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FLATIRON INSTITUTE
Center for Computational Quantum Physics

Dynamical Mean-Field Theory: What Have We Learned And What Lies Ahead?

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RPMBT XXI - Feenberg Medal

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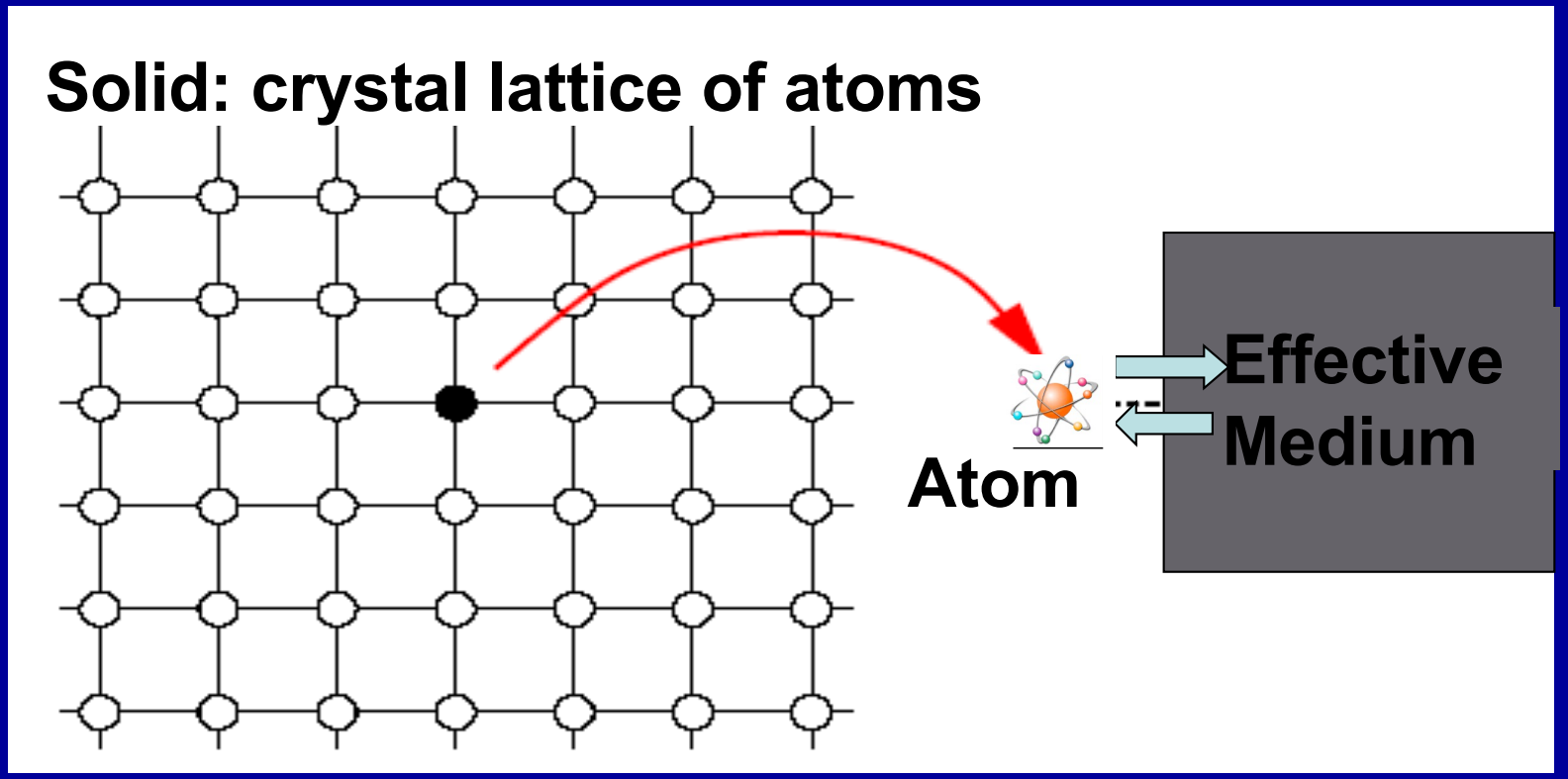
What is DMFT ?

- A *theoretical and computational method* to approach the many-body quantum problem. The method becomes exact in limiting cases and can be systematically improved in a controlled way.
- A *conceptual framework* to think about materials with strong electron correlations and understand their physics

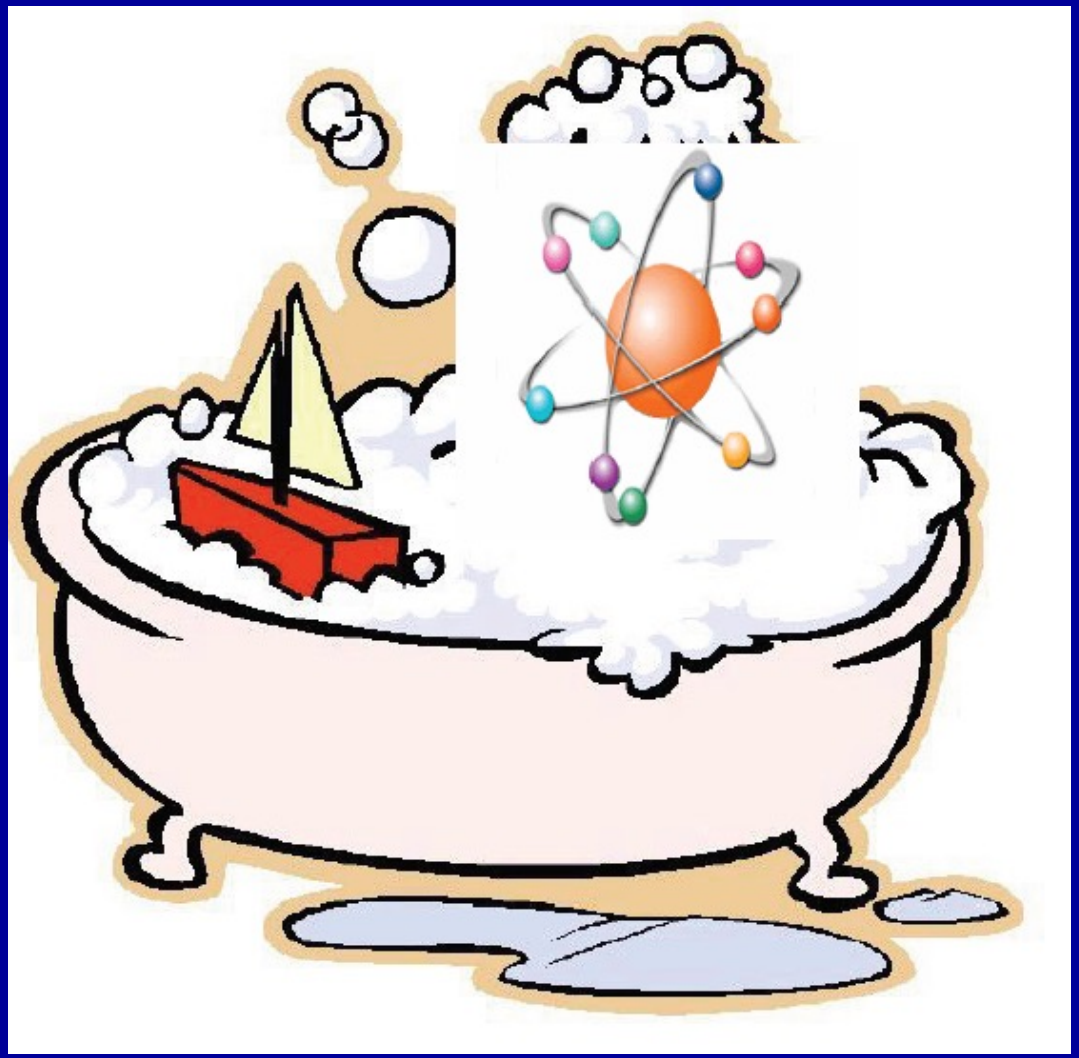
Dynamical Mean-Field Theory:

Viewing a material as an (ensemble of) **atoms** coupled to a **self-consistent effective medium**

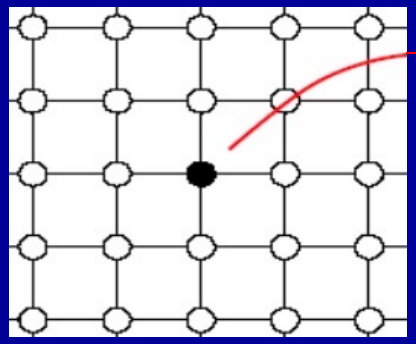


Correlated electrons in infinite dimensions W.Metzner & D.Vollhardt, 1989
Dynamical Mean-Field Theory A.G. & G.Kotliar, 1992

`Atom in a Bath’



The Embedding Concept



Observable: Local Green's function

$$G_{ii}(\tau - \tau') = -\langle T c_i(\tau) c_i^\dagger(\tau') \rangle$$

$\Delta(\tau - \tau')$

Effective Medium

(`Bath')

$\Delta(\tau$-τ'): Dynamical Mean-Field

Quantum generalization of Weiss field in Stat Mech

Chosen such as to reproduce the local G

Organizing Principle: Locality

The single-site DMFT approximation: local self-energy

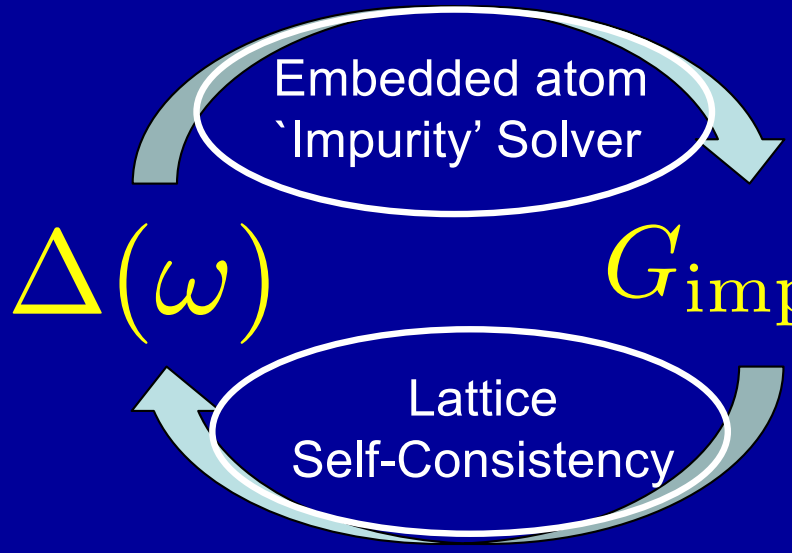
$$\Sigma_{\rm lattice}(\mathbf{k},\omega) \simeq \Sigma(\omega) \Leftrightarrow \Sigma_{ij}(\omega) \simeq \Sigma(\omega)\,\delta_{ij}$$

With $\Sigma(\omega)$ the self-energy of the embedded atom (`impurity')

A good approximation when correlation lengths
are SMALL (e.g. high temperature,
high doping, frustration, several competing fluctuations, etc.)

Can be improved in a systematic and controlled way
by enlarging the size of the embedded fragment:
Cluster Extensions of DMFT, Generalized Embedding Methods…

The DMFT Self-Consistency Loop



$$\Rightarrow \mathrm{Self-energy} : \Sigma(\omega)$$
$$\Rightarrow \mathrm{Local\ Vertex} : \Gamma(\omega_1, \omega_2, \omega_3)$$

Gives access to the lattice momentum-dependent Green's function and response functions:

$$G(\mathrm{k}, \omega) = [\omega + \mu - H_{\mathrm{k}} - \Sigma(\omega)]^{-1}$$
$$\chi(\mathrm{q}, \omega) \sim \chi_0 + \chi_0 \star \Gamma \star \chi$$

Weiss mean-field theory
Density-functional theory
Dynamical mean-field theory

} Share a similar conceptual basis

TABLE 2. Comparison of theories based on functionals of a local observable

Theory	MFT	DFT	DMFT
Quantity	Local magnetization m_i	Local density $n(x)$	Local GF $G_{ii}(\omega)$
Equivalent system	Spin in effective field	Electrons in effective potential	Quantum impurity model
Generalised Weiss field	Effective local field	Kohn-Sham potential	Effective hybridisation $\Delta(\omega)$

Total Energy Functional: E[G] or E[Σ]

The single-site DMFT construction is EXACT:

- For the non-interacting system

  U =0 → Σ = 0 - hence k-independent!

