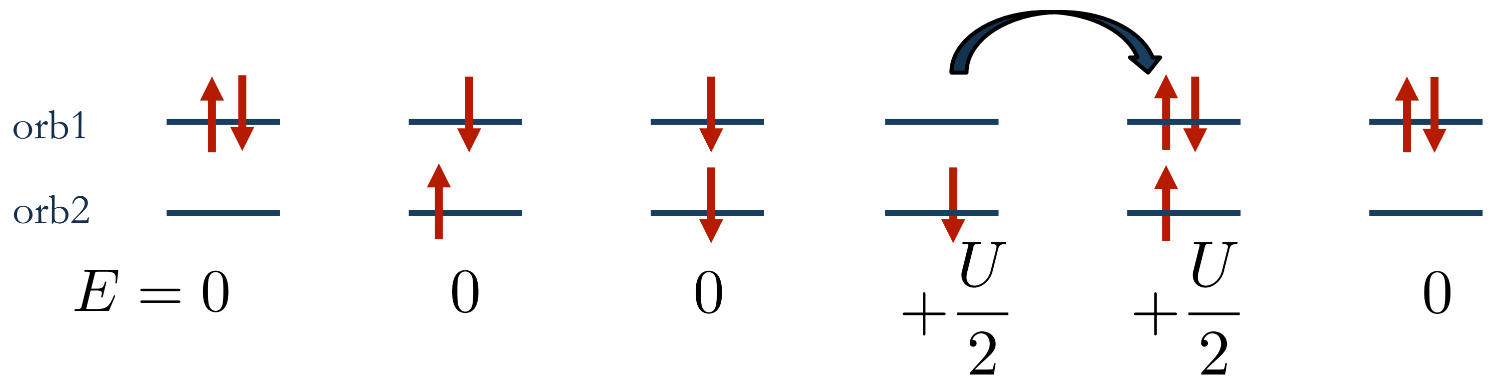
Mott gap for multiorbital interaction at J=0

At J=0, the interaction is proportional to the total charge on the site squared

$$H_{int} = \sum_i \frac{U}{2}\Big(\sum_{m\sigma}(n_{im\sigma} - \frac{1}{2})\Big)^2 = \sum_i \frac{U}{2}(n_i^{tot} - M)^2$$

M number of orbitals

all configurations with the same number of electrons on a site have the same interaction energy.



$$\Delta_{at} = E_{at}(n+1) + E_{at}(n-1) - 2E_{at}(n)$$

The atomic potential barrier is still U

$$\boxed{\Delta_{at} = U}$$

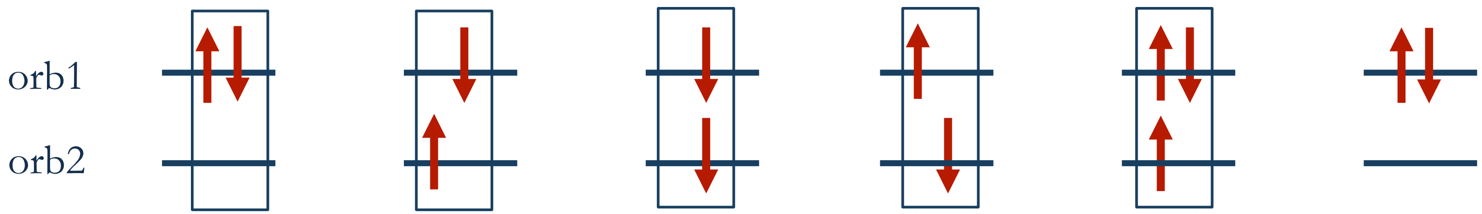
Hubbard bands at J=0

At J=0, the interaction is proportional to the total charge on the site

$$H_{int} = \sum_i \frac{U}{2}\left(\sum_{m\sigma}\left(n_{im\sigma} - \frac{1}{2}\right)\right)^2 = \sum_i \frac{U}{2}\left(n_i^{tot} - M\right)^2$$

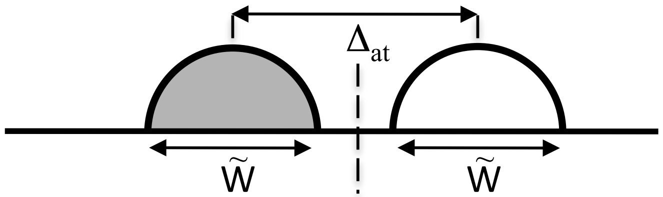
M number of orbitals

all configurations with the same number of electrons on a site have the same interaction energy.



More hopping channels, larger degeneracy-removal energy

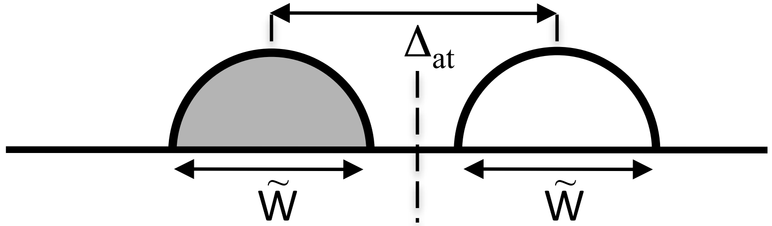
Wider Hubbard bands



$$\tilde{W} = \sqrt{M}\, W$$

Gunnarsson et al.
PRB 56, 1146 (1997)

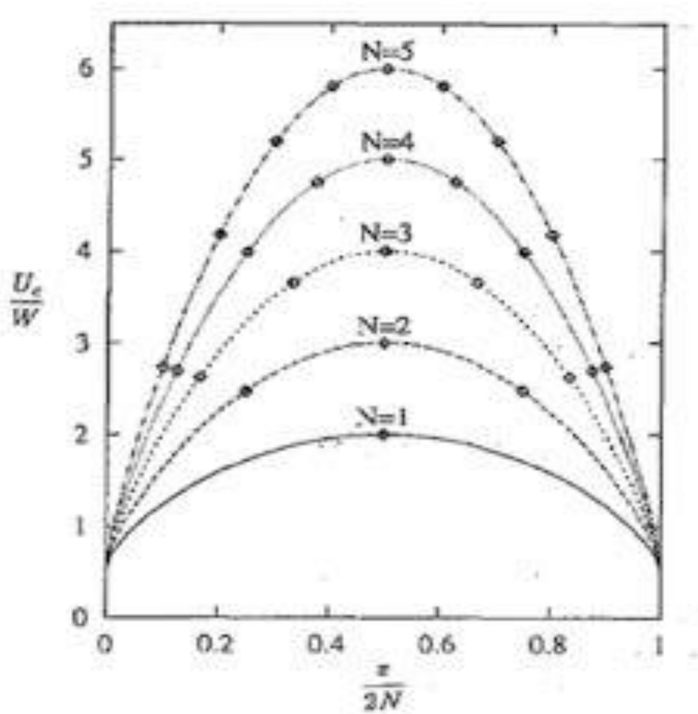
Mott transition in multi-orbital models



N orbitals

$$\Delta = U - \tilde{W} \quad \Rightarrow \quad \boxed{U_c = \tilde{W} = \sqrt{M}\, W}$$

Mott transitions at $\boxed{J=0}$



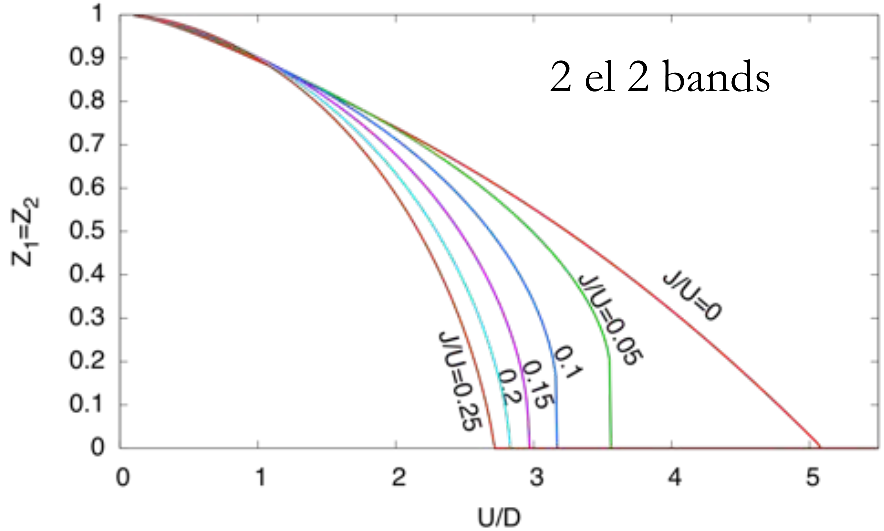
- Mott transition at <u>every integer filling</u>, Uc grows with M
- Uc is maximum at half-filling

