COLLÈGE DE FRANCE
1530

UNIVERSITÉ GRENOBLE ALPES

Chaire de Physique de la Matière Condensée

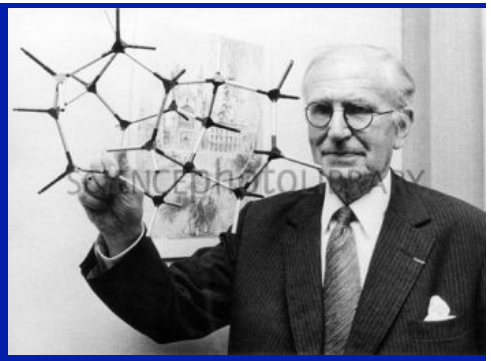
Matériaux et dispositifs à fortes corrélations électroniques

I.*3 Transition de Mott et théorie de champ moyen dynamique*

Antoine Georges

Cycle 2014-2015
4 mai 2015 – I.3

Mott Insulators



- Some materials (e.g. MANY oxides) are insulating despite having a PARTIALLY FILLED shell (even when number of electrons in the shell is ODD, e.g. d^1)
- Insulating behavior is observed well above magnetic ordering temperature, and (from the size of the gap) may involve a much larger energy scale
- Band-structure calculation in the non-magnetic state yields a metal

The Mott phenomenon at strong coupling (U >> t) HAS NOTHING TO DO with magnetism It is due to blocking of density/charge

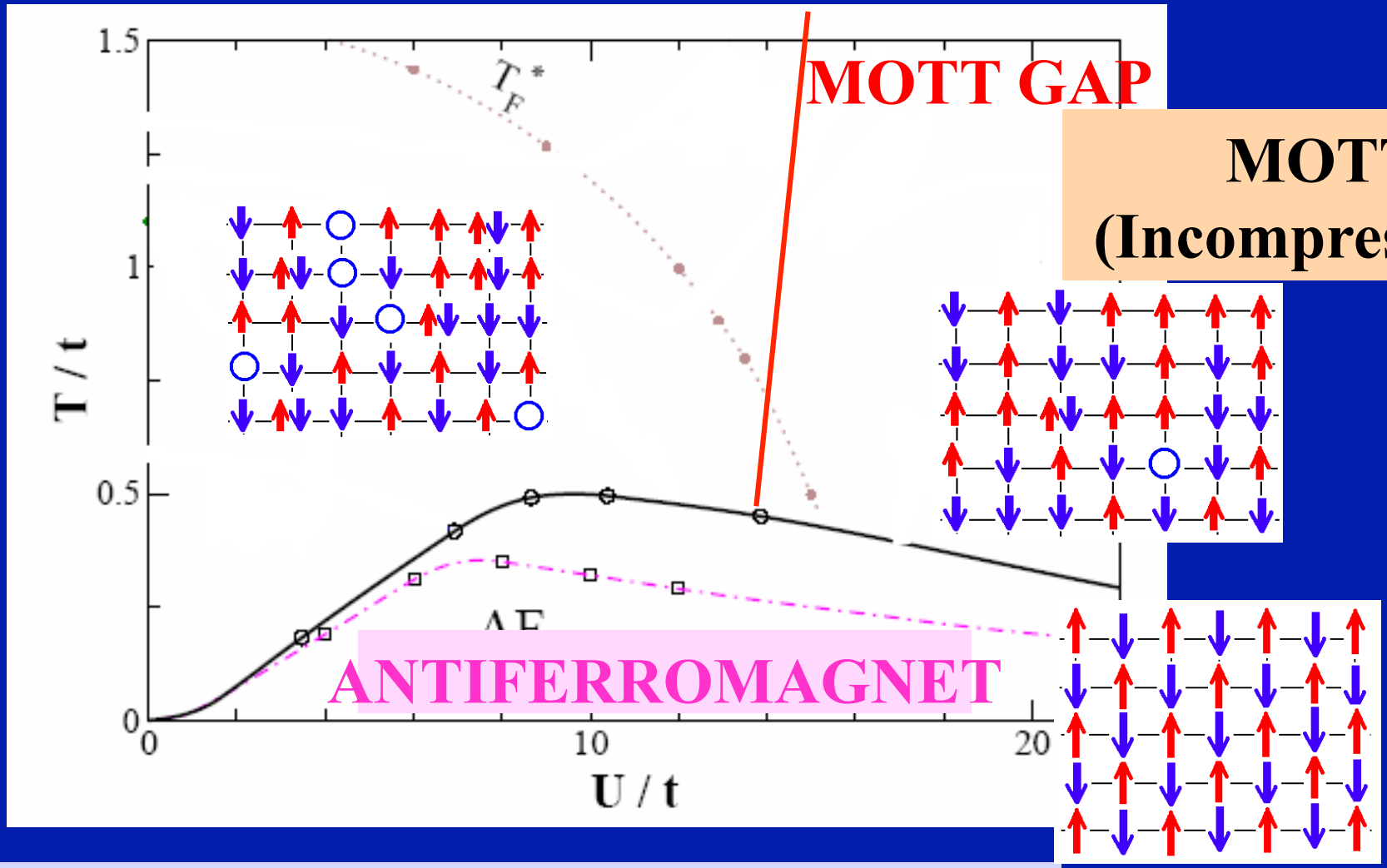
Energy scale for magnetism: superexchange $J \sim t^2/U$

Insulating gap: $\sim U > t >> J$

The system is basically an insulator even well above T_{Neel}

Ex: MANY oxides, e.g. NiO, YTiO3, cuprates etc…

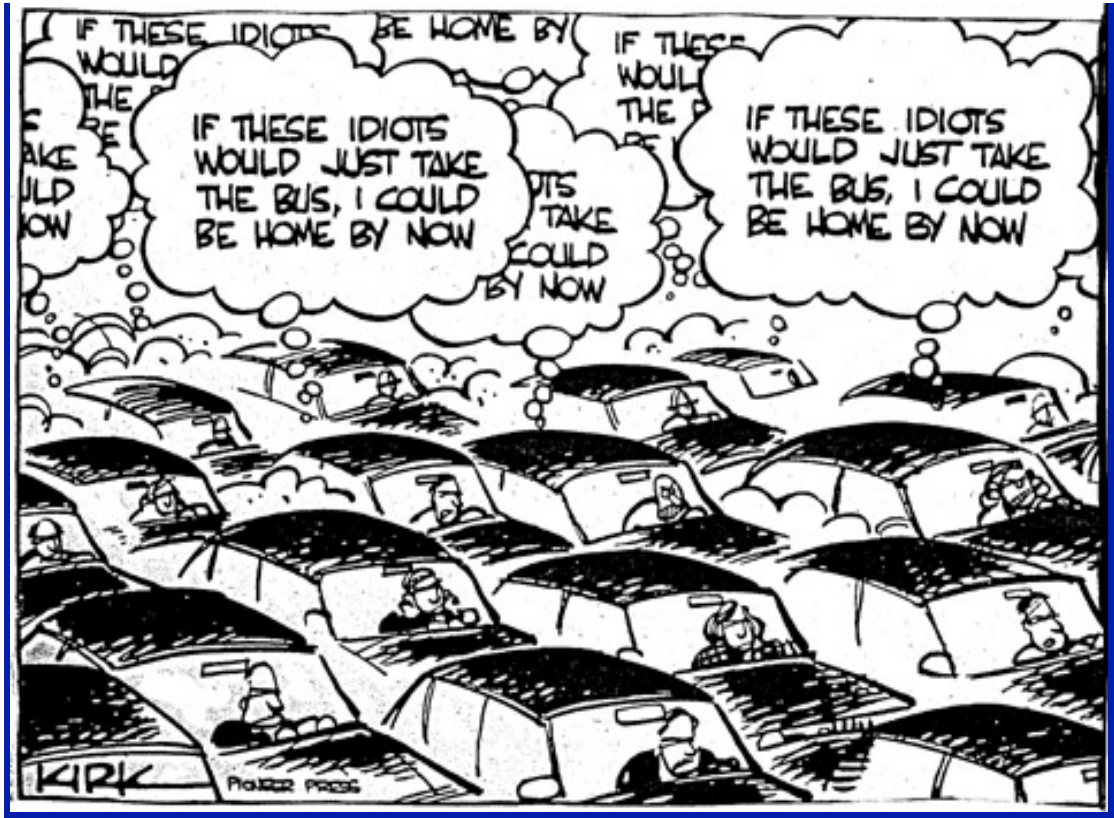
In contrast, LDA+U needs to assume ordering to describe the insulator



Critical boundary calculated for a 3D cubic lattice using:

-Quantum Monte Carlo (Staudt et al. Eur. Phys. J. B17 (2000) 411)

- Dynamical Mean-Field Theory approximation



A Mott insulator is an incompressible state of matter

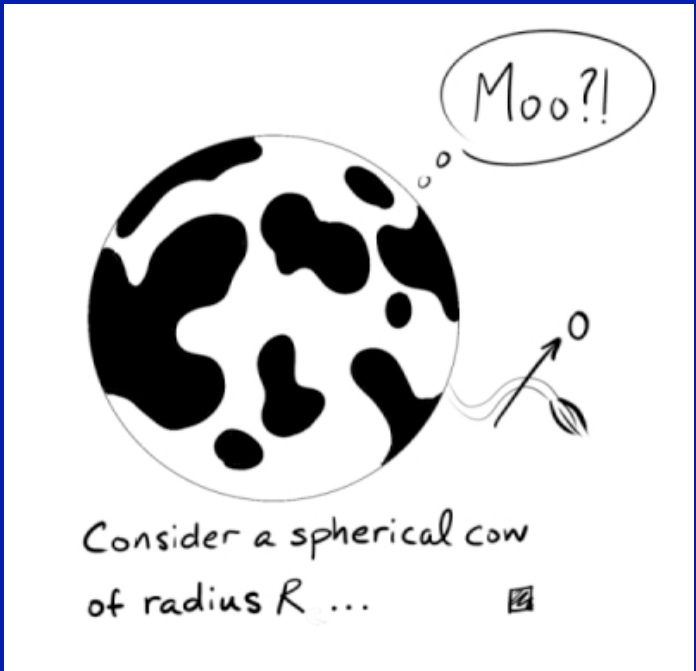
LA's highways

The RER-subway in Paris at rush hours

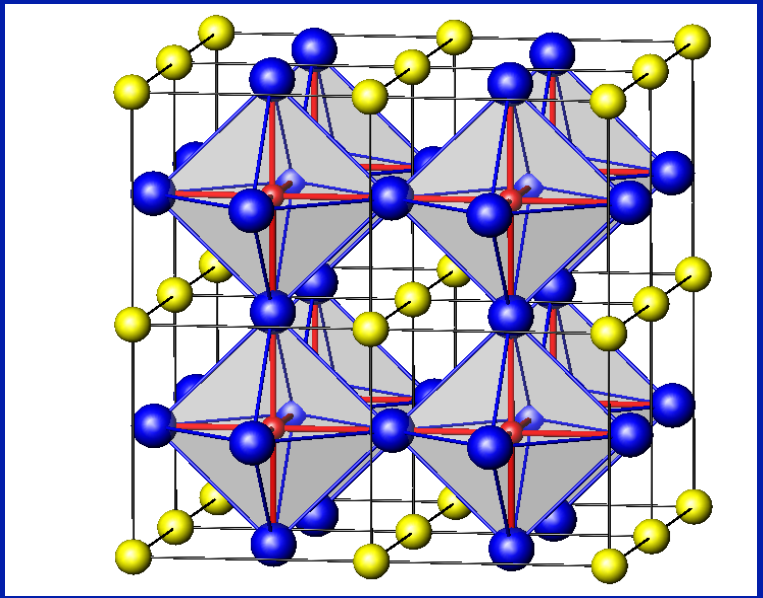
In fact, a Mott insulator is the simplest possible kind of insulator !