- For the isolated atom

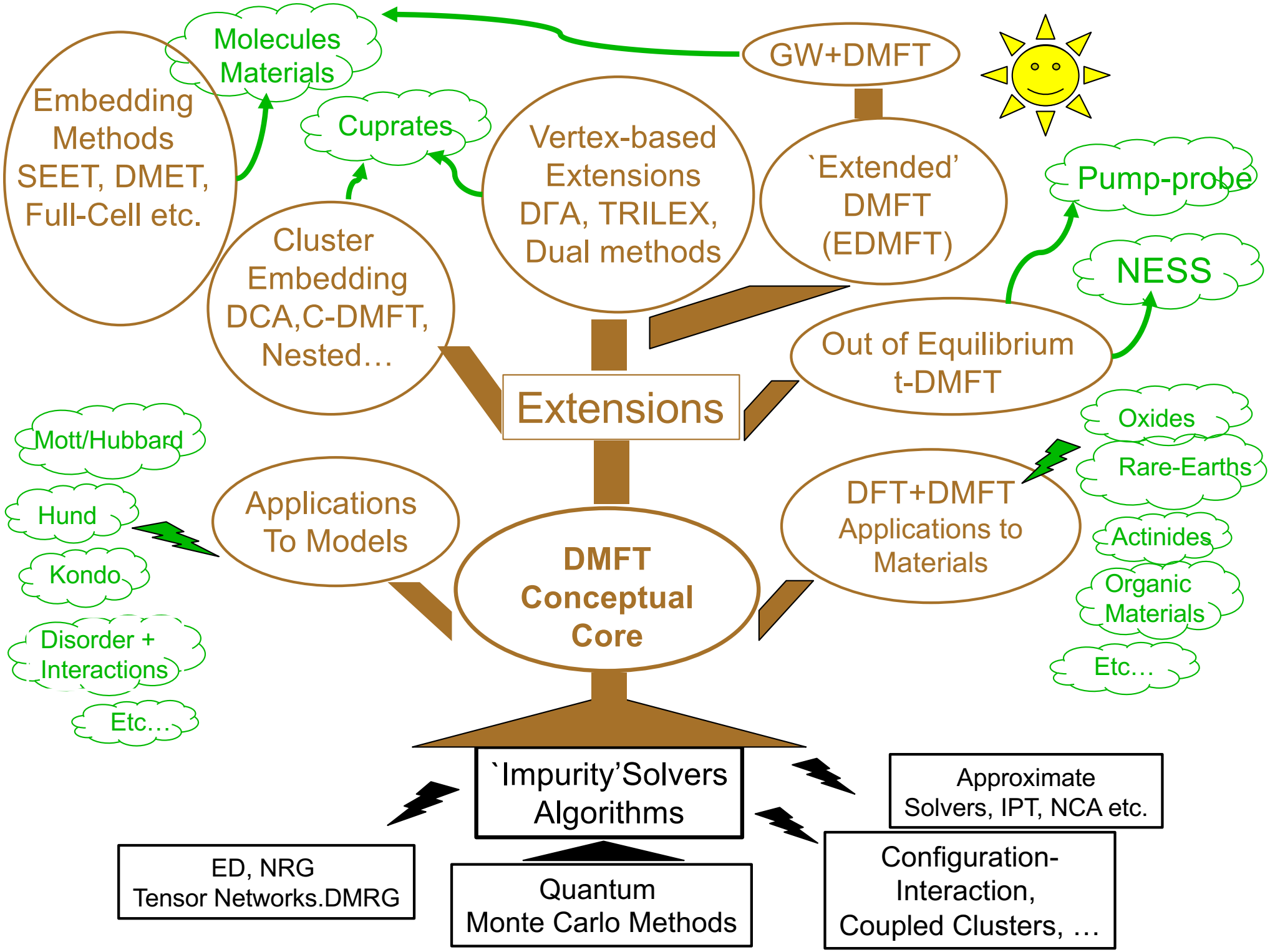
  `Atomic' limit t=0 → $\Sigma = \Sigma_{\text{atom}}(\omega)$

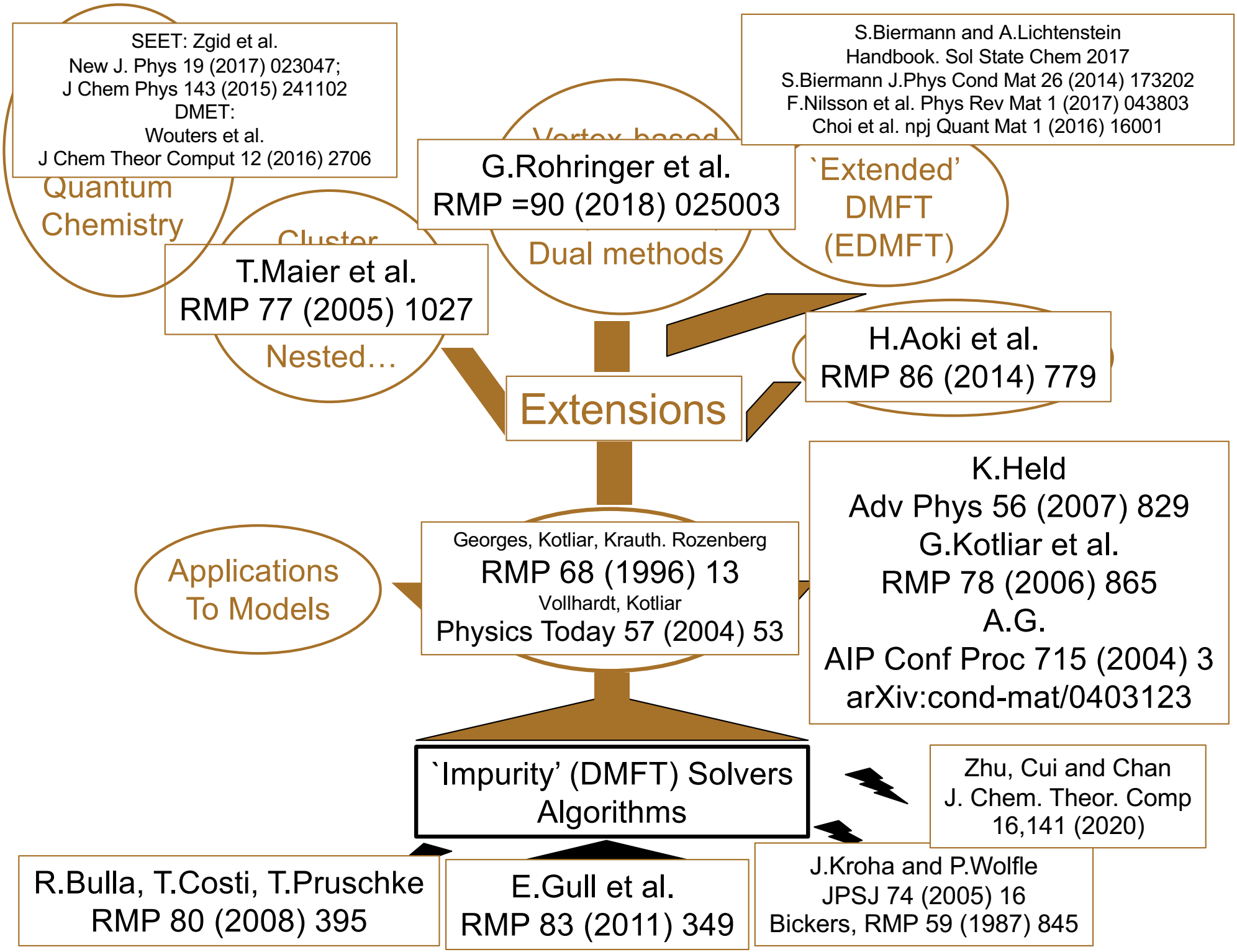
  Hence provides an interpolation

  from weak to strong coupling

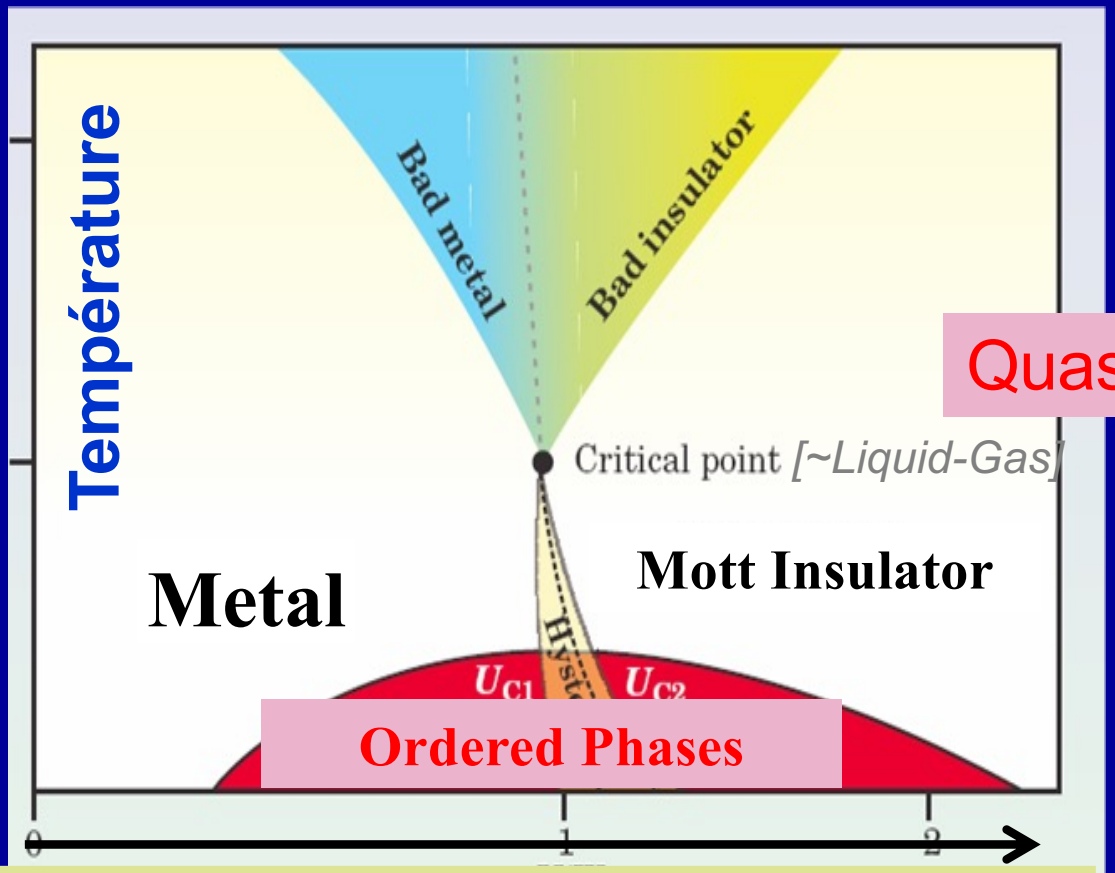
- In the formal limit of infinite dimensionality (infinite lattice coordination) [introduced by Metzner and Vollhardt, PRL 62 (1989) 324]

And, more relevant to physics:
it is a good approximation
when spatial correlations are not too long-range

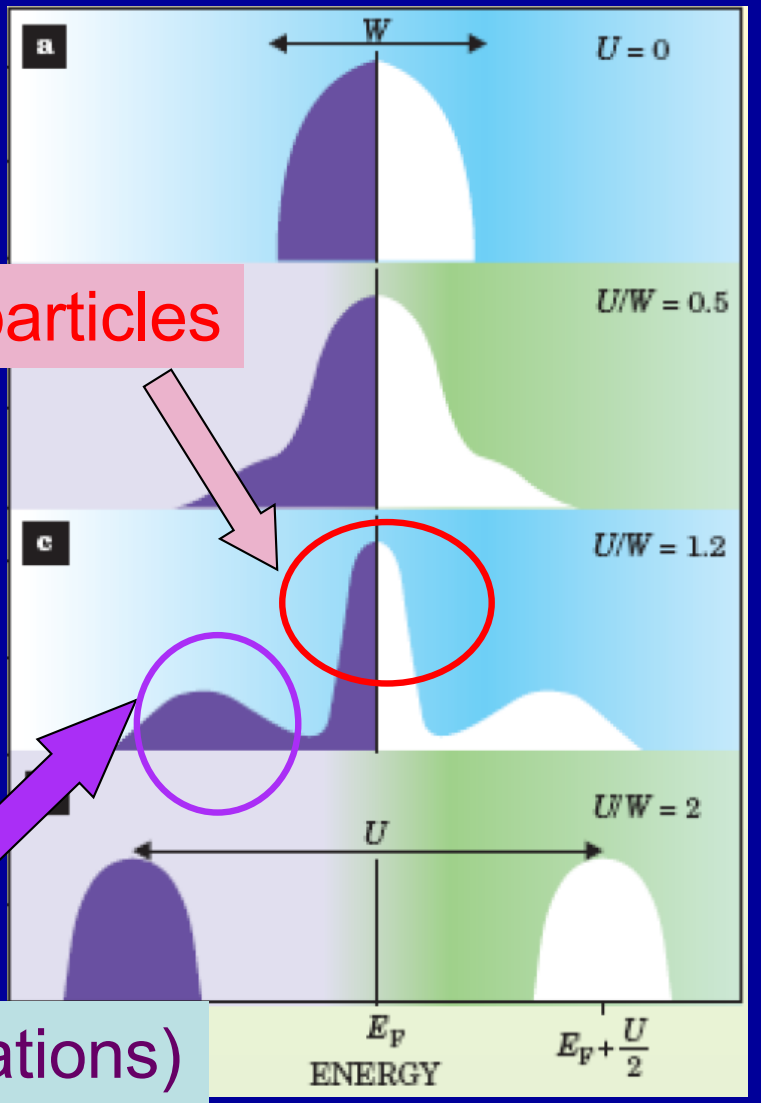

Molecules
Materials
Embedding
Methods
SEET, DMET,
Full-Cell etc.
Cuprates
Cluster
Embedding
DCA,C-DMFT,
Nested…
Vertex-based
Extensions
DΓA, TRILEX,
Dual methods
GW+DMFT
`Extended'
DMFT
(EDMFT)
Pump-probe
NESS
Out of Equilibrium
t-DMFT
Extensions
Mott/Hubbard
Hund
Kondo
Disorder +
Interactions
Etc…
Applications
To Models
DMFT
Conceptual
Core
DFT+DMFT
Applications to
Materials
Oxides
Rare-Earths
Actinides
Organic
Materials
Etc…
`Impurity'Solvers
Algorithms
ED, NRG
Tensor Networks.DMRG
Quantum
Monte Carlo Methods
Approximate
Solvers, IPT, NCA etc.
Configuration-
Interaction,
Coupled Clusters, …


