Many-body effects in iron pnictides and chalcogenides – separability of non-local and dynamical correlation effects

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"Emergent Quantum Phases in Condensed Matter", ISSP, Kashiwa

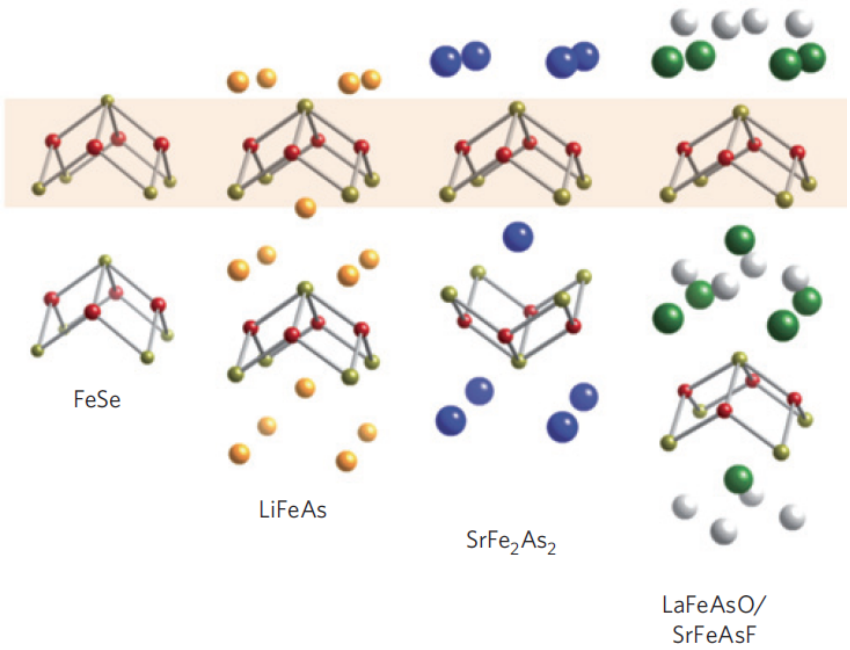
JMT, M. van Schilfgaarde & G. Kotliar, Phys. Rev. Lett. 109, 237010 (2012)

Iron Pnictides and Chalcogenides

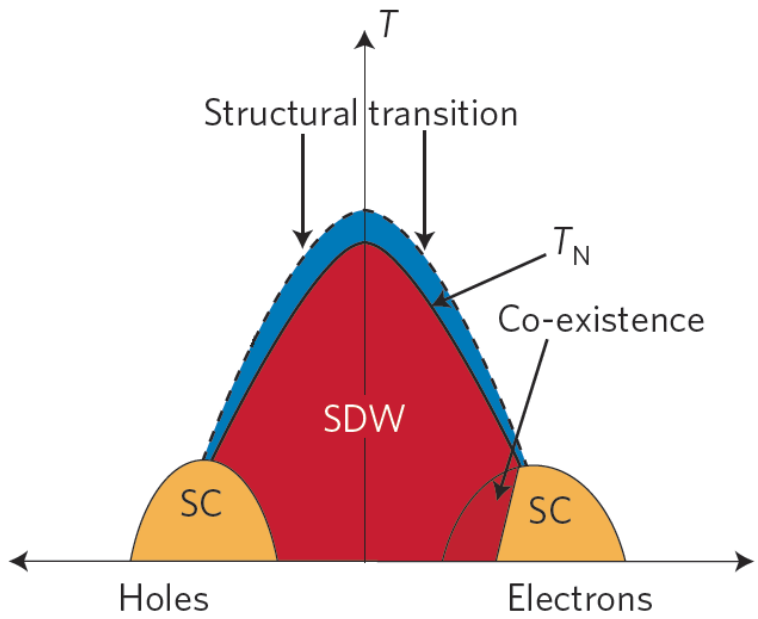
1. Intro: Pnictides and chalcogenides
2. electronic structure approach QS*GW*: what and why
3. QS*GW* results
   - Fermi surface of LiFeAs vis-à-vis ARPES
   - density of states FeSe vs. X-ray absorption
   - ARPES of $BaFe_2As_2$
4. Methodological insight
   - origins of effective masses
   - non-local correlations substantial
   - low energy dynamics of correlations is local
   - proposal: QS*GW*+dynamical mean field theory
5. Conclusions & Perspectives

JMT, M. van Schilfgaarde & G. Kotliar, Phys. Rev. Lett. 109, 237010 (2012)

Pnictides and chalcogenides – the family



[Paglione and Greene (2010)]



[Basov and Chubukov (2011)]

open issues

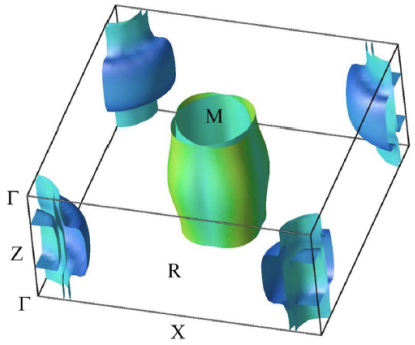
- superconductivity (of course...)
- origin of long range magnetic order (local vs itinerant picture)
- ...

Pnictides and chalcogenides – electronic structure

Density functional theory (DFT) in local density approximation (LDA)

Correct prediction of

- Fermi surfaces:
  LaFePO [Lebègue (2007)]
  LaFeAsO [Singh and Du (2008)] $\longrightarrow$
- striped AF spin ground state [Dong *et al.*(2008)]



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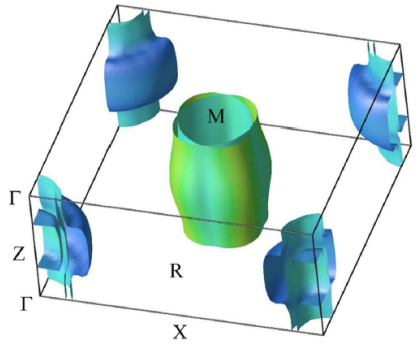
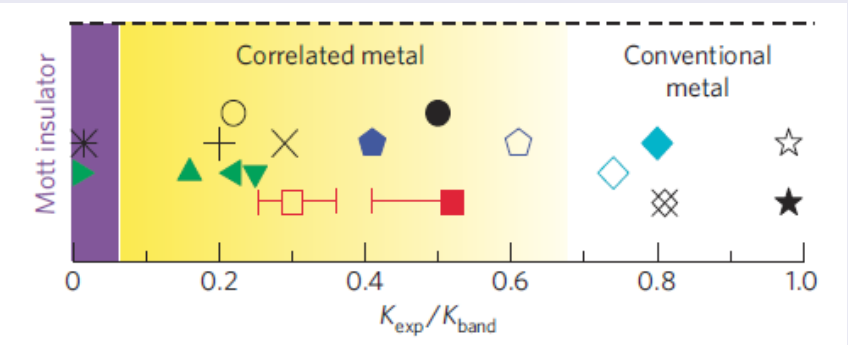
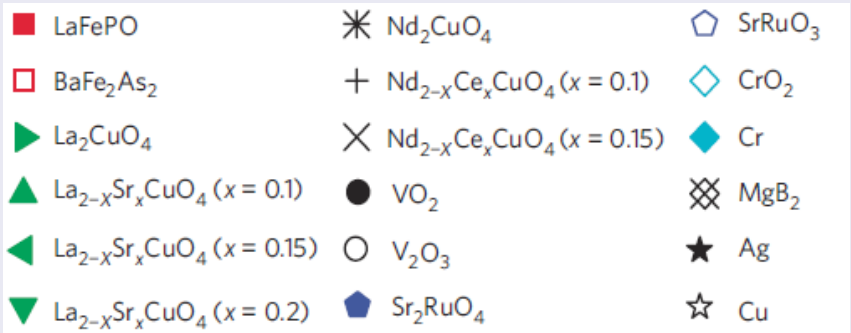


BUT: evidence for presence of **correlation effects** beyond band theory!

