A route to non-local correlations in electronic structure: *GW*+DMFT

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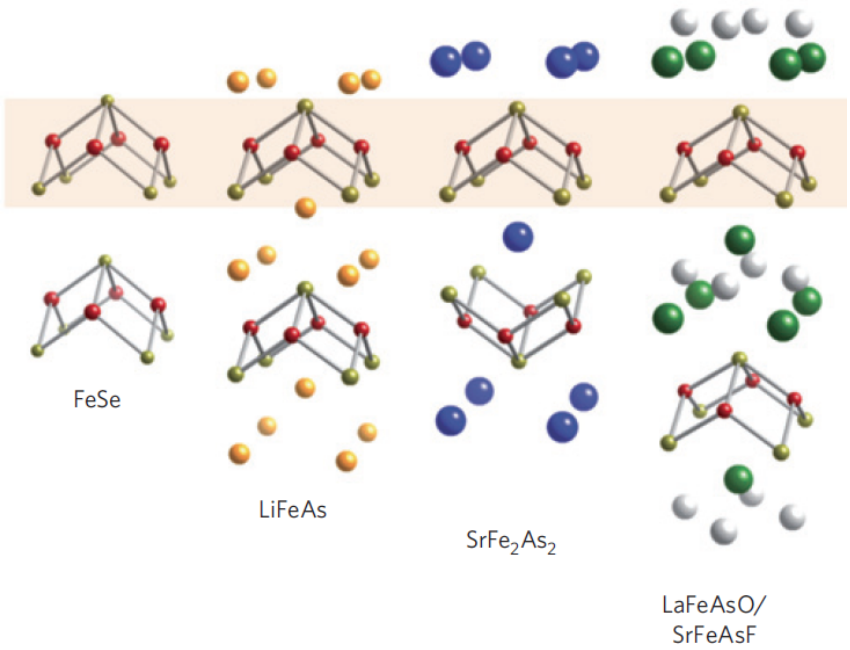
JMT, M. van Schilfgaarde & G. Kotliar, PRL 109, 237010 (2012)

JMT, M. Casula, T. Miyake, F. Aryasetiawan & S. Biermann, EPL 100, 67001 (2012)

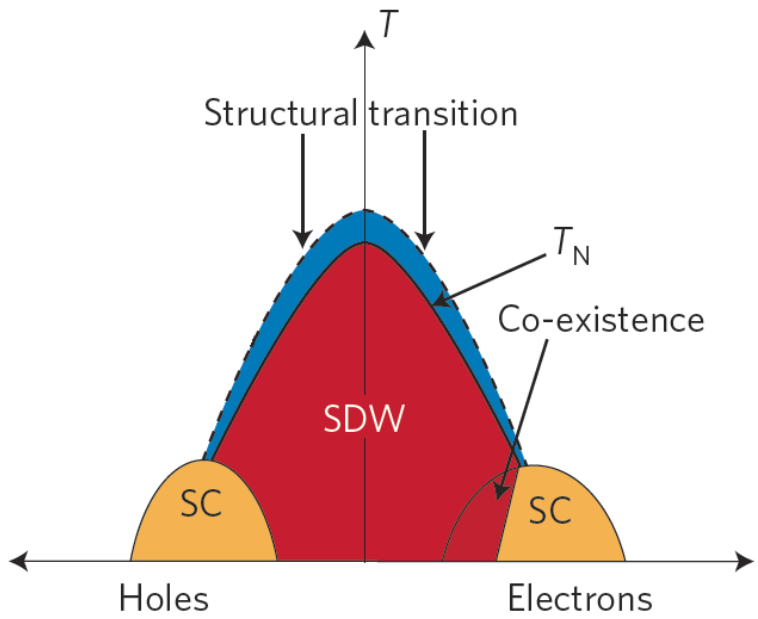
Local and non-local correlations

1. Introduction
   - Presence of correlation effects: example of iron pnictides
   - Dynamical mean field theory at a glance
   - Success and limitations of DMFT based approaches
   - Alternative: *GW*
2. Non-local correlations from the *GW* point of view
   - Iron pnictides and chalcogenides
3. The best of both worlds: *GW*+DMFT
   - Introduction
   - Application to $SrVO_3$
4. A simplified scheme: QS*GW*+DMFT

Pnictides and chalcogenides – the family



[Paglione and Greene (2010)]



[Basov and Chubukov (2011)]

open issues

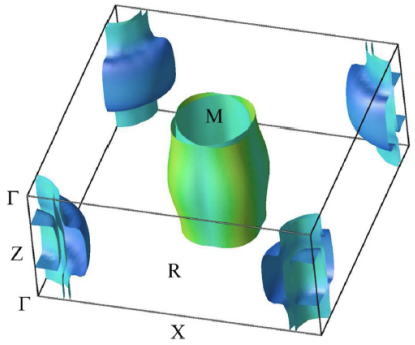
- superconductivity (of course...)
- origin of long range magnetic order (local vs itinerant picture)
- ...

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)]



Pnictides and chalcogenides – electronic structure

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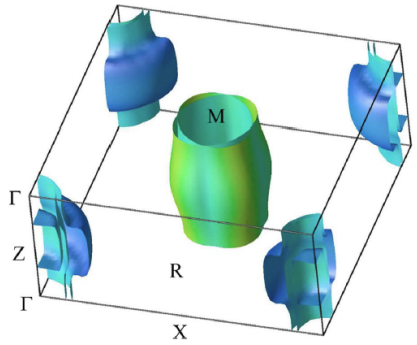
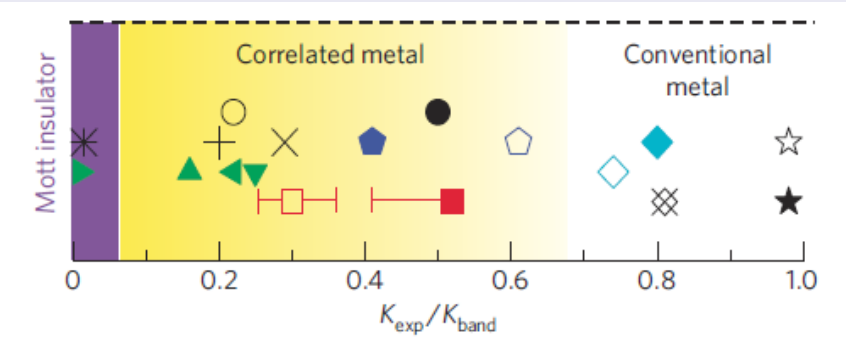
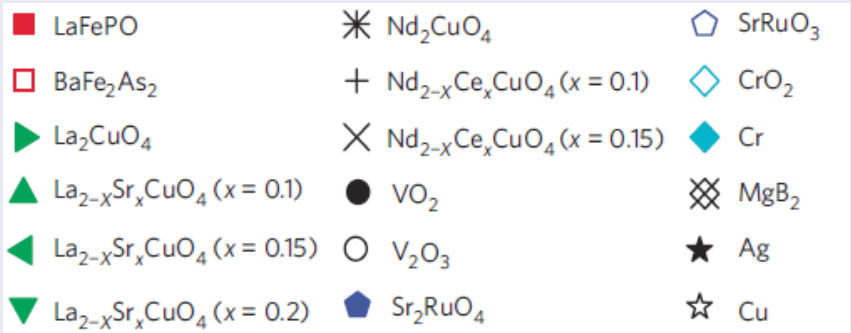


BUT: evidence for presence of **correlation effects** beyond band theory!

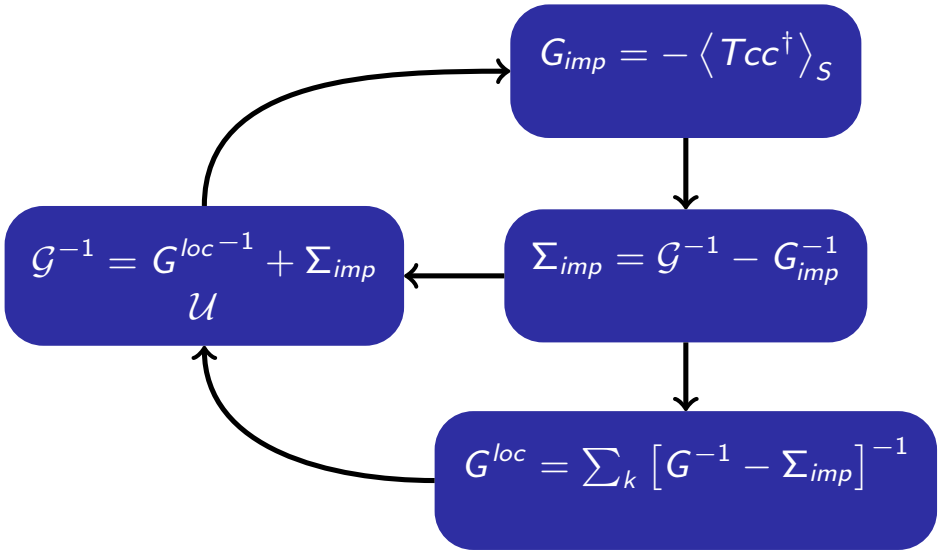
Example: reduction of kinetic energy K wrt band-theory [Qazilbash *et al.*]