"A child of five would understand this.
Send someone to fetch a child of five."
—Groucho Marx

From spherical cow models…



… to real materials

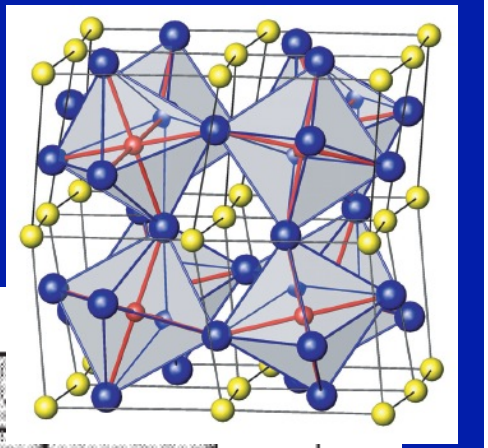


$SrVO_3$

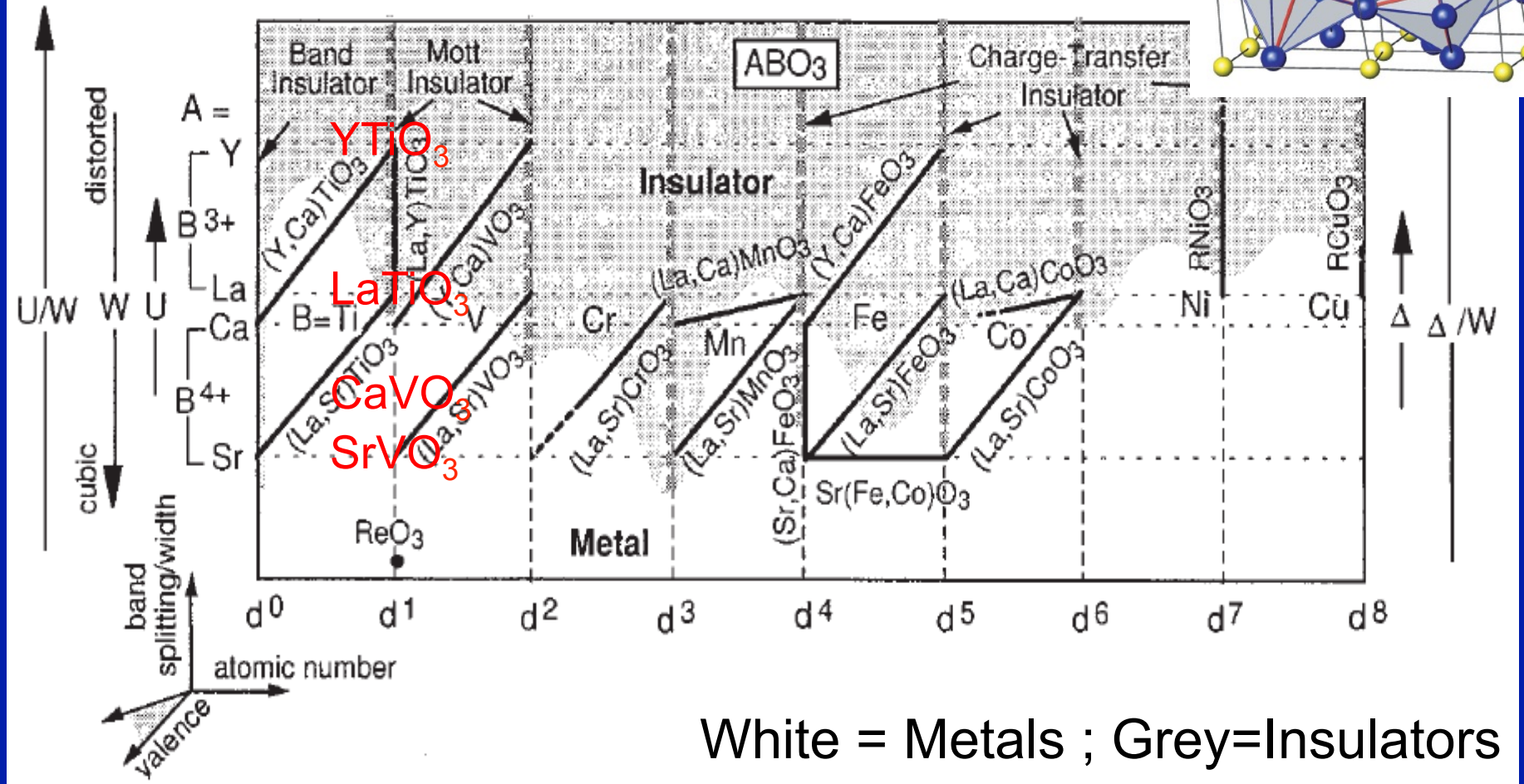
``Atsushi Fujimori's map of ABO_3 perovskites''

J.Phys Chem Sol. 53 (1992) 1595

Imada, Fujimori, Tokura, Rev.Mod.Phys (1998)



A=rare-earth site (yellow) B=metal site (red)

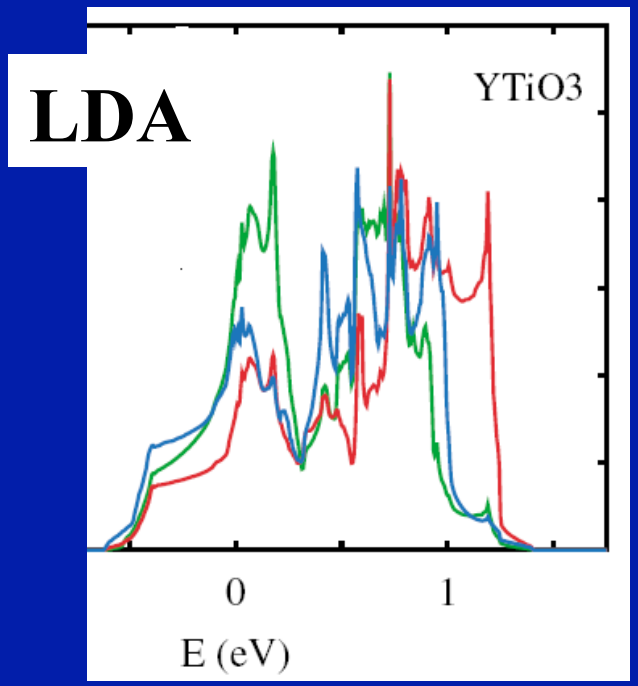


White = Metals ; Grey=Insulators

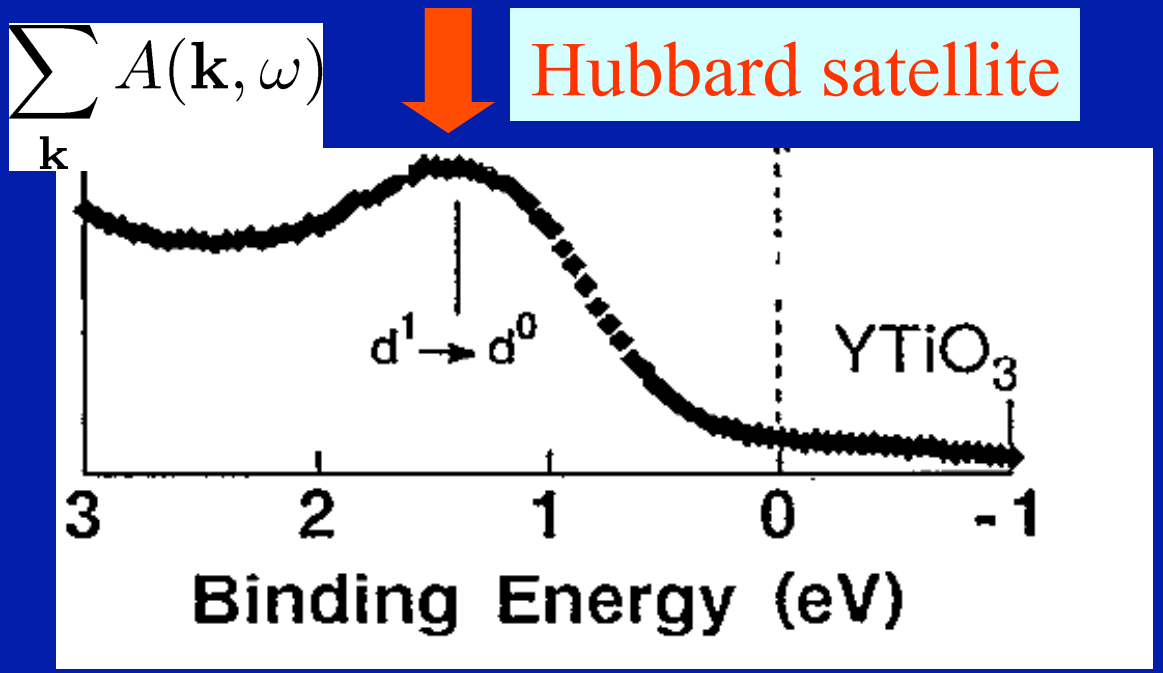
Mott insulators :

Their excitation spectra contain atomic-like excitations

Band structure calculations (interpreting Kohn-Sham spectra as excitations) is in serious trouble for correlated materials !



Metallic LDA (KS) spectrum !

