Electronic correlations in LaFeAsO: Insights from LDA+DMFT calculations

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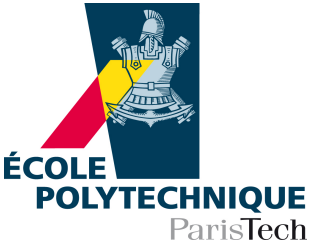
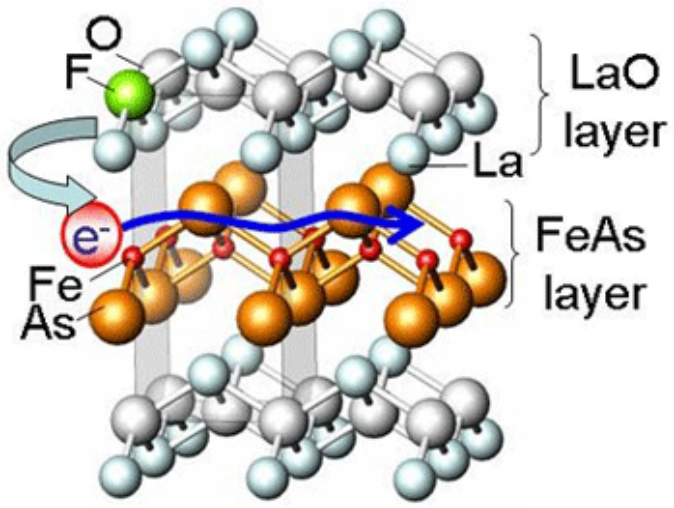


The Pnictide superconductors



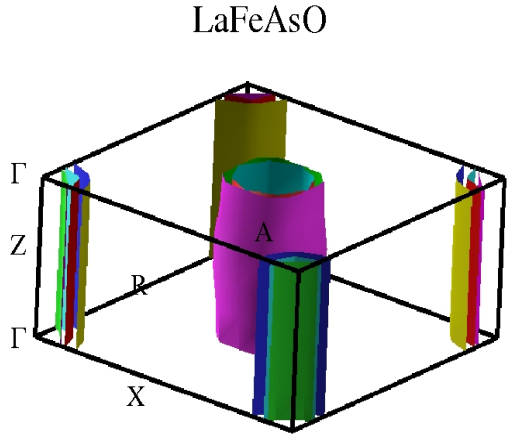
[J. Phys. Soc. J. 77 (2008) 053709]

Several families: 1111: e.g. LaOFeAs
122: e.g. $BaFe_2As_2$
11: e.g. $Fe_{1+x}Se$

'High' T_C of about 55 K (SmOFeAs doped)

Layered structure
charge transfer from LaO layer to FeAs layer

low-energy bands: Fe-3*d*
Multiorbital physics!



Is the 3d band in FeAs superconductors as strongly correlated as in the cuprates?

What's it about

How correlated are the Pnictide superconductors?

Previous calculations give contrary answers:
From strongly correlated to weakly correlated