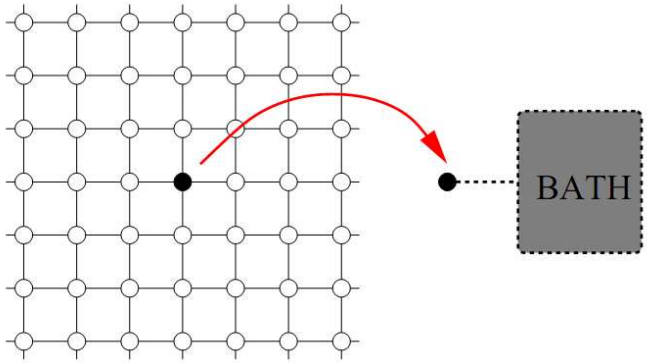




- band renormalizations/effective masses (photoemission, de-Haas-van-Alphen, optics)
- magnitude of ordered moments
- size of Fermi surfaces

Dynamical mean field theory (DMFT) [Georges & Kotliar, Metzner & Vollhardt]





- map solid onto effective atom coupled to bath and subject to local interactions

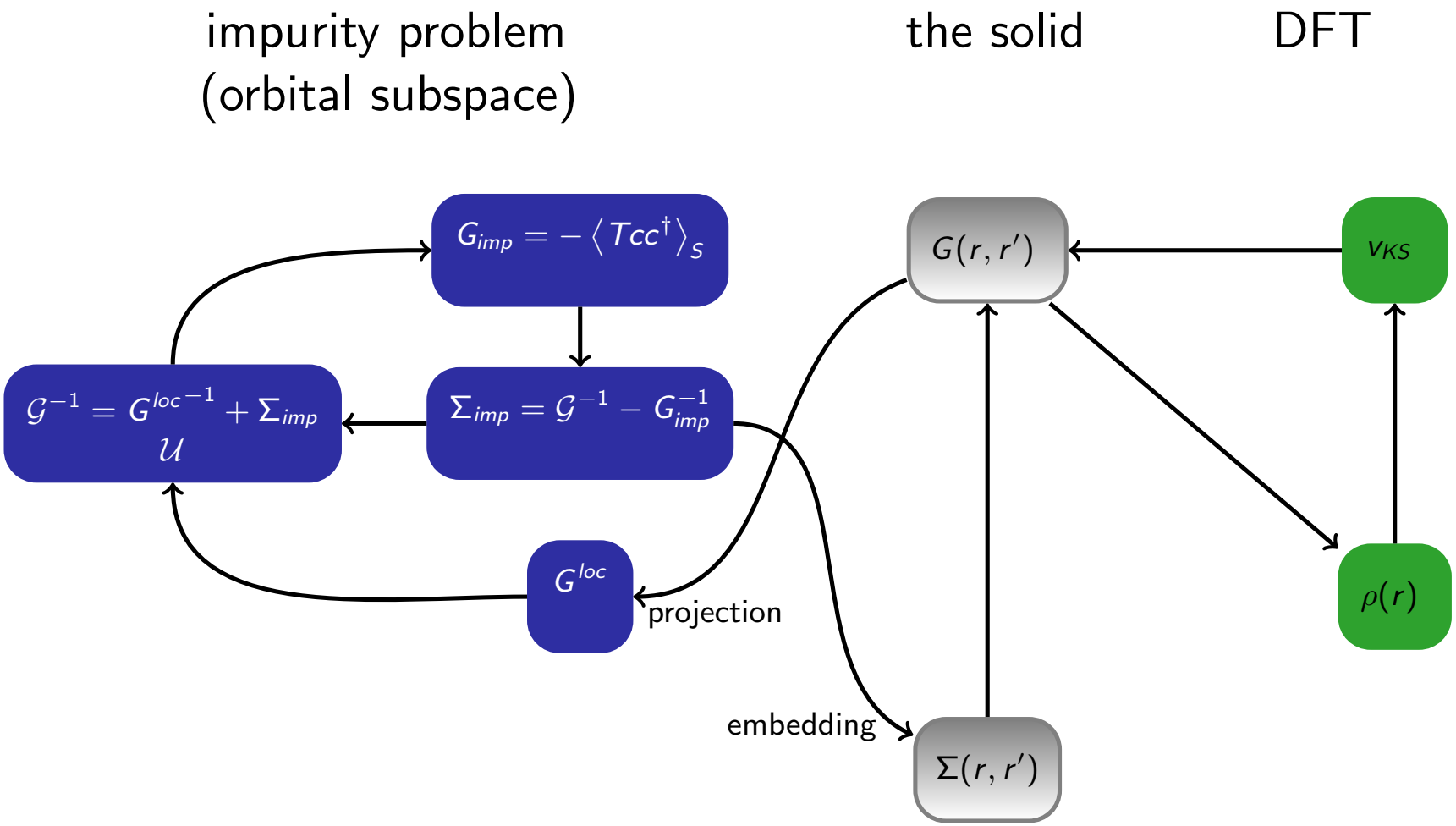
$$S = -\int_0^\beta d\tau \int_0^\beta d\tau' \sum_\sigma c_\sigma^\dagger(\tau)\mathcal{G}^{-1}(\tau-\tau')c_\sigma(\tau') + U\int_0^\beta d\tau n_\uparrow n_\downarrow$$

- relate the effective atom to solid $\rightarrow$ self-consistency condition

$$G_{imp} \stackrel{!}{=} G^{loc}$$

- exact in infinite dimension (mean-field), non-perturbative in interaction

DFT+DMFT [Katsnelson and Lichtenstein, Anisimov et al.]



Beyond the density functional picture

Successes of modern many-body theory (iron pnictides)

- DFT+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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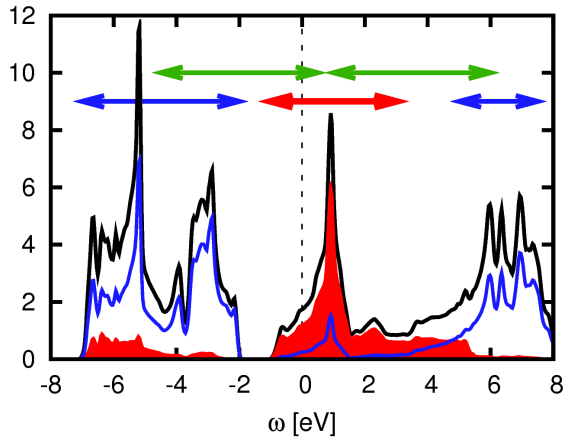
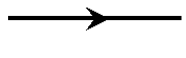
BUT: are the *ad hoc* assumptions warranted?

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

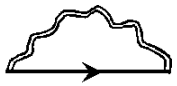
Also: double counting issue...



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

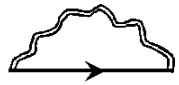
the good...

- 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
QSGW $\rightarrow$ static non-local effective $v_{xc}^{\mathrm{QS}GW}$ [van Schilfgaarde *et al.*]

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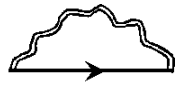
self-energy $\Sigma = G_0 W =$ 

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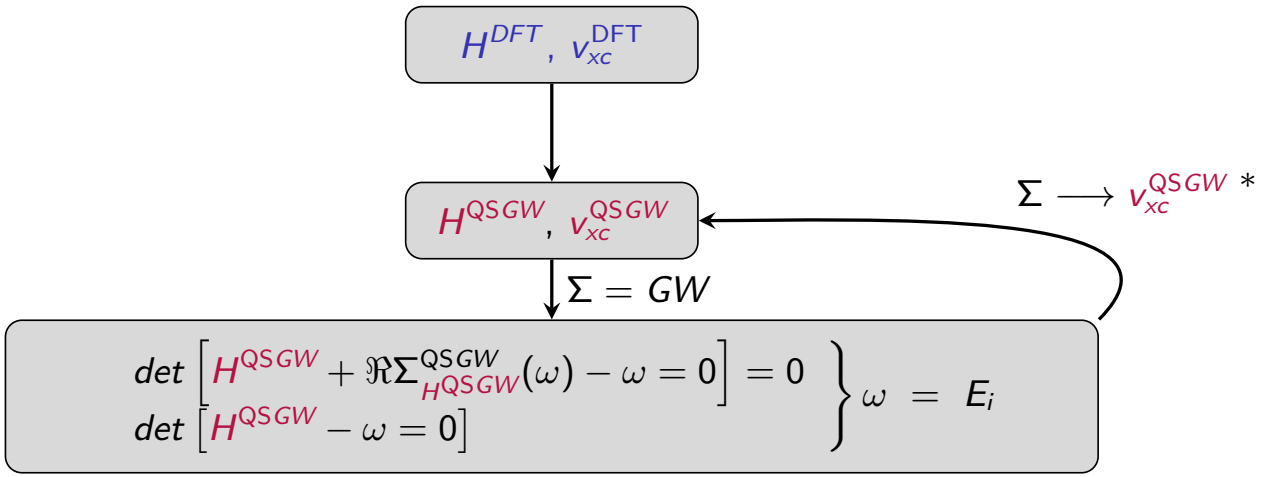