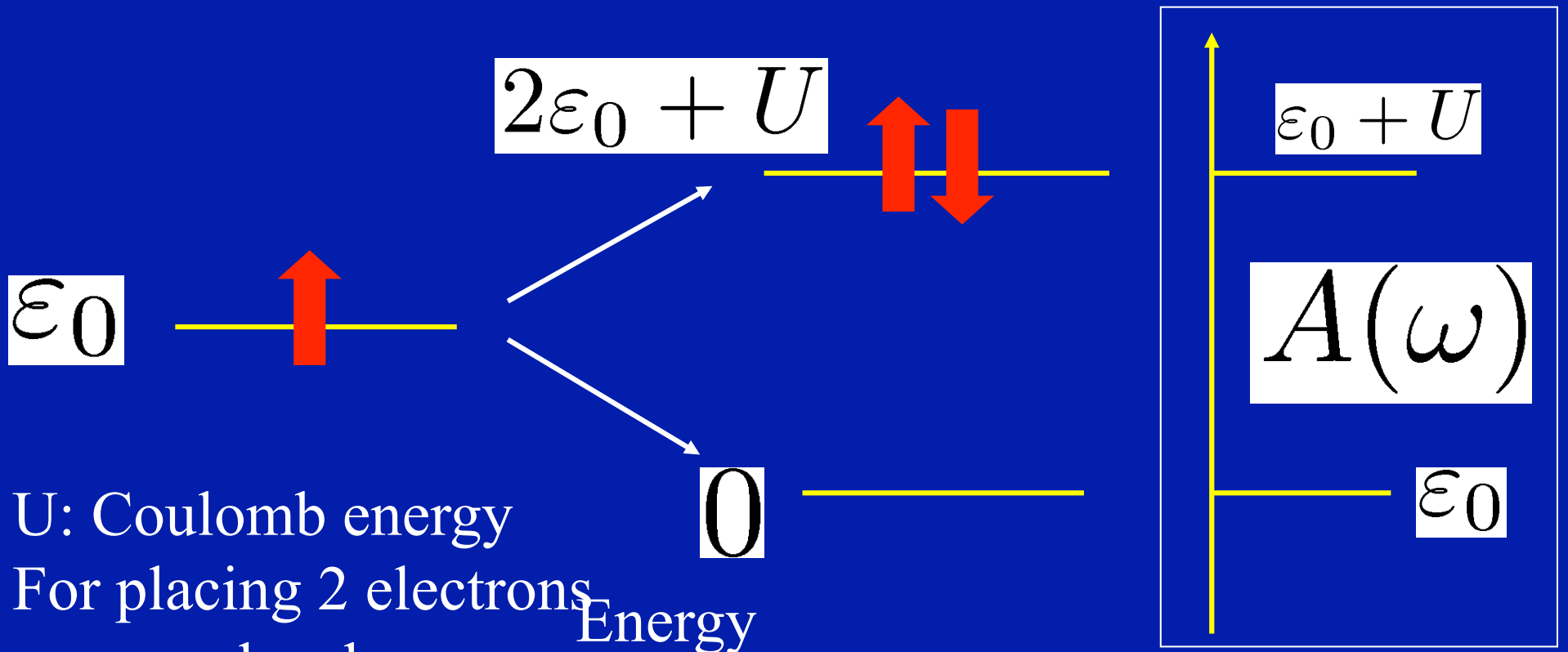


Photoemission: Fujimori et al., PRL 1992

A ``Hubbard satellite'' is nothing but an *atomic transition*

(broadened by the solid-state environment)

Imagine a simplified atom with a single atomic level



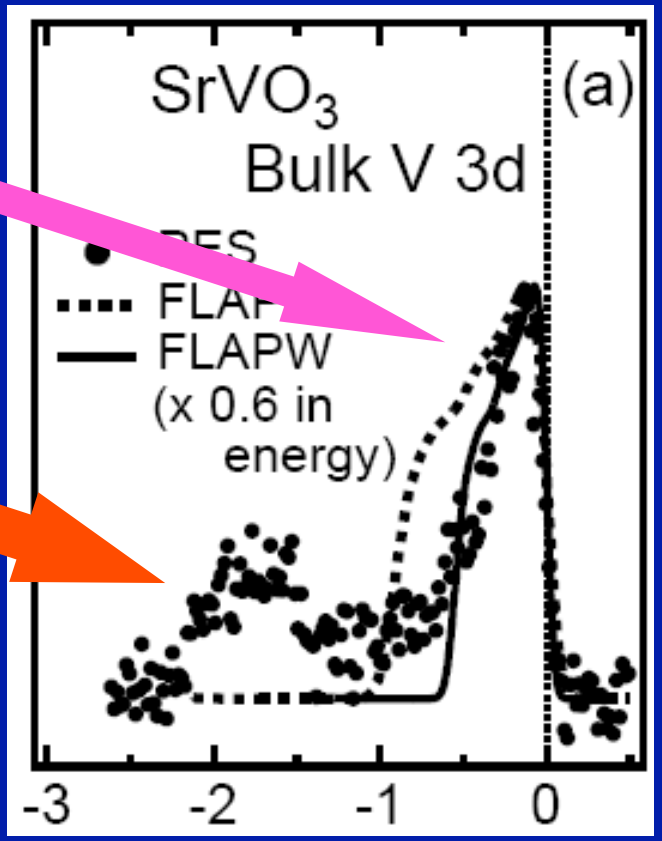
U: Coulomb energy
For placing 2 electrons on same level

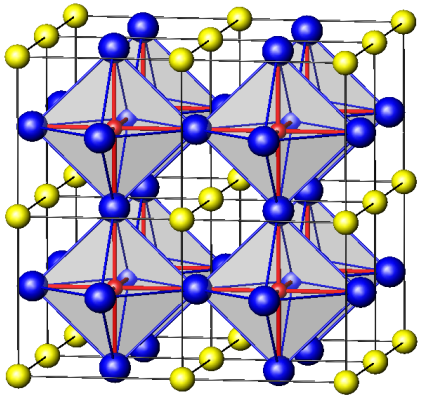
Note: Energetics of the Mott gap requires an accurate description of the many-body eigenstates of single atoms (`multiplets')

→Hund's coupling crucial, see later

Correlated metals: atomic-like excitations at high energy, quasiparticles at low energy

- **Narrowing of quasiparticle bands** due to correlations (the Brinkman-Rice phenomenon)
- **Hubbard satellites** (i.e extension to the solid of atomic-like transitions)





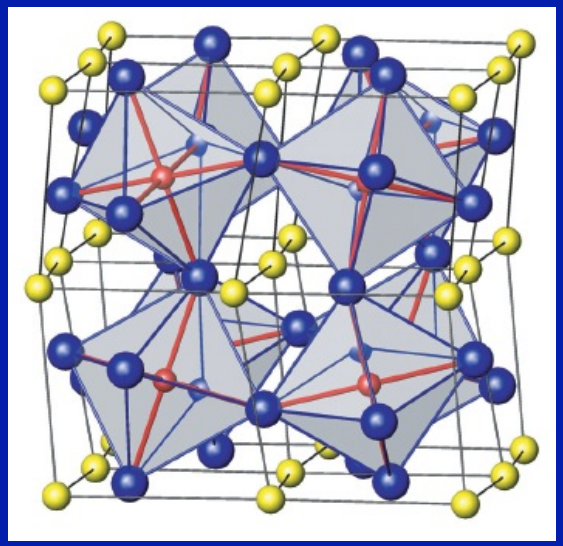
$SrVO_3$

Dashed line:
Spectrum obtained from Conventional band-structure methods (DFT-LDA)

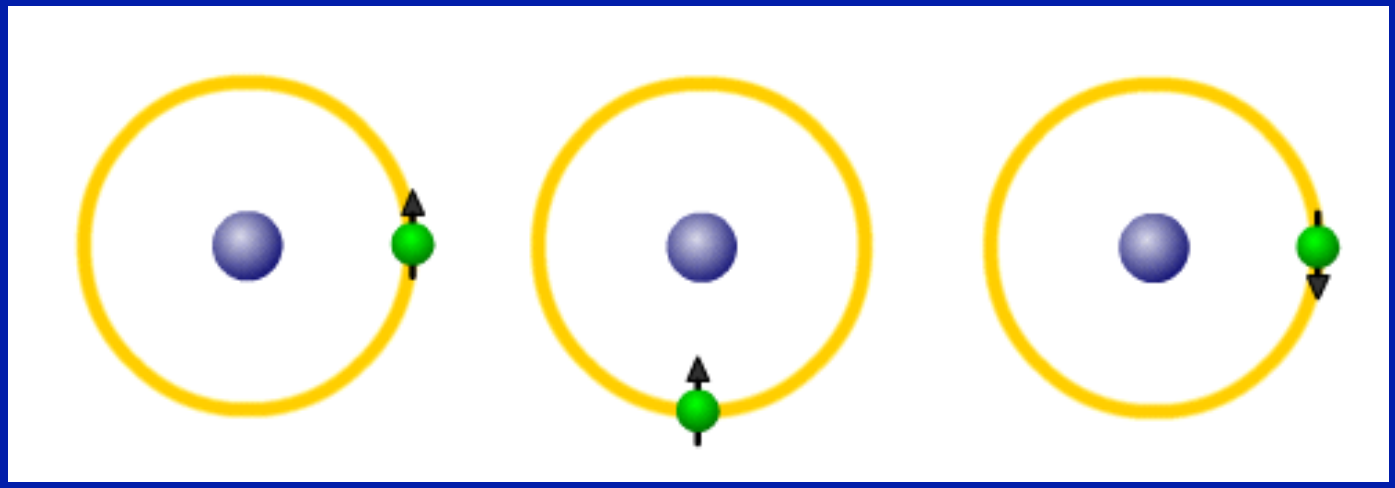
Sekiyama et al., PRL 2004

From weak to strong correlations in d^1 oxides [Fujimori et al. PRL 69, 1796 (1992)]

Puzzle:
Why is $SrVO_3$ a metal and $LaTiO_3$, $YTiO_3$ Mott insulators ?