SEET: Zgid et al.
New J. Phys 19 (2017) 023047;
J Chem Phys 143 (2015) 241102
DMET:
Wouters et al.
J Chem Theor Comput 12 (2016) 2706
Quantum Chemistry
S.Biermann and A.Lichtenstein
Handbook. Sol State Chem 2017
S.Biermann J.Phys Cond Mat 26 (2014) 173202
F.Nilsson et al. Phys Rev Mat 1 (2017) 043803
Choi et al. npj Quant Mat 1 (2016) 16001
G.Rohringer et al.
RMP =90 (2018) 025003
Dual methods
`Extended' DMFT (EDMFT)
T.Maier et al.
RMP 77 (2005) 1027
Nested…
H.Aoki et al.
RMP 86 (2014) 779
Extensions
K.Held
Adv Phys 56 (2007) 829
G.Kotliar et al.
RMP 78 (2006) 865
A.G.
AIP Conf Proc 715 (2004) 3
arXiv:cond-mat/0403123
Applications To Models
Georges, Kotliar, Krauth. Rozenberg
RMP 68 (1996) 13
Vollhardt, Kotliar
Physics Today 57 (2004) 53
`Impurity' (DMFT) Solvers Algorithms
Zhu, Cui and Chan
J. Chem. Theor. Comp
16,141 (2020)
R.Bulla, T.Costi, T.Pruschke
RMP 80 (2008) 395
E.Gull et al.
RMP 83 (2011) 349
J.Kroha and P.Wolfle
JPSJ 74 (2005) 16
Bickers, RMP 59 (1987) 845

An early success of DMFT (1992-1999)
Theory of the Mott transition



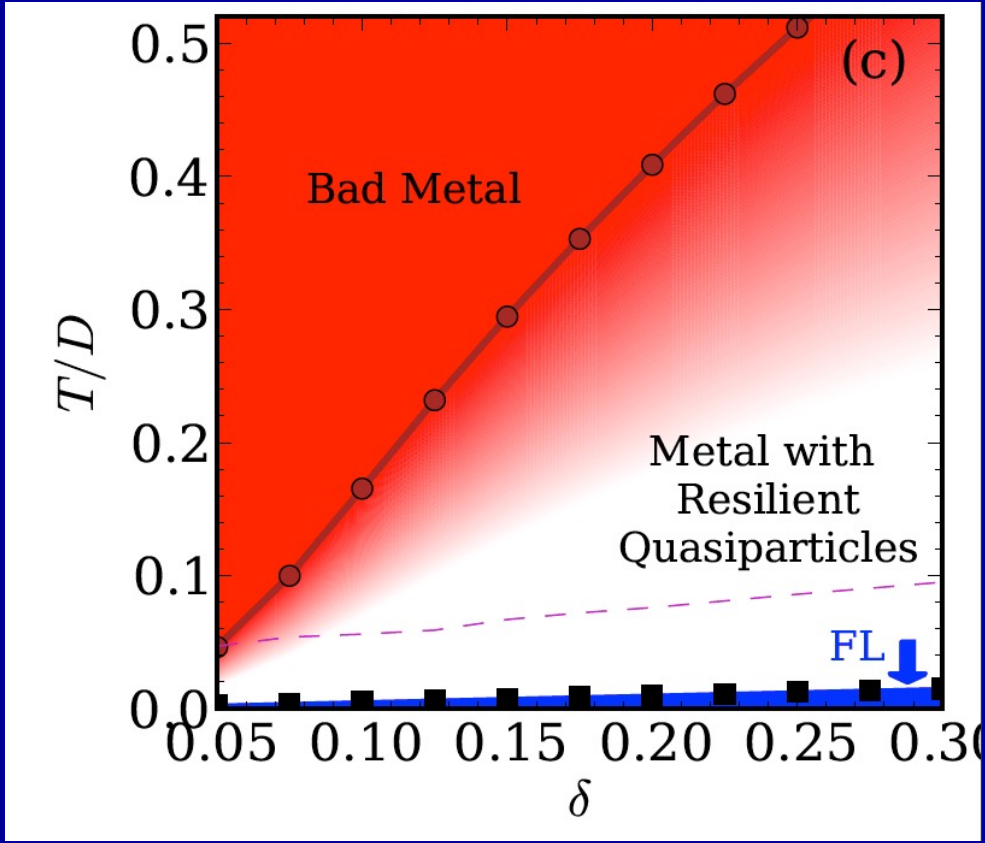
Quasiparticles



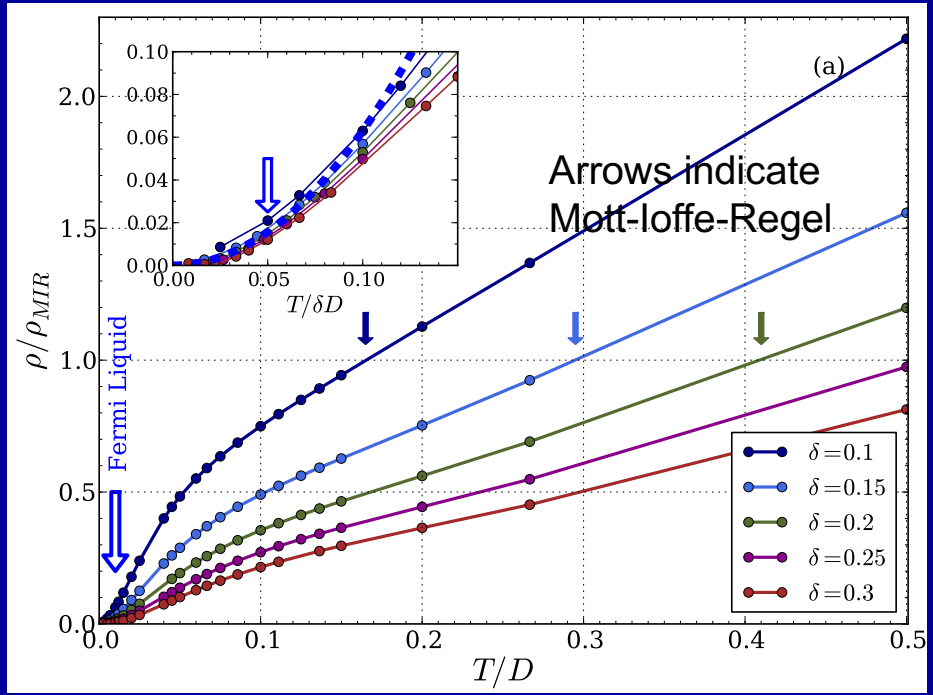
Hubbard `bands' (Quasi atomic excitations)

DMFT insight into a long-standing problem: "How bad metals become good"

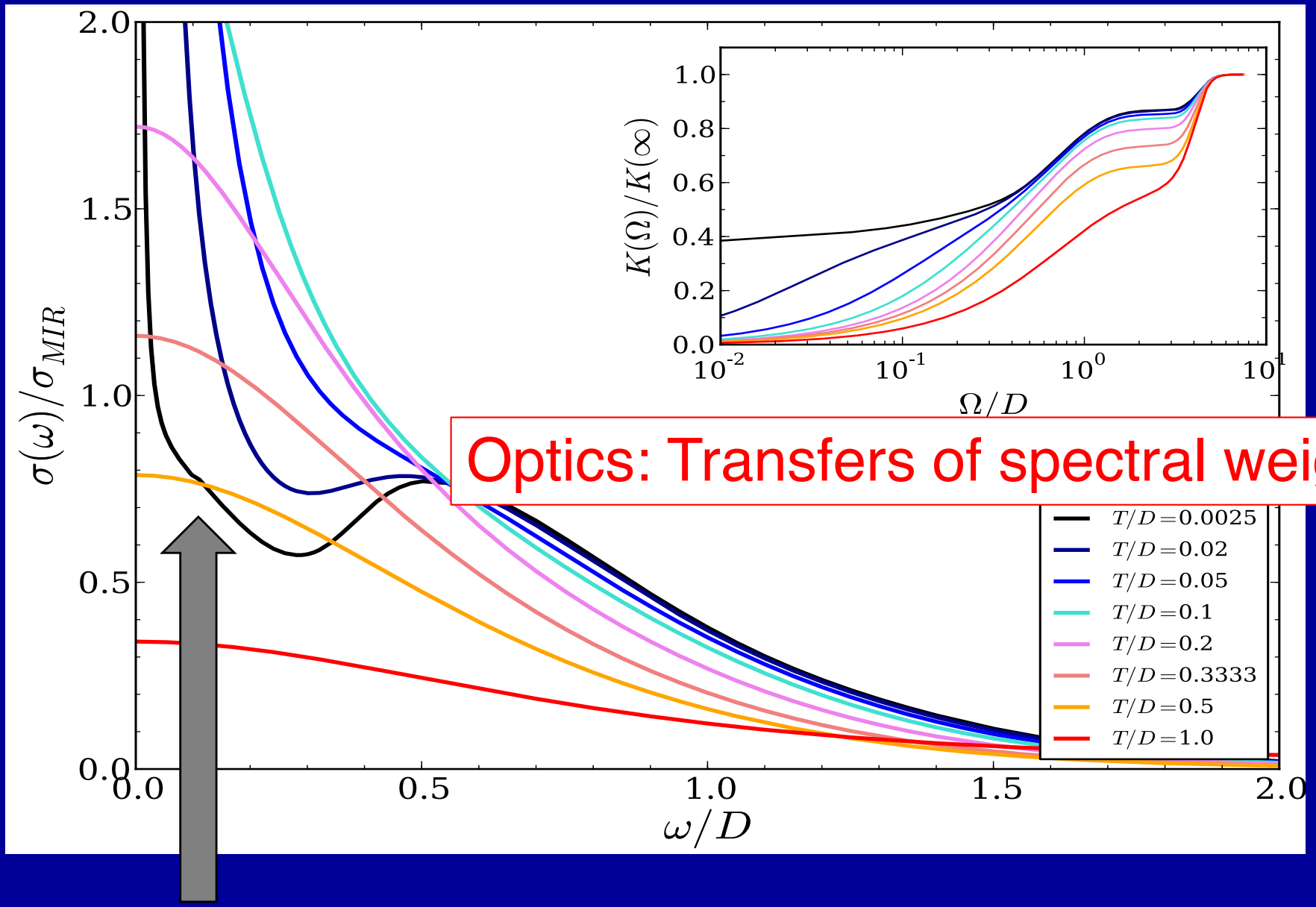
`Resilient' *quasiparticles* beyond Landau Theory



Resistivity: from a Fermi Liquid
to a bad metal above Mott-Ioffe Regel

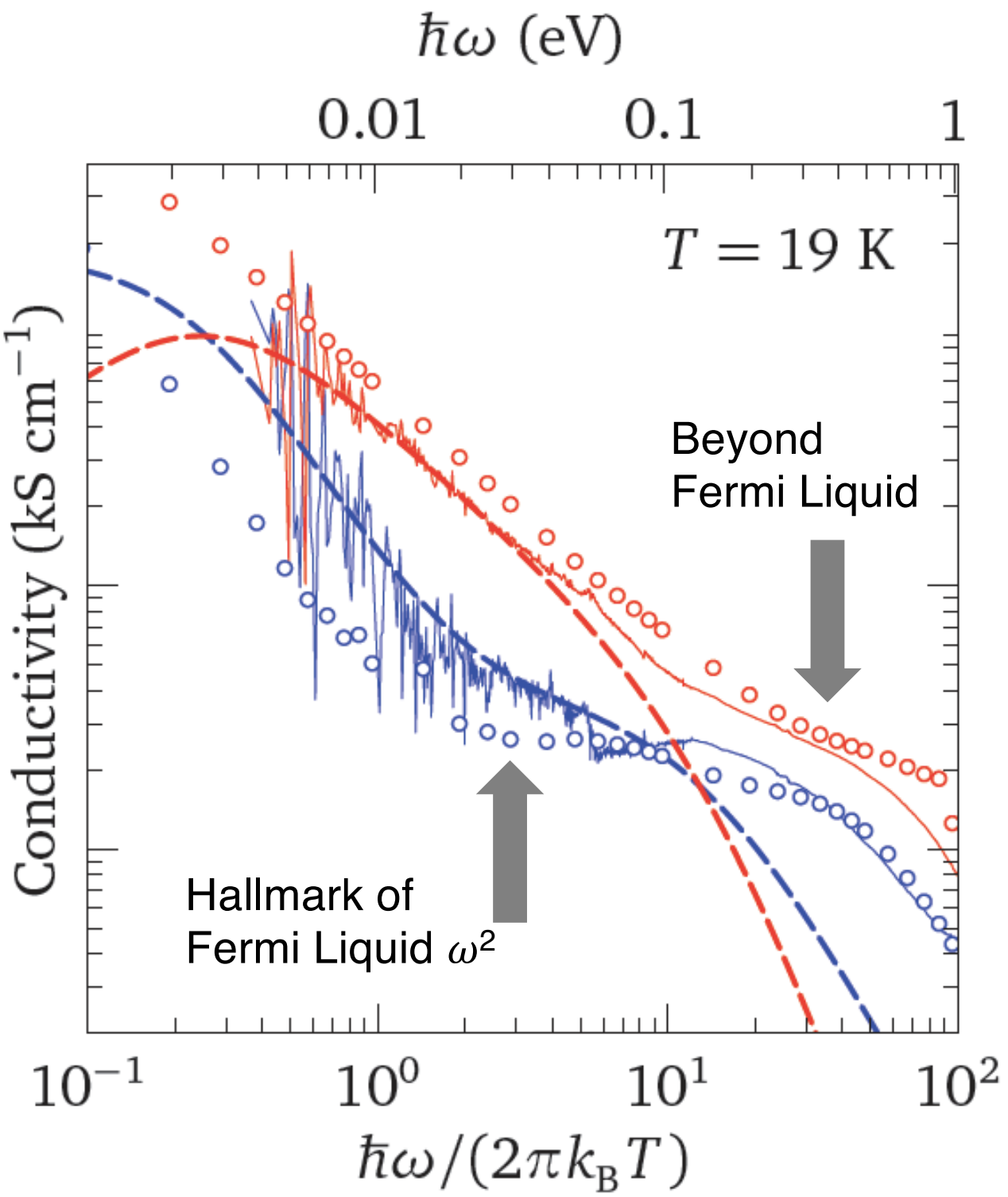


Deng et al. PRL 110 (2013) 086401 ; Xu et al. PRL 111 (2013) 036401



Optics: Transfers of spectral weight

This non-Drude ``foot" is actually the signature of Landau's Fermi liquid (ω^2) in the optical spectrum