* in practice: $v_{xc}^{\mathrm{QS}GW} = \frac{1}{2}\sum_{ij\mathbf{k}} |\Psi_{\mathbf{k}i}\rangle \Re\left[\Sigma_{ij}^{\mathrm{QS}GW}(\mathbf{k}, E_{\mathbf{k}i}) + \Sigma_{ji}^{\mathrm{QS}GW}(\mathbf{k}, E_{\mathbf{k}j})\right] \langle\Psi_{\mathbf{k}j}|$

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

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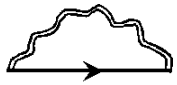
self-energy $\Sigma = G_0 W =$ 

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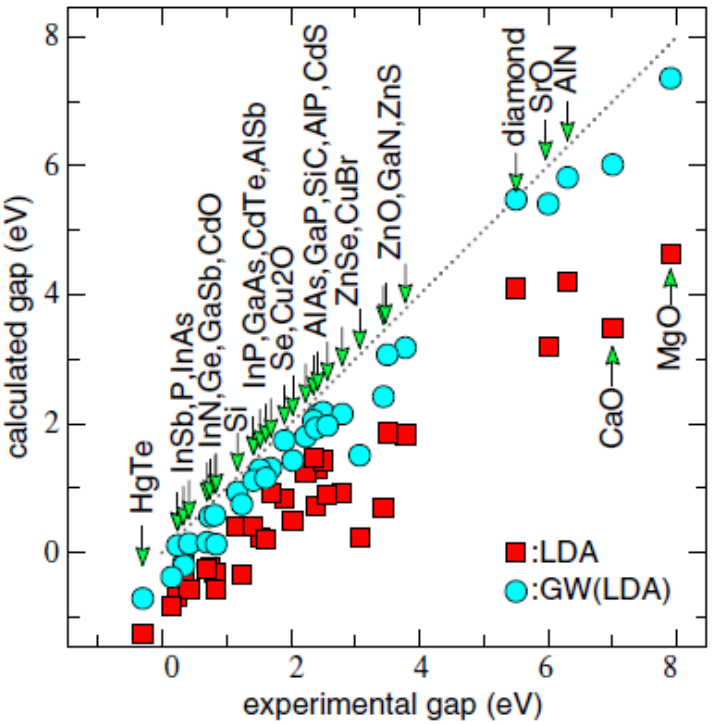
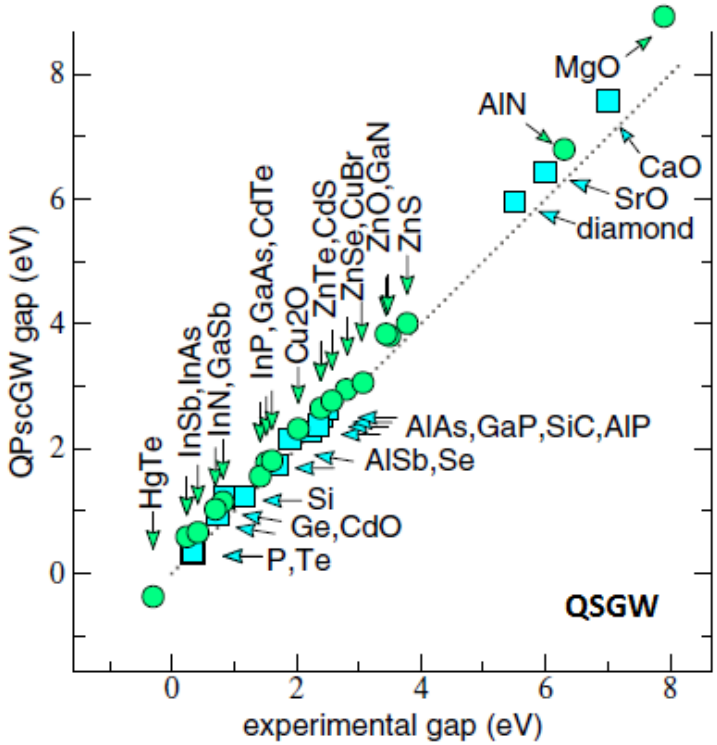
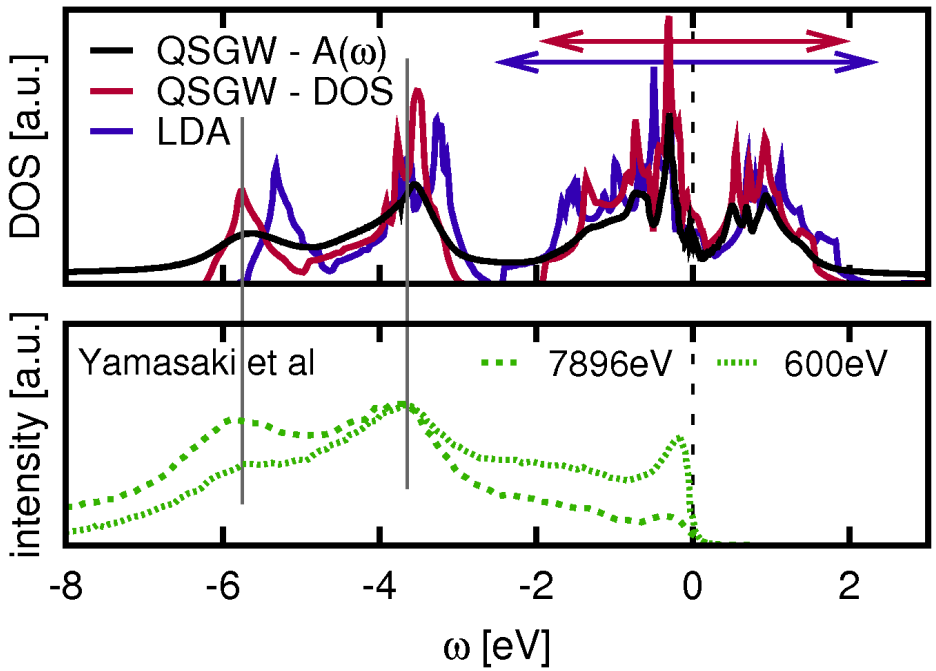




Results: Iron selenide FeSe

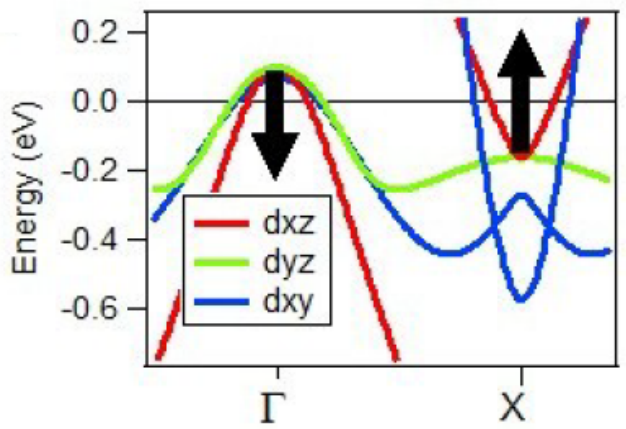
1. correlations beyond the 3d-shell

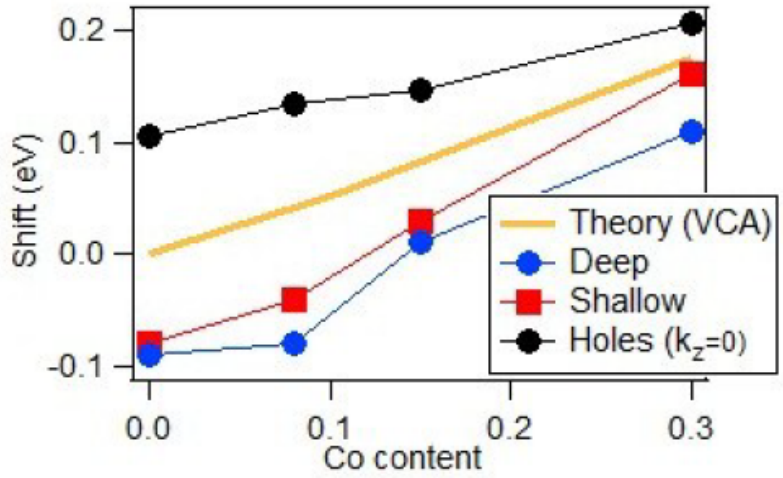


- **correction of high energy excitations** ✓ : effect of Σ_{pp}, Σ_{pd}, beyond DMFT
- **band-narrowing of 22%** ✗
  not strong enough (photoemission: $m^*/m^{\mathrm{LDA}} \approx 3.6$) $\rightarrow$ need DMFT?