Electrons “hesitate”
between being localized
on short-time-scales
and itinerant on long time-scales



We see this from spectroscopy…

In materials with strong correlations
LOCAL ATOMIC PHYSICS
is crucial

The `standard model' (according to most textbooks) :

a solid is a kind of gas of ~ independent electrons subject to the periodic potential of ions

→ Bloch wavefunctions, energy bands

Interactions are important for sure but treated « on average »

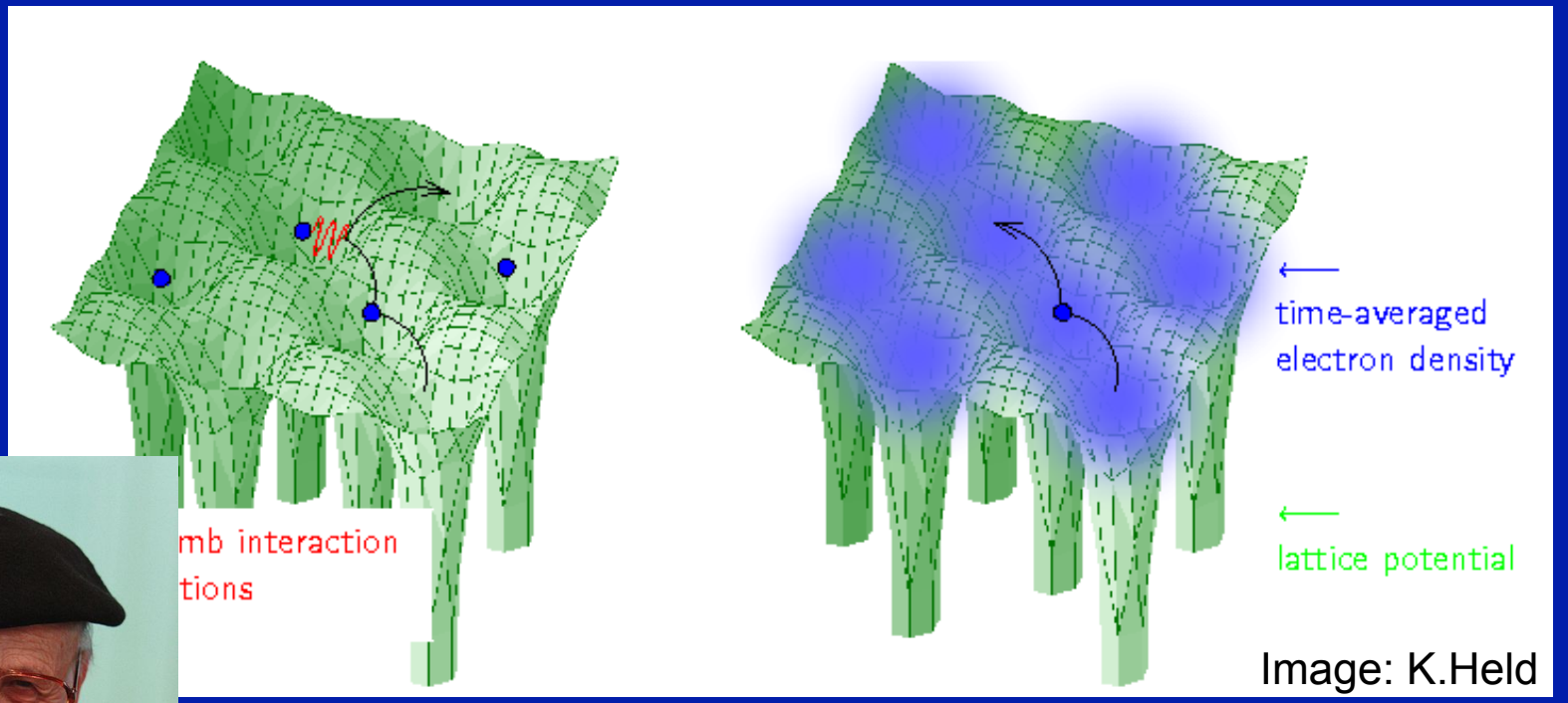


Image: K.Held

Modern (and most useful) incarnation: DFT-LDA

$$E[\rho(r)] = E_{kin}[\rho(r)] + E_{Hartree}[\rho(r)] + E_{xc}[\rho(r)]$$

We need to change our theoretical description
and computational tools
in order to deal with these
« strongly-correlated electron materials »

- Think in terms of atoms, not in terms of an electron gas ! [closer to a chemist point of view]
- Each atom is an interacting (many-body) problem
- Atomic orbitals overlap but motion of electrons is opposed by energy cost for changing the valence of each atom

A theoretical description of the solid-state based on ATOMS rather than on an electron-gas picture: « ***Dynamical Mean-Field Theory*** »

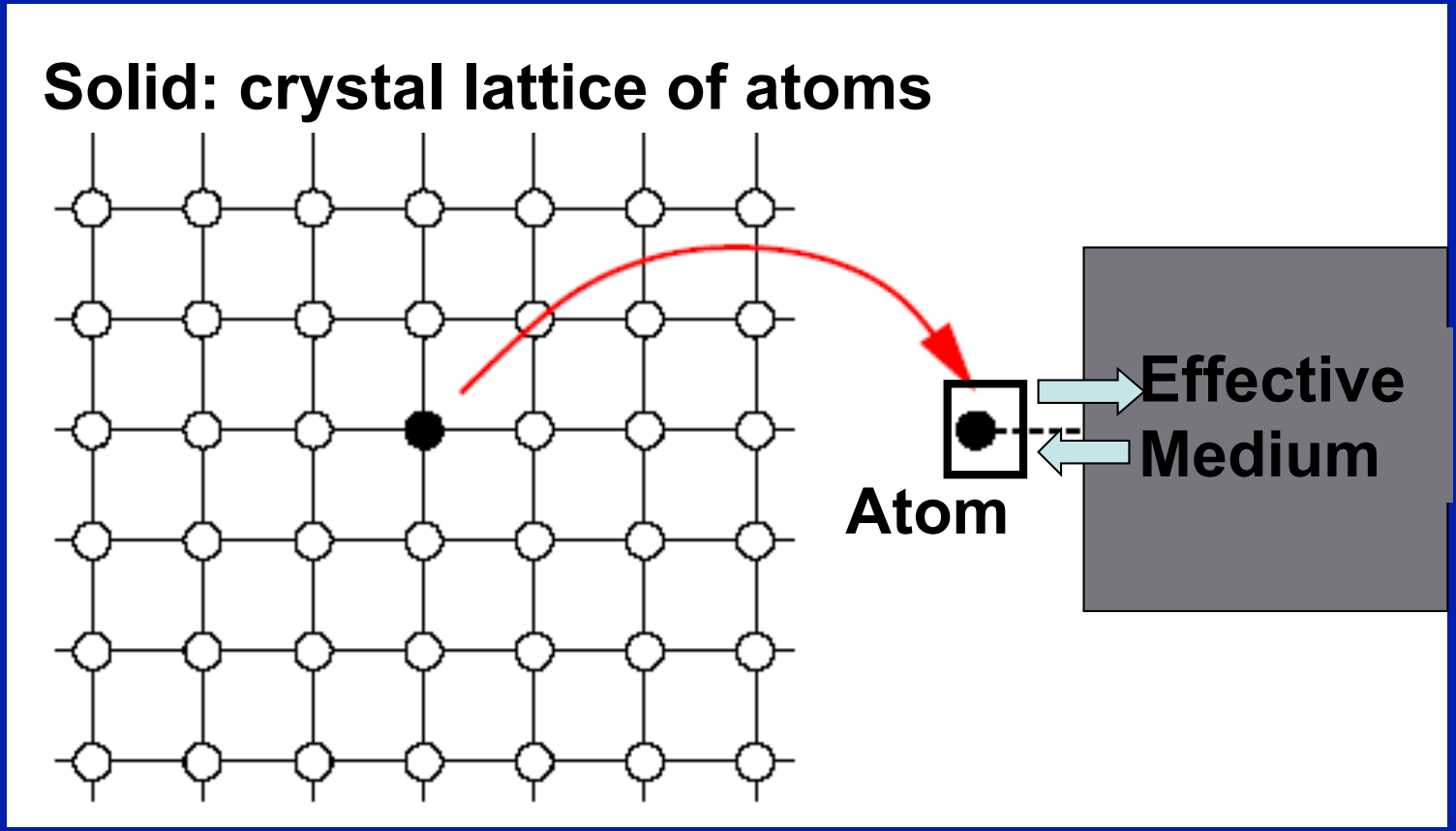
Dynamical Mean-Field Theory:
A.G. & G.Kotliar, PRB 45, 6479 (1992)
Correlated electrons in large dimensions:
W.Metzner & D.Vollhardt, PRL 62, 324 (1989)

Important intermediate steps by: Müller-Hartmann, Schweitzer and Czycholl, Brandt and Mielsch, V.Janis

Early review: Georges et al. Rev Mod Phys 68, 13 (1996)

Dynamical Mean-Field Theory:

viewing a material as an (ensemble of) atoms coupled to a self-consistent medium



"O God! I could be bounded in a nutshell,
and count myself king of infinite space,
were it not that I have bad dreams !"

William Shakespeare (in: Hamlet)

Example: DMFT for the Hubbard model (a model of coupled atoms)

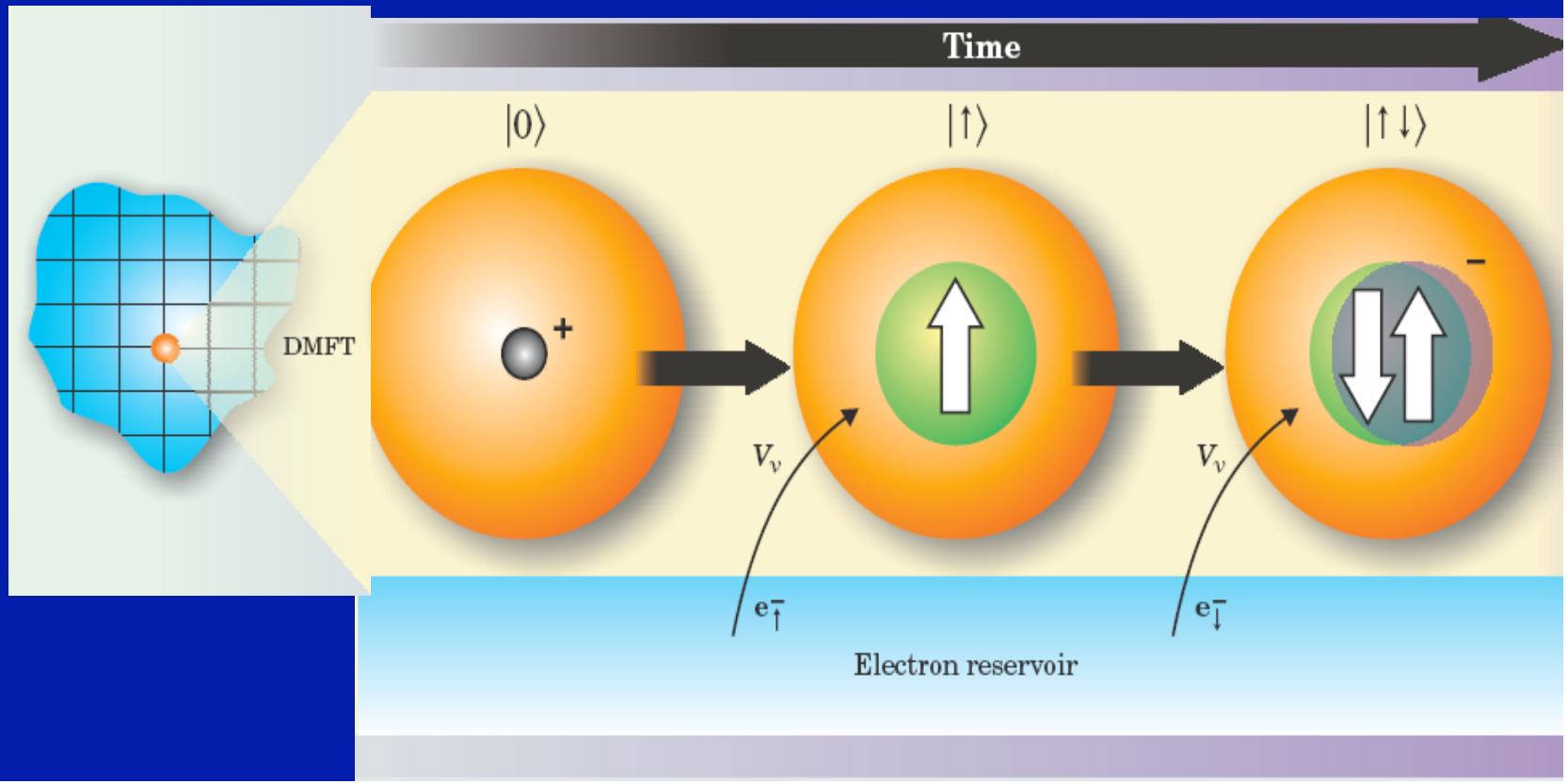
$$H = -\sum_{\mathbf{R}\mathbf{R}'} t_{\mathbf{R}\mathbf{R}'} d^{\dagger}_{\mathbf{R}\sigma} d_{\mathbf{R}'\sigma} + \sum_{\mathbf{R}} H^{\mathbf{R}}_{atom}$$