Sr_2RuO_4
Re σ(ω)
Im σ(ω)

Plain Lines:
Experiment

Dashed Line:
Fermi Liquid Theory

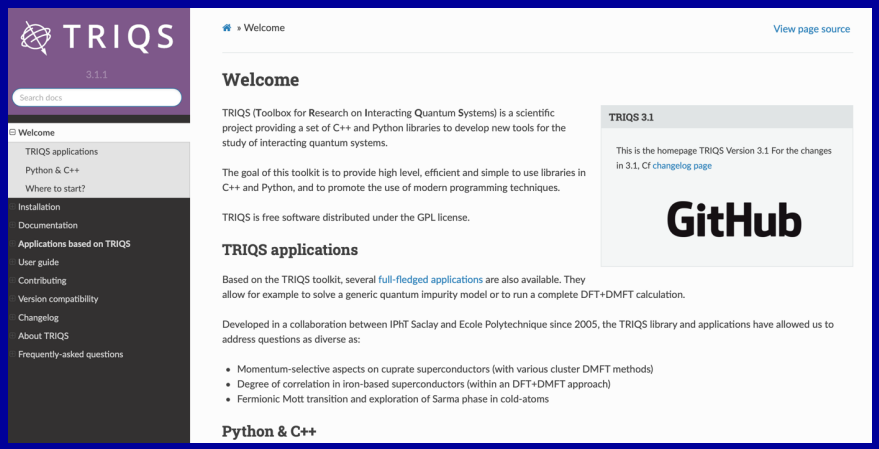
Dots:
Theoretical
Calculation
(LDA+DMFT)

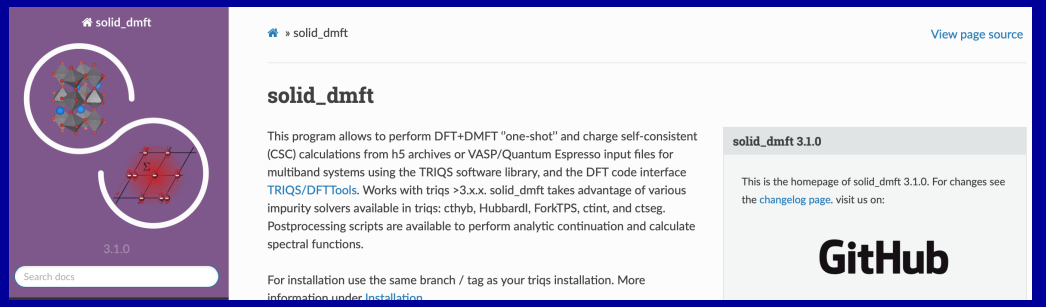
D.Stricker et al.
PRL 113, 0874040
(2014)

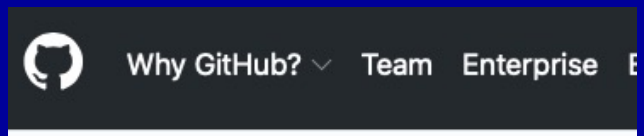
Under the Hood:
Development of Efficient `Impurity Solver' Algorithms is CRUCIAL

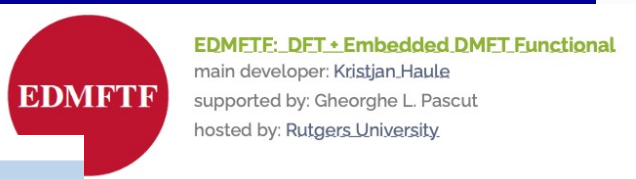
- Solvers working directly with a continuous bath
  Typically: Quantum Monte Carlo (various kinds)
- Solvers requiring a discretization (Hamiltonian form) of the bath - Exact Diagonalisation, Wilson Numerical Renormalisation Group, Fork Tensor Product States, Configuration Interaction, Coupled Cluster, etc.
- Approximation Schemes e.g. IPT, NCA, OCA, …

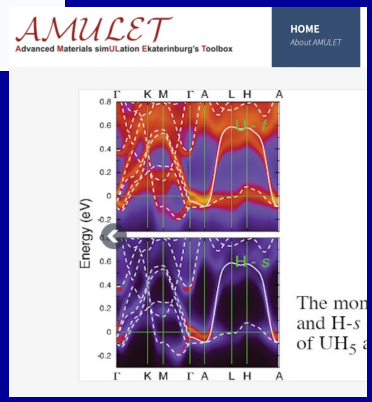
A Vital Community Endeavor: Efficient and Sustainable Open-Source Software Libraries

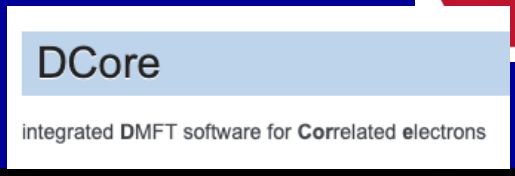


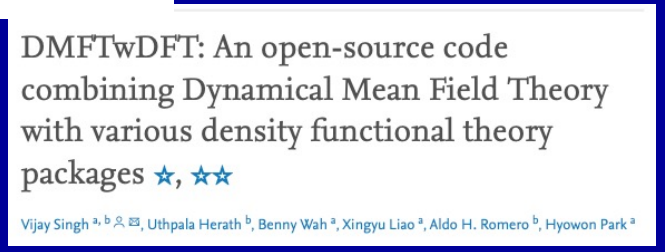


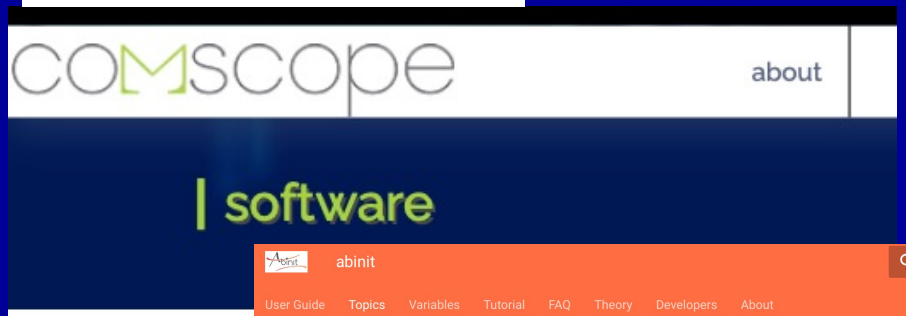


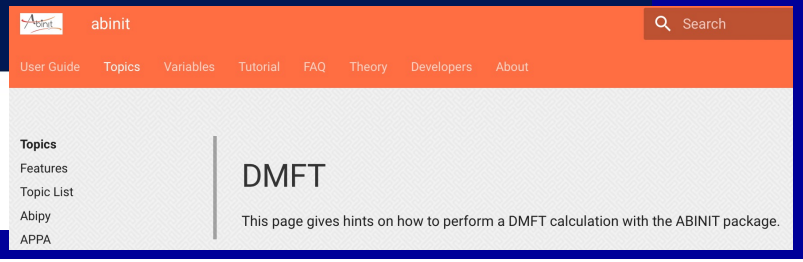


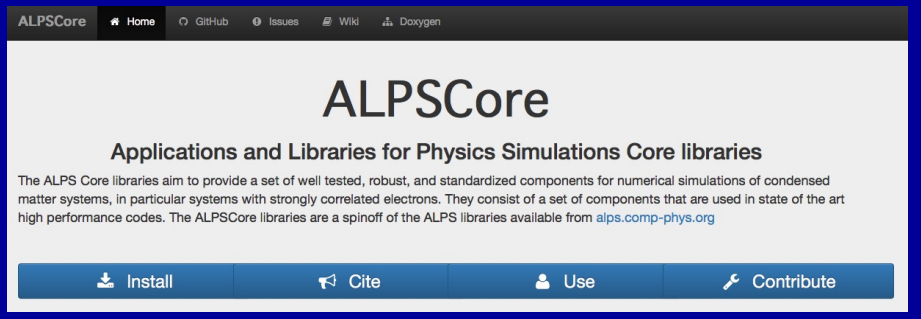












QMC algorithmic breakthroughs

Early days: Hirsch-Fye Algorithm (1986)
First application to DMFT (1992):
Mark Jarrell; Rozenberg and Kotliar; AG and W.Krauth

Continuous-time quantum Monte Carlo (CT-QMC): 2005 → Today

- Interaction expansion(CT-INT) Rubtsov (2005)
- Hybridization expansion (CT-HYB)
  P. Werner, M.Troyer, A.Millis et al 2006; Haule 2007
- Auxiliary field (CT-AUX) E.Gull O.Parcollet 2008

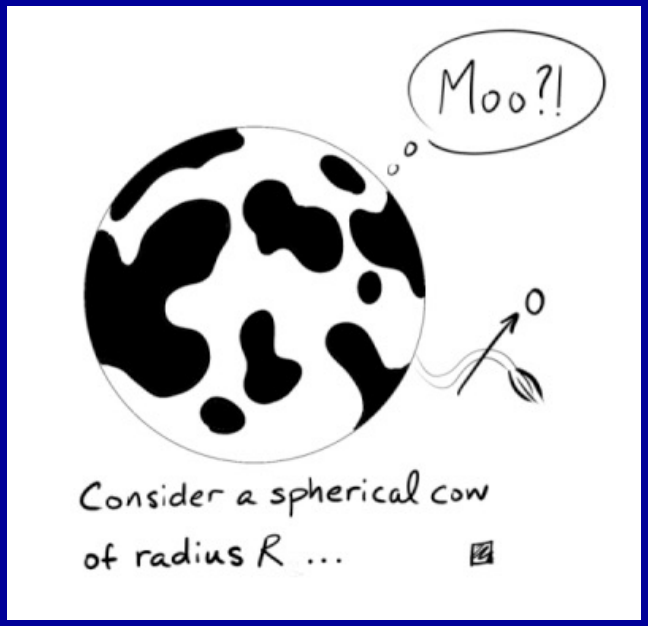
Review: Gull et al. Rev Mod Phys 83, 349 (2011)

- Inchworm: Cohen, Gull et al. 2015→
- Real-time Diagrammatic MC: Waintal, Parcollet, Messio,Profumo, Bertrand, Dumitrescu et al (2017→)

The Happy Marriage of DMFT With Electronic Structure (DFT,GW,…)

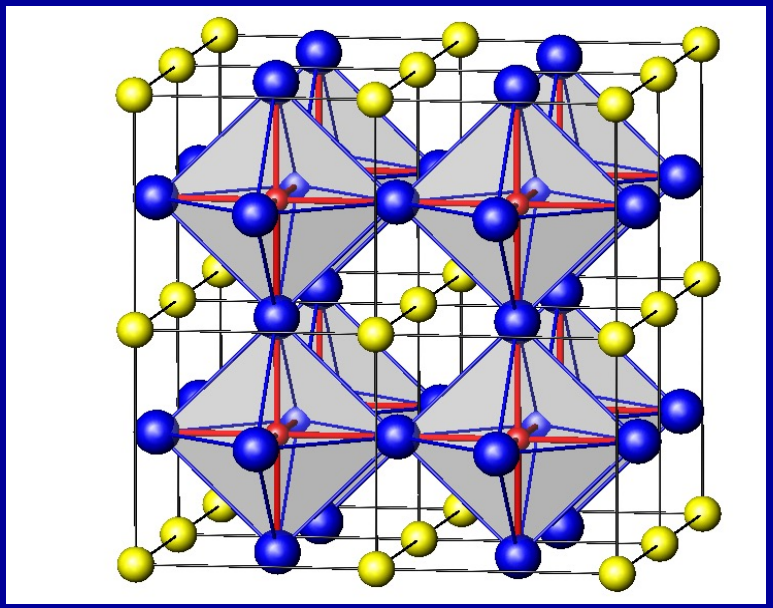
An interdisciplinary collective effort started ~ 1996 and still continuing today

Anisimov, Kotliar et al. J.Phys Cond Mat 9, 7359 (1997)
Lichtenstein and Katsnelson Phys Rev B 57, 6884 (1998)