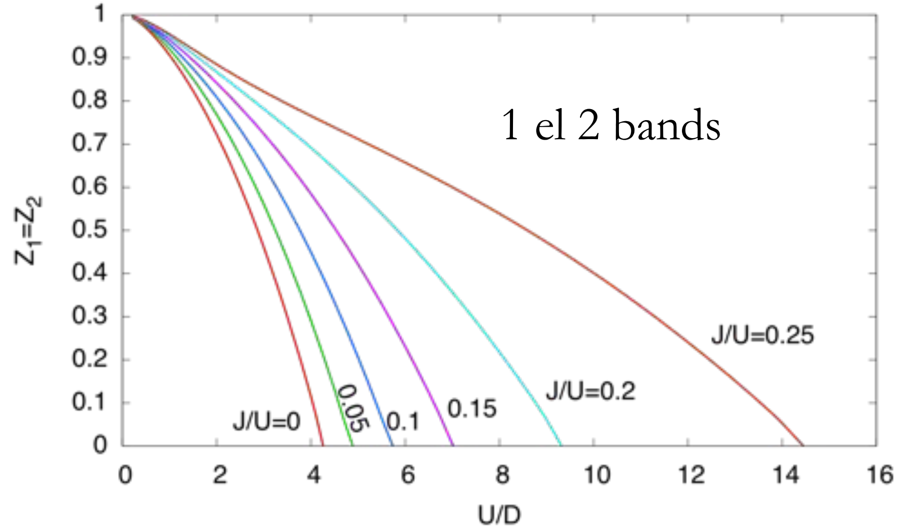
Lu PRB 49 5687 (1994), Rozenberg PRB 55 R4855 (1997), Florens et al. 66 205102 (2002)

M-orbital degenerate Hubbard model: Slave-spins

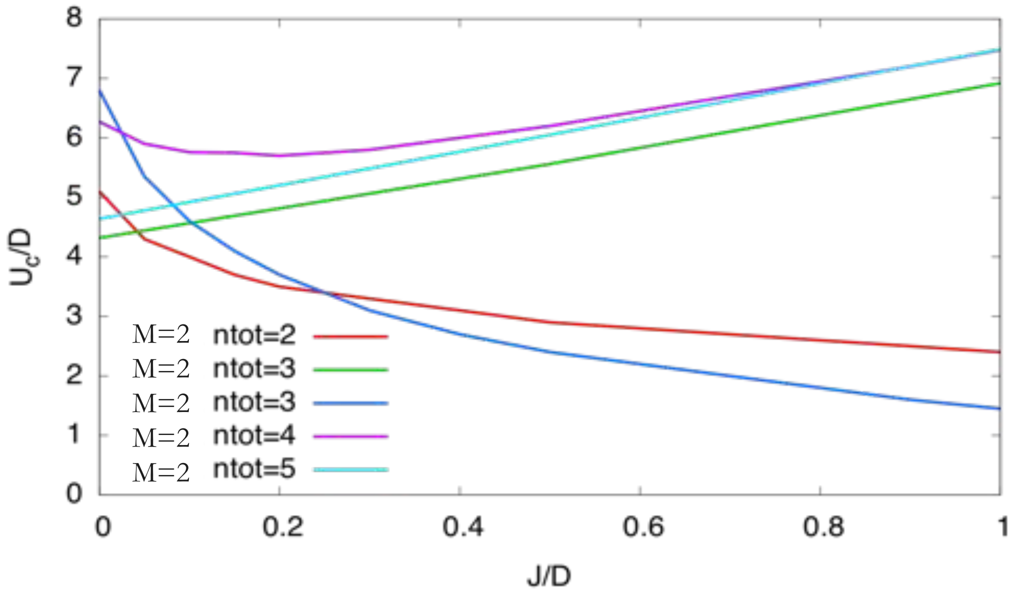
effect of Hund's J

Quasiparticle renormalization factor $Z=(m^*/m)^{-1}$





Uc is lowered by J only for half-filling. In all other cases is <u>enhanced</u>

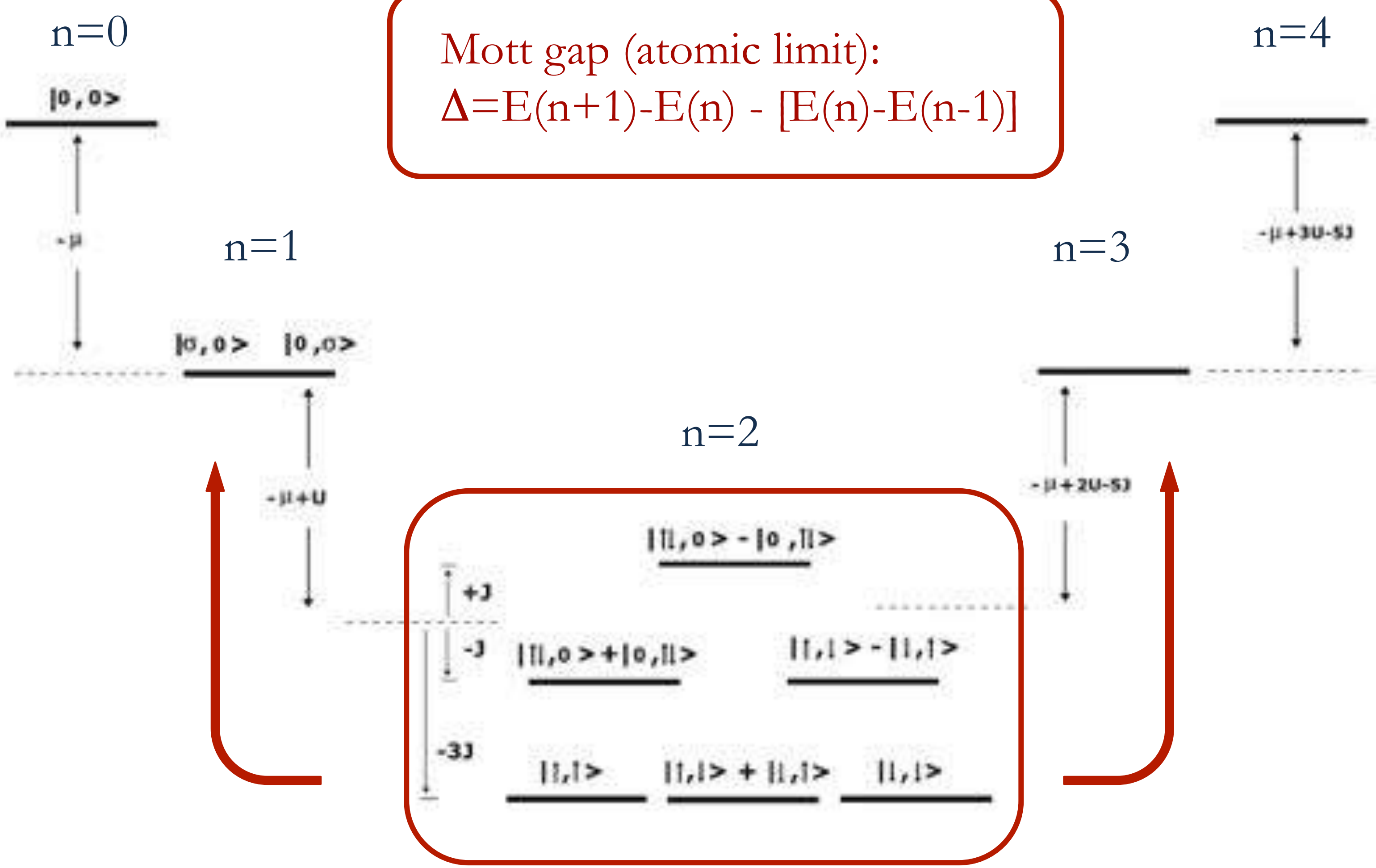


L. de' Medici, PRB 83, 205112 (2011)
see also
Werner, Gull and Millis PRB 79, 115119 (2009)

Hund's coupling changes the Mott gap: increased at ½ filling

Mott gap (atomic limit):