$$H_{atom} = \varepsilon_d \sum_{\sigma} n_{\sigma} + U n_{\uparrow} n_{\downarrow}$$

Focus on a given lattice site:

"Atom" can be in 4 possible configurations: $|0\rangle, |\uparrow\rangle, |\downarrow\rangle, |\uparrow\downarrow\rangle$

Describe ``history'' of fluctuations between those configurations



Imaginary-time effective action describing these histories:

$$
\begin{aligned}
S &= S_{\rm at} + S_{\rm hyb} \\
S_{\rm at} &= \int_0^\beta d\tau \sum_\sigma d_\sigma^\dagger(\tau) \left(-\frac{\partial}{\partial \tau} + \varepsilon_d \right) d_\sigma(\tau) + U \int_0^\beta d\tau n_\uparrow n_\downarrow \\
S_{\rm hyb} &= \int_0^\beta d\tau \int_0^\beta d\tau' \sum_\sigma d_\sigma^\dagger(\tau') \Delta(\tau - \tau') d_\sigma(\tau)
\end{aligned}
$$

The amplitude Δ(τ) for hopping in and out of the selected site is self-consistently determined: it is the quantum-mechanical Generalization of the Weiss effective field.

$$\mathcal{G}_0^{-1} \equiv \omega + \mu - \Delta(i\omega)$$ Effective `bare propagator'

Hamiltonian formulation: Anderson impurity model

$$H_c = \sum_{l\sigma} E_l a^+_{l\sigma} a_{l\sigma}$$

$$H = H_{\rm c} + H_{\rm at} + H_{\rm hyb}$$

Conduction electron host (``bath'', environment)

$$H_{\rm at} = \varepsilon_d \sum_{\sigma} d^\dagger_\sigma d_\sigma + U\, n^d_\uparrow n^d_\downarrow$$

Single-level ``atom''

$$H_{hyb} = \sum_{l\sigma} [V_l a^+_{l\sigma} d_\sigma + {\rm h.c.}]$$

Transfers electrons between bath and atom – Hybridization, tunneling

Local effective action:

Focus on dynamics of impurity orbital: integrate out conduction electrons → Effective action for impurity orbital:

$$
\begin{aligned}
S &= S_{\rm at} + S_{\rm hyb} \\
S_{\rm at} &= \int_0^\beta d\tau \sum_\sigma d_\sigma^\dagger(\tau) \left(-\frac{\partial}{\partial \tau} + \varepsilon_d \right) d_\sigma(\tau) + U \int_0^\beta d\tau n_\uparrow n_\downarrow \\
S_{\rm hyb} &= \int_0^\beta d\tau \int_0^\beta d\tau' \sum_\sigma d_\sigma^\dagger(\tau') \Delta(\tau - \tau') d_\sigma(\tau)
\end{aligned}
$$

$$
-\frac{1}{\pi} {\rm Im} \Delta(\omega + i0^+) = \sum_l |V_l|^2 \, \delta(\omega - E_l)
$$

$$
\mathcal{G}_0^{-1} \equiv \omega + \mu - \Delta(i\omega)
$$

Effective `bare propagator'

Focus on energy-dependent local observable :

$$G_{RR}(\omega) \equiv G_{\rm loc}$$

On-site Green's function (or spectral function) of the `solid'

Use atom-in-a-bath as a reference system to represent this observable:

→ IMPOSE that ε_d and Δ should be chosen such that:

$$G_{\rm imp}[\omega; \varepsilon_d, \Delta(\omega)] = G_{\rm loc}(\omega)$$

At this point, given G_{loc} of the lattice Hubbard model,
we have just introduced an exact local representation of it

G_{RR} is related to the exact self-energy of the lattice (solid) by:

$$G_{\mathbf{RR}}(\omega) = \sum_{\mathbf{k}} \frac{1}{\omega + \mu - \varepsilon_{\mathbf{k}} - \Sigma(\mathbf{k}, \omega)} = G_{\mathrm{loc}}(\omega)$$

In which ε_k is the tight-binding band (FT of the hopping $t_{RR'}$)

High-frequency → $\varepsilon_d = -\mu + \sum_k \varepsilon_k \; (= -\mu)$

Let us now make the **APPROXIMATION** that the lattice self-energy is **k-independent** and coincides with that of the effective atom (impurity problem):

$$\Sigma(\mathbf{k}, \omega) \simeq \Sigma_{\mathrm{imp}}(\omega)$$

This leads to the following self-consistency condition:

$$G_{\mathrm{imp}}[i\omega; \Delta] = \sum_{\mathbf{k}} \frac{1}{G_{\mathrm{imp}}[i\omega; \Delta]^{-1} + \Delta(i\omega) - \varepsilon_{\mathbf{k}}}$$

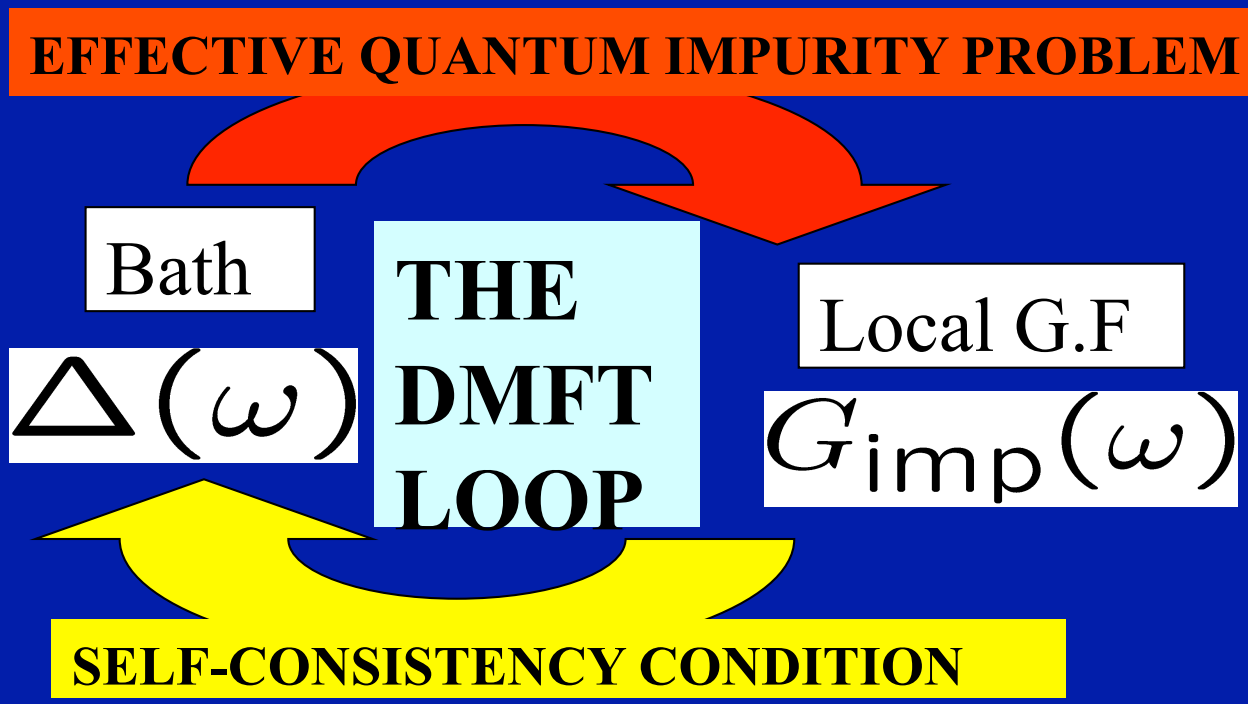
e.g. semi-circular d.o.s width 2D: $\Delta(i\omega) = \frac{D^2}{4} G(i\omega)$

The self-consistency equation and the DMFT loop

Approximating the self-energy by that of the local problem : $\Sigma(\mathbf{k},\omega) \simeq \Sigma_{imp}(\omega)$

→ fully determines both the local G and Δ:

$$G_{\mathrm{imp}}[i\omega;\Delta] = \sum_{\mathbf{k}} \frac{1}{G_{\mathrm{imp}}[i\omega;\Delta]^{-1} + \Delta(i\omega) - \varepsilon_{\mathbf{k}}}$$



The DMFT construction is EXACT:

- For the non-interacting system
  (U =0 → Σ = 0 !)
- For the isolated atom
  (strong-coupling limit t=0 → Δ = 0)

→ Hence provides an interpolation from weak to strong coupling

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

Proofs: LW functional, Cavity construction (more on board)

Δ(ω): generalizing the Weiss field to the quantum world

Pierre Weiss
1865-1940
« Théorie du Champ Moléculaire »
(1907)