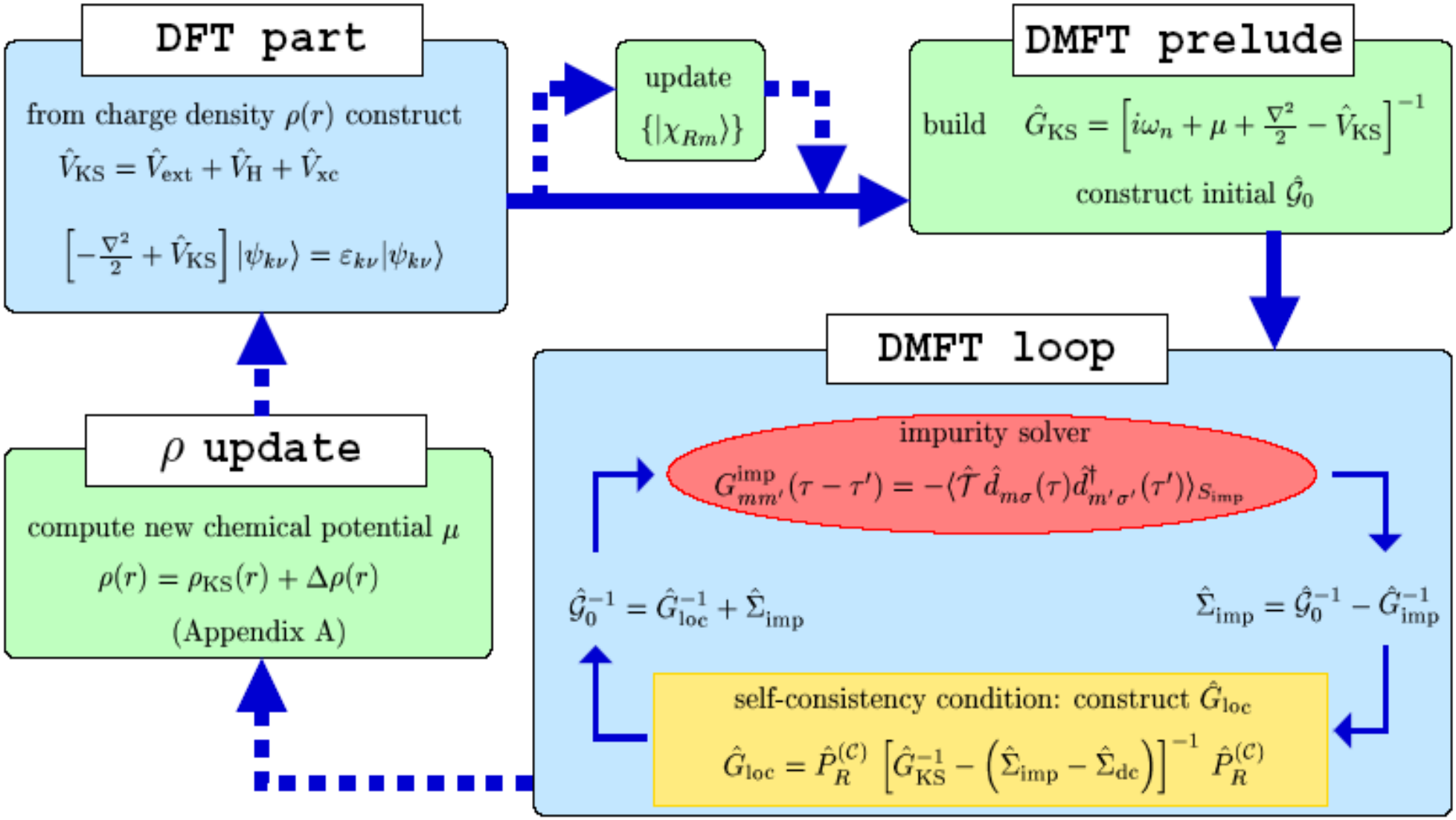


Compute

Understand

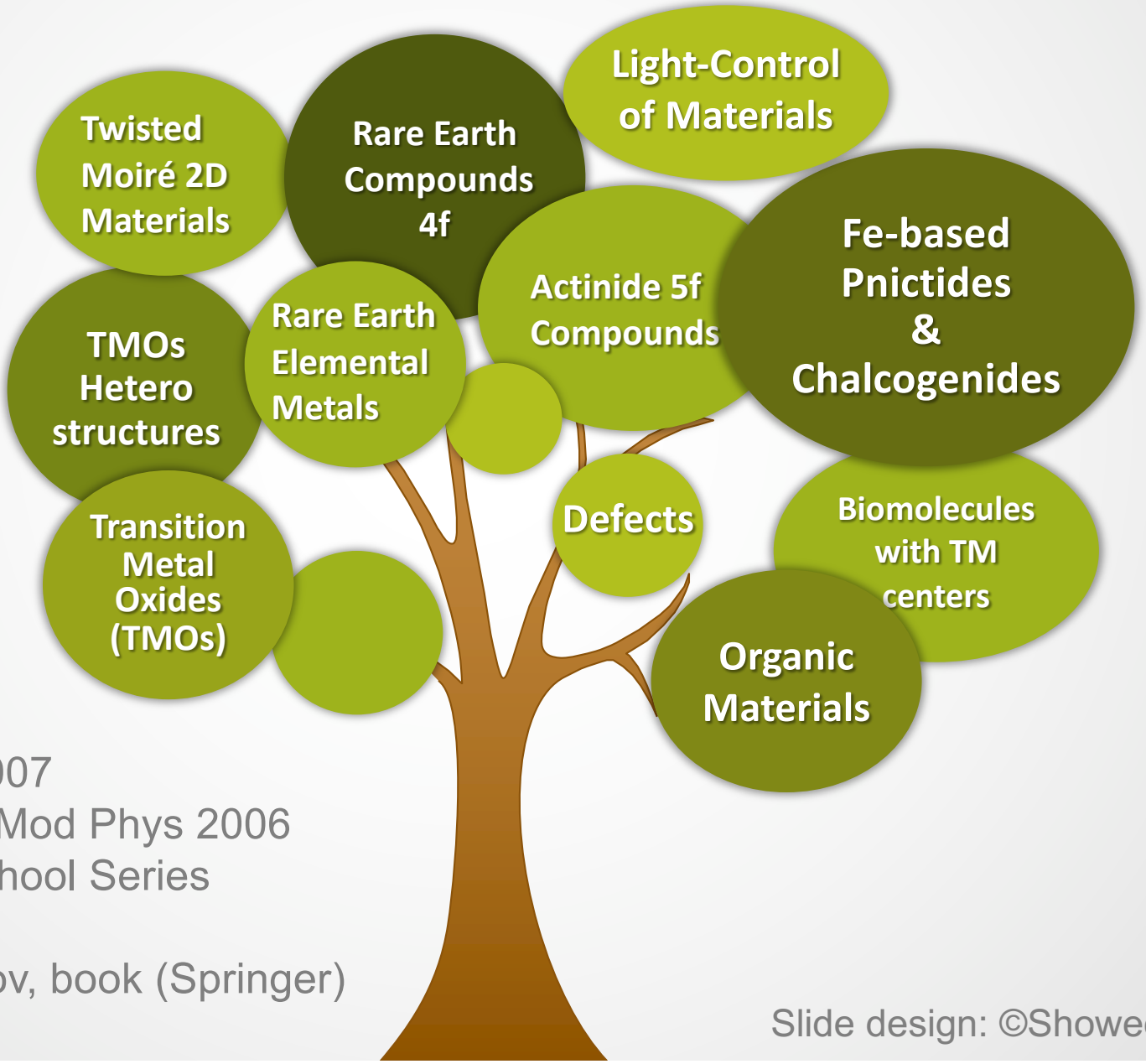


Realistic DMFT, in a nutshell…



Total Energy Functional: $E\left[\rho(r), G^{loc}_{mm'}(\omega)\right]$

Electronic Structure with DMFT: A Multitude of Materials



Reviews:

-Held Adv Phys 2007

-Kotliar et al. Rev Mod Phys 2006

-Jülich Autumn School Series (Pavarini et al.)

-Anisimov&Izyumov, book (Springer)

Etc.

Self-Energy: The DMFT *ansatz*

For a multi-band/multi-orbital material

$|\chi_m^{\mathbf{k}}\rangle$: A set of localized orbitals with many-body interactions $U_{m1m2m3m4}$ are added: correlated Hilbert space

$|\psi_\nu^{\mathbf{k}}\rangle$: The (usually larger) set of Bloch bands (e.g. Kohn-Sham states) describing the material (larger Hilbert space)

$$\Sigma_{\nu\nu'}(\omega,\mathbf{k}) = \sum_{mm'} \langle\psi_\nu^{\mathbf{k}}|\chi_m^{\mathbf{k}}\rangle\, \Sigma_{mm'}(\omega)\, \langle\chi_{m'}^{\mathbf{k}}|\psi_{\nu'}^{\mathbf{k}}\rangle$$

Self-energy `upfolded' to the whole system (k-dependent)

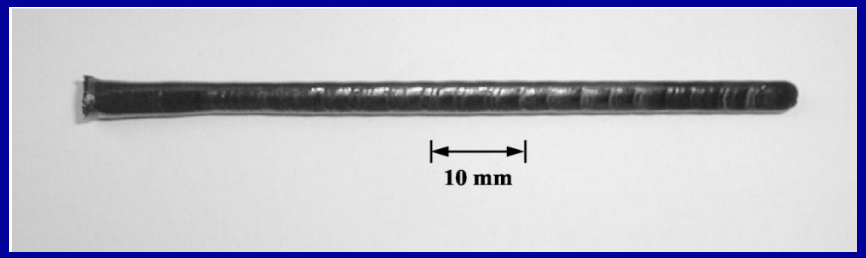
Orbital content of Bloch states (k-dep)

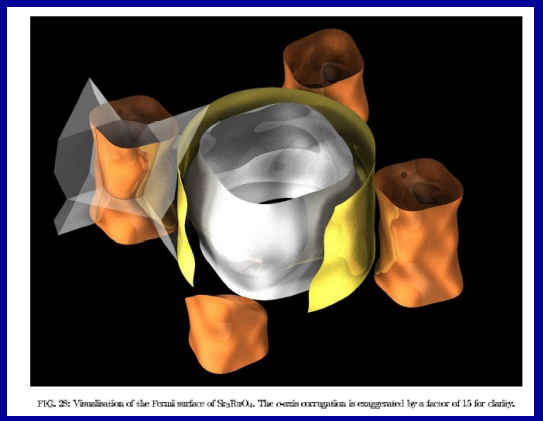
Local self-energy `upfolded' to the whole system (k-dependent)

Putting this directly to the test from Hi-Res ARPES: Sr_2RuO_4

A.Tamai et al. Phys Rev X 9, 021048 (2019)

The `fruit-fly' of Transition-Metal Oxides!

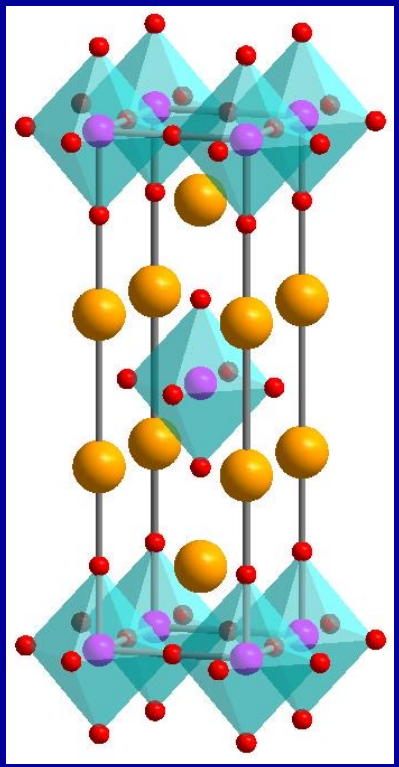




Large clean single-crystals

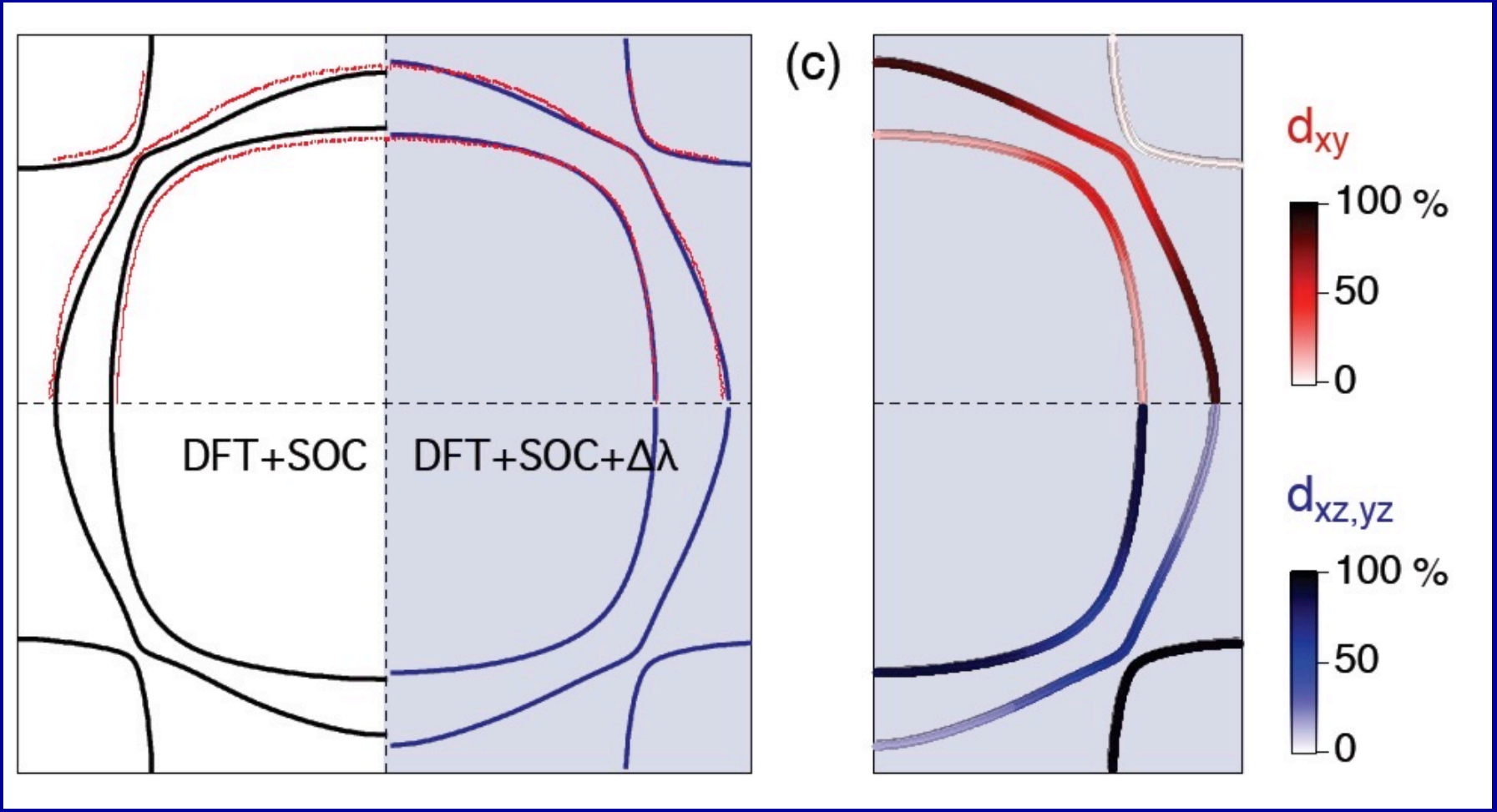
→ Investigated with basically <u>all techniques</u> in the experimentalist's toolbox