Example: reduction of kinetic energy K wrt band-theory [Qazilbash *et al.*]





- band renormalizations/effective masses (photoemission, de-Haas-van-Alphen, optics)
- magnitude of ordered moments
- size of Fermi surfaces

Beyond the density functional picture

Successes of modern many-body theory

- LDA+DMFT (realistic dynamical mean field theory) [Georges *et al.*, Anisimov, Lichtenstein]
  - correct effective masses [Yin *et al.*, Aichhorn *et al.*, Ferber *et al.*....]
  - magnitudes of ordered moments [Yin *et al.*]
  - good structures (relaxation) [Aichhorn *et al.*]
- Gutzwiller: good structures [Wang *et al.*, ...]

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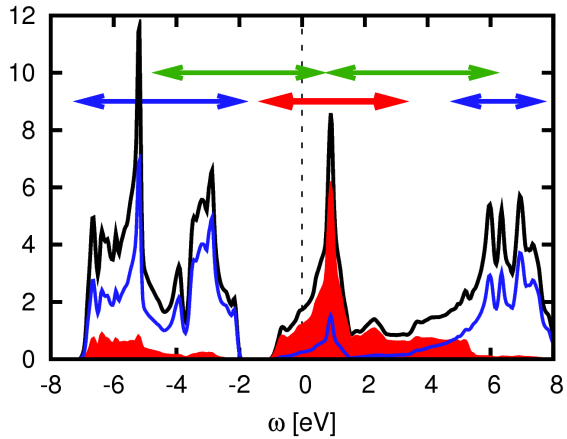
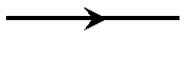
BUT: are the *ad hoc* assumptions warranted?

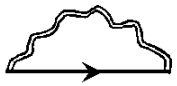
1. treatment of band/orbital **subspace** sufficient?
   inter and out of subspace renormalizations?
2. **correlations local ?** $\Sigma \approx \Sigma^{\mathrm{DMFT}}(\omega)|RL\rangle\langle RL'|$?
3. starting point dependence (LDA, GGA, ...)



The GW approximation [Hedin]

(starting) Greens function $G_0 =$ 

(RPA) screened interaction $W =$ $= \left[V^{-1}_{Coulomb} -\right.$  $\left.\right]^{-1}$

self-energy $\Sigma = G_0 W =$ 

many-body correction $G^{-1} = G_0^{-1} - \left[\Sigma - v_{xc}\right]$

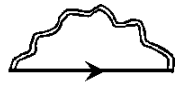
the good...

- no assumption on locality
- dynamical screening
- all electron approach [no (or very large) orbital subspace]

the bad...

- 1st order perturbation (in W) only
- starting point dependence (what is G_0? LDA, ...) $\longrightarrow$ self-consistency?

QSGW = quasi-particle self-consistent GW [van Schilfgaarde *et al.*]

self-energy $\Sigma = G_0 W =$ 

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require same poles in G_0 and $G \rightarrow$ quasi-particle self-consistency
QS$GW \rightarrow$ static non-local effective v_{xc}^{QSGW} [van Schilfgaarde *et al.*]

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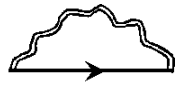
self-energy $\Sigma = G_0 W =$ 

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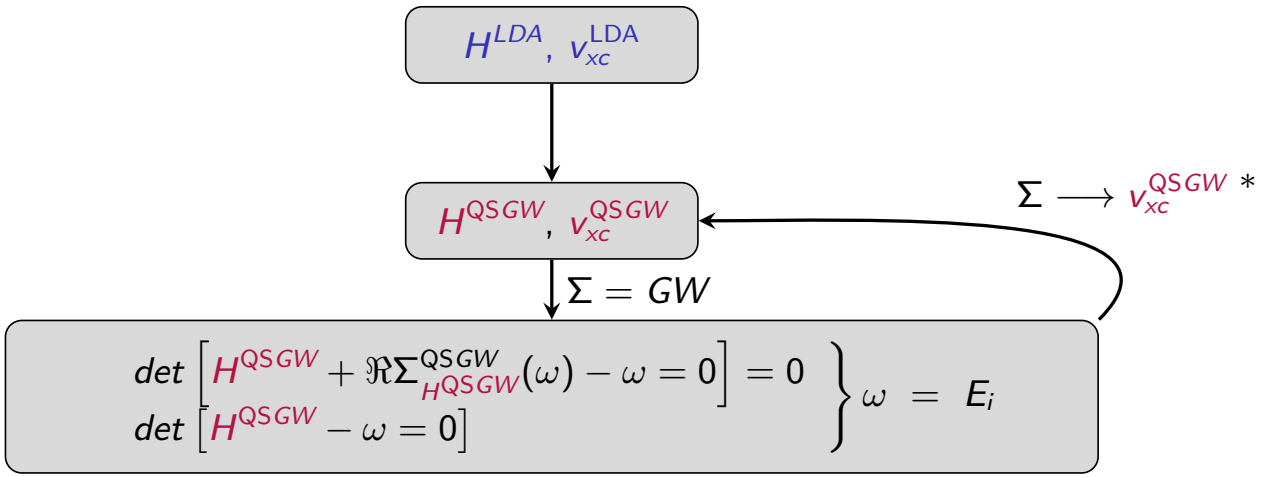

* in practice: $v_{xc}^{QSGW} = \frac{1}{2}\sum_{ij\mathbf{k}} |\Psi_{\mathbf{k}i}\rangle \Re\left[\Sigma_{ij}^{QSGW}(\mathbf{k}, E_{\mathbf{k}i}) + \Sigma_{ji}^{QSGW}(\mathbf{k}, E_{\mathbf{k}j})\right] \langle\Psi_{\mathbf{k}j}|$

$\longrightarrow$ no dependence on the starting point H^{LDA}

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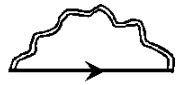
self-energy $\Sigma = G_0 W =$ 