Einstein, Paul Ehrenfest, Paul Langevin, Heike Kammerlingh-Onnes, and Pierre Weiss at Ehrenfest's home, Leyden, the Netherlands. From Einstein, His Life and Times, *by Philipp Frank (New York: A.A. Knopf, 1947). Photo courtesy AIP Emilio Segrè Visual Archives.*

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

} rely on 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

- Exact energy functional of local observable

- Exact representation of local observable:
- Generalized ``Weiss field''
- Self-consistency condition, later approximated

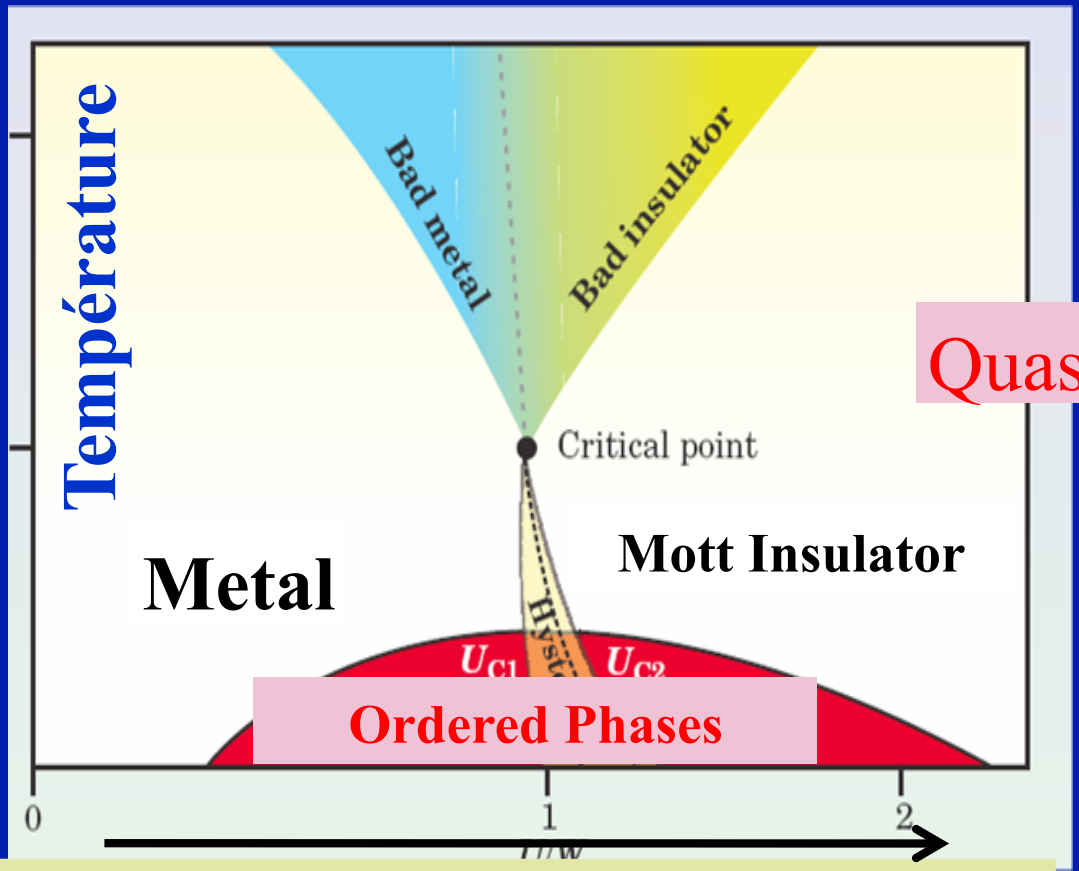
see e.g:
A.G
arXiv cond-mat
0403123

Low-frequency behavior of $\Delta(\omega)$ determines nature of the phase

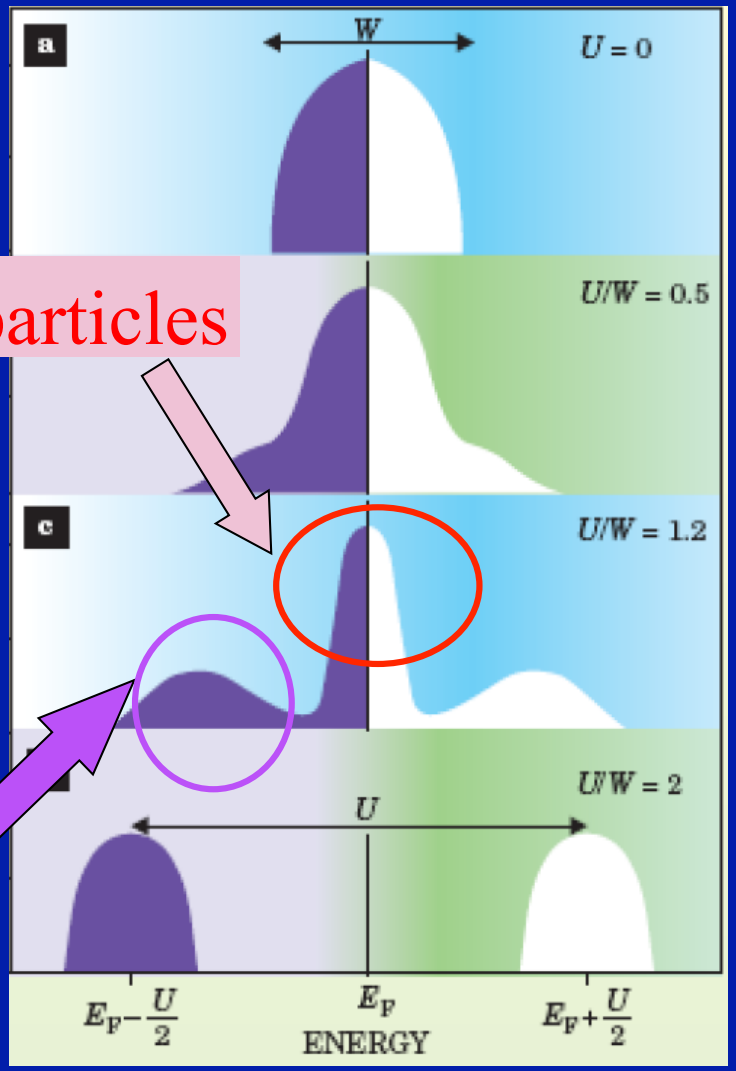
- $\Delta(\omega \to 0)$ finite → local moment is screened. `Self-consistent' Kondo effect. Gapless metallic state.

- $\Delta(\omega)$ gapped → no Kondo effect, degenerate ground-state, insulator with local moments