A.Mackenzie, Y.Maeno Rev Mod Phys 75, 657 (2003)



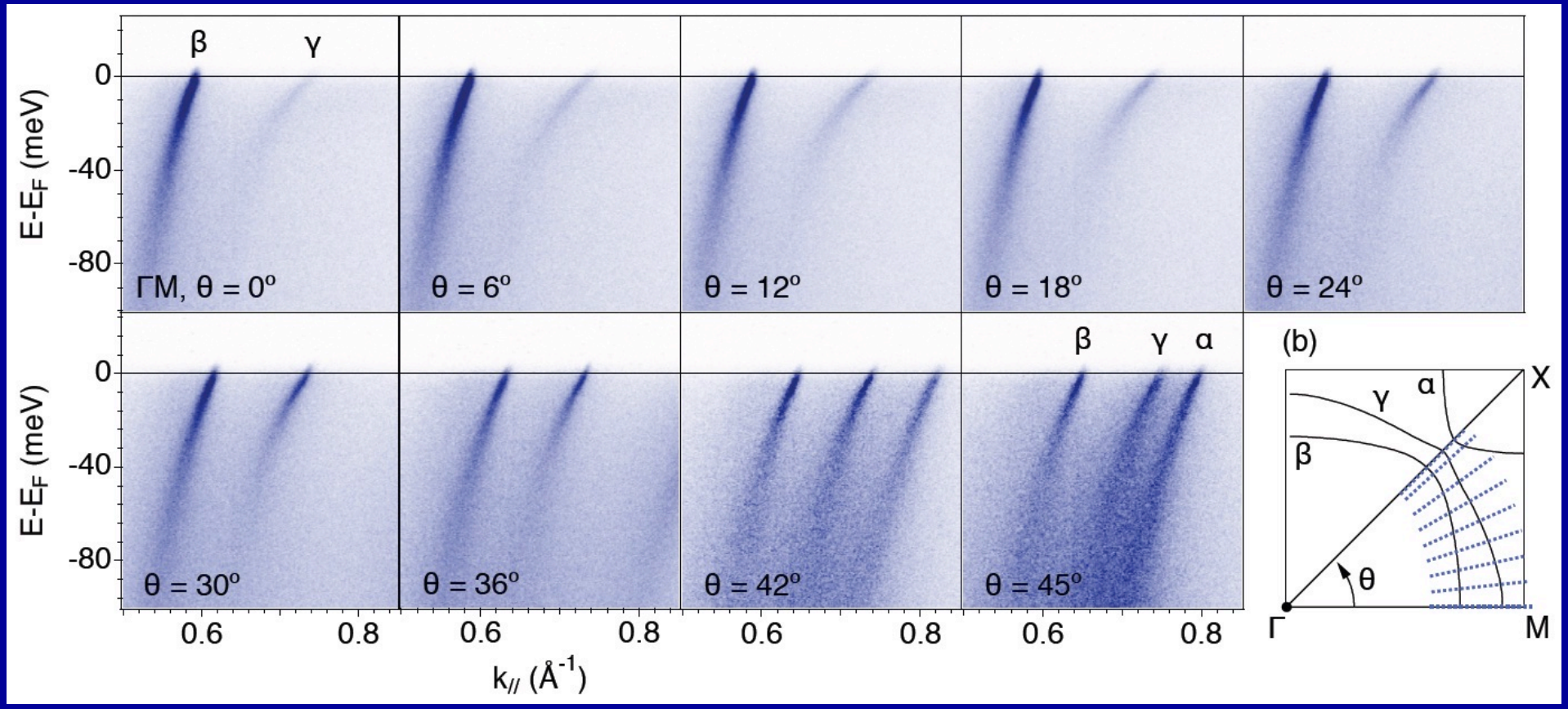
Simple Structure

Orbital Content of Quasiparticle States is strongly angular dependent due to spin-orbit



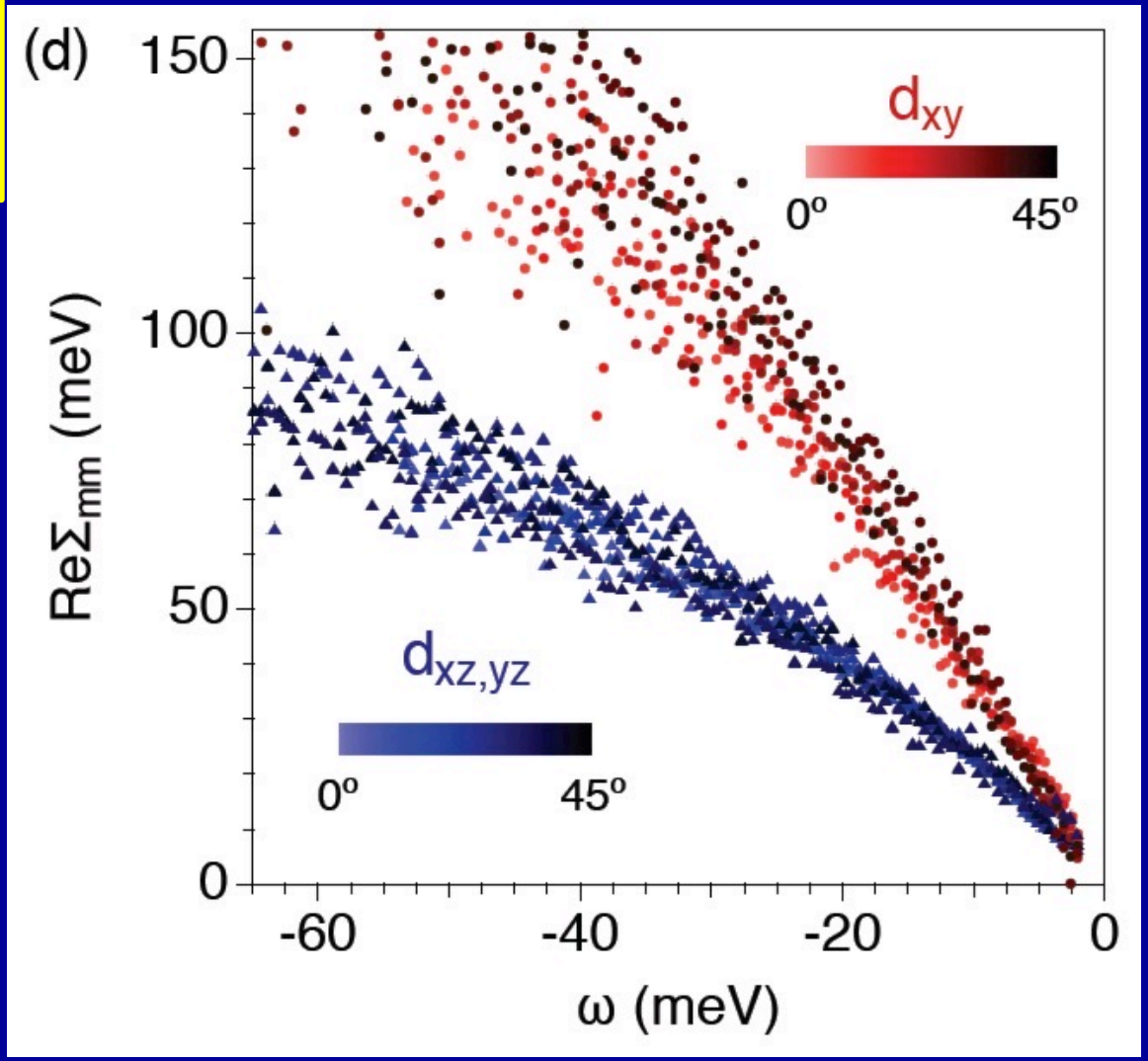
DMFT prediction (Pavarini et al; Kim et al.): Effective enhancement $\Delta\lambda$ of SOC

From ARPES MDC data: Extract self-energy in orbital basis for each angle θ

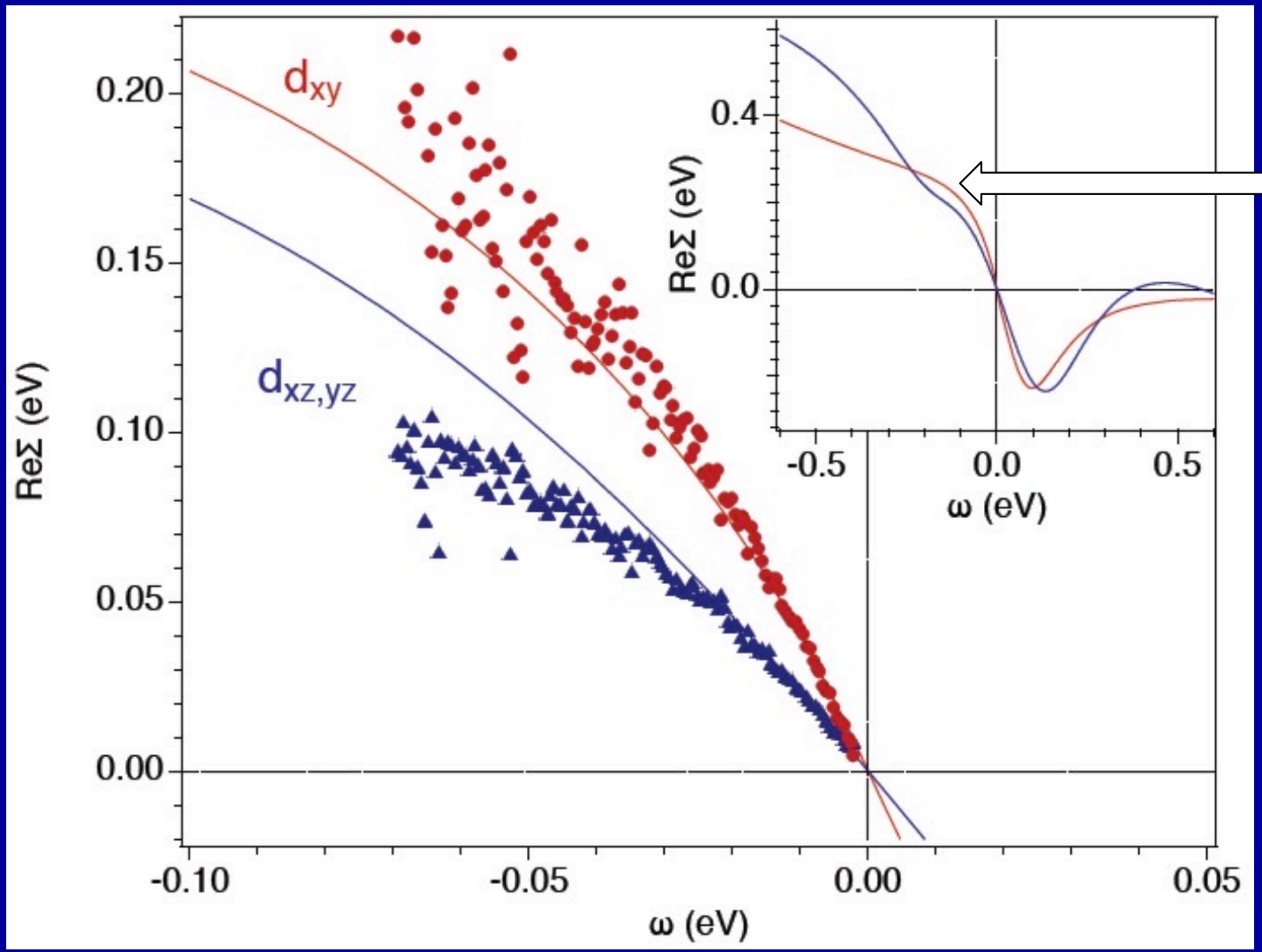


~ Collapse of data corresponding to different angles !