LDA+DMFT in a nutshell

Renormalization and Self energy

Spectral function

Discussion

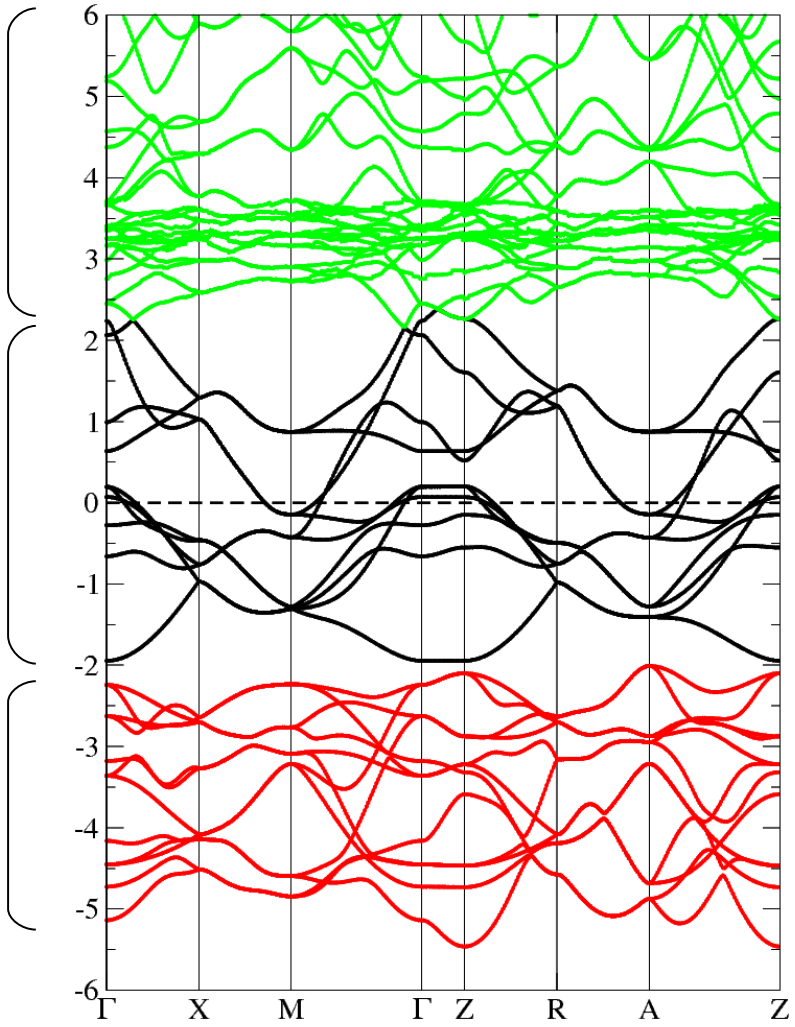
LDA Bands structure and energy window

Unoccupied states (La)

Fe *d*: *W* = 4 eV

As *p*

O *p*



dpp-model:
Energy [-6,+2] eV

quite localized Wanniers

Parameters (cRPA):

$$U_{\mathrm{av}} = 2.79\,\mathrm{eV}$$
$$J = 0.79\,\mathrm{eV}$$

LDA+DMFT in a nutshell

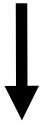
DFT part (Wien2K):

Solve Kohn-Sham Equations

$\Rightarrow H_\nu(\mathbf{k}), |\psi_{\mathbf{k}\nu}\rangle$

Interfacing:

Construct local orbitals and projectors $P^{\alpha,\sigma}_{L\nu}(\mathbf{k})$



DMFT part:

Imp. Solver: $G^{\mathrm{imp}}_{LL'}(i\omega) \Rightarrow \Sigma^{\mathrm{imp}}_{LL'}(i\omega)$