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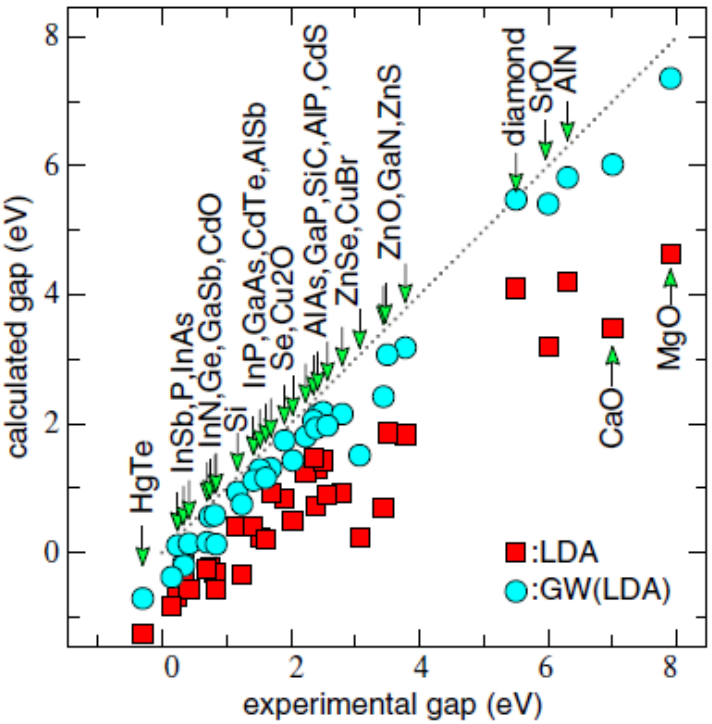
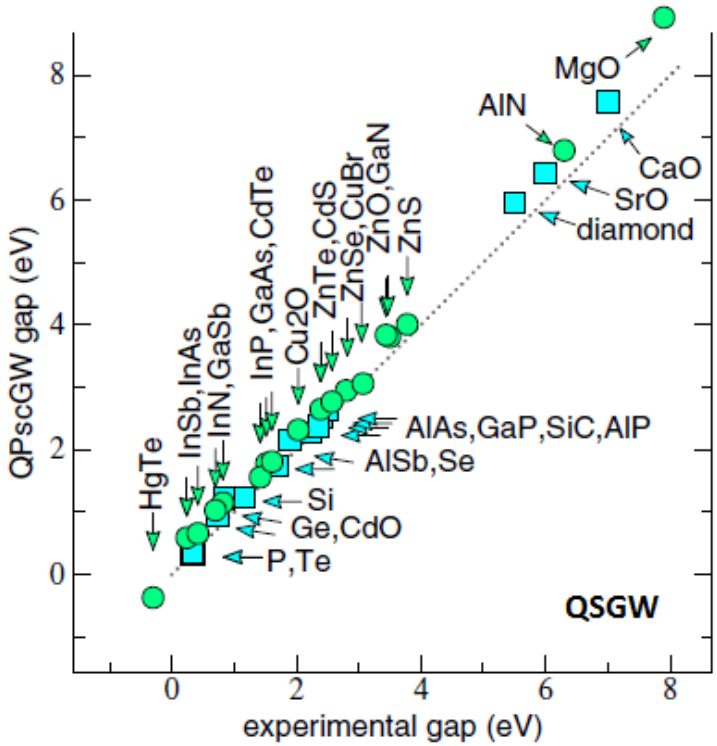
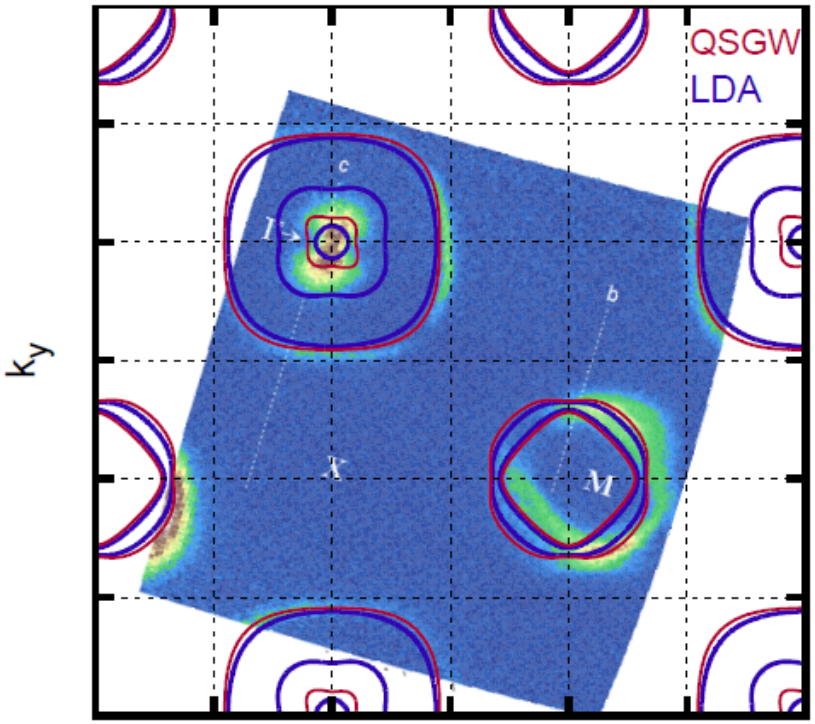




Results: The Fermi surface of LiFeAs

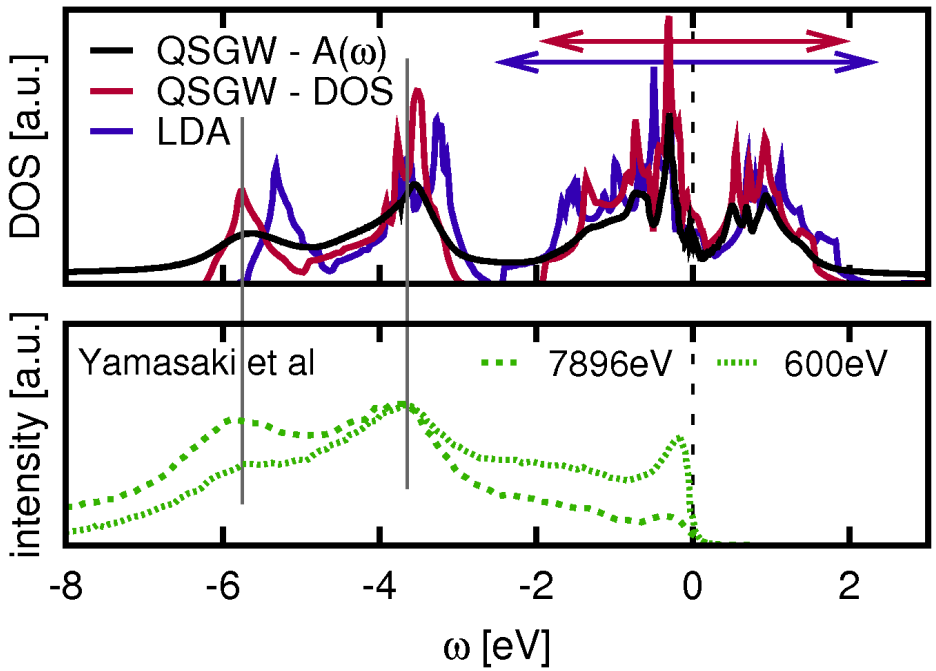


QSGW

- excellent agreement with experiment [Borisenko *et al.*]
- at Γ point: xy grows slightly & xz,yz shrinks substantially