→ DMFT `Locality *ansatz*' is a good approximation



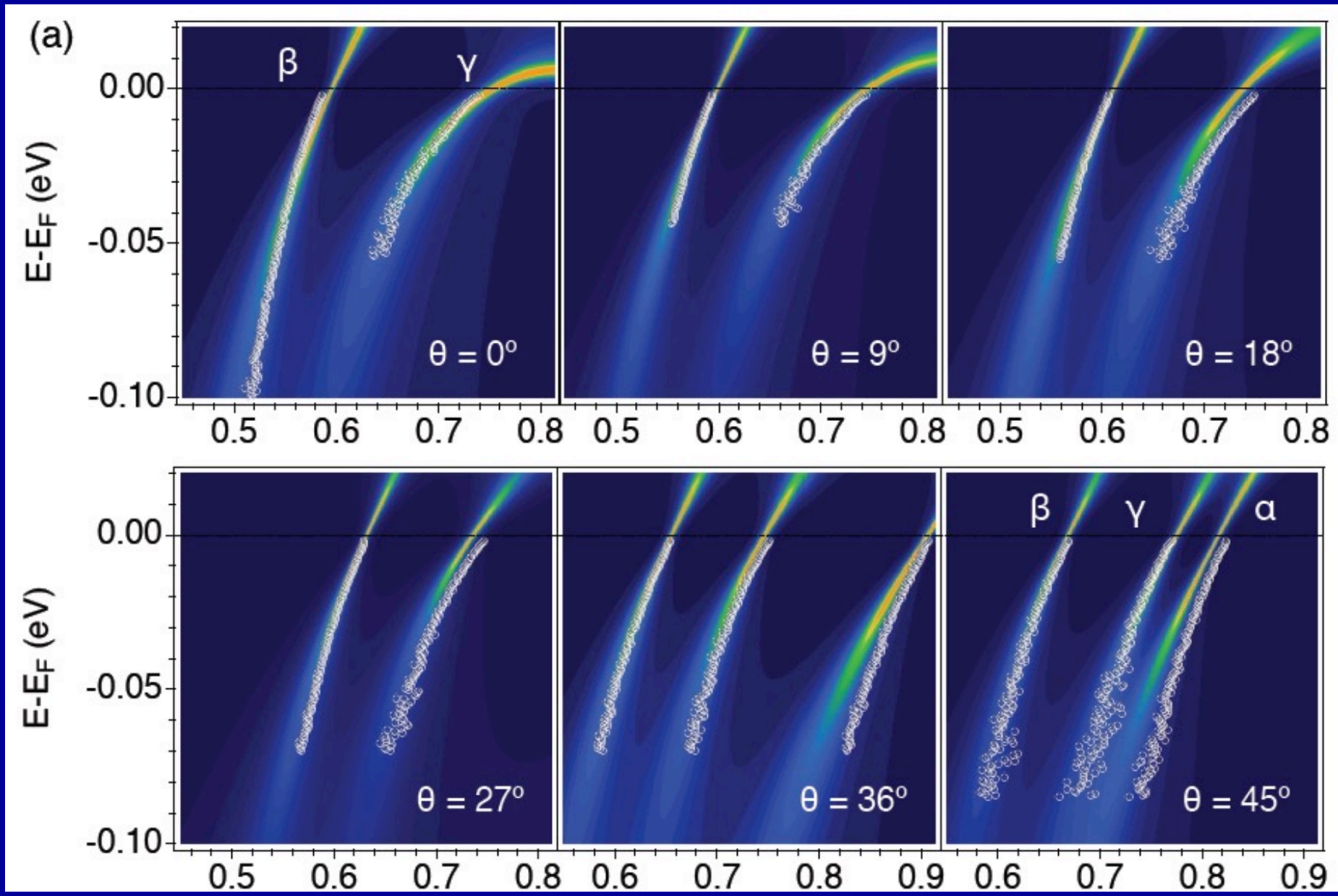
Comparison to LDA+DMFT self-energies



Kink (<u>electronic</u> origin at ~ 100meV)

Comparing DMFT to ARPES

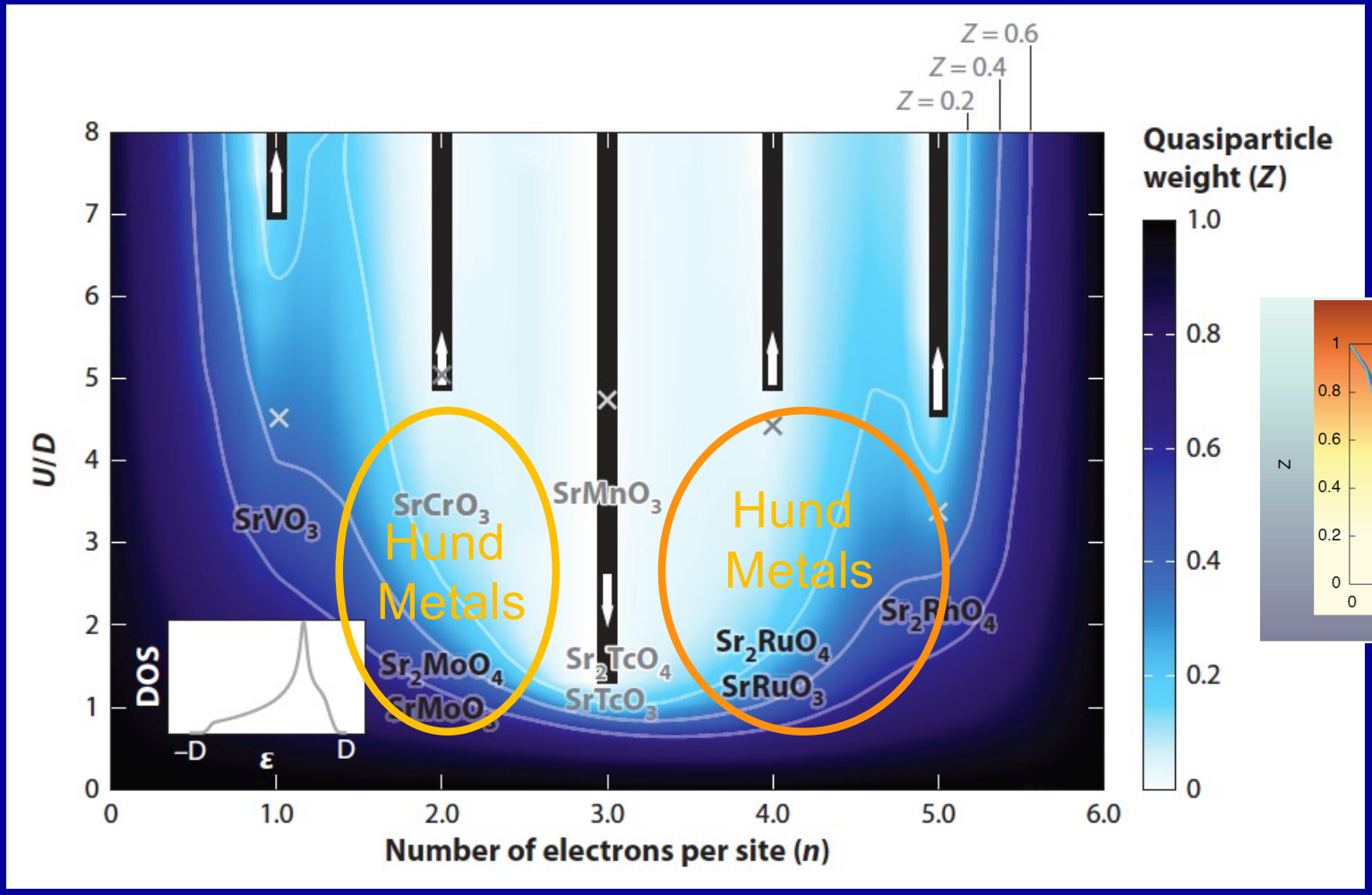
(Dots: ARPES MDCs. Colors: DMFT spectral intensity)

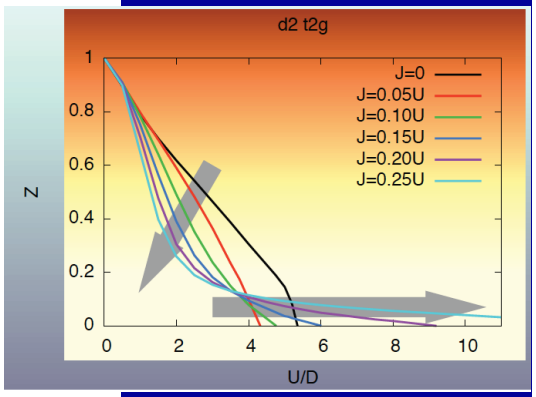


Sr_2RuO_4 is a member of the big and happy family of `Hund Metals'

- Iron-Based Superconductors → Gabi's talk
- Oxides of 4d Transition Metals
- In the case of Sr2RuO4, proximity to van Hove singularity also plays an important role, cf. comparison to Sr_2MoO_4 Karp et al. 125, 166401 (2020)
- Hund Metals: Haule and Kotliar New J. Phys. 11, 025021 (2009); Werner, Gull, Troyer and Millis, PRL 101, 166405 (2008); Mravlje et al. PRL106, 096401 (2011); Yin, Haule and Kotliar Nat Mat 10, 932 (2011); de'Medici et al. PRL 107, 256401 (2011); AG, de'Medici and Mravlje, Ann Rev Cond. Mat. Phys Vol 4 (2013), and many more…

4d Transition-Metal Oxides:
Strong Correlations far from the Mott Transition





3d oxides: U/D ~ 4; 4d oxides: U/D ~ 2 ; D: ½ bandwidth

Hallmark of Hundness: Coherence of Spin and Orbital Degrees of Freedom Occurs at Distinct Scales

~ 1000K

Fermi Liquid

Metal with fluctuating spins
No orbital fluctuations

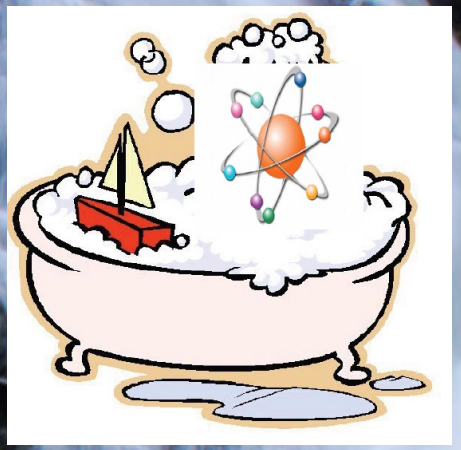
Incoherent

Now beautifully understood from a Renormalization Group perspective, cf. recent work by von Delft, Lee, Weichselbaum et al., Aron, Kotliar et al., Horvat, Žitko, Mravlje, Kugler et al.,

→ See Gabi's talk

High energy
High temperature
Short time scales
Short distances
Large lattice spacing
LOCAL
INCOHERENT

Atomic configurations/Multiplets
Intra-shell interactions+crystal fields



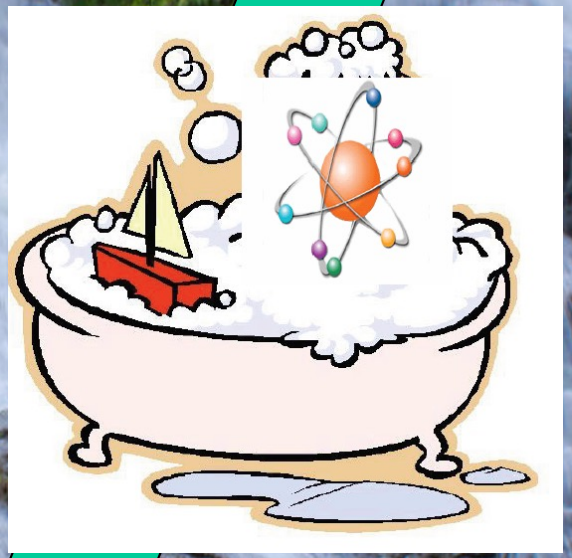
Environment Lifts degeneracies…

Low energy
Low temperature
Long time scales
Long distances
Small lattice spacing
NON-LOCAL
COHERENT

Collective ground-state
Low-energy excitations
Effective low-energy theory

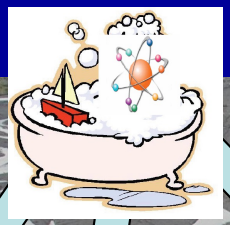
This is very much how we think about materials
In DMFT:
Start from local atomic configurations
and follow the flow down into collective behaviour
Initially, spatial correlations are short-range
At lower energy, spatial correlations build up
→ Need to go beyond single-site DMFT

Atomic configurations/Multiplets
Intra-shell interactions+crystal fields



?

Including Spatial Fluctuations: Beyond Single-Site DMFT



EDMFT,
GW+DMFT,
Generalized
Embedding,…

Including
Long-wavelength
fluctuations w/ vertex
DΓA, TRILEX,
Dual Fermions/Bosons,…

Cluster
Extensions
of DMFT:
CDMFT, DCA,…

Embedding Methods Are Controlled

Cluster extensions of single-site DMFT

→ `Molecular' mean-field

(cf. Bethe-Peierls, Kikuchi)

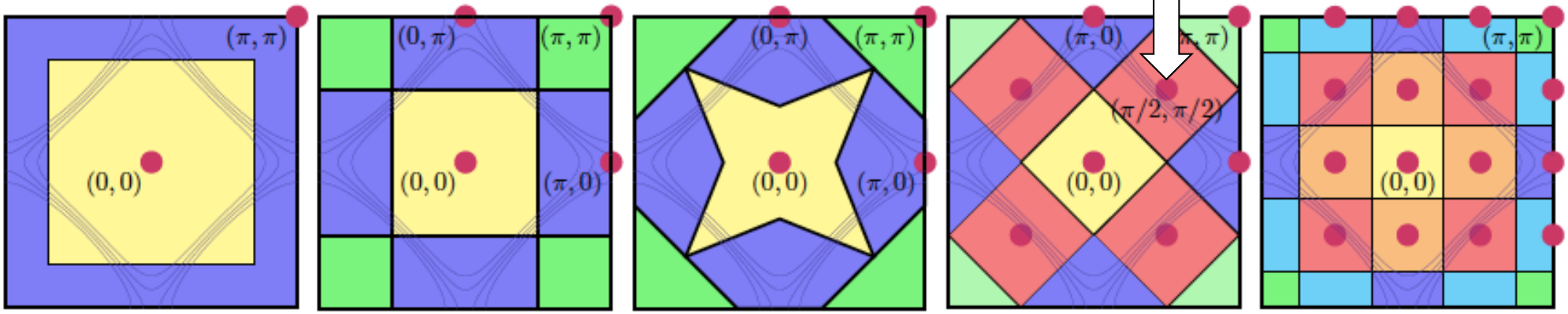
Several flavors, e.g. DCA: Patching momentum-space, cluster used to calculate self-energy at cluster momenta.

Self-energy approximated as piecewise constant in momentum space:

$$\Sigma(k,\omega) \simeq \Sigma(K,\omega) \quad (k \in P_K)$$

Antinode

Node



Numerous works by several groups in the last ~ 20 years

For reviews see:

[27] T. Maier, M. Jarrell, T. Pruschke, and M. H. Hettler, Rev. Mod. Phys. 77, 1027 (2005).

[28] G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianett, Rev. Mod. Phys. 78, 865 (2006).

[29] A. M. S. Tremblay, B. Kyung, and D. Senechal, Low Temp. Phys. 32, 424 (2006).