$\Delta = E(n+1) - E(n) - [E(n) - E(n-1)]$

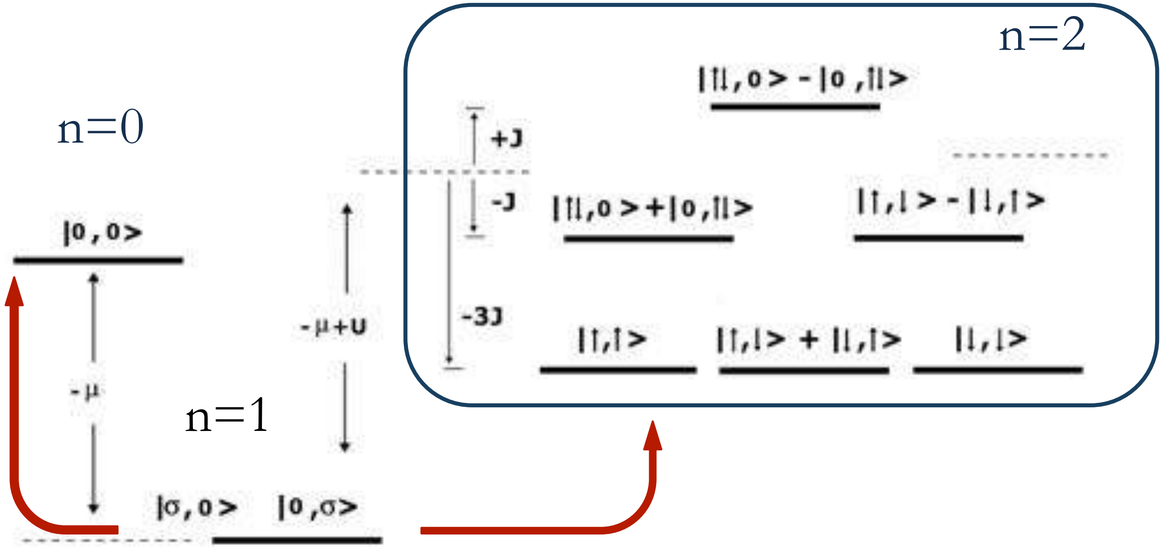


At half-filling $\Delta = U + J$ → The gap increases with J

Needs a smaller U for the Mott transition: Uc decreases

Hund's coupling changes the Mott gap: decreased for a generic filling

For <n>=1 the situation is inverted

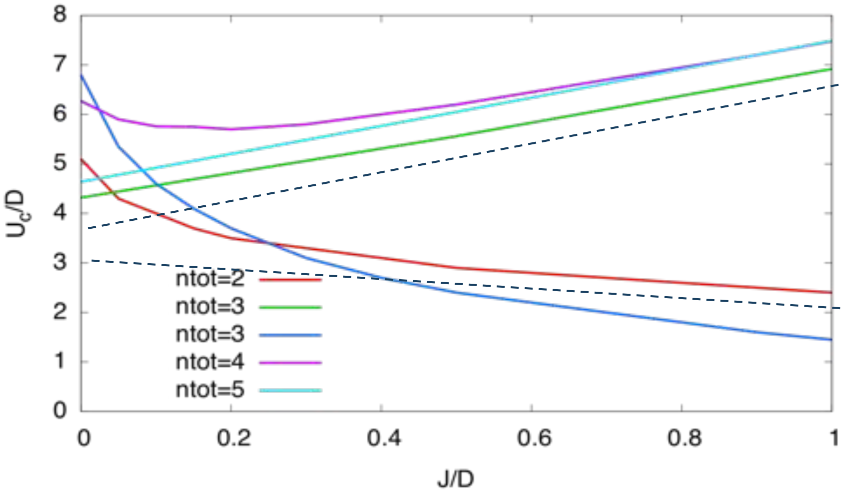


Δ=U-3J

The gap is reduced by J

Needs a <u>larger</u> U for the Mott transition:

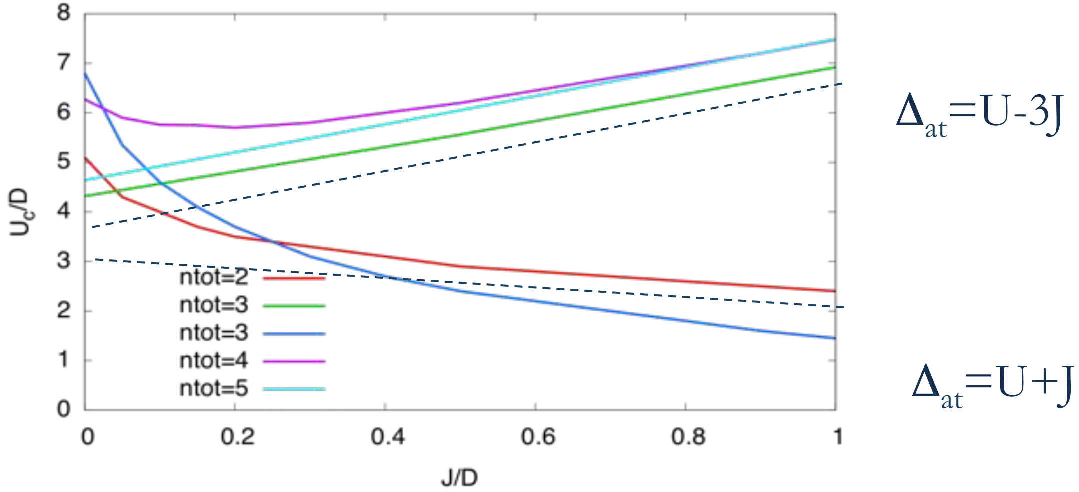
Uc increases



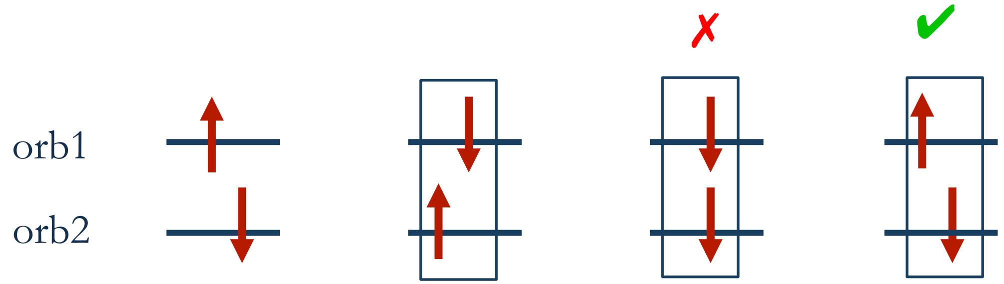
Δ=U-3J

Δ=U+J

Suppression of orbital fluctuations by J



In all cases different from one electron (n=1) or one hole (n=2M-1) filling, there is a fast suppression of Uc at small J



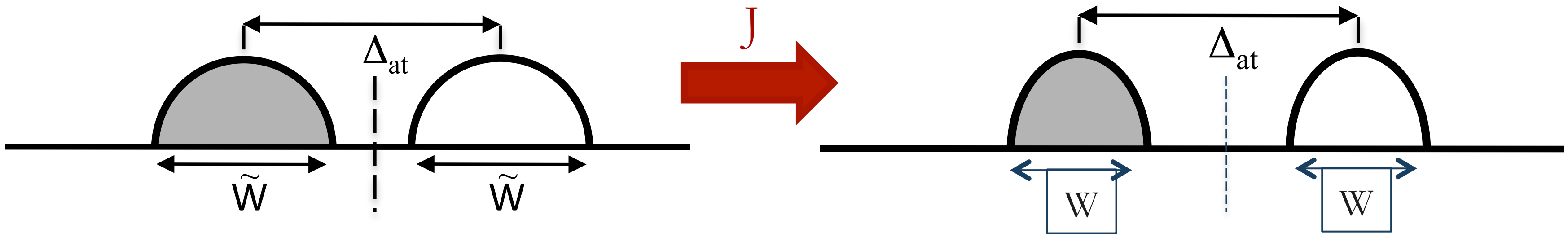
$$\Delta = U + J - W$$

$$\Rightarrow U_c = W - J$$

$$\boxed{= U_c(N=1) - J}$$

J limits again the hopping channels, smaller dispersion

$$\tilde{W} = \sqrt{M}\, W$$



Ground-state degeneracy and hopping processes

Both the Hubbard band widths and the quasiparticle mass are governed by the same hopping processes which are the less effective the lower the ground-state degeneracy

Hund's coupling lowers the ground-state degeneracy

$\Rightarrow$

- reduces the width Hubbard band
- reduces the quasiparticle weight (enhances the effective mass)

2 orbitals		
n	ground state degeneracy	
	J=0	finite J
0 (or 4)	1	1
1 (or 3)	4	4
2	6	3

3 orbitals		
n	ground state degeneracy	
	J=0	finite J
0 (or 6)	1	1
1 (or 5)	6	6
2 (or 4)	15	9
3	20	4

orbital decoupling mechanism

LdM and Capone in "The Iron-pnictide superconductors", Springer book 2017
see also Fanfarillo and Bascones, PRB 92 075136 (2015)

$\boxed{J\neq 0}$

2 orbitals at half-filling – atomic limit

$\boxed{J=0}$

Energy

ground state
6 x degenerate

$|\uparrow\downarrow, 0\rangle \qquad |0, \uparrow\downarrow\rangle$

$(|\uparrow,\downarrow\rangle - |\downarrow,\uparrow\rangle)/\sqrt{2}$

$|\downarrow,\downarrow\rangle \qquad (|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle)/\sqrt{2} \qquad |\uparrow,\uparrow\rangle$

Fermi-liquid coherence (DMFT perspective): 2-orbital Anderson Impurity model
Schrieffer –Wolff transformation -> Kondo model -> Kondo Temperature T_K

The low-energy, high-spin sector, allowed t^2 processes are only diagonal:
T_K is diagonal in orbital space (i.e. it differs if $t_1\neq t_2$)!

Schrieffer, J. Appl. Phys. 1967

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LdM and Capone in "The Iron-pnictide superconductors", Springer book 2017
see also Fanfarillo and Bascones, PRB 92 075136 (2015)

2 orbitals at half-filling – atomic limit

$\boxed{J=0}$

ground state
6 x degenerate

$\boxed{J\neq 0}$

$|\uparrow\downarrow, 0\rangle \quad |0, \uparrow\downarrow\rangle$

$(|\uparrow,\downarrow\rangle - |\downarrow,\uparrow\rangle)/\sqrt{2}$

$|\downarrow,\downarrow\rangle \quad (|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle)/\sqrt{2} \quad |\uparrow,\uparrow\rangle$

Energy

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T_K is diagonal in orbital space (i.e. it differs if $t_1 \neq t_2$)!