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

2. non-local correlations: experimental evidence





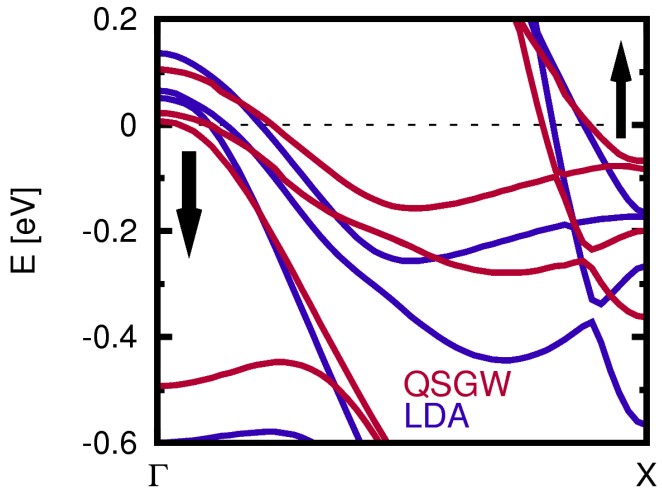
" We show that there is a k-dependent energy shift compared to density functional calculations "

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].

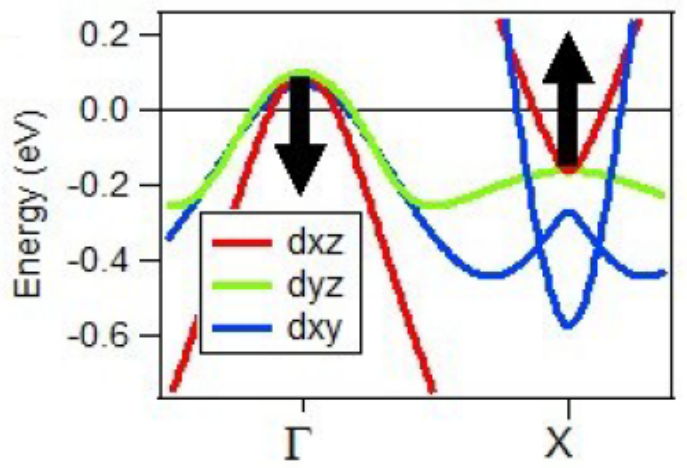
[Brouet *et al.*, PRL 110, 167002 (2013), also: Dhaka *et al.*arXiv:1205.6731v1]

$BaFe_2As_2$: trends with respect to DFT

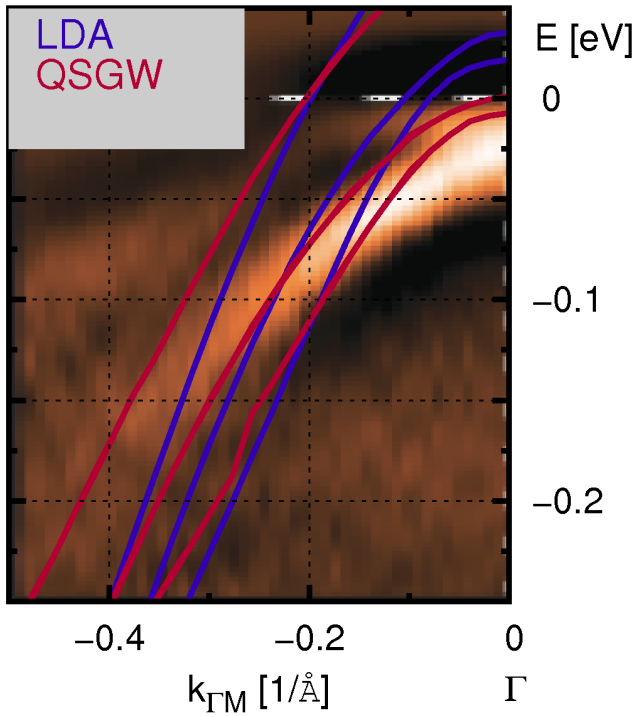
QSGW [JMT *et al.* (2012)]



[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: $\frac{m^{\mathrm{QS}GW}}{m^{\mathrm{LDA}}} = \frac{dE^{LDA}_{\mathbf{k}i}}{dk_\alpha} \Big/ \frac{dE^{\mathrm{QS}GW}_{\mathbf{k}i}}{dk_\alpha}$

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

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.*.]

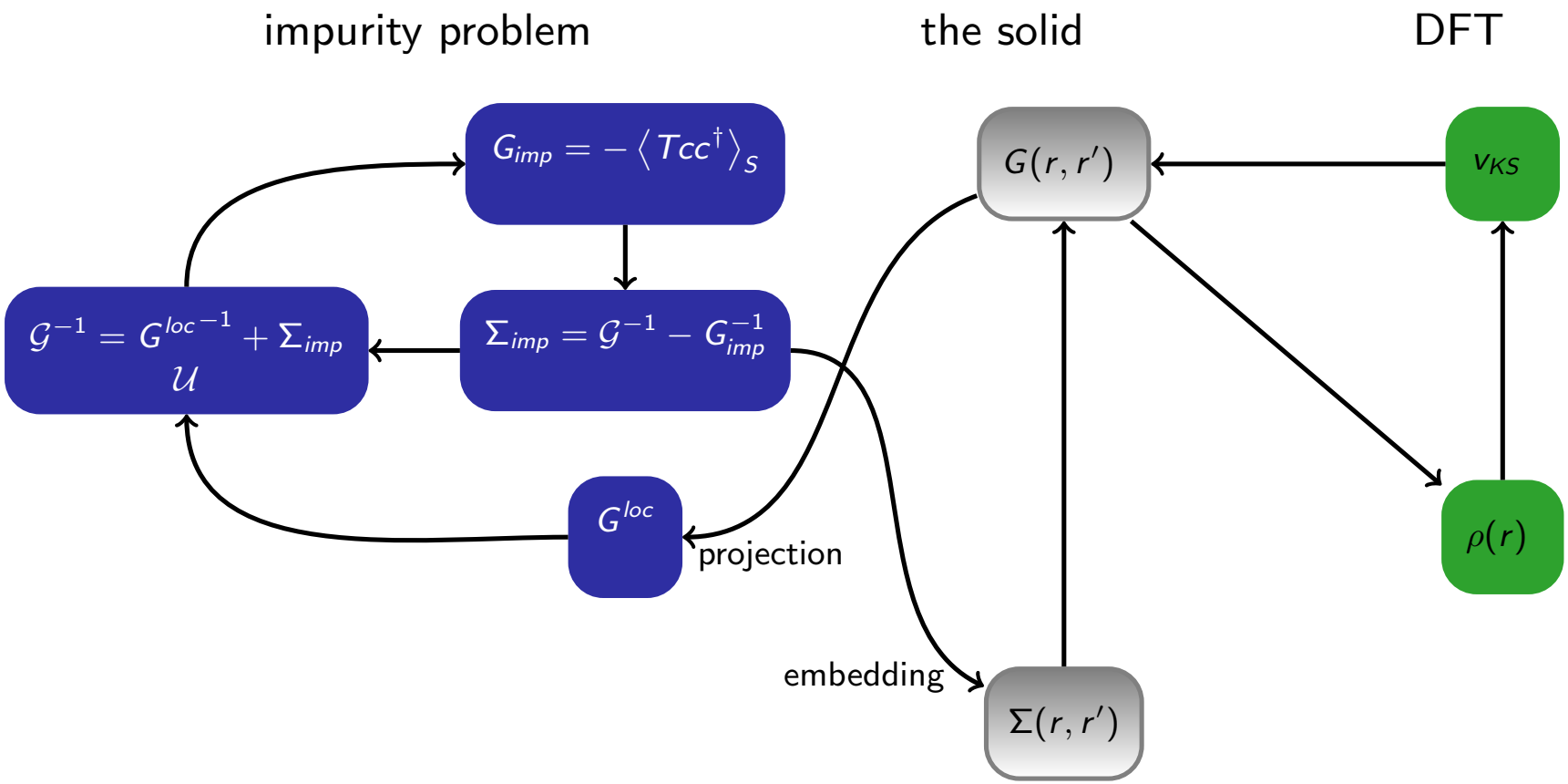
Motivation

- *GW*: non-local correlations substantial
- DMFT: small(er) quasi-particle weights