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



Quasiparticles

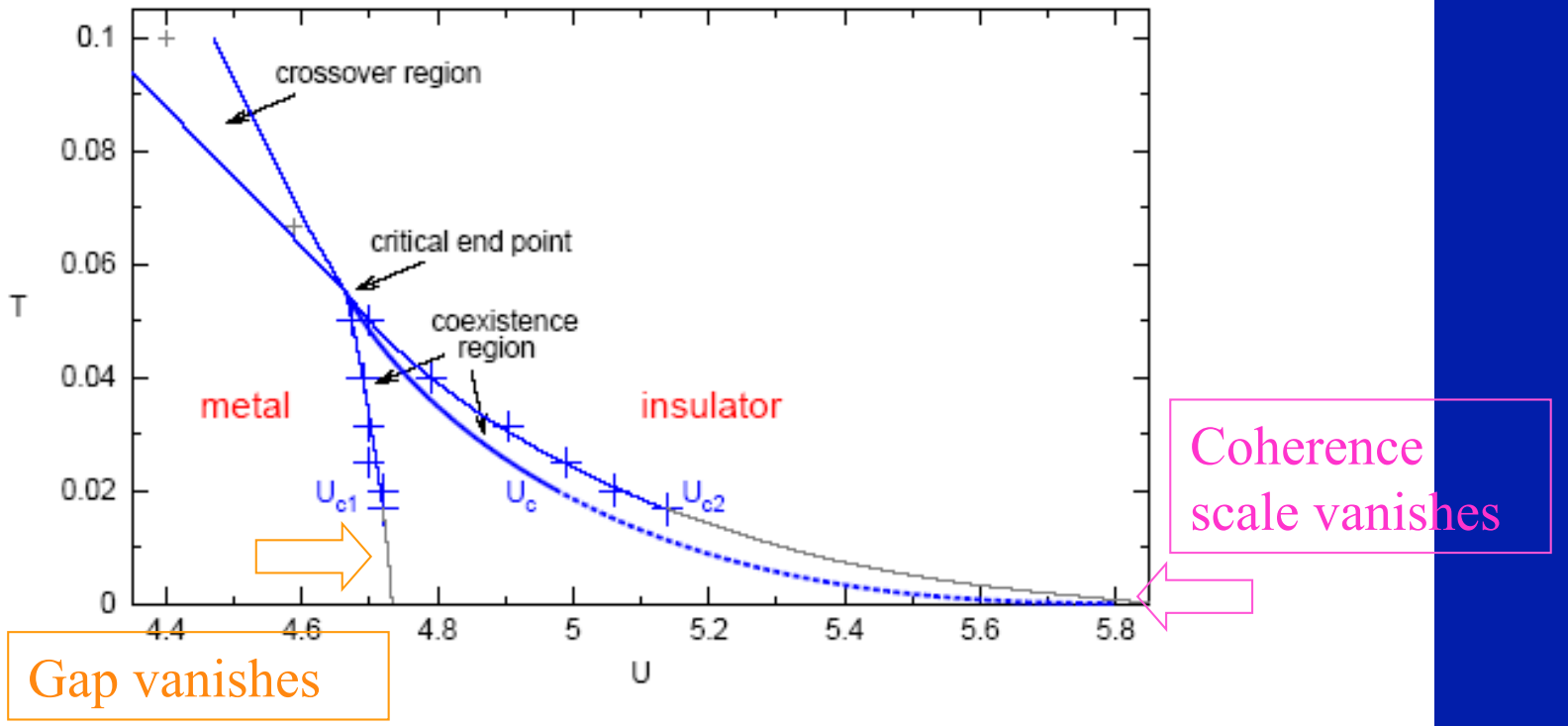


Quasi atomic excitations

Phase diagram : zoom on paramagnetic solutions

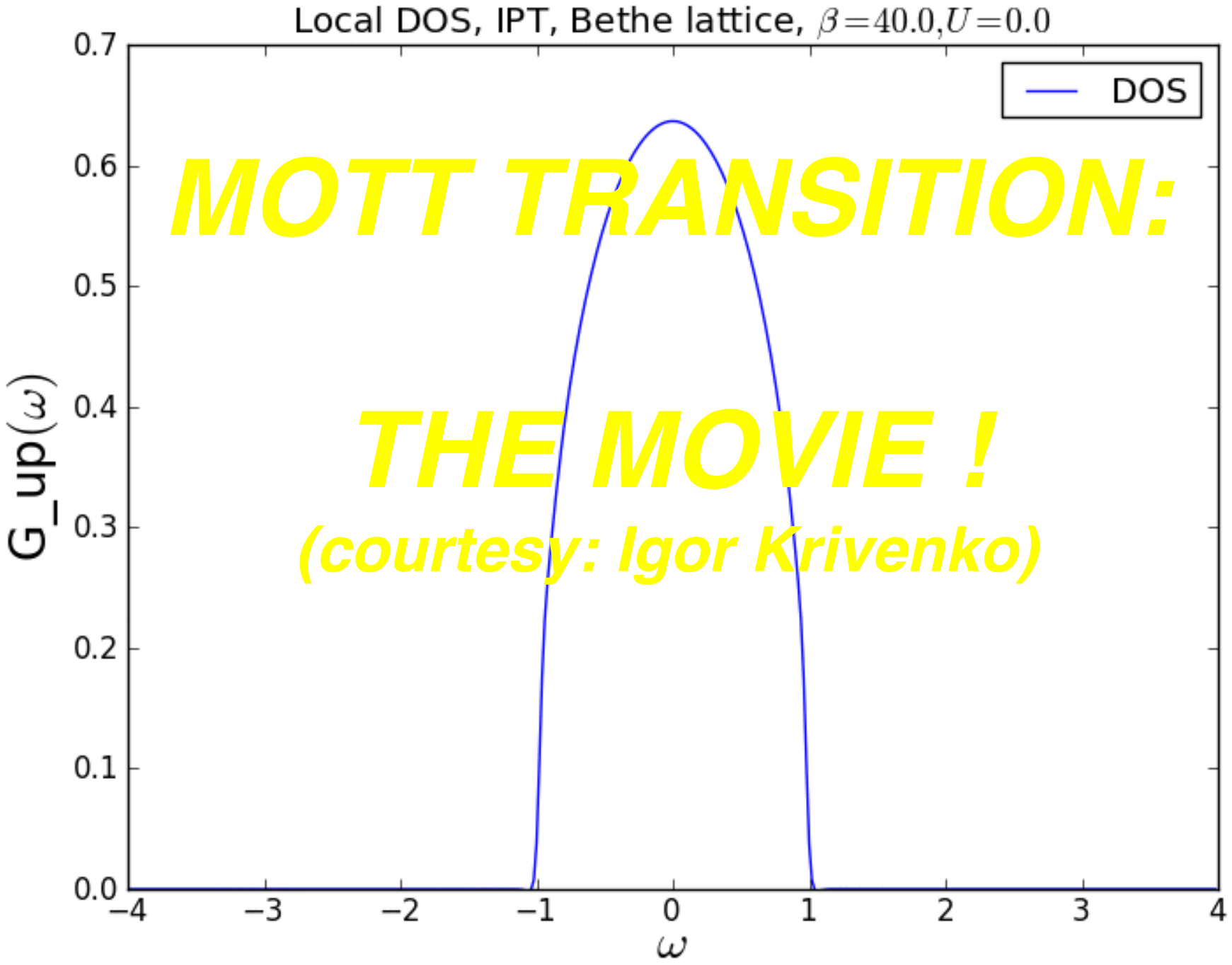
Hubbard model, Bethe lattice, homog. phase, $n = 1$, e.g., DMFT(QMC)

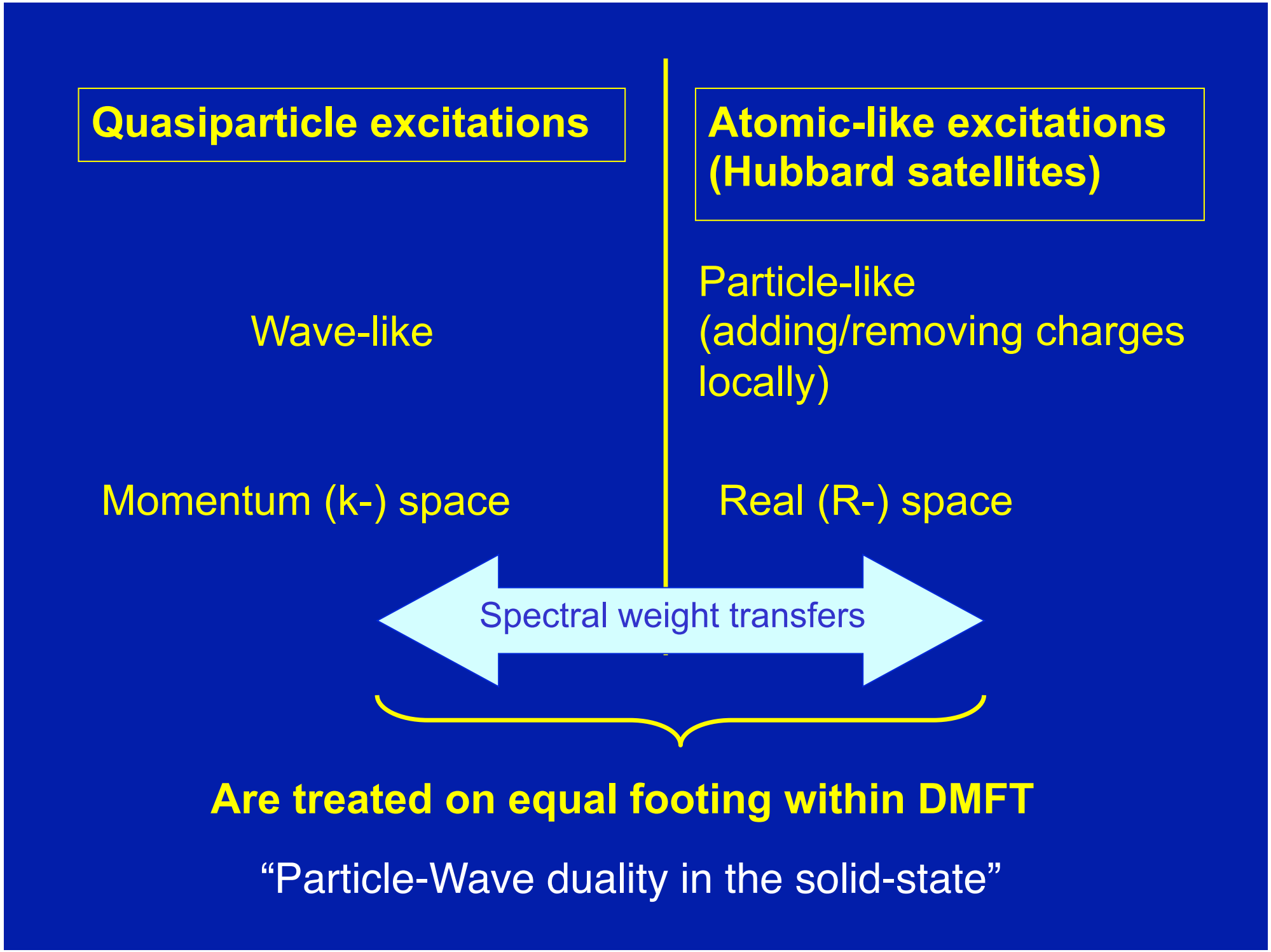
[Blümer '02]



- coexistence region $[U_{c1}; U_{c2}]$, first-order transition
- crossover above critical region

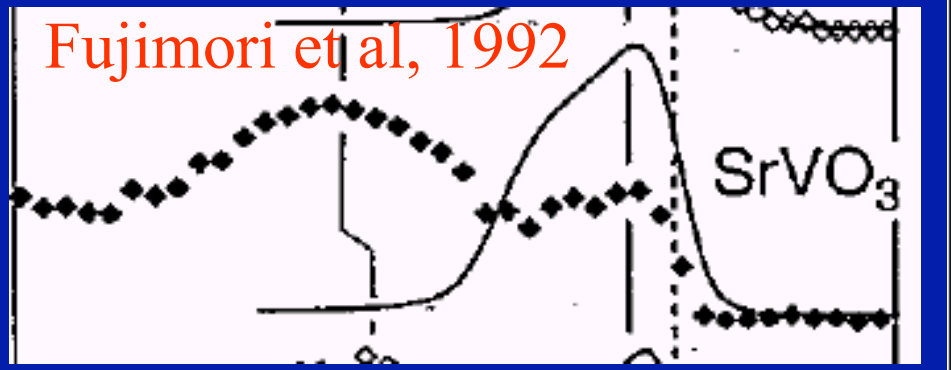
Blümer et al. Units here are 4D=2*bandwidth


Local DOS, IPT, Bethe lattice, $\beta = 40.0, U = 0.0$
DOS
G_up(ω)
ω
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
−4
−3
−2
−1
0
1
2
3
4
MOTT TRANSITION:
THE MOVIE !
(courtesy: Igor Krivenko)

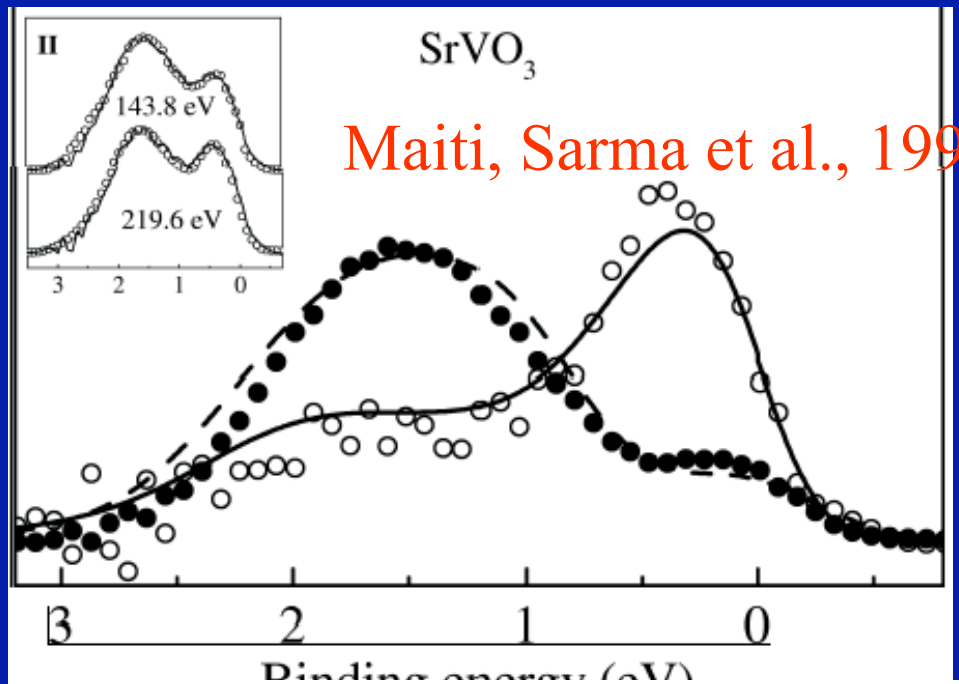

Quasiparticle excitations
Atomic-like excitations (Hubbard satellites)
Wave-like
Particle-like (adding/removing charges locally)
Momentum (k-) space
Real (R-) space
Spectral weight transfers
Are treated on equal footing within DMFT
"Particle-Wave duality in the solid-state"