Lattice Green function: $G^{\mathrm{loc}}_{LL'}(i\omega)$

SC condition: $G^{\mathrm{loc}}_{LL'} = G^{\mathrm{imp}}_{LL'}$

No re-interfacing with electronic structure calculation

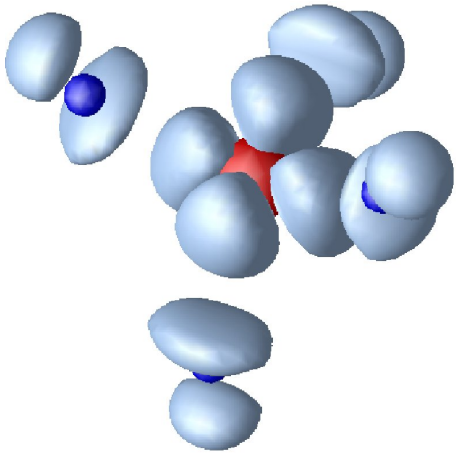
No full self-consistency

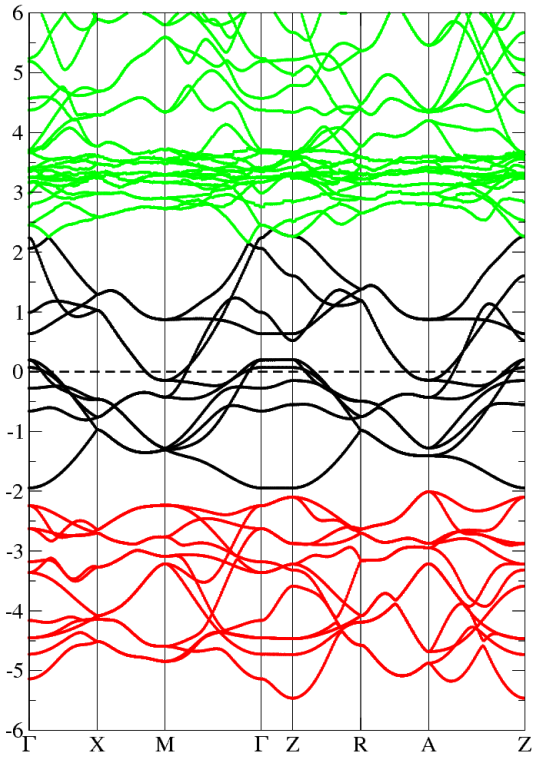
One-shot LDA+DMFT

Local orbitals

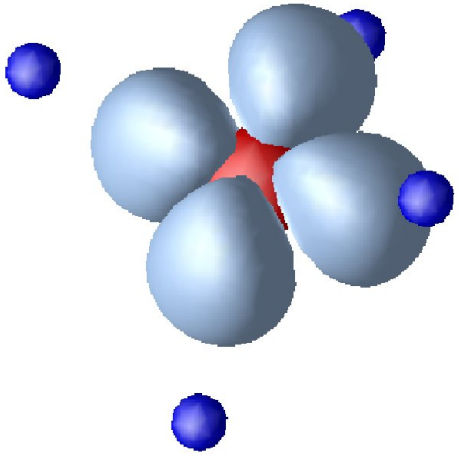
Example: LaFeAsO, Fe- $d_{x^2-y^2}$ orbital

Fe-d bands only





Fe-d, As-p, O-p



More bands included ⟹ more localized local orbitals

Wannier orbitals from: V.I. Anisimov et al., J. Phys.: Cond. Matter 21, 075602 (2009)

Impurity Hamiltonian and Solver

Interaction Hamiltonian (density-density):

$$H_{\mathrm{int}} = \sum_{mm'\sigma\sigma'} U^{\sigma\sigma'}_{m,m'} n_{m\sigma} n_{m'\sigma'}$$

$$U^{\sigma\bar{\sigma}}_{mm'} = U_{mm'mm'}, \qquad U^{\sigma\sigma}_{mm'} = U_{mm'mm'} - U_{mm'm'm}$$

4-index *U*-matrix constructed from Slater integrals:

$$F^0 = U, \quad J = (F^2 + F^4)/14, \quad F^4/F^2 = 0.625$$

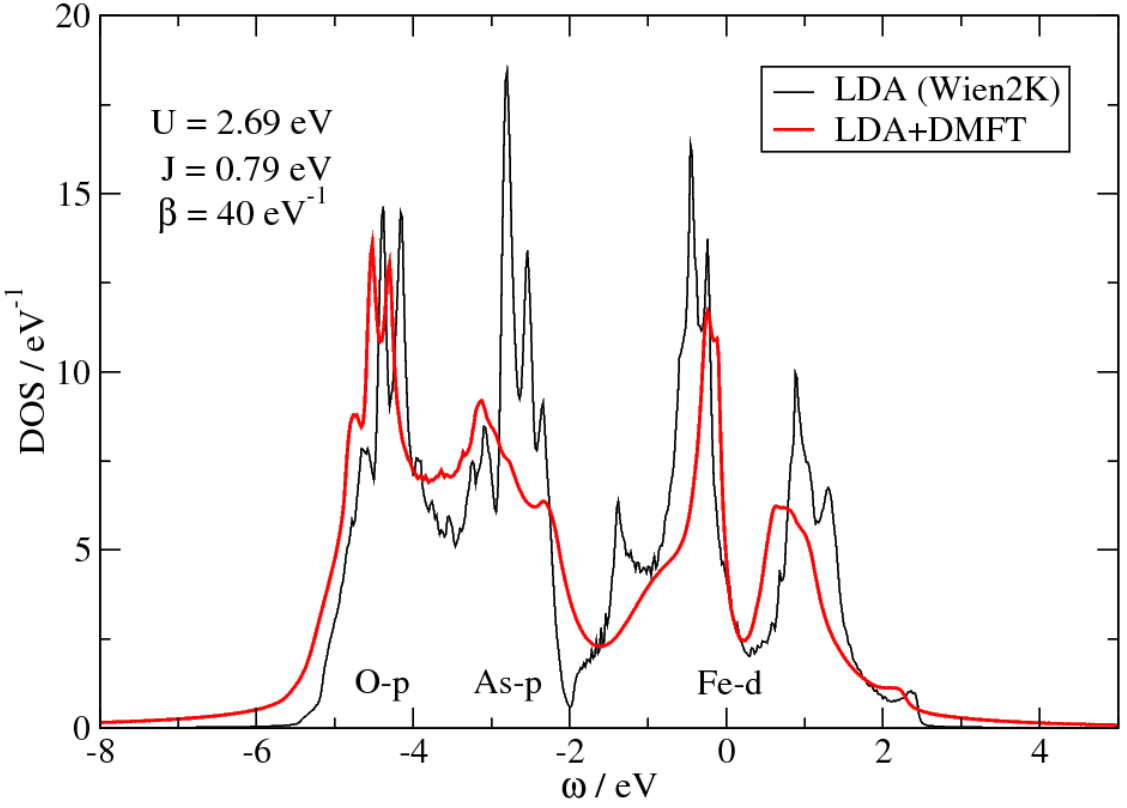
Impurity Solver: **Continuous Time Quantum Monte Carlo**