Schrieffer, J. Appl. Phys. 1967

Analytical Slave-spin results

$$\bar{\epsilon}_0 \equiv \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} \langle f^\dagger_{\mathbf{k}\sigma} f_{\mathbf{k}\sigma} \rangle_f$$ bare kinetic energy

- 1-band Hubbard $U_c = -16\bar{\epsilon}_0$ (=3.39 for Bethe W=2D)
- 2-band Hubbard (J=0) $U_c = -24\bar{\epsilon}_0$ (=5.09 for Bethe W=2D)
- N-band Hubbard (J=0) $U_c = -8(N+1)\bar{\epsilon}_0$
- 2-band Hubbard (J≠0, large J):
  - Kanamori interaction (w/ spin-flip and pair-hopping terms)

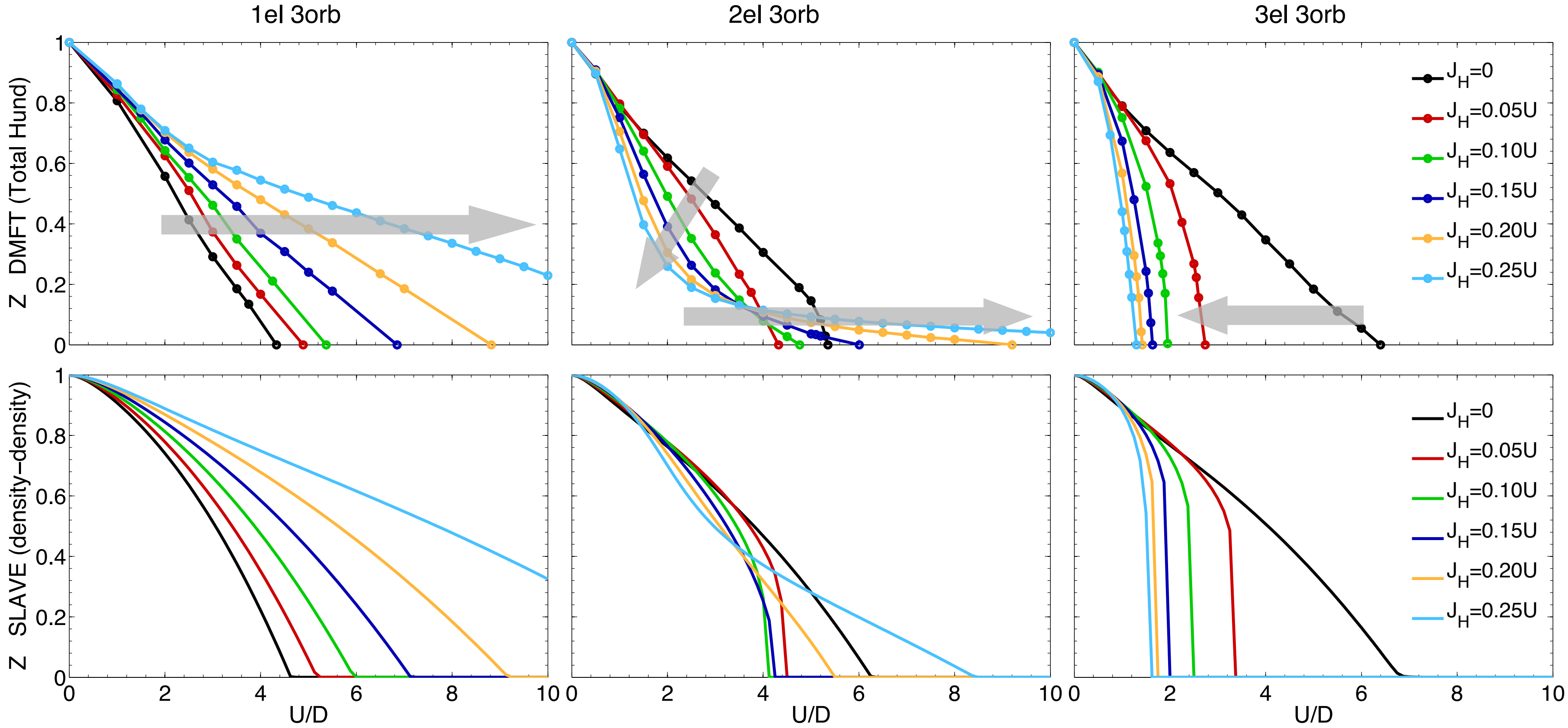
    $$U_c(N=2) = -16\bar{\epsilon}_0 - J = U_c(N=1) - J$$
  - density-density interaction

    $$U_c(N=2) = -8\bar{\epsilon}_0 - J = U_c(N=1)/2 - J$$

L. de' Medici and M. Capone in *The Iron Pnictide Superconductors*, Springer Series in Solid-State Sciences, Vol. 186, pp. 115-185, Mancini, F., Citro, R. (Eds) 2017.

3-band Hubbard model (semicircular DOS)

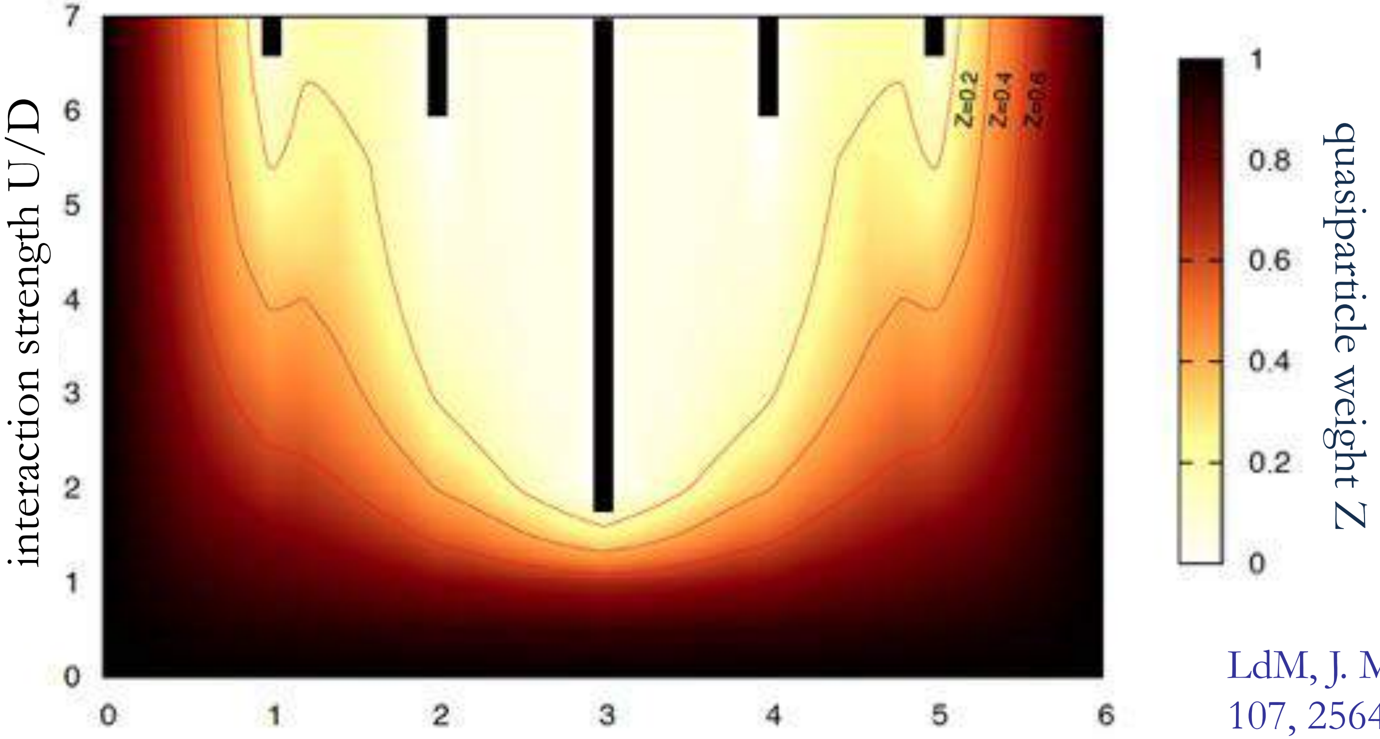
M=3 orbitals (relevant for t_{2g} materials)



Favored: metal bad metal Mott insulator

LdM, J. Mravlje, A. Georges, PRL 107, 256401 (2011)
Fanfarillo and Bascones PRB 92, 075136 (2015)

3-band Hubbard model (semicircular DOS)