Cincinatti/Baton Rouge (Jarrell et al.), Rutgers (Kotliar, Haule et al.), Sherbrooke (Tremblay, Senechal et al., Kyung, Sordi), Columbia (Millis et al.)., Michigan (Gull et al.) Oak Ridge (Maier et al.), Tokyo (Imada, Sakai et al.) Hamburg(Lichtenstein et al.), Rome (Capone et al.) Paris/Saclay/Orsay (Parcollet, Ferrero, AG, Civelli et al.), Stuttgart (Gunnarsson) etc…

To quote only one achievement:

These approaches have established that the Pseudogap in the doped 2D Hubbard model is caused by spin correlations

(not pair or CDW fluctuations)

Many groups and authors 2005 → 2020

See e.g. PRX 8, 021048 for references

Recent `handshake' with Tensor Network Methods:

Wietek et al. PRX 11, 031007 (2021)

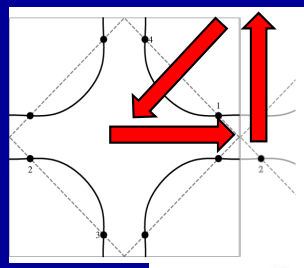
Controlled results, converged to infinite cluster size, are possible in part of the PG regime

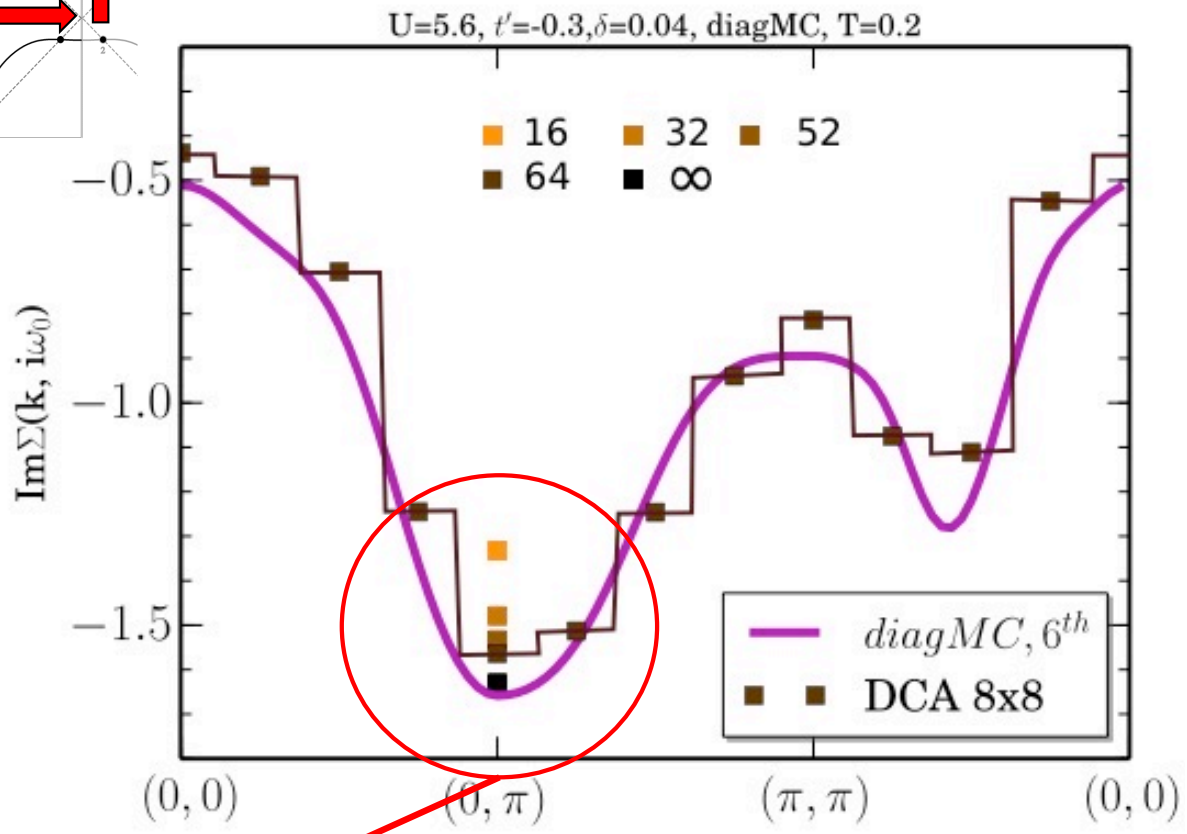
Wei Wu, Ferrero, AG, Kozik PRB 96, 041105R (2017)

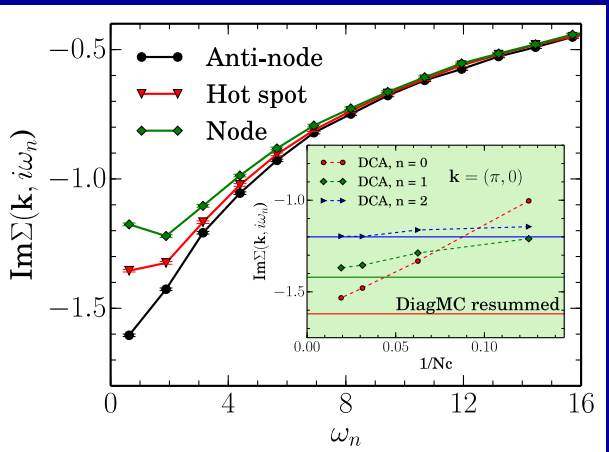
- For U/t=5.6 , t'/t=-0.3 and doping p=0.04 (reference `Wei point' ☺)
- CONVERGE the self energy at T=0.2t with <u>two independent methods</u>:
- DCA w/ convergence in cluster size
- Diagrammatic Monte Carlo on the Infinite Lattice
- Recently significant improvements to the DiagMC method (RDET) have allowed to reach T/t=0.1 Rossi,Simkovic, Ferrero EPL 132 (2020) 11001

DCA and DiagMC: quantitative agreement

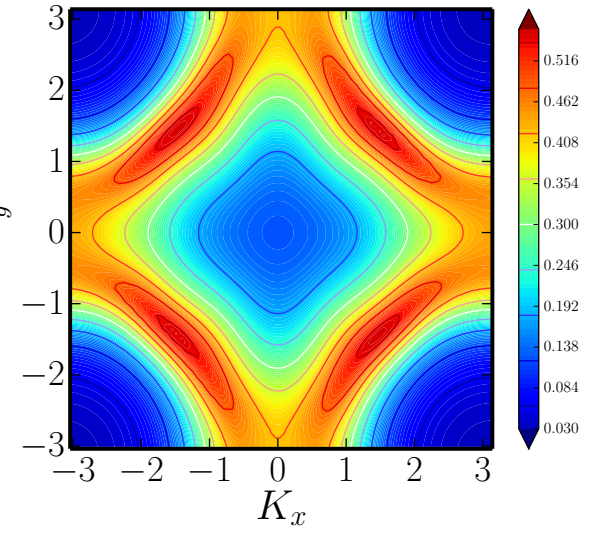
→ Computational solution of the 2D Hubbard model in this regime !







Nodal/Antinodal Dichotomy



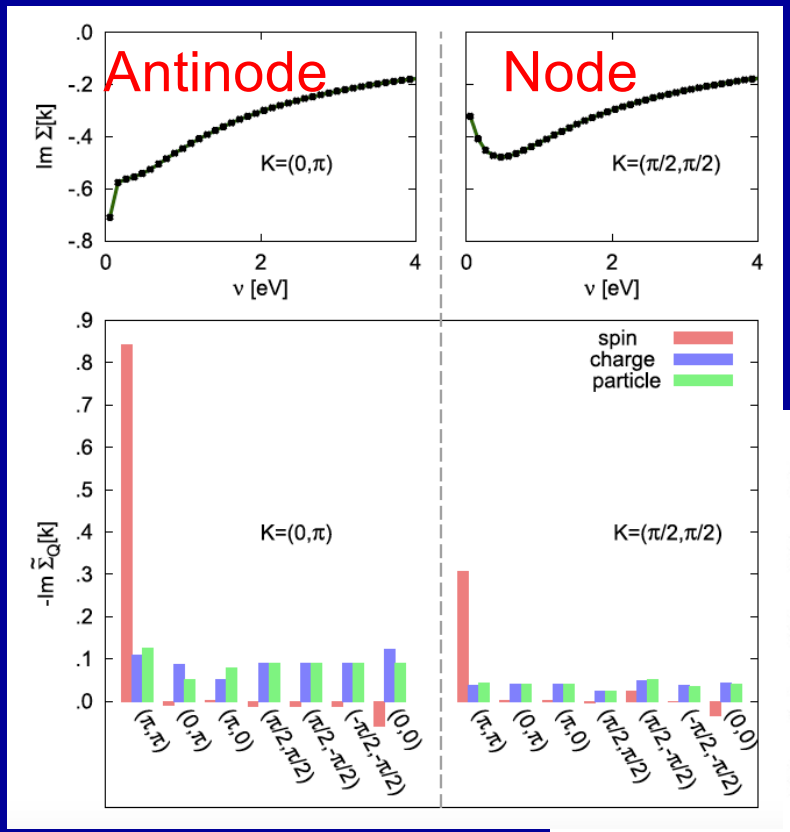
ImΣ becomes LARGE at antinode !

`Fluctuation Diagnostics'

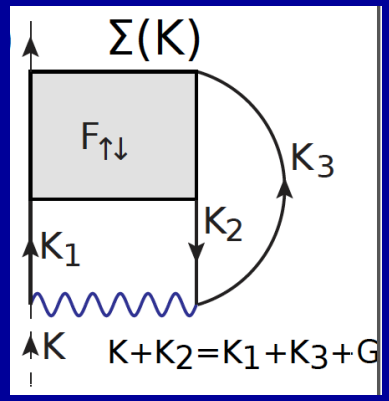
PRL 114, 236402 (2015) PHYSICAL REVIEW LETTERS week ending 12 JUNE 2015

Fluctuation Diagnostics of the Electron Self-Energy: Origin of the Pseudogap Physics

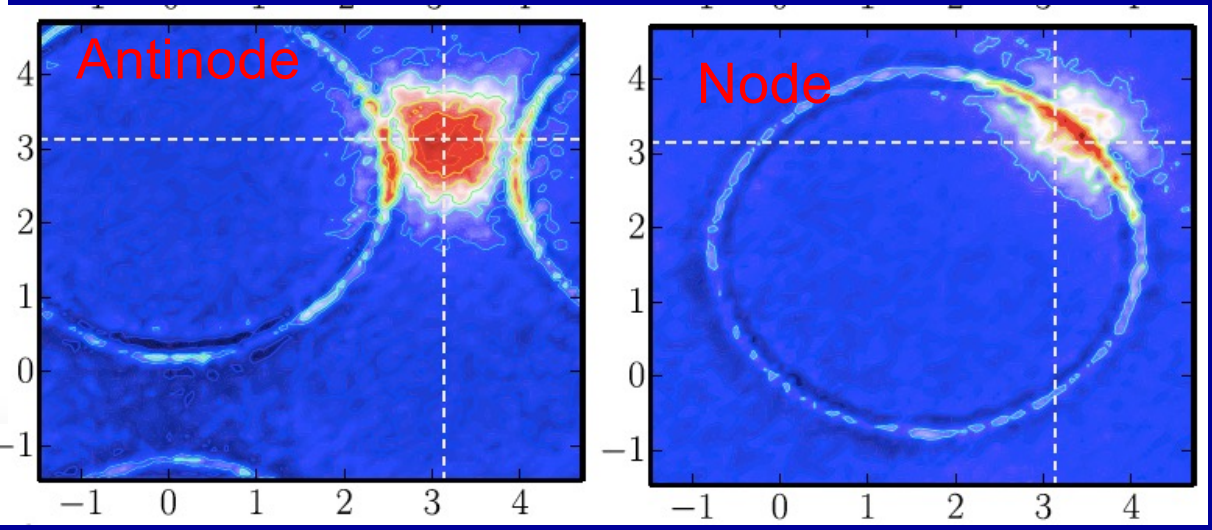
O. Gunnarsson,[1] T. Schäfer,[2] J. P. F. LeBlanc,[3,4] E. Gull,[4] J. Merino,[5] G. Sangiovanni,[6] G. Rohringer,[2] and A. Toschi[2]



DCA Gunnarsson et al



Wei et al. (2017) - DiagMC



Pseudogap: Take-home Message

- The PG is associated with spin correlations (longer range at weak coupling, shorter range at strong coupling)
- All cluster DMFT studies agree on this and a consistent picture emerges, also in agreement with DiagMC and recent METTS
- NOT particle-particle correlations (aka preformed pairs)
- NOT charge sector (e.g. precursor of CDW)
- In the range of parameters investigated in the above study: commensurate (π,π) wavevector
- At lower T, incommensurate SDW correlations set in
- At T=0 and low-doping: stripe ordering is a strong competitor - DMRG, AFQMC and DMFT are in agreement about this
  cf.Peters and Kawakami, PRB 89, 155134 (2014)

Computational Methods: The Handshake Challenge

`Embedding' Methods: DMFT and beyond
Organizing principle: Locality

Handshake: current challenge/frontier !

Wave-function Compression Methods: MPS, Tensor Networks etc.
Organizing principle: Entanglement

Temperature

Coupling

Looking Ahead…

- Looking towards the next big advance on `impurity solvers'. Promising candidates: Inchworm, Real-time (quasi)MC, Fork Tensor Product States, METTS,…
- Long-range interactions and spatial correlations: GW+DMFT, Making vertex-based extensions more efficient, Combinations with lattice DiagMC,…
- Designing new embedding schemes: `full-cell' embedding, SEET,…

A heartfelt ‘THANK YOU!’ to collaborators and friends over the years, and especially to:

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