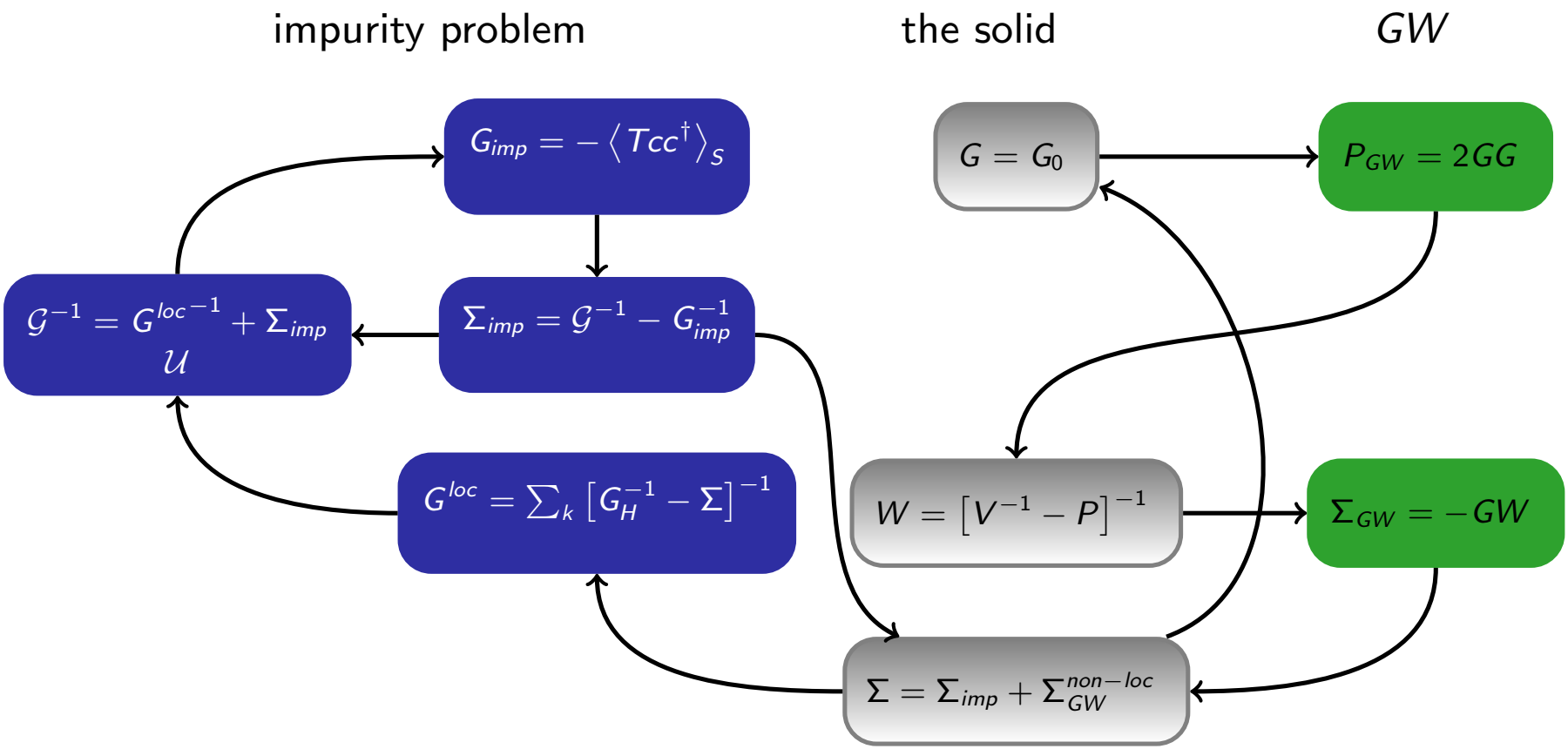
Q Can we combine the best of both methods?

A *GW*+DMFT!

DFT+DMFT


impurity problem
the solid
DFT
$G_{imp} = -\langle Tcc^{\dagger} \rangle_S$
$\mathcal{G}^{-1} = G^{loc\,-1} + \Sigma_{imp}$
$\mathcal{U}$
$\Sigma_{imp} = \mathcal{G}^{-1} - G_{imp}^{-1}$
G^{loc}
projection
embedding
$G(r, r')$
$\Sigma(r, r')$
v_{KS}
$\rho(r)$

"*GW*+DMFT"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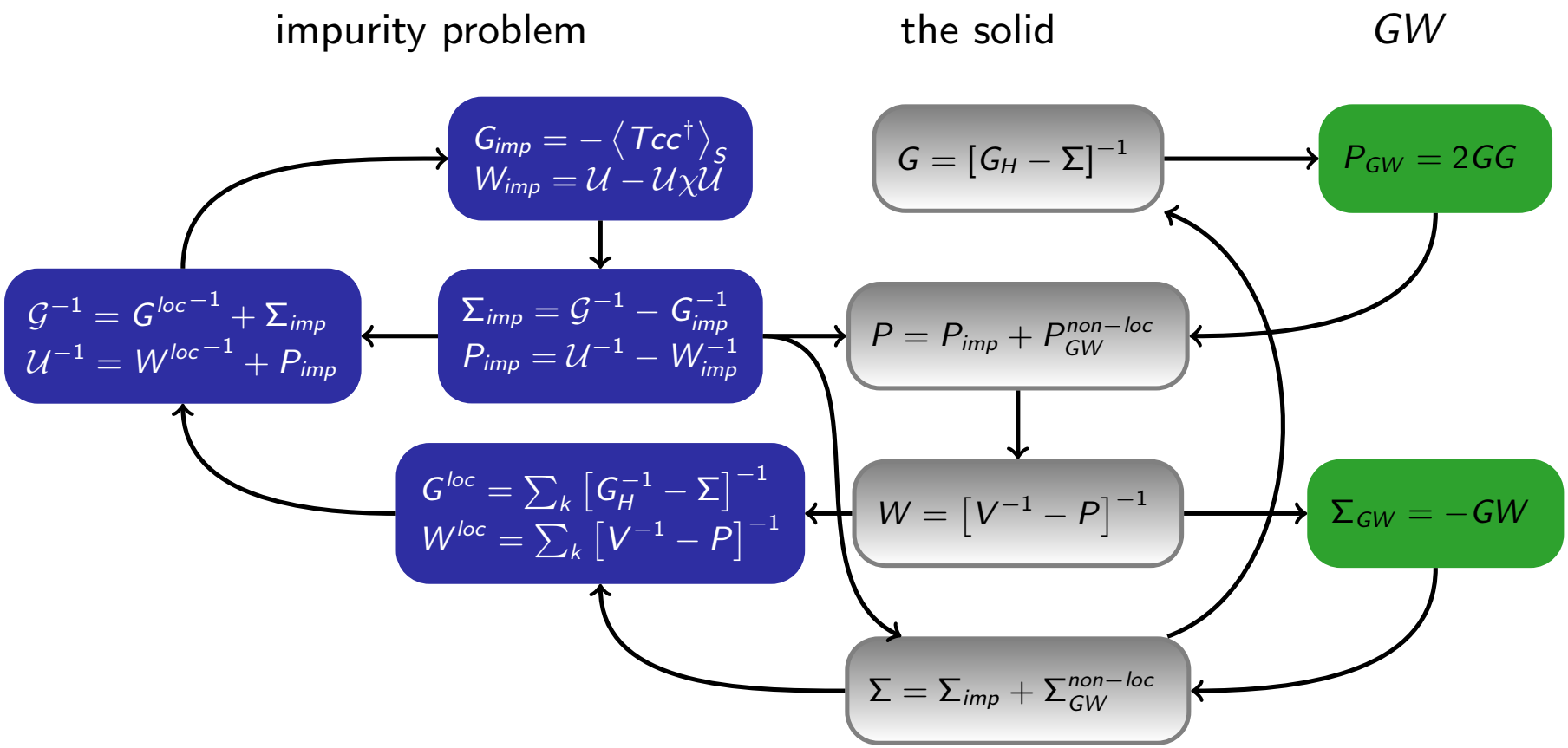
$\mathcal{U}$
$\Sigma_{imp} = \mathcal{G}^{-1} - G_{imp}^{-1}$
$G^{loc} = \sum_k \left[G_H^{-1} - \Sigma \right]^{-1}$
$G = G_0$
$P_{GW} = 2GG$
$W = \left[V^{-1} - P \right]^{-1}$
$\Sigma_{GW} = -GW$
$\Sigma = \Sigma_{imp} + \Sigma_{GW}^{non-loc}$