J/U=0.15

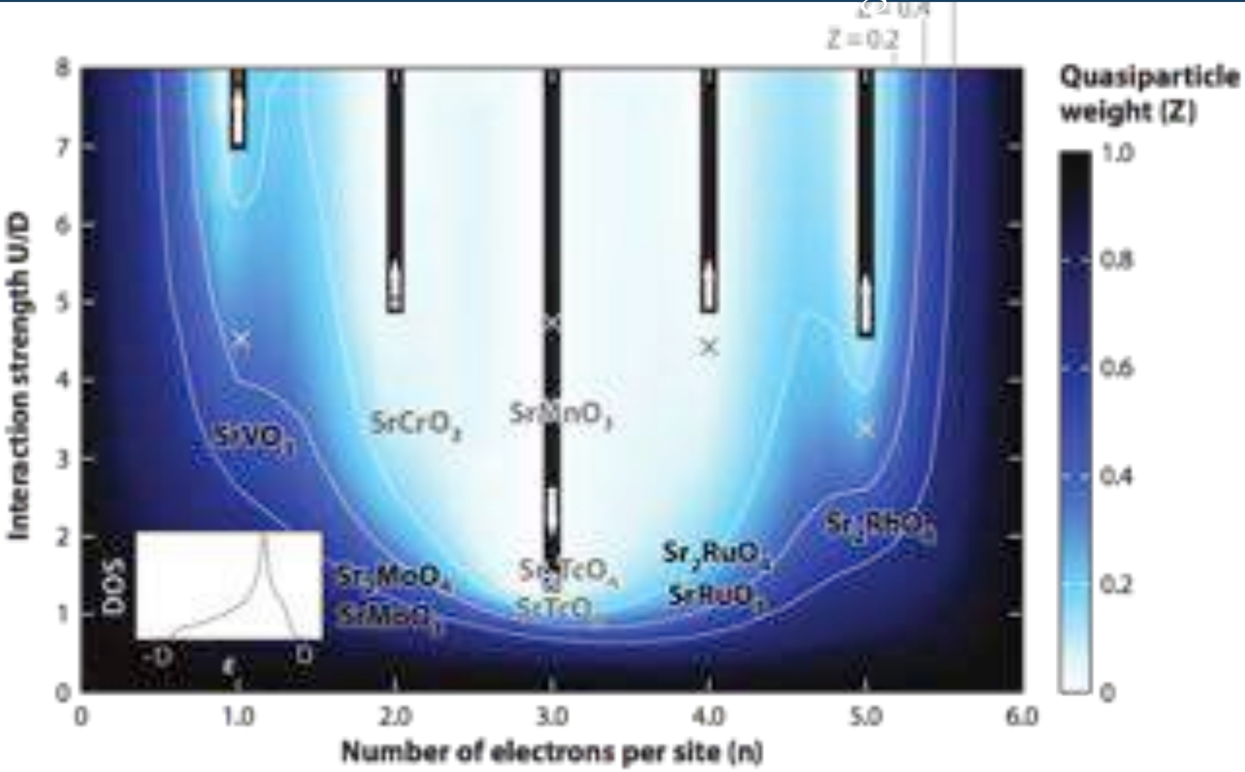
DMFT

LdM, J. Mravlje, A. Georges, PRL 107, 256401 (2011)

Number N of electrons in M orbitals	Degeneracy of atomic ground-state	Mott gap	Correlations	Materials behaviour promoted by J
one electron or one hole ($N = 1, 2M - 1$)	unaffected	reduced	diminished	metallic
half-filled ($N = M$)	reduced	increased	increased	insulating
All other cases ($N \neq 1, M, 2M - 1$)	reduced	reduced	Conflicting effect (see text)	bad metallic

Table I: The effects of an increasing Hund's rule coupling on the degree of correlations.

3-band Hubbard model (t_{2g} density of states)

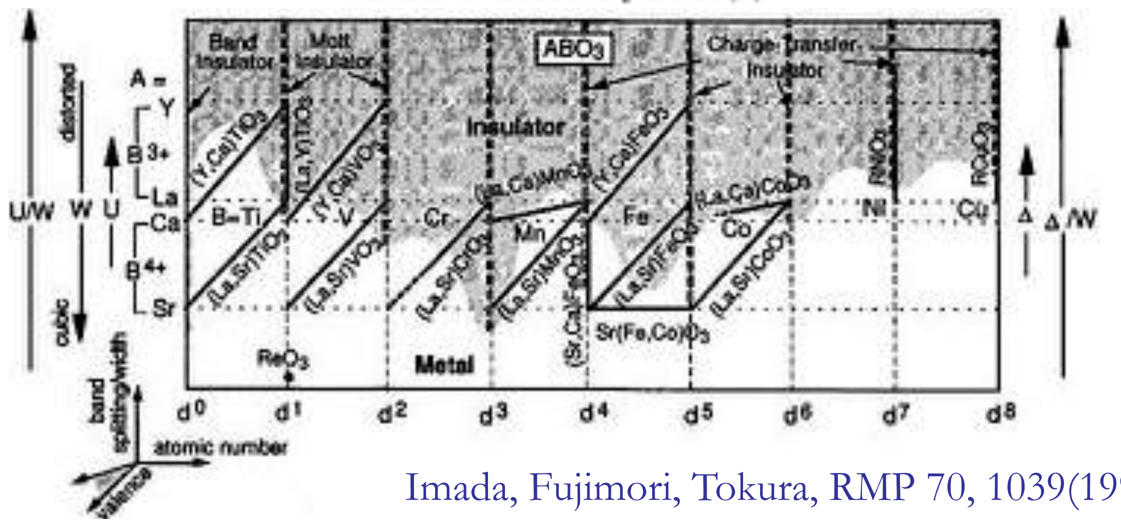


J/U=0.15

DMFT

- 103 cubic
- 214 tetragonal

(further splitting)



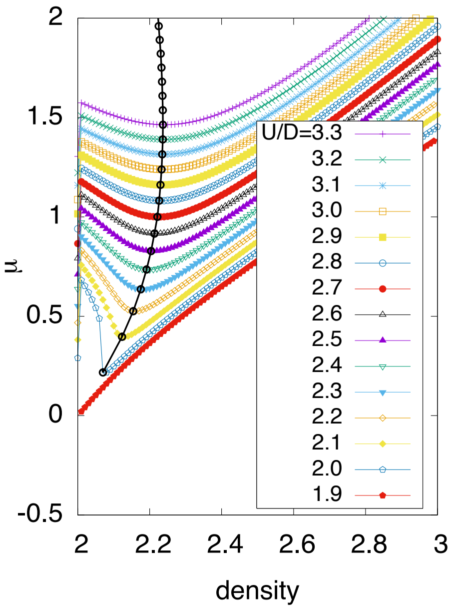
A. Georges, LdM, J. Mravlje, Annual Reviews Cond. Mat. 4, 137 (2013)

Imada, Fujimori, Tokura, RMP 70, 1039(1998)

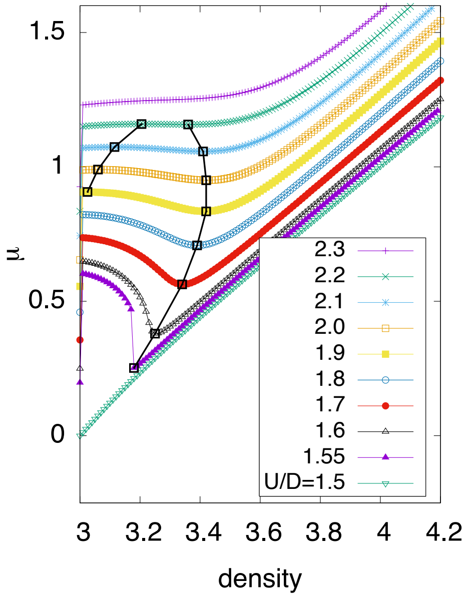
Compressibility in 2/3/5-orbital Hubbard model

J/U=0.25

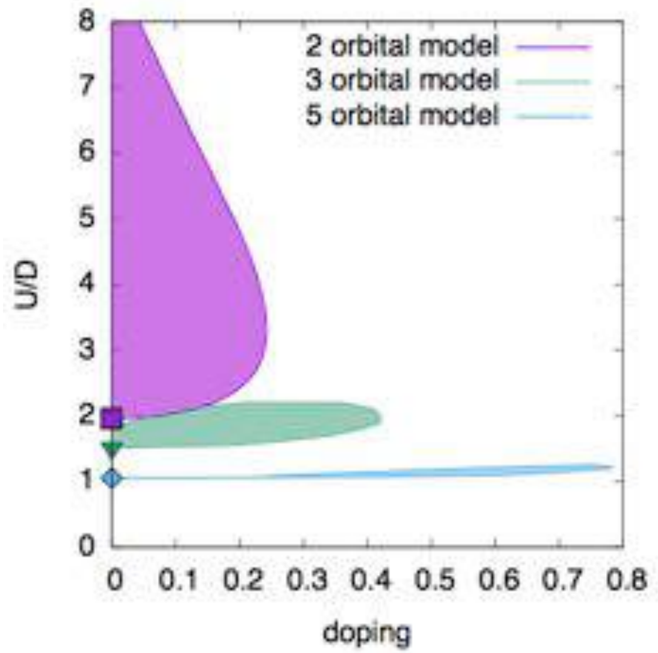
LdM, PRL 118 (2017)



2 orbitals



3 orbitals
(5 orb analogous)