Hybridisation expansion (P. Werner et al., 2006):
Very efficient solver for quantum impurity problems

Density-Density interactions:
Maximum number of conserved quantities
Efficient update scheme (“Segment picture”)

Inverse temperature $\beta = 40\mathrm{eV}^{-1}$ without problems

DMFT results: total DOS



dpp-hamiltonian:
W = [-5.5,2.5] eV
Fe-*d*, As-*p*, O-*p*

$$U_{\mathrm{av}} = 2.79\,\mathrm{eV}$$
$$J = 0.79\,\mathrm{eV}$$

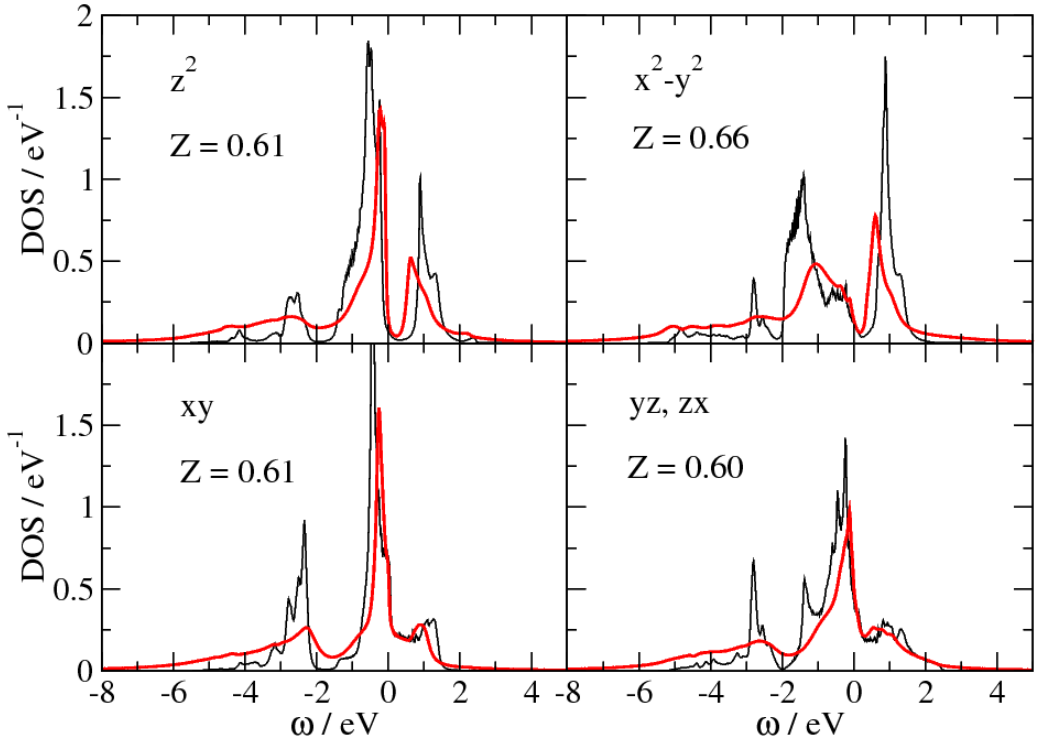
Analytic continuation of the Self energy

Fe-*d* bands renormalized, As-*p* affected by correlations
O-*p* almost unchanged.

Moderate correlation regime: ***Z* = 0.6**

No indication for upper or lower Hubbard bands!

DMFT results: orbitally resolved



All orbitals show similar renormalization:
average ***Z* = 0.62,** effective mass ***m** = 1.62**