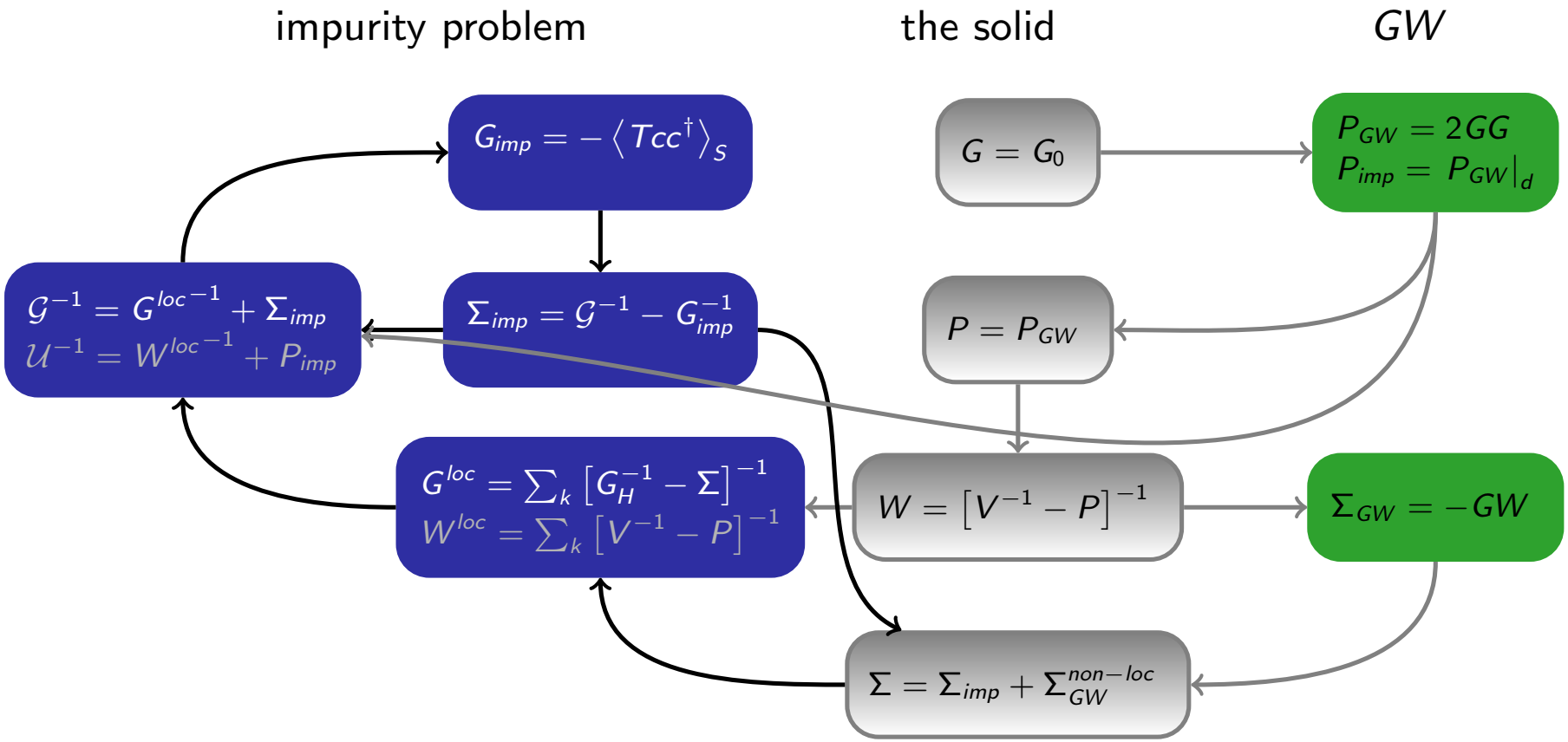
GW+DMFT [Biermann, Aryasetiawan and Georges]



- extended DMFT: $W^{loc} \stackrel{!}{=} W_{imp}$ [Si and Smith, Kajueter, Sengupta and Georges, Sun and Kotliar]
- derivable from a free energy functional $\Gamma[G, W]$, fully *ab initio*
- no starting point (DFT) dependence, no double counting problem

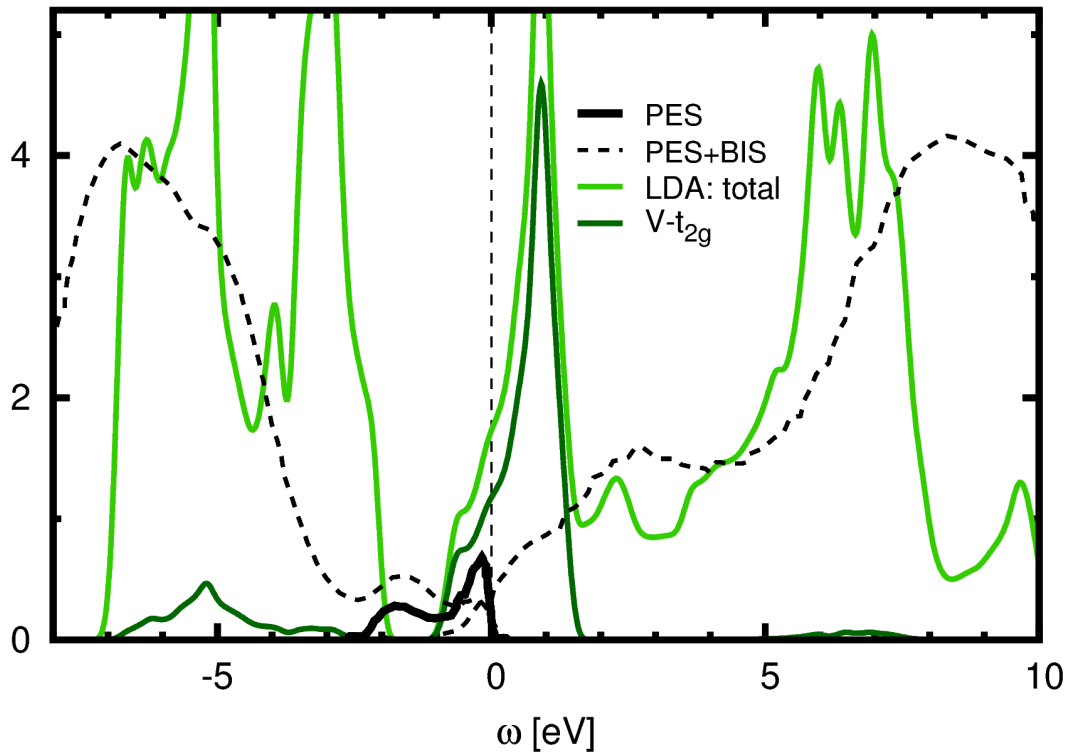
Applications: ad atoms [Hansmann et al.], model calculations [Ayral et al.]

simplified *GW*+DMFT [used in: JMT et al.]



- one-shot *GW* fixes $\Sigma^{non-loc}(\mathbf{k}, \omega)$
- constraint RPA sets dynamical Hubbard $\mathcal{U}(\omega)$
- starting point dependence in $\Sigma^{non-loc}$ and $\mathcal{U}$