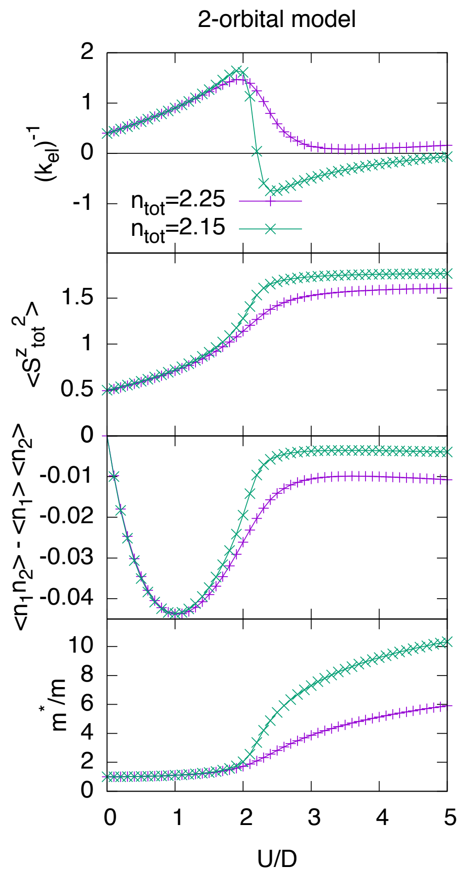


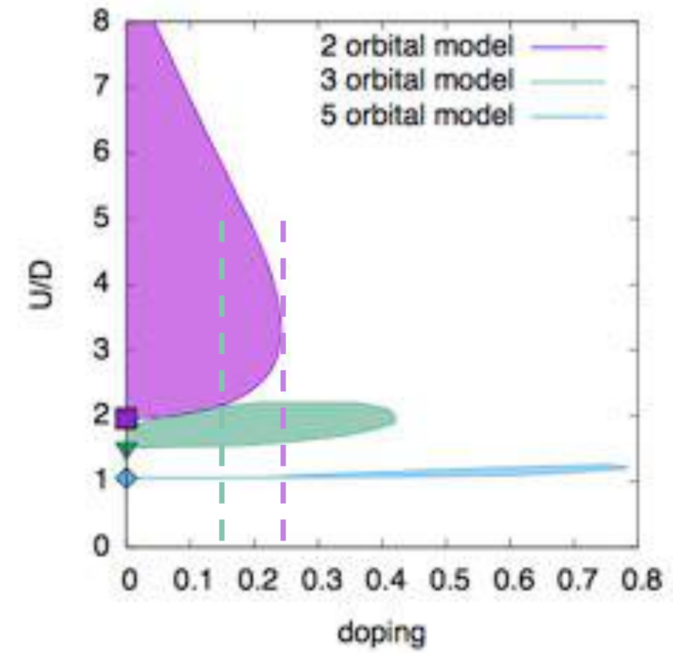
$$\chi = \frac{dn}{d\mu}$$

Divergence of the compressibility on a cross-over line departing from the Mott transition at half filling

Hund's metal frontier and enhanced compressibility

LdM, PRL 118 (2017)

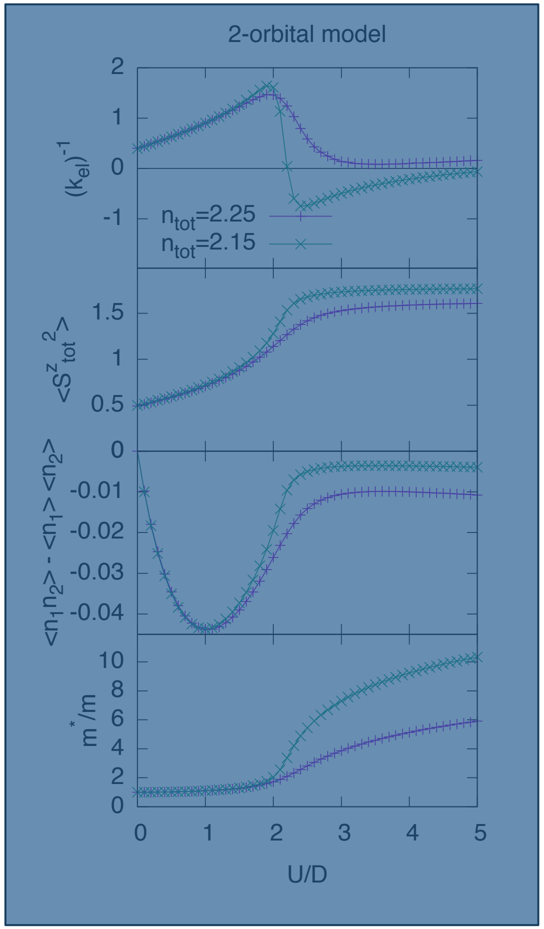


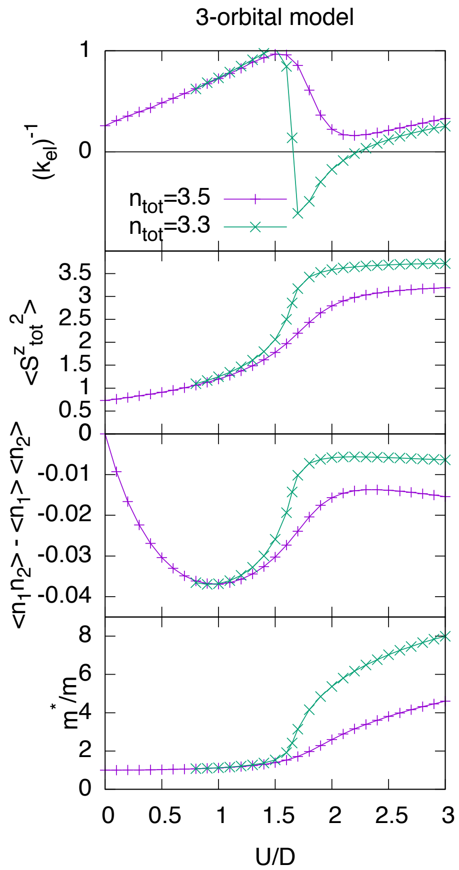


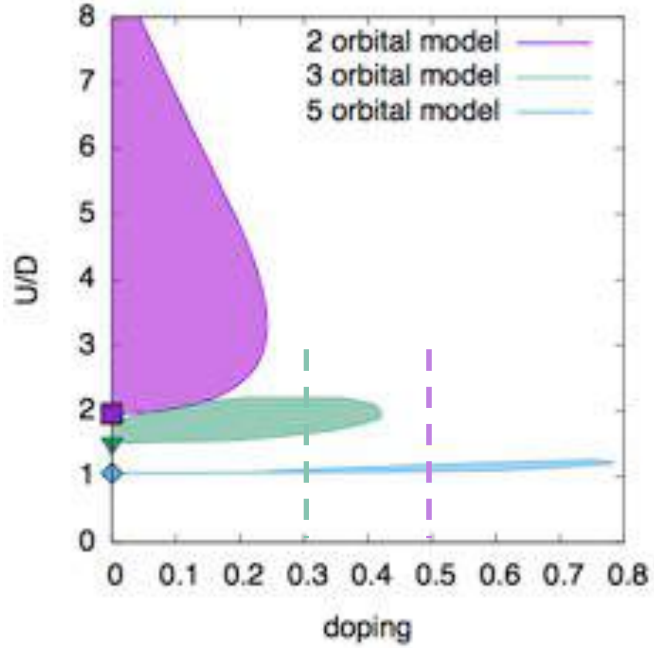
The enhanced/divergent compressibility always occurs <u>near (just inside) the Hund's metal frontier</u>

Hund's metal frontier and enhanced compressibility

LdM, PRL 118 (2017)







The enhanced/divergent compressibility always occurs <u>near</u>
<u>(just inside) the Hund's metal frontier</u>

Enhanced compressibility and superconductivity

In a Fermi liquid:

- $$\chi = \frac{\chi_0/Z}{1+F_0^s}$$

  If χ diverges for a finite Z → $F_0^s < 0$
  → attraction (q=0, ω -> 0) between quasiparticles

- in presence of some electron-boson coupling:
  (Ward identity for the density vertex)

  $$\Lambda(q \to 0, \omega = 0) = \frac{1}{Z(1+F_0^s)}$$

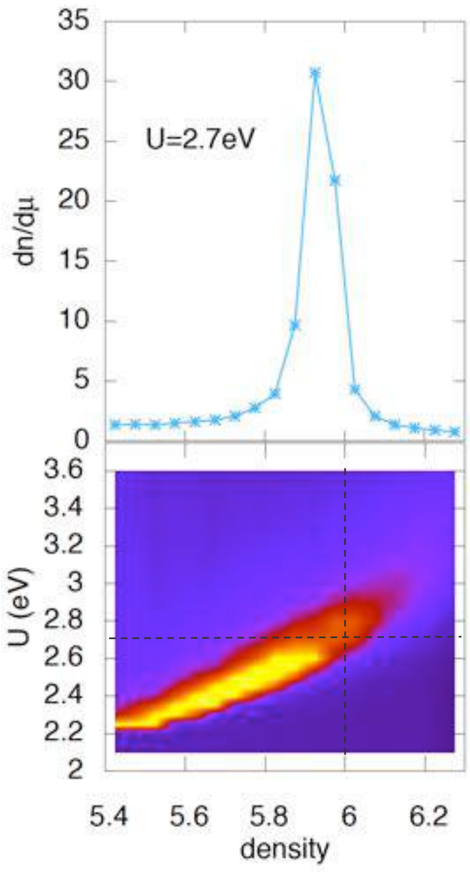
  → enhanced ~ χ

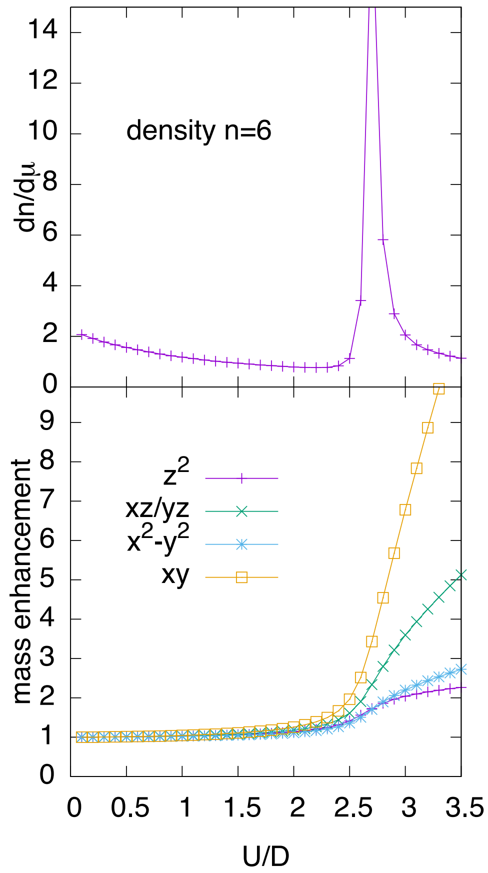
- Phase separation → superconductivity scenario very much studied in the 90's for Cuprates
  cfr: Emery, Kivelson and Lin, PRL 64, 475 (1990)
  Grilli et al. PRL 67, 259 (1991)
  Castellani, Di Castro and Grilli, PRL 75, 4650 (1995) , …

In this region not only the quasiparticle energies are renormalized non-trivially, but also their interactions (mutual and with low-energy bosons)!