Results: Iron selenide FeSe

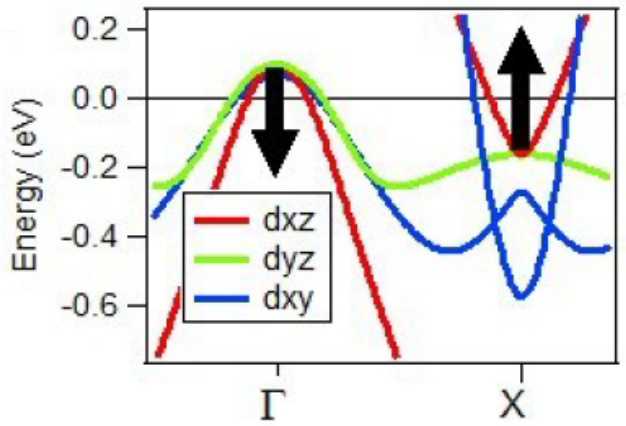
1. correlations beyond the 3d-shell

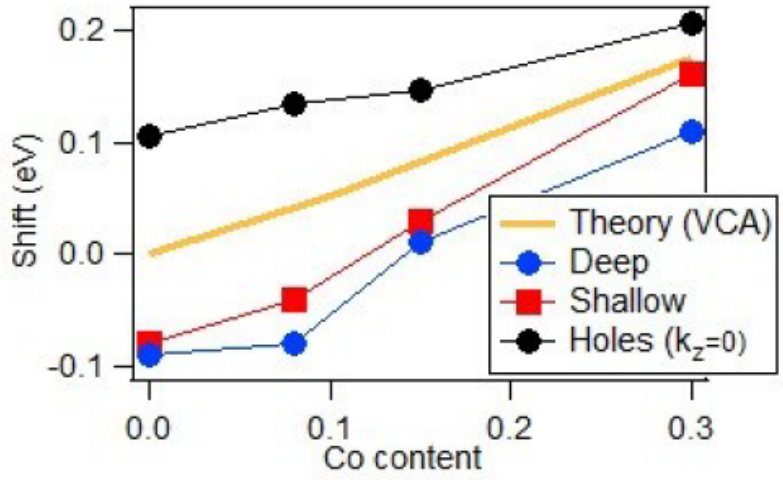


- **correction of high energy excitations** ✓ : effect of Σ_{pp}, Σ_{pd}, beyond DMFT
- **band-narrowing of 22%** ✗
  not strong enough (photoemission: $m^*/m^{\mathrm{LDA}} \approx 3.6$) $\rightarrow$ need DMFT?

$Ba(Fe_{1-x}Co_x)_2As_2$: photoemission

2. non-local correlations: experimental evidence





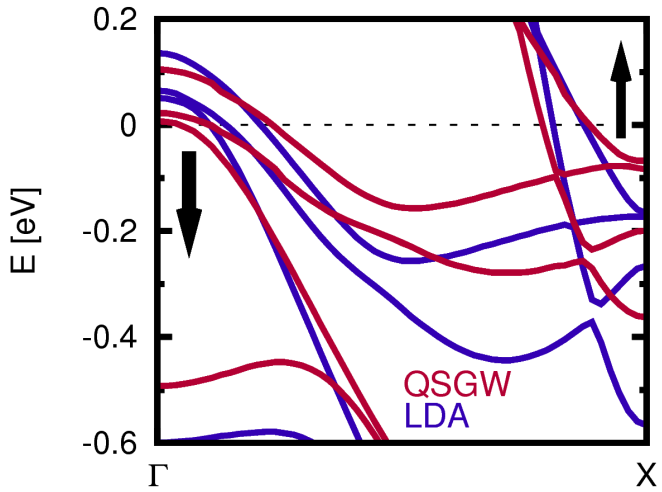
" We show that there is a k-dependent energy shift compared to density functional calculations "

It would be very interesting to obtain a full theoretical description of this effect. It has not been seen in DMFT based electronic structure calculations up to now, possibly because it requires an extremely high energy precision to be resolved (of the order of 10 meV). Alternatively, one may speculate that it is due to non-local self-energy effects not included at the DMFT level. A very recent GW study may be interpreted in this sense [26].

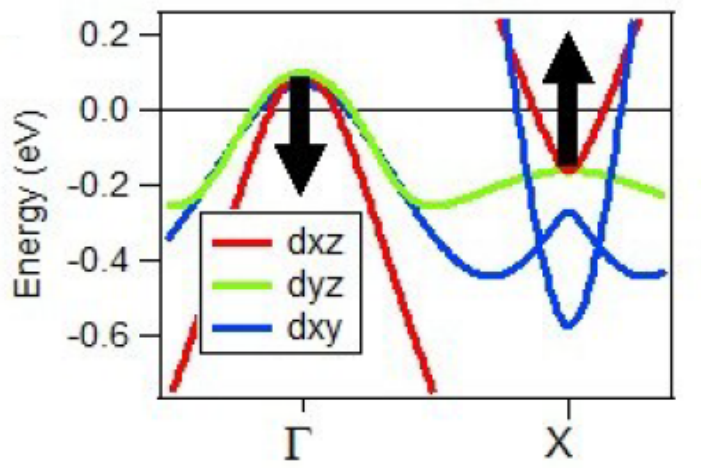
[Brouet *et al.*, PRL 110, 167002 (2013), also: Dhaka *et al.*arXiv:1205.6731v1]

$BaFe_2As_2$: trends with respect to LDA

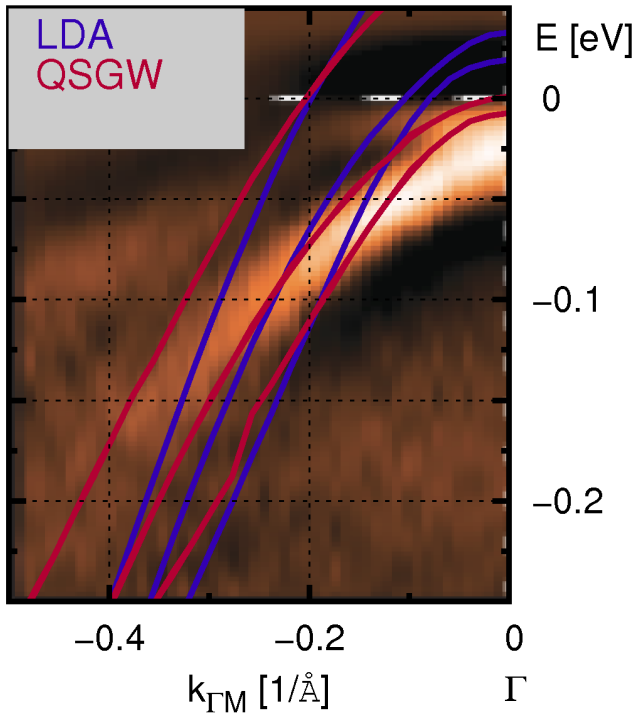
QSGW [JMT *et al.*]