$SrVO_3$: photoemission and DFT

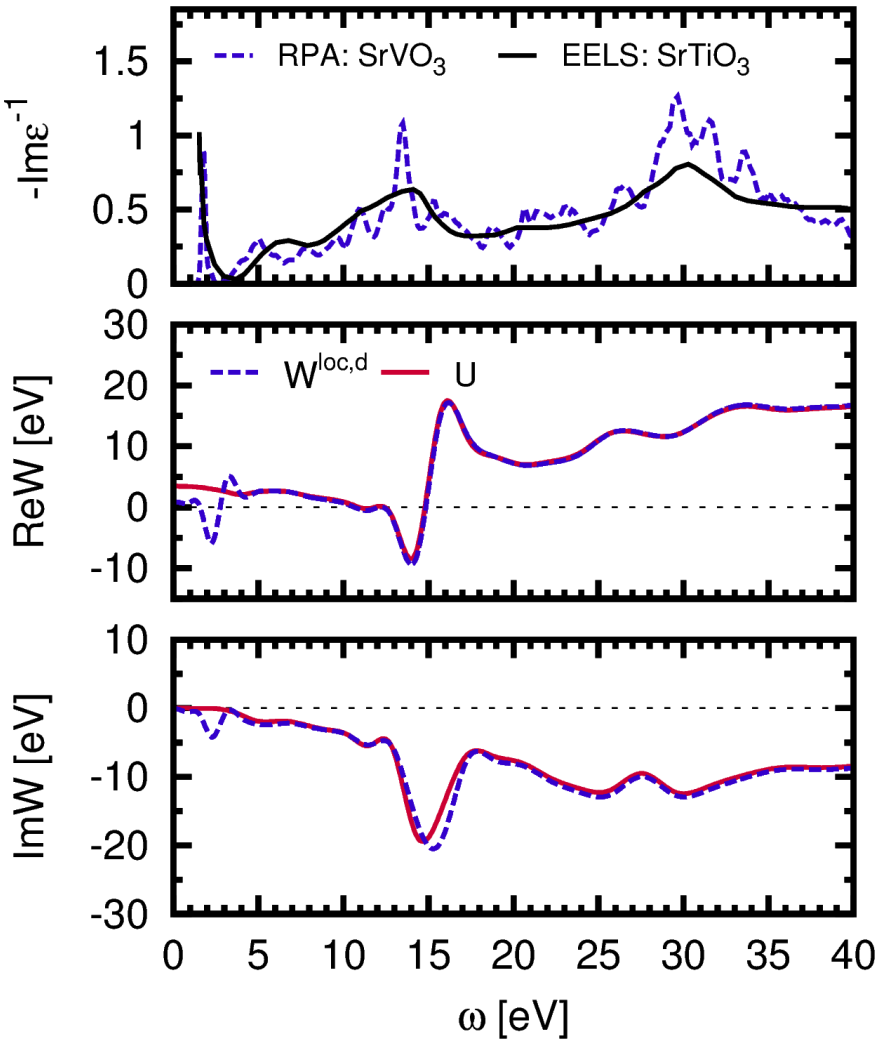


LDA [wien2k], photoemission [Sekiyama et al], (inverse) photoemission [Morikawa et al.]

DFT vs. Photoemission vs. DFT:

- bigger bandwidth (m^*/m)
- satellite absent: lower Hubbard band (LHB)
- separation between O2p-V3d-Sr5d too small

$SrVO_3$: RPA



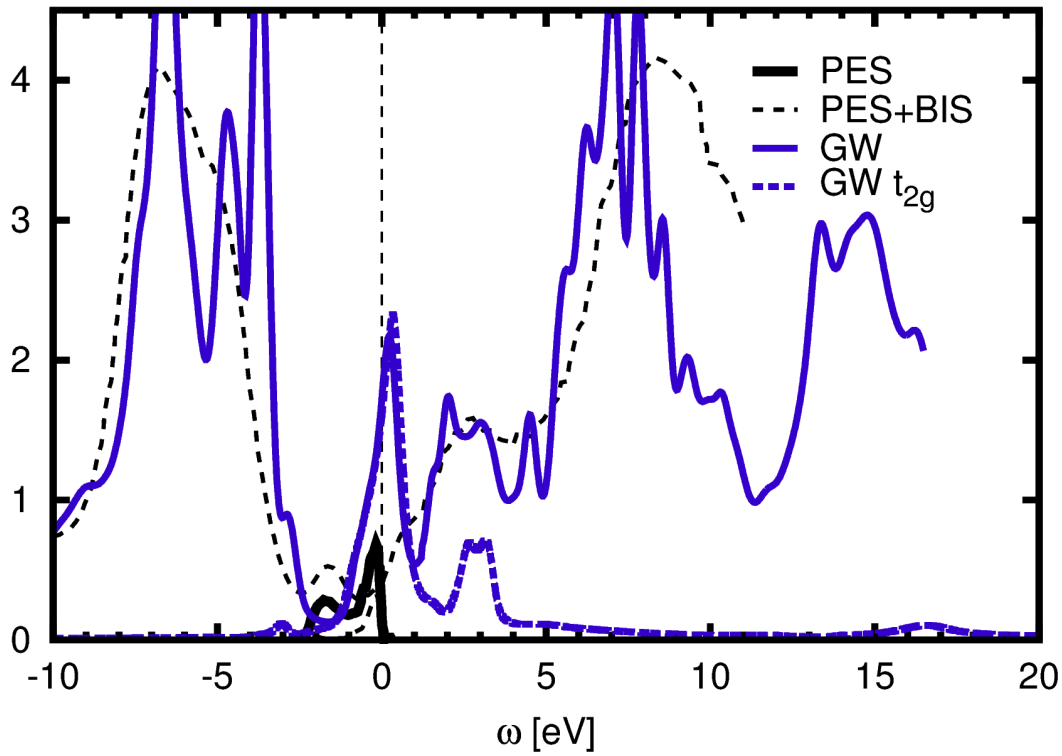
low energy mass enhancement:

- previous *static* DMFT needed $U \approx 5.0$eV for similar spectral transfers
- here $\mathcal{U}(\omega = 0) = 3.5$eV $\longrightarrow$ Hubbard bands at smaller distance
- but frequency dependence $\longrightarrow$ (plasmonic) satellites/weight

satellites:

- $\Im\Sigma(\omega) = \sum_{kn} \psi_{kn} \Im W(\epsilon_{kn} - \omega) \psi^*_{kn}$ peak at ω_0 in $\Im W$ $\longrightarrow$ weight at $\epsilon_{kn} \pm \omega_0$

$SrVO_3$: *GW*

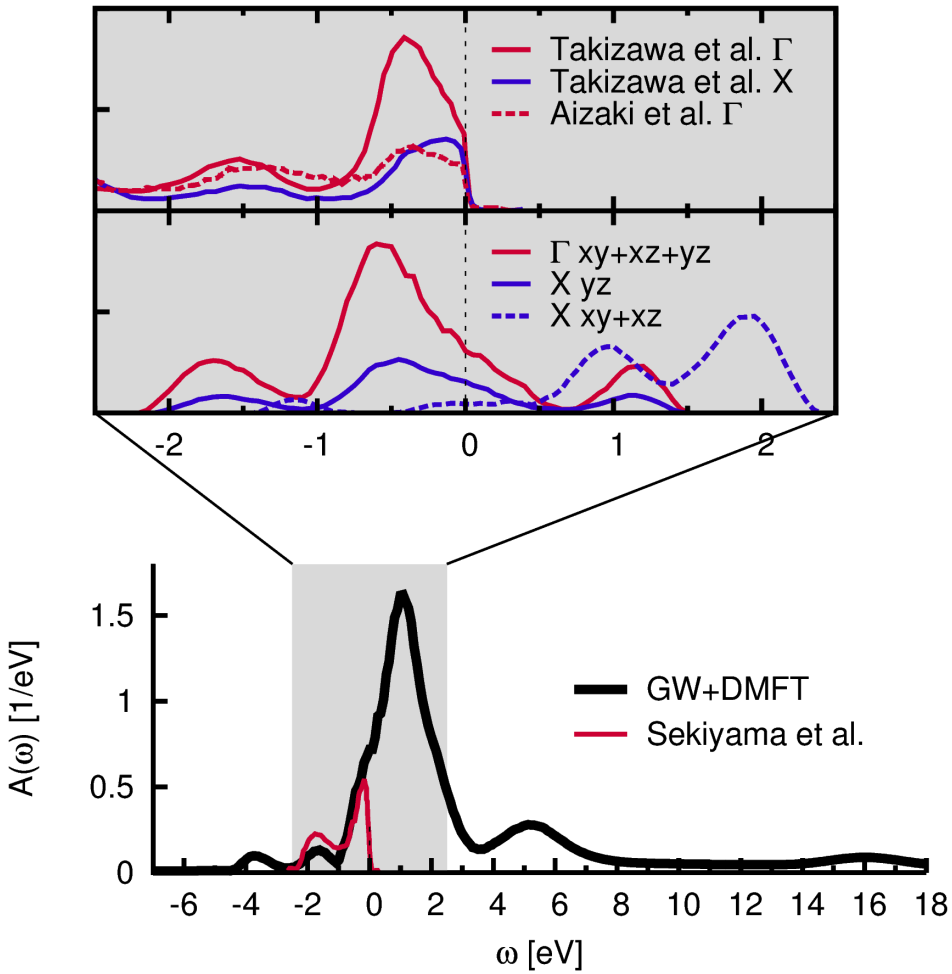


GW [FPLMTO/Kotani], photoemission [Sekiyama et al], (inverse) photoemission [Morikawa et al.]

GW vs. photoemission

- still no lower Hubbard band, but e_g states!
- O2p and Sr5d almost right
- satellites at ± 3 eV, plasmon at 16eV

$SrVO_3$: *GW*+DMFT



- plasmon peak @ 15eV
- *GW* feature moved from 3 to 5eV
- LHB at correct position
- UHB at 2eV, merged with QP dispersion
- IPES feature at 2.7eV from e_g states congruent with lattice cluster calculations [Mossanek et al.]

For *GW*+DMFT for $SrVO_3$ with static U see also [Taranto et al.]
For DMFT@LDA + GW for $SrVO_3$ see [Sakuma et al.]

A simplified scheme?

full *GW*+DMFT scheme rather sophisticated...