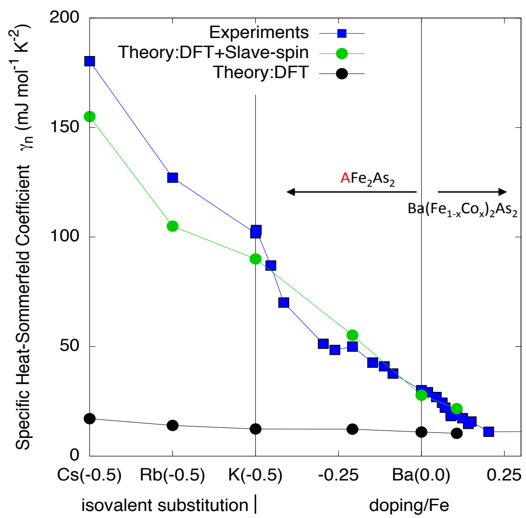
Enhanced compressibility in $BaFe_2As_2$

LdM, PRL 118 (2017)





U=2.7eV, J/U=0.25
(interaction parameters that capture the Sommerfeld coefficient in the whole 122 family)



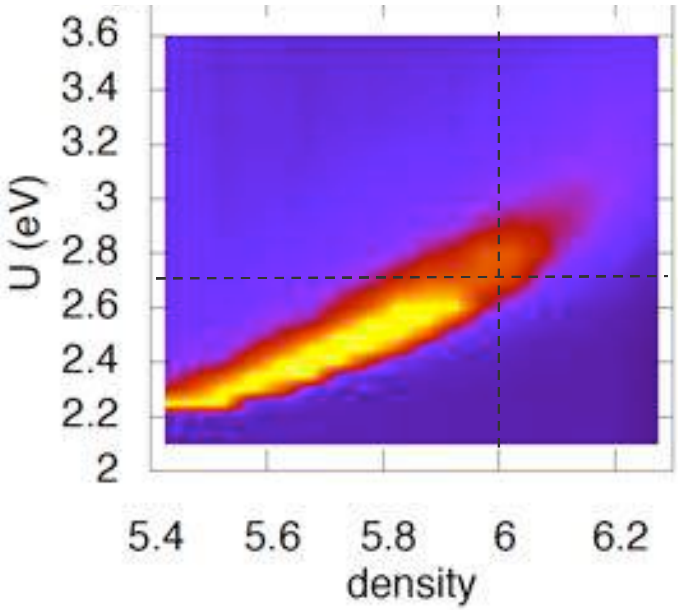
F. Hardy et al. PRB 94, 205113 (2017)

compressibility enhanced:

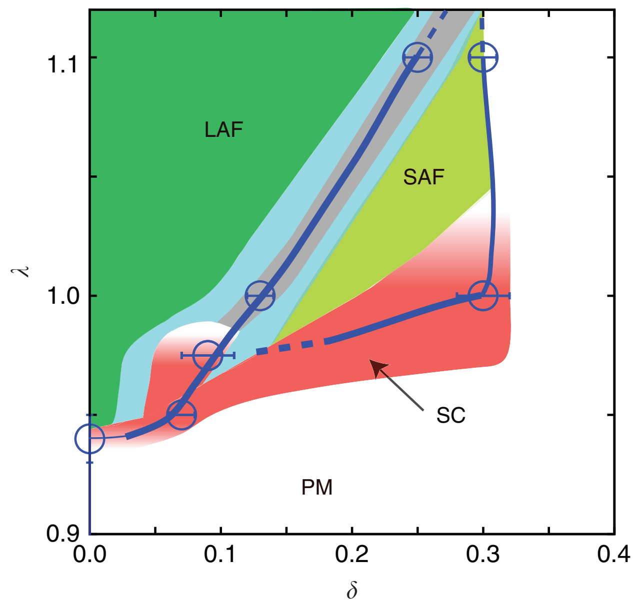
- in the doping zone where high-Tc happens
- at the entrance of the Hund's metal zone

Phase separation in VQMC

$BaFe_2As_2$



LaFeAsO



LdM, PRL 118, 167003 (2017)

Misawa and Imada, Nat. Comm 5, 5738 (2014)

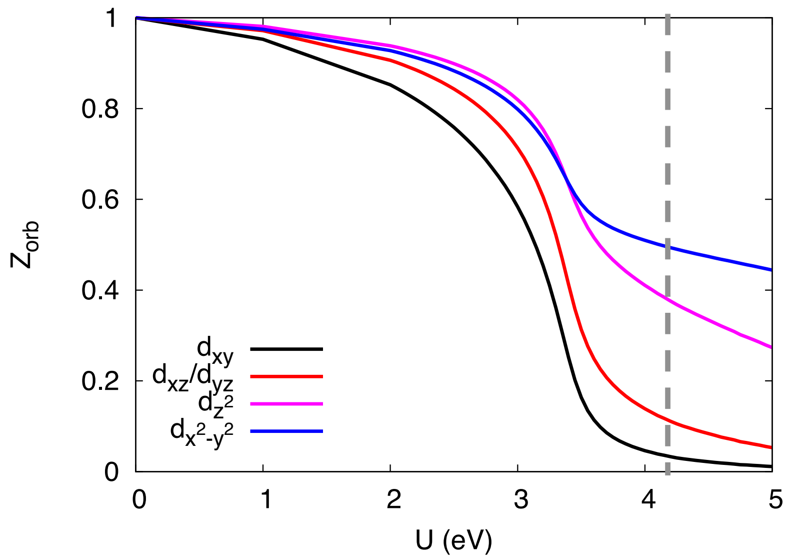
In slave-spin mean-field it is simply a Fermi-liquid instability

- independent of any symmetry breaking
- caused by Hund's coupling
- universal feature of Hund's metals