[Brouet *et al.*, PRL 110, 167002 (2013)]



$BaFe_{1.85}Co_{0.15}As_2$ [ARPES: [Zhang *et al.*]]



QSGW

- sizable **shrinking of pockets** ✓
- size of **Fermi surface** in good agreement with experiments [pure Ba122, not shown] ✓
- band-width narrowing of 16% (wrt LDA)
  BUT: experimental dispersion still lower $\longrightarrow$ effective mass too small! ✗

Origin of effective masses

mass enhancement wrt band-theory: $\frac{m^{\mathrm{QS}GW}}{m^{\mathrm{LDA}}} = \frac{dE^{LDA}_{\mathbf{k}i}}{dk_\alpha} \Big/ \frac{dE^{\mathrm{QS}GW}_{\mathbf{k}i}}{dk_\alpha}$

$$\frac{dE^{\mathrm{QS}GW}_{\mathbf{k}i}}{dk_\alpha} = \frac{\langle \Psi_{\mathbf{k}i} | \partial_{k_\alpha} \left(H^{\mathrm{QS}GW} + \Re\Sigma^{\mathrm{QS}GW}(\omega = 0) \right) | \Psi_{\mathbf{k}i} \rangle}{\left[1 - \langle \Psi_{\mathbf{k}i} | \partial_\omega \Re\Sigma^{\mathrm{QS}GW} | \Psi_{\mathbf{k}i} \rangle \right]_{\omega=0}}$$

Hence mass renormalization through:

- $Z_{\mathbf{k}i} = \left[1 - \langle \Psi_{\mathbf{k}i} | \partial_\omega \Re\Sigma^{\mathrm{QS}GW} | \Psi_{\mathbf{k}i} \rangle \right]^{-1}_{\omega=0}$ $\rightarrow$ **dynamics!**
- momentum dependence of correlations $\rightarrow$ **non-locality!**
- change in charge density

Origin of effective masses

	QSGW $\frac{m^{QSGW}}{m^{LDA}}$	QSGW $1/Z^{QSGW}$ @ Γ		[AR]PES m^*/m^{LDA}		DMFT $1/Z^{DMFT}$ [Yin, Ferber, Aichhorn]	
		xy	xz/yz	xy	xz/yz	xy	xz/yz
$CaFe_2As_2$	1.05	2.2	2.1	2.5[1]		2.7	2.0
$SrFe_2As_2$	1.13	2.3	2.0	3.0[2]		2.7	2.6
$BaFe_2As_2$	1.16	2.2	2.2	2.7	2.3 [3]	3.0	2.8
LiFeAs	1.15	2.4	2.1	3.0[4]		3.3/2.8	2.8/2.4
FeSe	1.22	2.4	2.2	3.6[5]		3.5/5.0	2.9/4.0
FeTe	1.17	2.6	2.3	6.9[6]		7.2	4.8

- trends along series captured
- $\frac{m^{QSGW}}{m^{LDA}} < 1/Z^{QSGW} \longrightarrow \partial_k \Sigma$ delocalizes (cf. electron gas)

  k-dependence non-negligible! (not included in DMFT)
- "dynamical" masses $1/Z$ too small by factor of 2 or more

[[1] Wang *et al.*, [2] Yi *et al.*, [3] Brouet *et al.*, [4] Borisenko *et al.*, [4] Yamasaki *et al.*, [6] Tamai *et al.*.]

Non-locality of self-energy

Momentum variance of X within maximally localized Wannier basis:

$$(\delta_k X)^2 = 1/{N_L}^2 \sum_{\mathbf{k}LL'} \left| \Re X^{\mathbf{k}}_{LL'} - \Re X^{loc}_{LL'} \right|^2$$

static corrections of QSGW beyond LDA:

$$\widetilde{\Sigma}(\omega = 0) = G^{\mathrm{QS}GW} W - v_{xc}^{\mathrm{LDA}}$$

	$\delta_k \widetilde{\Sigma}$ [eV]	$\delta_k v_{xc}^{\mathrm{LDA}}$ [eV]
$BaFe_2As_2$	0.1	0.22
FeSe	0.12	0.2

- **k**–dependence non-negligible on relative scale

$\longrightarrow$ **DMFT assumption (local self-energy) not fully warranted**

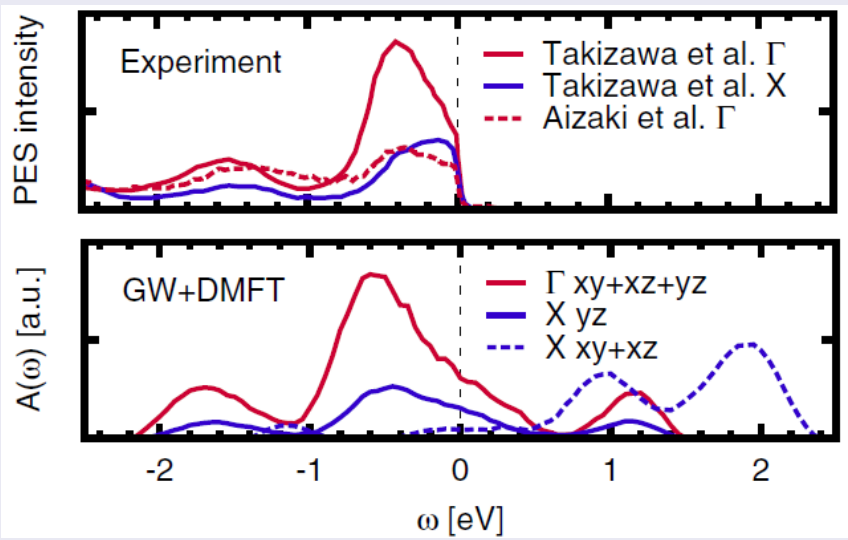
Combine GW and DMFT

- dynamical correlations insufficient in QS*GW*
- non-local correlations substantial

$\Big\}$ $\longrightarrow$ combine *GW* and DMFT

GW+DMFT [Biermann *et al.*, Sun *et al.*]

$$\Sigma(\mathbf{k},\omega) \quad = \quad \Sigma^{DMFT}(\omega) + \Sigma^{GW}_{non-local}(\mathbf{k},\omega)$$



$SrVO_3$: *GW*+DMFT with dynamical $U(\omega)$

JMT, M. Casula, T. Miyake, F. Aryasetiawan, S. Biermann

EPL 100, 67001 (2012)

BUT difficult to implement!

no self-consistent *ab initio* implementation in existence.

Can we propose a more workable scheme?

What is the nature of local and non-local correlations?

Locality of quasi-particle dynamics

How **frequency dependent** are the non-local correlations?