Experimentally: Mass renormalization of ***m** = 1.8 - 3.0**

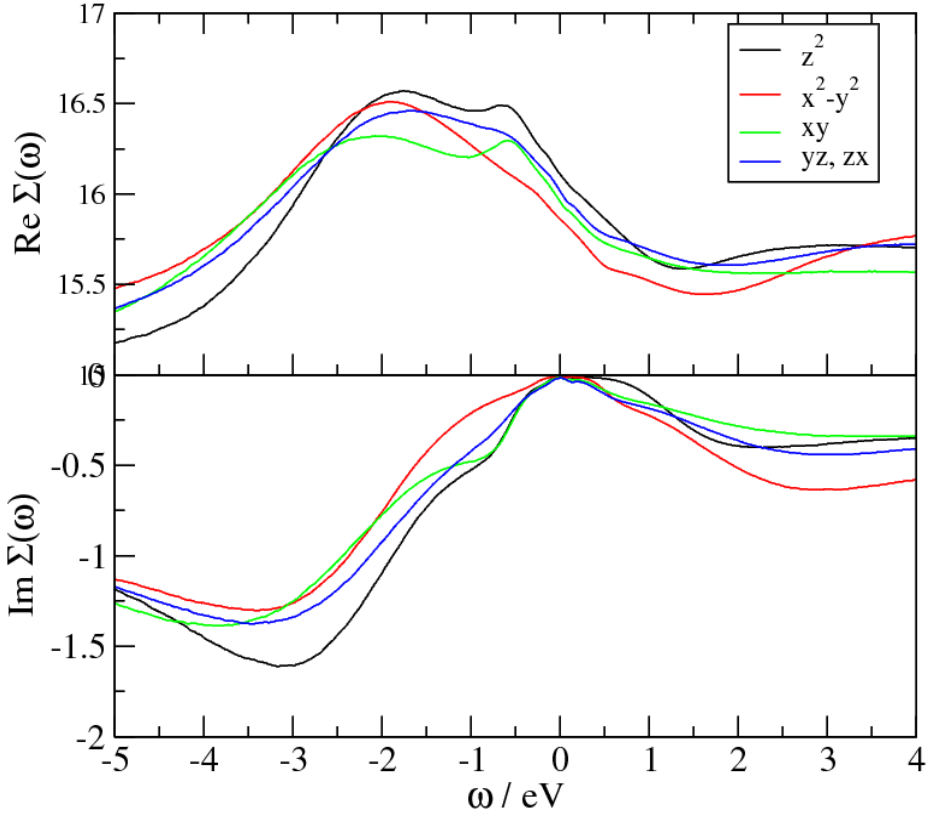
Difference: Neglect of SDW state, no spatial spin fluctuations in single site DMFT

Note: DMFT Spectra at ω = 0 coincide with LDA DOS → Fermi liquid

DMFT results: Self-energy

Analytic continuation of the self energy: $\Sigma(i\omega) \rightarrow \Sigma(\omega + i\delta)$

By stochastic Maximum Entropy (K.S.D. Beach, arXiv:cond-mat/0403055)



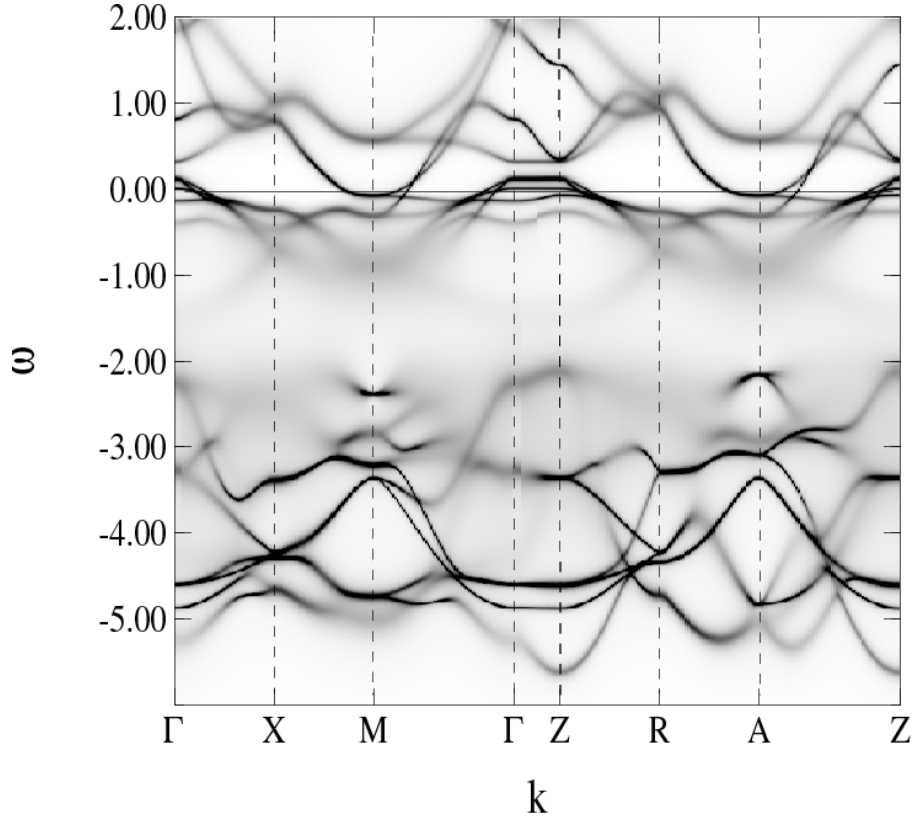
Around ω = 0:

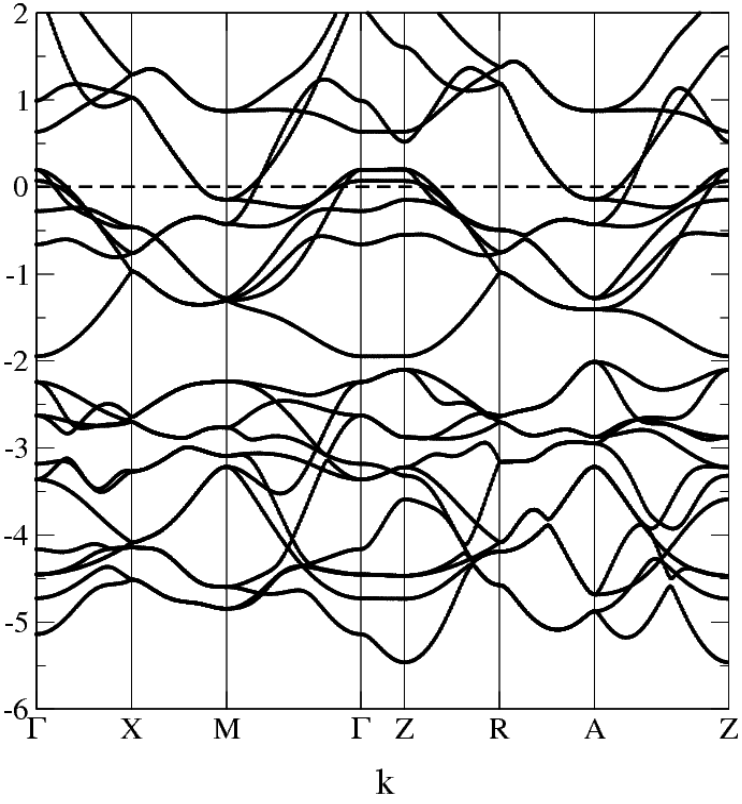
Linear behavior of real part

Small imaginary part, dispersing quadratically

Indication of Fermi liqud behavior

DMFT results: Spectra





Bloch spectral function:

$$A(\mathbf{k},\omega) = -\frac{1}{\pi}\mathrm{Im}\mathrm{Tr}\; G_{\nu\nu'}(\mathbf{k},\omega+i\delta)$$

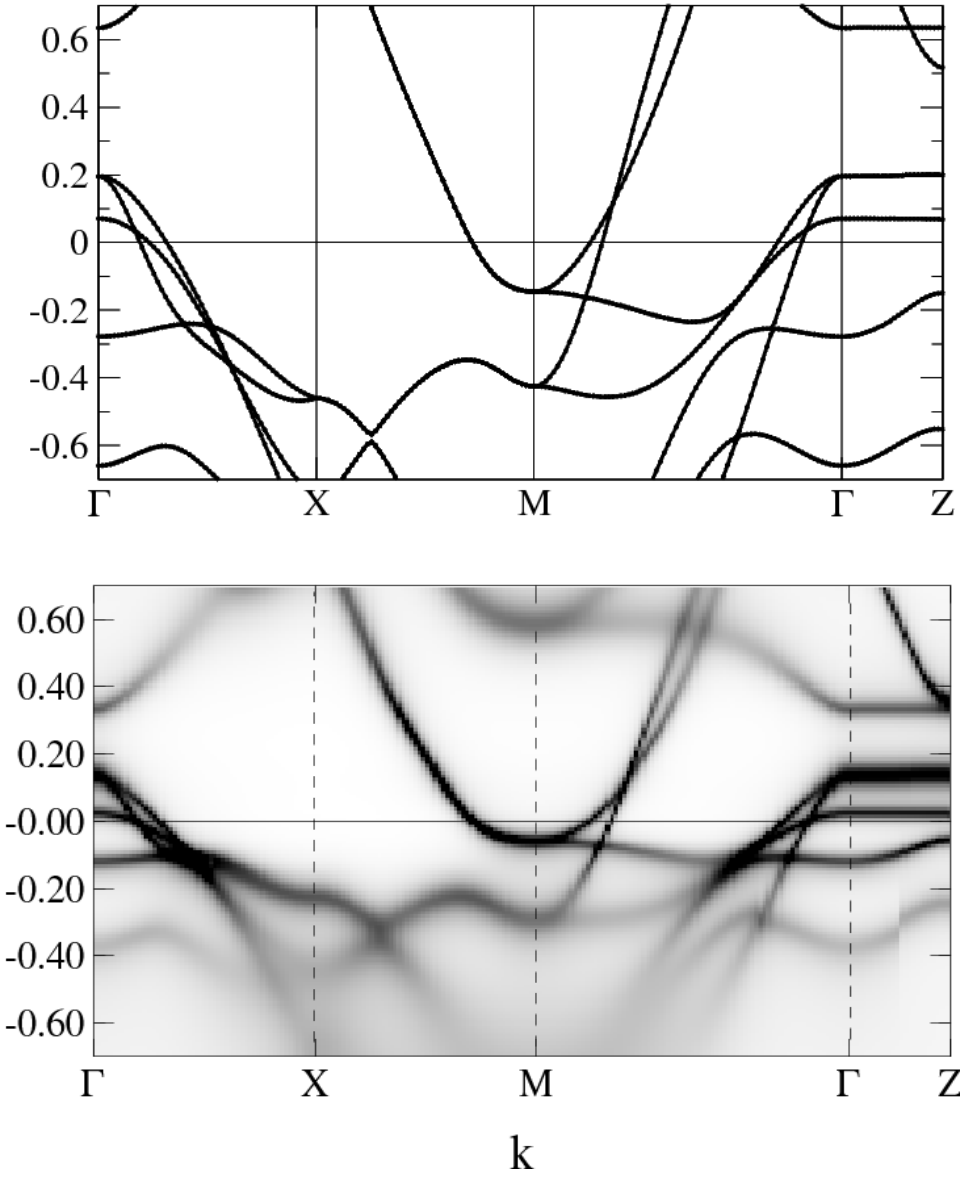
LDA bands:

$\epsilon_{\mathbf{k}\nu}$ from Wien2K

well-defined quasi-particles around Fermilevel

Spread-out excitation for ω < -0.4 eV due to increased scattering rate

DMFT results: Spectra



Differences LDA / DMFT:

Life-time effects

Additional crystal-field splitting

e.g.: d_{xy} and d_{z^2} bands shift significantly to Fermi level

No good experimental data
low-T LaFeAsO is magnetic

⇒ Comparison difficult

Parameters of the calculations

Size of Hamiltonian, energy window

Results so far: Hamiltonian included **Fe-d, As-p, O-p**

Larger Hamiltonian (60 bands): Z **= 0.60**

Convergence in size of Hamiltonian!

Double Counting correction

FLL double counting: Z **= 0.60** m^* **= 1.6**

AMF double counting: Z **= 0.45** m^* **= 2.3**

Sets the reliability ('error bars') of the calculation

Conclusions

Calculations for LaFeAsO:

Moderatly correlated metal, no signature for Hubbard bands

Effective mass around $m^* = 2.0 +/- 0.4$

Reliable results for large enough energy window

localized Wanniers (*dpp* hamiltonian or even larger)

For more quantitative comparison with experiments:

other compounds (LaFeAsP, '122', FeSe)

M. Aichhorn et al., Phys. Rev. B 80, 085101 (2009)

Dependence on parameters and window

Calculations for several parameter sets (*dpp* hamiltonian):

U = 2.69 eV, J = 0.79 eV:	**<Z> = 0.62**	(from cRPA)
U = 3.69 eV, J = 0.58 eV:	**<Z> = 0.67**	
U = 3.69 eV, J = 0.80 eV:	**<Z> = 0.53**	
U = 5.00 eV, J = 0.80 eV:	**<Z> = 0.42**	

⇒ Not close to a Mott transition

Using Wannier functions constructed from a huge number of bands:

(U and J expected to be slightly larger):

U = 3.69 eV, J = 0.80 eV:	**<Z> = 0.56**	
U = 3.00 eV, J = 0.80 eV:	**<Z> = 0.60**	(cRPA?)
U = 3.00 eV, J = 0.60 eV:	**<Z> = 0.73**	

Very consistent picture, similar dependence in parameters
Saturated as function of Bloch bands