Q Short-cut that retains most appealing features of *GW*+DMFT?

A Investigate nature of correlation effects!

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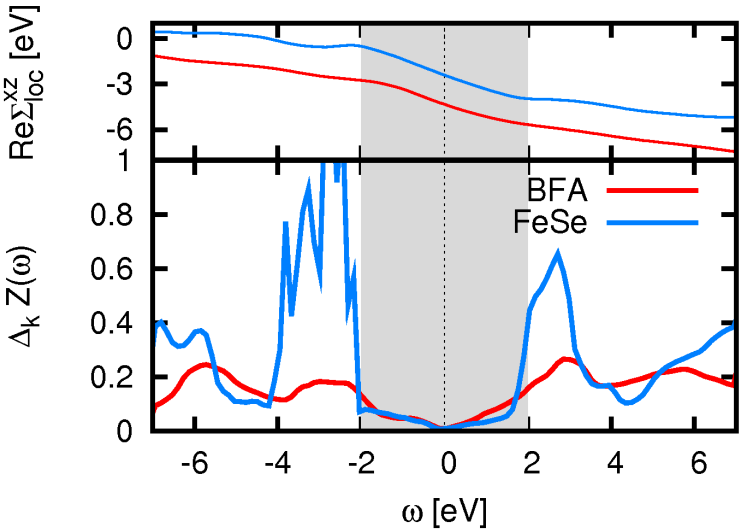
$\Delta_k Z(\omega = 0) = \sqrt{\sum_{\mathbf{k}L} |Z_L^{\mathbf{k}} - Z_L^{loc}|^2} \approx 0.5\% \ \forall$ *GW* calculations here.



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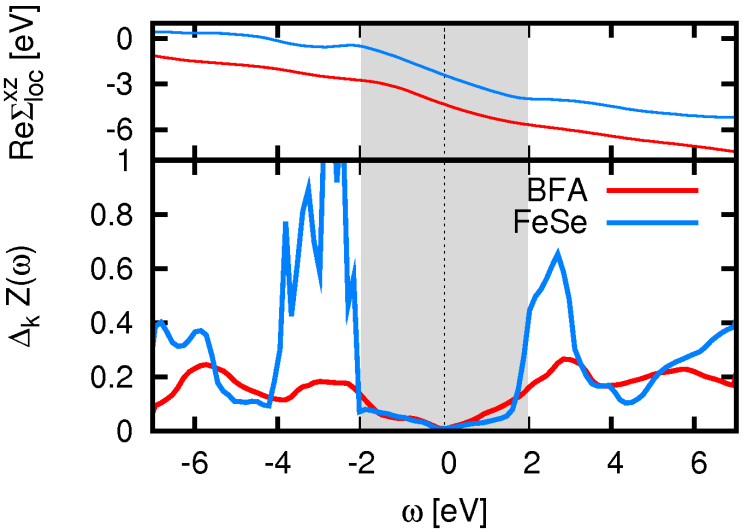

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$\longrightarrow$ **quasiparticle dynamics is local**

$\longrightarrow$ 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.*]

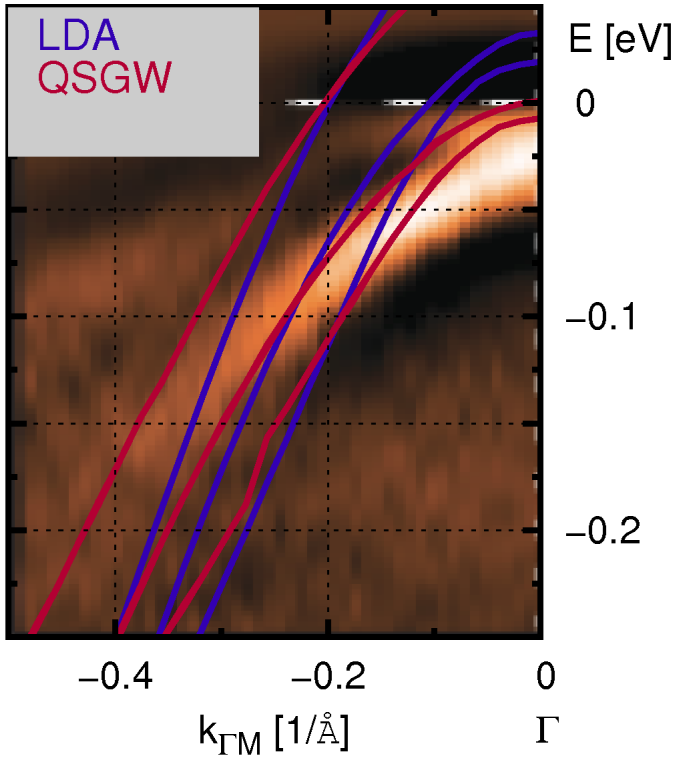
empirically **non-local correlations static** $\longrightarrow$ use **static** QSGW potential v_{xc}^{QSGW}

Proposal: DMFT on top of $H^{QSGW}[v_{xc}^{QSGW}]$

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

- 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 ✓

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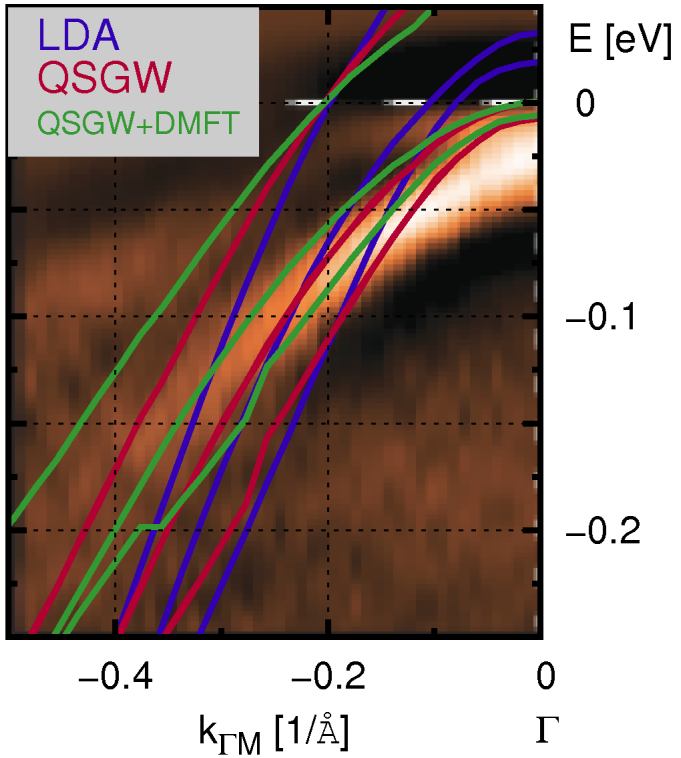


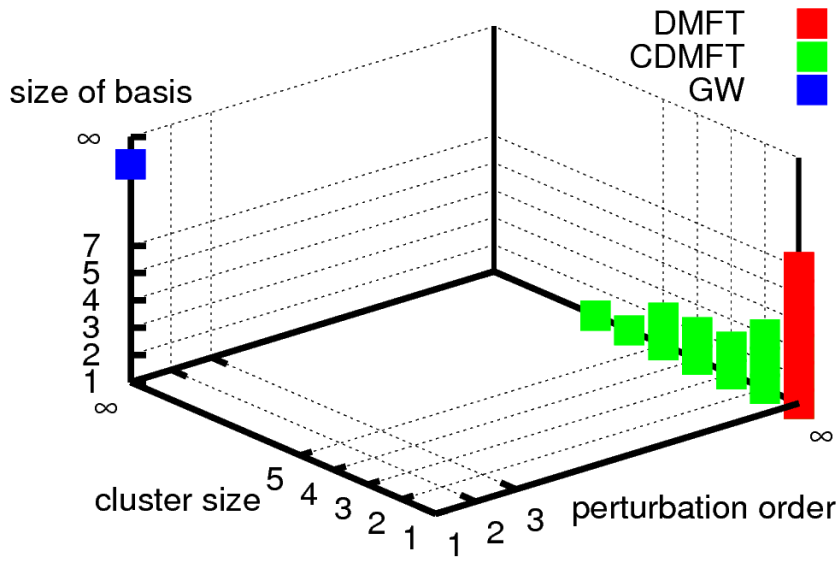
QSGW

- 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"

Conclusions



Methodology

- non-local correlations can be sizeable
  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**
- Combining both merits: *GW*+DMFT
- quasiparticle dynamics **local** & non-locality **static** $\longrightarrow$ **QS***GW***+DMFT**

Thank you for your attention

JMT, M. van Schilfgaarde & G. Kotliar, PRL 109, 237010 (2012)

JMT, M. Casula, T. Miyake, F. Aryasetiawan & S. Biermann, EPL 100, 67001 (2012)