Measure: **k**–variance of qp-weight $Z_L^{\mathbf{k}} = \left[1 - \langle \Psi_{\mathbf{k}L} | \partial_\omega \Re\Sigma^{\mathrm{QS}GW}(\mathbf{k},\omega) | \Psi_{\mathbf{k}L} \rangle\right]^{-1}$

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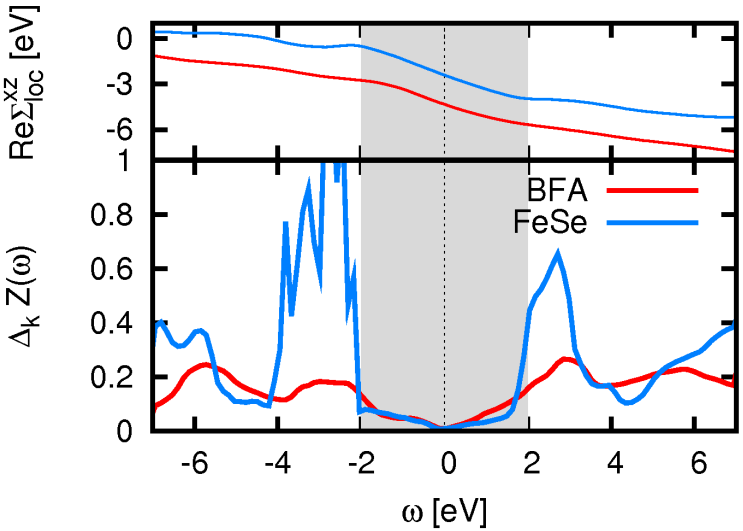
$\Delta_k Z(\omega = 0) = \sqrt{\sum_{\mathbf{k}L} |Z_L^{\mathbf{k}} - Z_L^{loc}|^2} \approx 0.5\% \ \forall$ *GW* calculations here.



QS*GW* empirically: $\partial_k Z_k \approx 0$

$\Delta_k Z(\omega) \ll Z/10$ for $|\omega|$<2eV: momentum variance **very small** in Fermi liquid regime

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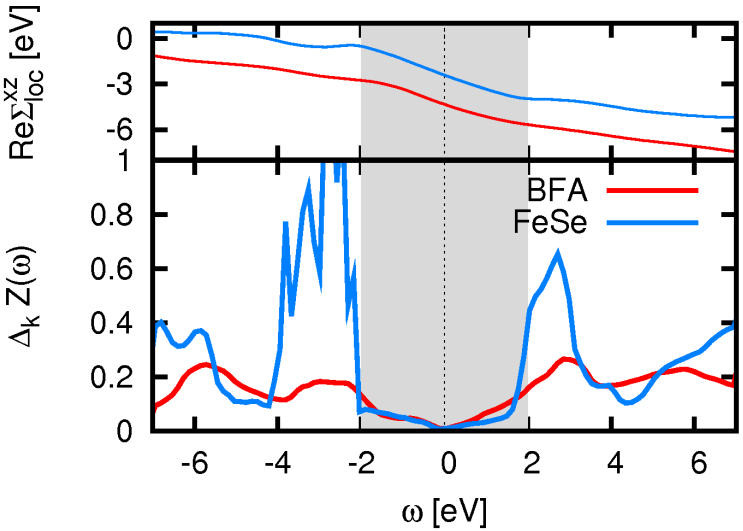

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$\longrightarrow$ **quasiparticle dynamics is local**

$\longrightarrow$ non-local and dynamical correlations are **separable**!

$$\Sigma(\mathbf{k},\omega) = \Sigma^{non-local}(\mathbf{k},\cancel{\omega}) + \Sigma^{loc}(\omega) \qquad (|\omega|<2\mathrm{eV})$$

Proposal: QSGW+DMFT

GW+DMFT [Biermann *et al.*]

$$\Sigma(\mathbf{k},\omega) = \Sigma^{DMFT}(\omega) + \Sigma^{GW}_{non-local}(\mathbf{k},\omega)$$

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QSGW+DMFT [JMT *et al.*]

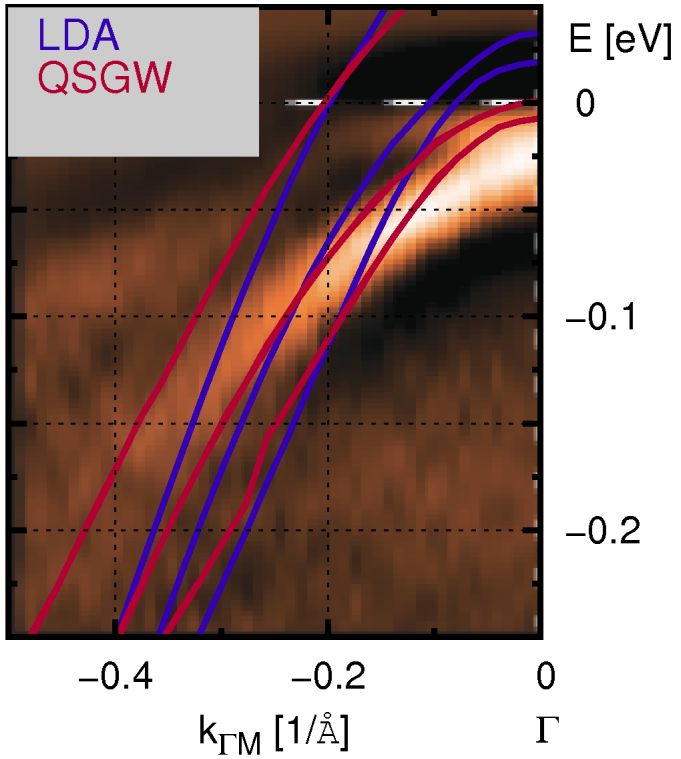
empirically (for iron pnictides/chalcogenides, but also for transition metal oxides ($SrVO_3$, ...))
non-local correlations are static $\longrightarrow$ capturable by **static** QSGW potential v_{xc}^{QSGW}

Proposal: DMFT on top of H^{QSGW}

$$\Sigma^{QSGW+DMFT} = \Sigma^{DMFT}(\omega) - [G^{QSGW} W]_{loc}(\omega)$$

- no remnant of DFT
- double counting well defined
- (quasiparticle) self-consistency limited to GW

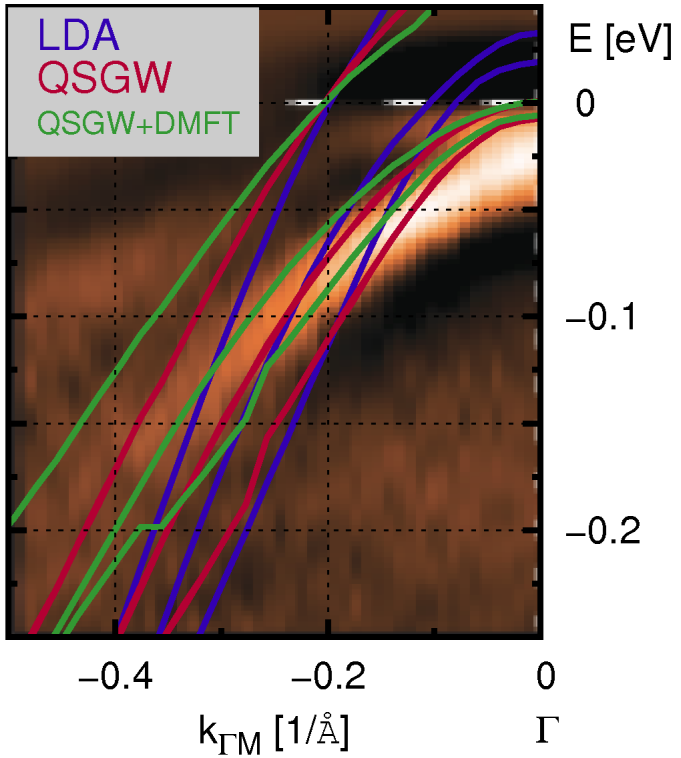
$BaFe_2As_2$ / $BaFe_{1.85}Co_{0.15}As_2$ [ARPES: $BaFe_{1.85}Co_{0.15}As_2$ [Zhang *et al.*]]