Small window calculations

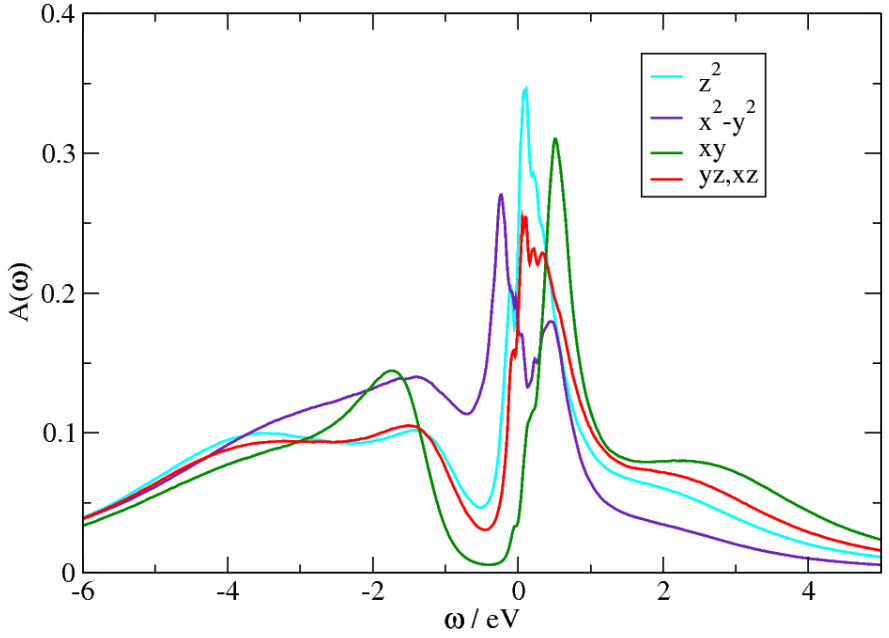
Problem: Small energy window → Spread Wannier functions

Leakage on neighboring atoms

→ Highly anisotropic interaction matrix

Is it justified to use

- Single-site DMFT?
- Spherical approximation to *U?*



U = 4.0 eV, *J* = 0.7 eV:
Z = 0.1 - 0.35
(Consistent with Haule et al.)

Other parameter sets:

U = 4.0 eV, *J* = 0.2 eV:
Z = 0.65 - 0.75

U = 2.9 eV, *J* = 0.43 eV:
Z = 0.5 - 0.6

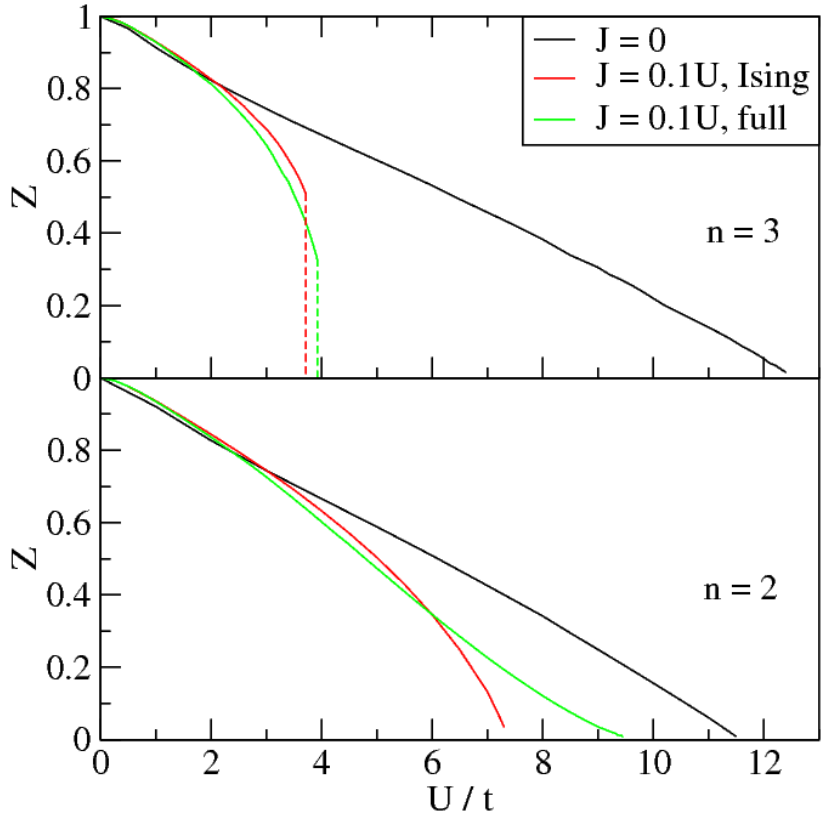
Strong sensitivity to parameters (***J***) → Reliability of results ???

Remarks on the Hund's rule exchange

Neglected terms:

$$H_{\mathrm{sf+ph}} = -\frac{J}{2}\sum_{mm'}\left(c^{\dagger}_{m\uparrow}c_{m\downarrow}c^{\dagger}_{m'\downarrow}c_{m'\uparrow} + c^{\dagger}_{m\uparrow}c^{\dagger}_{m\downarrow}c_{m'\uparrow}c_{m'\downarrow} + \mathrm{h.c.}\right)$$

spin-flip pair-hopping



Model calculations using SFT:
Bethe lattice, $M = 3$

Negligible influence for moderate correlations, $Z = 0.5$

Important near phase transition, $Z = 0.1$-0.2!

No change in physical picture of the Pnictide superconductors