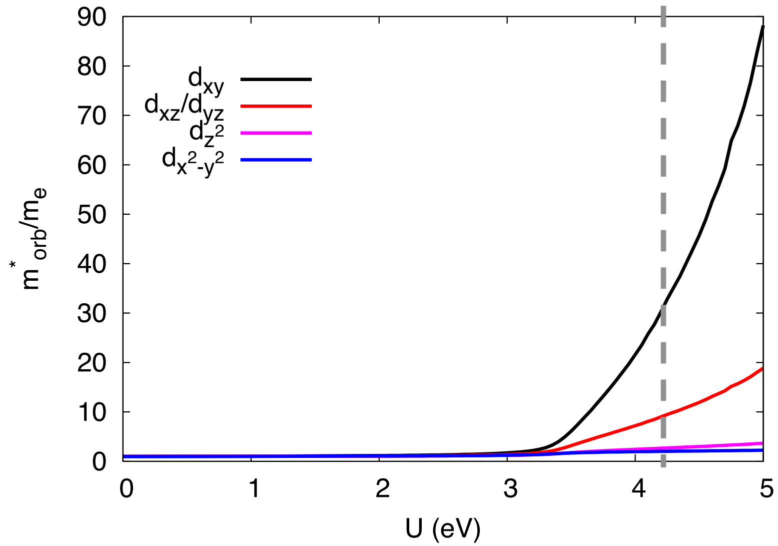
FeSe within DFT+Slave-Spin mean-field

P. Villar Arribi
ESRF

quasiparticle weights



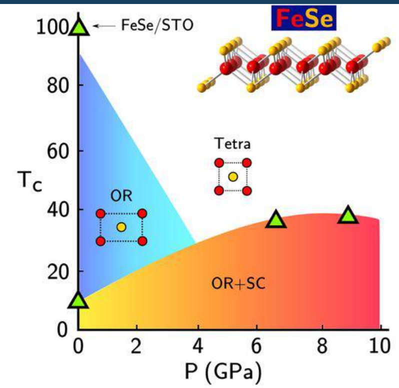
mass enhancements

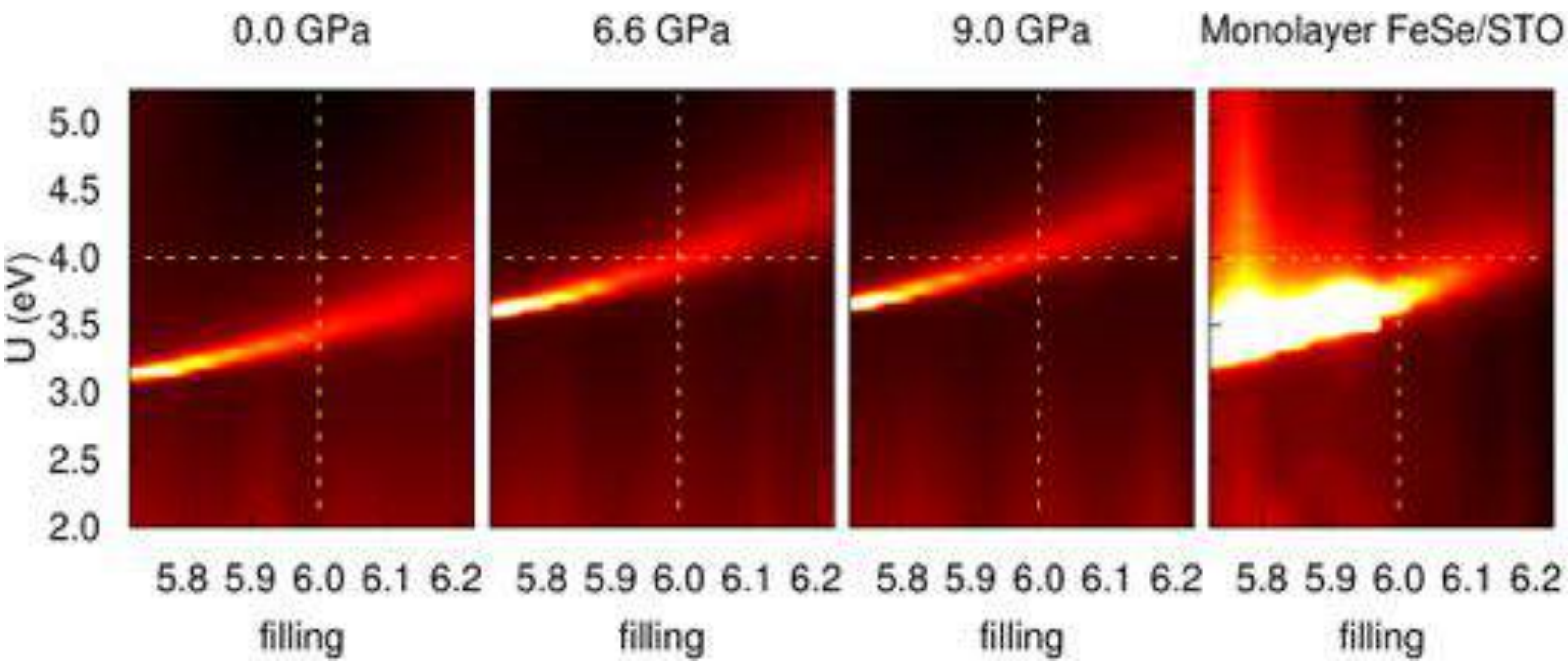


RPA-estimated U : bulk material deep within the "Hund's metal" phase

FeSe/pressure & monolayer

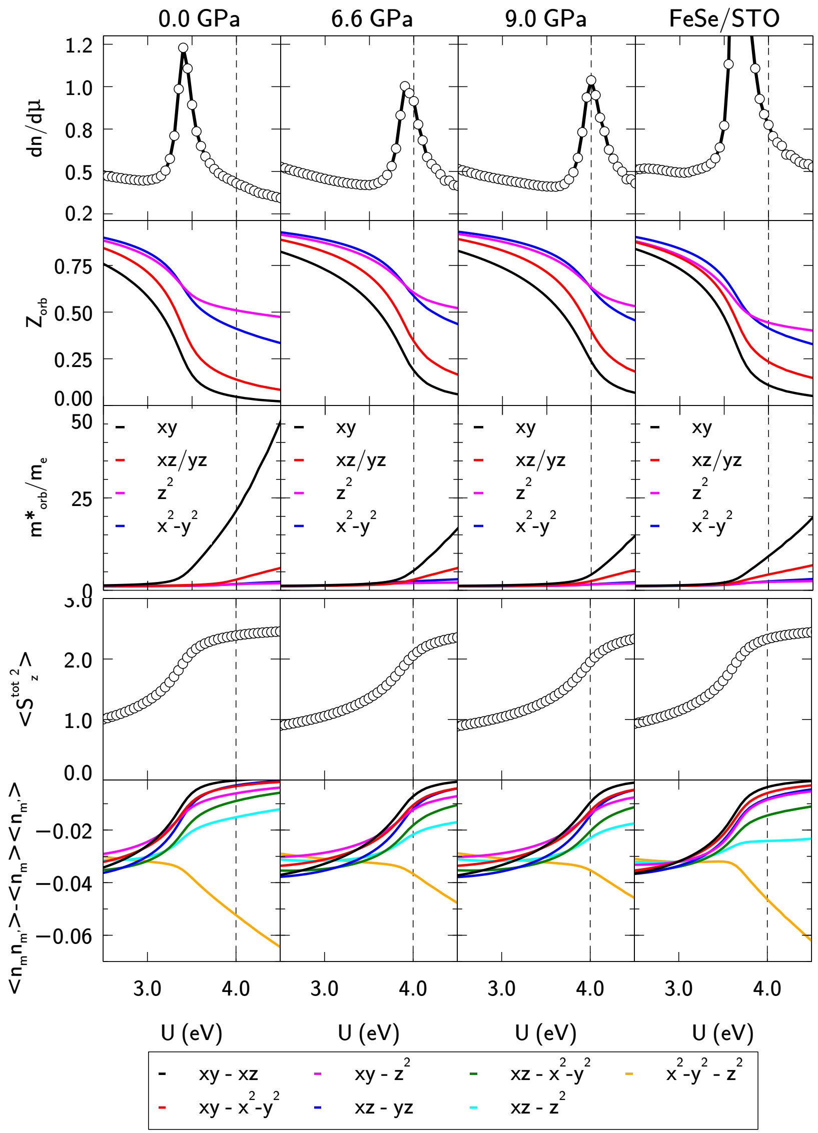
FeSe: Tc grows under pressure
FeSe monolayer: highest claimed T_c

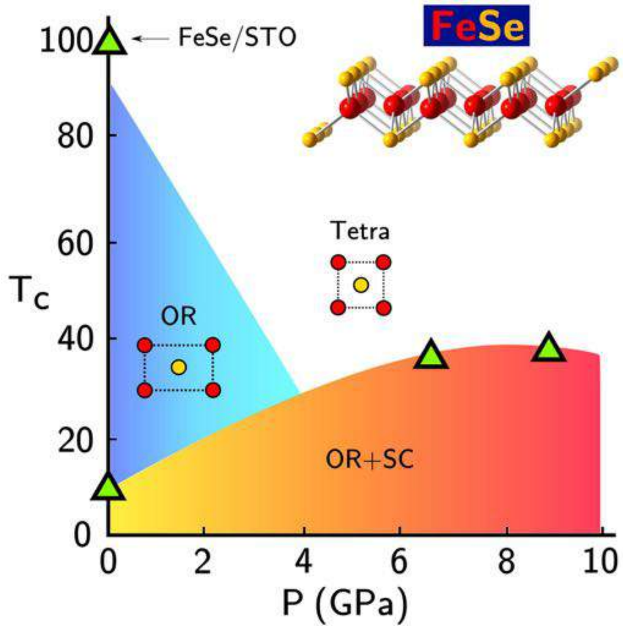




Pablo Villar-Arribi and LdM, unpublished

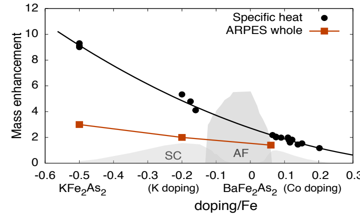
FeSe/pressure & monolayer





Pablo Villar-Arribi and LdM, unpublished

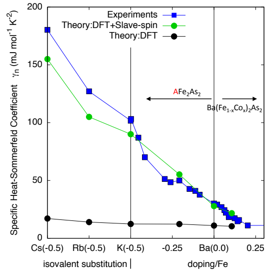
Conclusions and References



- **Hund's metals: high local moments, enhanced correlations, selective**
- **Onset of orbital selectivity easily highlights the Hund's metal frontier**

LdM, G. Giovannetti and M. Capone, PRL 112, 177001 (2014)

Selective Mott Physics as a key to Iron superconductors

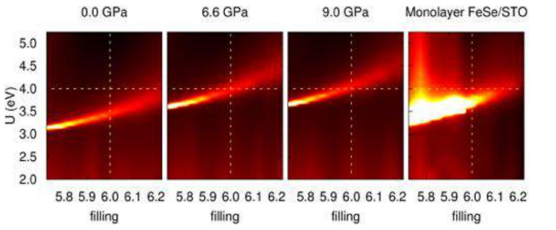


LdM, *Weak AND strong correlations in Fe Superconductors*,

in "Iron-based Superconductivity", Springer Series in Material Sciences, Vol 211, pp 409-441 (2015)

- **Hund's induced phase-separation/enhanced qp interactions at Hund's metal frontier**
- **The mechanism can be associated to orbital decoupling**
- **This can favor superconductivity**

LdM, *Hund's induced Fermi-liquid instabilities and enhanced quasiparticle interactions*
PRL 118, 167003 (2017)



Pablo Villar-Arribi and LdM, *Enhanced compressibility in FeSe induced by Hund's coupling*, unpublished

- **Slave-spins**:

L. de' Medici and M. Capone in *The Iron Pnictide Superconductors*, Springer Series in Solid-State Sciences, Vol. 186, pp. 115-185, Mancini, F., Citro, R. (Eds) 2017.