QSGW

- non-local shift of the pocket ✓

$BaFe_2As_2$ / $BaFe_{1.85}Co_{0.15}As_2$ [ARPES: $BaFe_{1.85}Co_{0.15}As_2$ [Zhang *et al.*]]



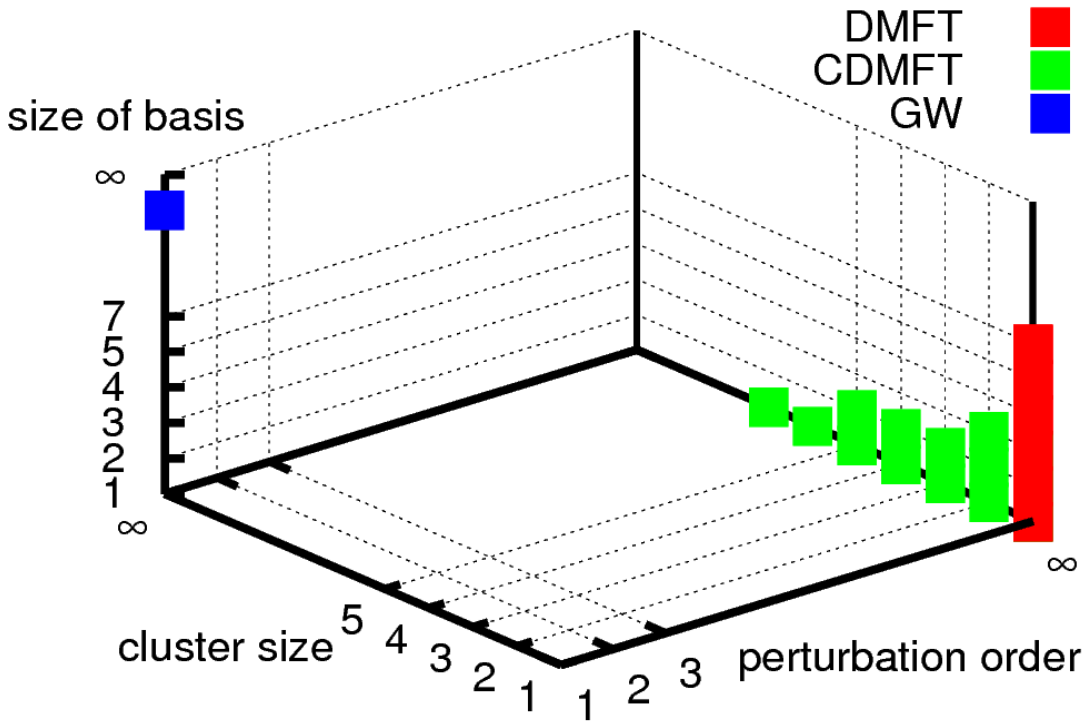
QSGW

- non-local shift of the pocket ✓

"QS*GW*+DMFT" (scaling with $Z^{DMFT}/Z^{QSGW} = 1.4(1.3)$ for xy (xz/yz))

- correct effective masses, good dispersions ✓ $\longrightarrow$ "the best of both worlds"

Many-body theory



DMFT, GW, ... (QS)*GW*+DMFT (for pnictides and chalcogenides)

QS*GW*

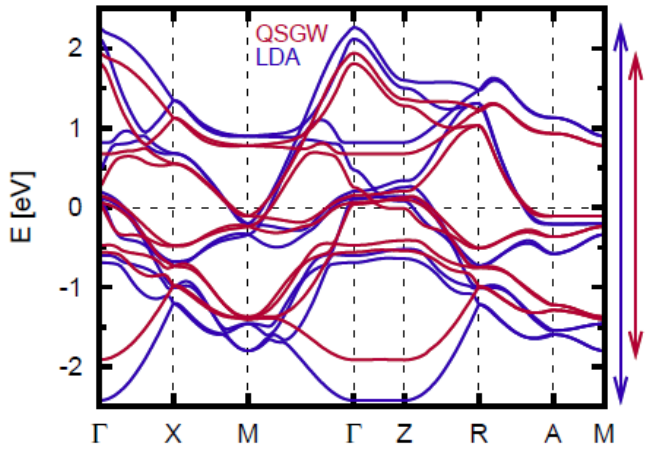
- good Fermi surfaces ✓
- excellent correction of higher energy excitations ✓
- non-local shifts in pockets ✓
- (dynamical) effective masses too small ✗

Methodology

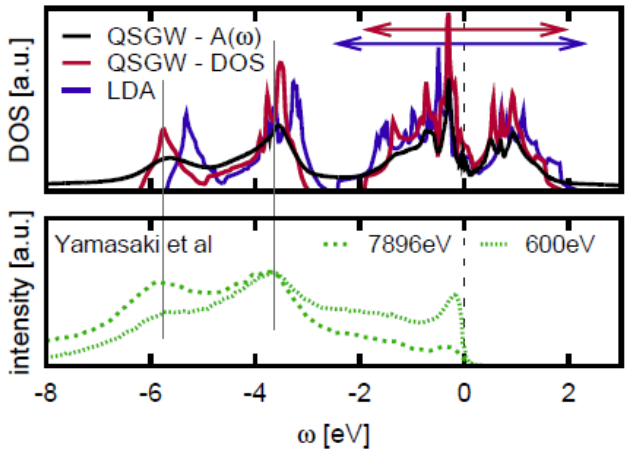
- non-local correlations can be sizable
  but: correlations beyond *GW* **local** [Zein *et al.*, Khodel *et al.*] $\longrightarrow$ **justification for** *GW*
- dynamics insufficient in *GW*
  but: dynamics is **local** $\longrightarrow$ **justification for DMFT**
- dynamics **local** & non-locality **static** $\longrightarrow$ **QS*GW*+DMFT**

JMT, M. van Schilfgaarde & G. Kotliar, Phys. Rev. Lett. 109, 237010 (2012)

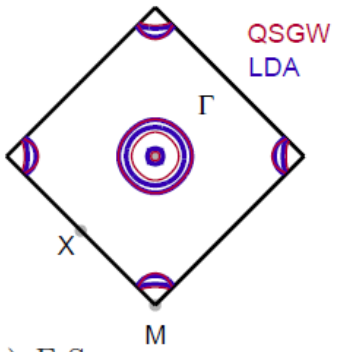
FeSe/FeTe



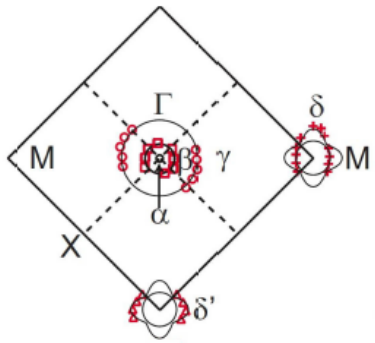
(a) band-structure of FeSe



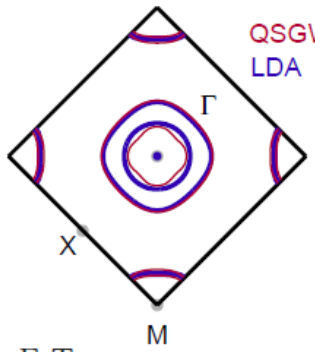
(b) FeSe density of states vs. photoemission



(c) FeSe



(d) $Fe_{1.04}Te_{0.66}Se_{0.34}$

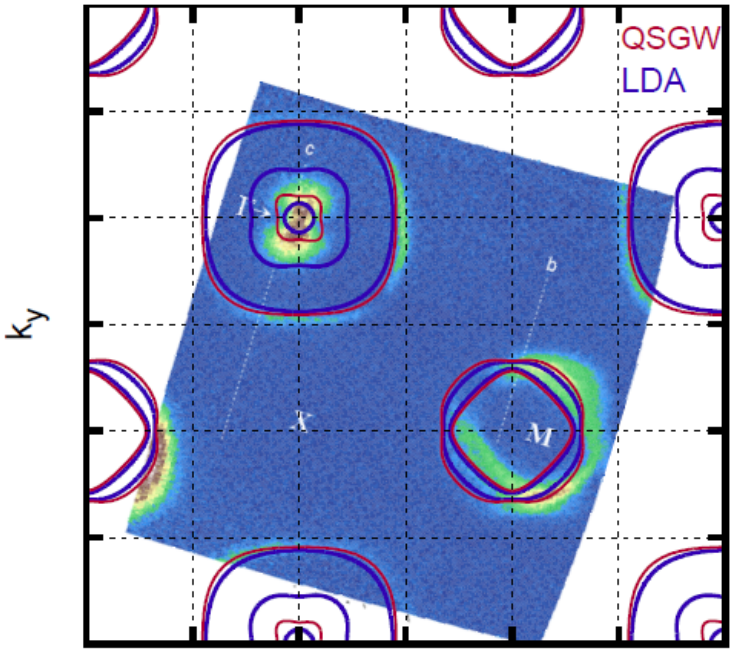


(e) FeTe

- band-narrowing of 22% [again not enough, need DMFT]
- shifting of high energy excitations (Se 4p) [omitted in LDA+DMFT, beyond pure Fe3d-Σ]
- good Fermi surfaces

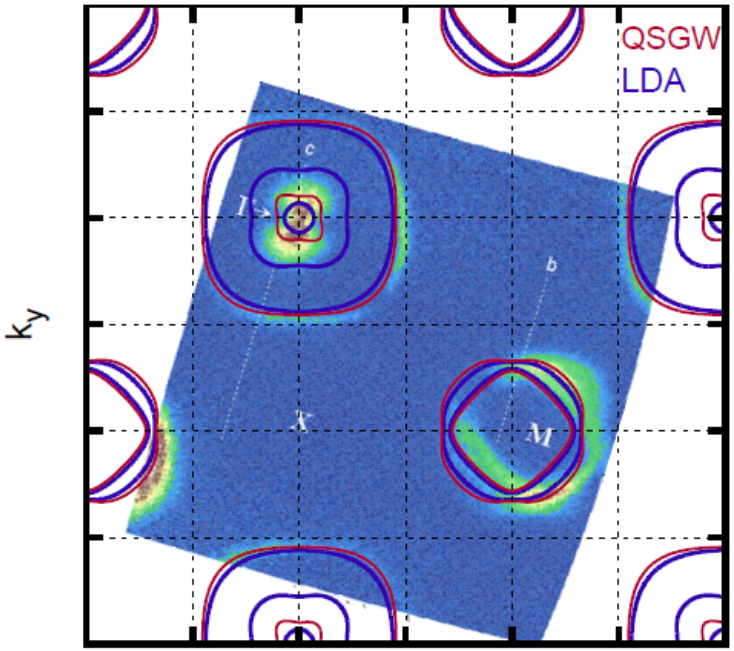
k_F [Å^{-1}]	FeSe	$Fe_{1.04}Te_{0.66}Se_{0.34}$	FeTe
γ	0.29	0.3	0.43
α	0.04	0.03	-

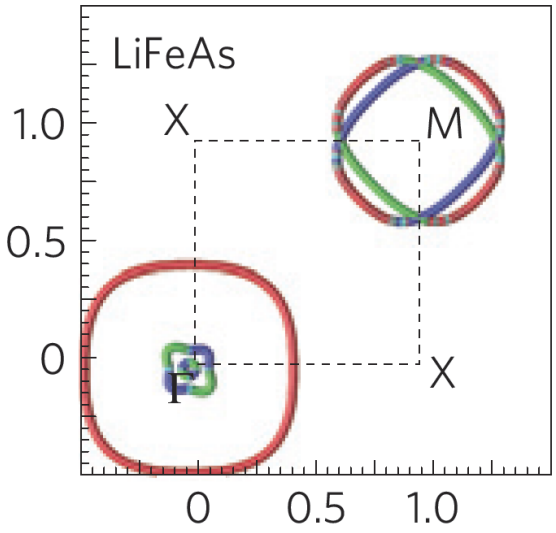
The Fermi surface of LiFeAs



- excellent agreement with experiment [Borisenko *et al.*]
- at Γ point: xy grows, xz,yz shrinks

The Fermi surface of LiFeAs





[Yin et al]

- excellent agreement with experiment [Borisenko *et al.*]
- at Γ point: xy grows, xz,yz shrinks
- trends similar to LDA+DMFT [Yin *et al.*, Ferber *et al.*]
  (orbital shifts more important for FS than momentum dependence)

Consequences for methodology

- correlations not purely local $\longrightarrow \Sigma = \Sigma(\mathbf{k}, \omega)$!
- corrections *beyond GW* local [Zein *et al.*, Khodel *et al.*]

$\longrightarrow$ use *GW* for non-local correlations

- dynamical correlations not sufficiently treated in *GW*

$\longrightarrow$ use DMFT for local dynamics

$\longrightarrow$ Idea of combining *GW* and DMFT $\longrightarrow$ *GW*+DMFT [Biermann *et al.*, Sun *et al.*]

$$\begin{aligned} \Sigma(\mathbf{k}, \omega) &= \Sigma^{local}(\omega) + \Sigma^{non-local}(\mathbf{k}, \omega) \\ &= \Sigma^{DMFT}(\omega) + \left[\Sigma^{GW}(\mathbf{k}, \omega) - \sum_{\mathbf{k}} \Sigma^{GW}(\mathbf{k}, \omega) \right] \end{aligned}$$

Current non-selfconsistent implementations suffer of double counting and starting point dependence [Biermann *et al.*, JMT *et al.*]