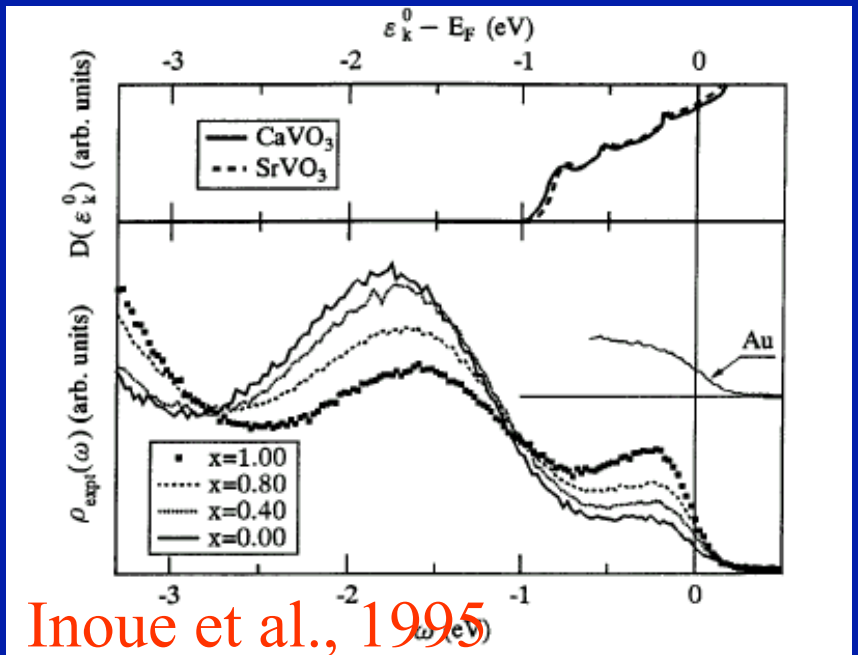
PHOTOEMISSION…
A 12 years (success) story

Fujimori et al, 1992



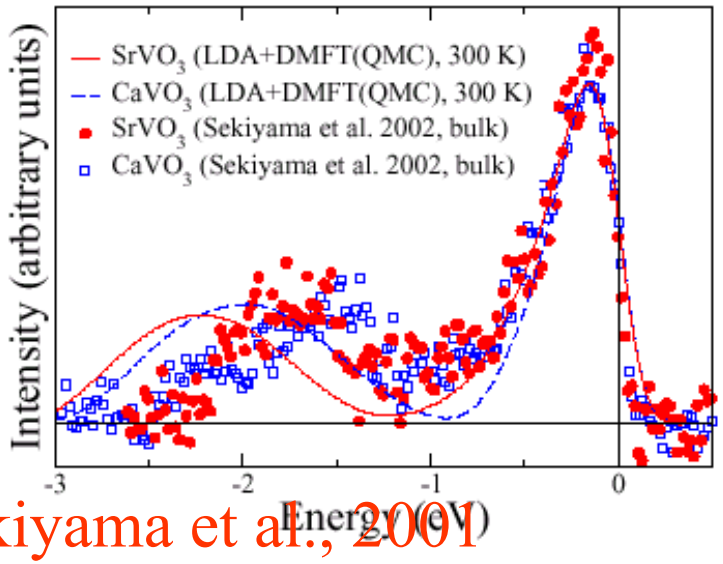
Maiti, Sarma et al., 1999





Inoue et al., 1995

FIG. 1. Top: DOS $D(\varepsilon_k^0)$ of $CaVO_3$ and $SrVO_3$ calculated by the APW method. Bottom: measured photoemission spectra $\rho_{expt}(\omega)$ of $Ca_{1-x}Sr_xVO_3$ taken with $h\nu = 50$ eV. A spectrum of Au is also shown as a reference to E_F and the instrumental resolution.



Sekiyama et al., 2001

FIG. 4: Comparison of the calculated, parameter-free LDA+DMFT(QMC) spectra of $SrVO_3$ (solid line) and $CaVO_3$ (dashed line) with bulk-sensitive high-resolution PES ($SrVO_3$: circles; $CaVO_3$: rectangles) [4]. Horizontal line: experimental subtraction of the background intensity.

Simple approximations:

- Ignoring low-energy Kondo effect/quasiparticles → ~ Hubbard-like approximations
- Focusing on quasiparticles only → similar to Brinkman-Rice/slave bosons

$$G(\omega) \simeq \frac{1}{2}\left[\frac{1}{\omega - \Delta(\omega) - U/2} + \frac{1}{\omega - \Delta(\omega) + U/2}\right]$$

$$\Delta = \frac{D^2}{4} G$$

$$\Rightarrow \boxed{D^4 G^3 - 8D^2 \omega G^2 + 4(4\omega^2 + D^2 - U^2)G - 16\omega = 0}$$

Hubbard-like (III) approximation: ignore Kondo-like processes/ quasiparticles

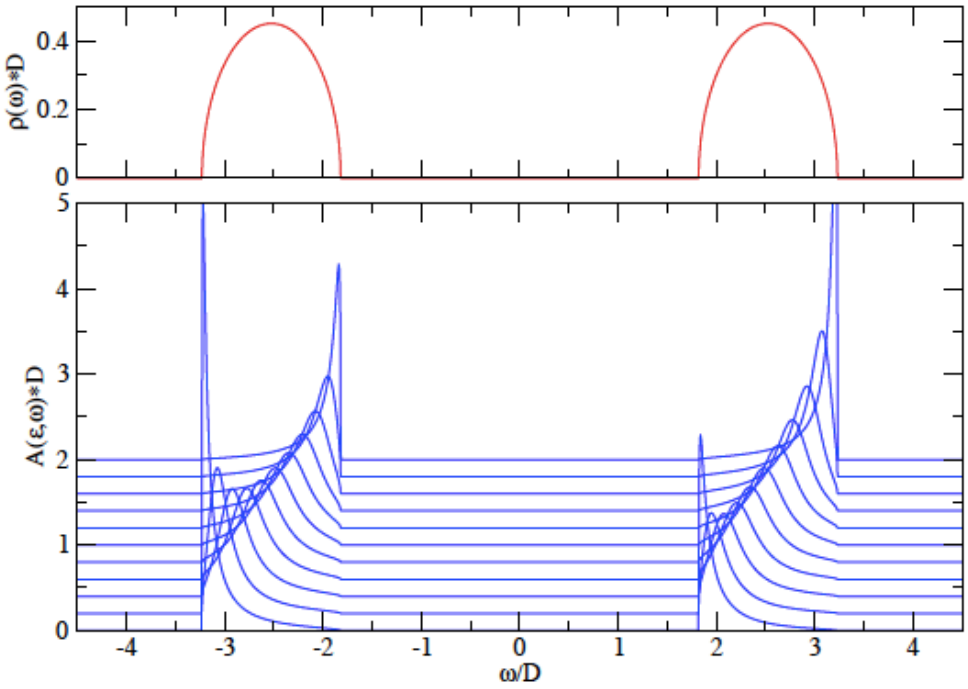


FIG. 10: Spectral density $\rho(\omega)$(a) and ε-resolved spectral function $A(\varepsilon, \omega)$ for several ε (from bottom to the top, $\varepsilon = -D, \ldots D$ with a step 0.2) (b), with $U/D = 4.0$ and $T = 0$. The results are from Hubbard III approximation.

1-pole approximation ~ Gutzwiller/BR

M.Potthoff PRB64, 165114 (2001)

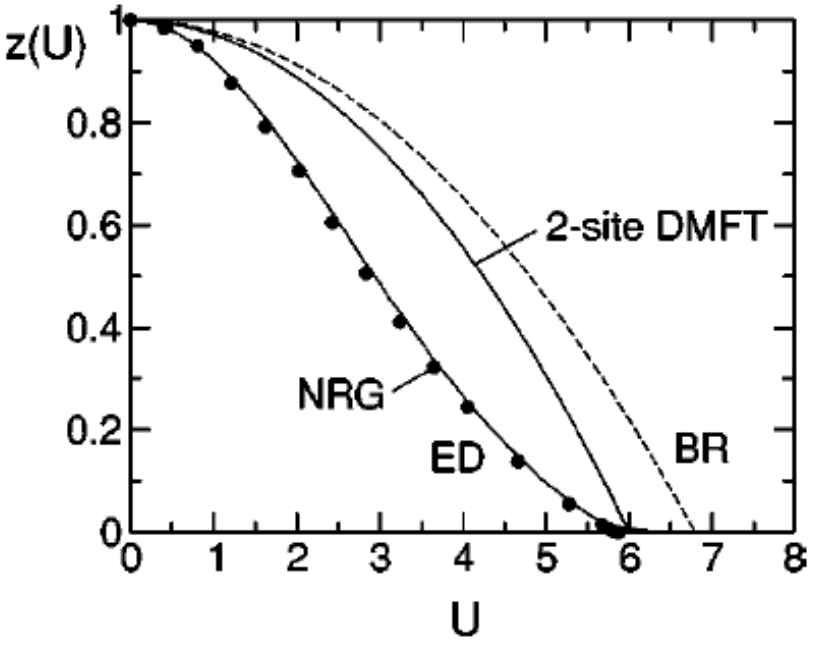
$$\Delta(\omega) = \frac{V^2}{\omega}$$

$$\Rightarrow \Sigma = \frac{U^2}{8}\left[\frac{1}{\omega - 3V} + \frac{1}{\omega + 3V}\right]$$

$$\Rightarrow Z^{-1} \equiv 1 - \frac{\partial \Sigma}{\partial \omega}|_{\omega=0} = 1 + \frac{U^2}{36V^2}$$

$$V^2 = \frac{U^2}{36}\frac{Z}{1-Z}$$

$$\Delta = \frac{D^2}{4}G \Rightarrow \frac{U^2}{36}\frac{Z}{1-Z} = \frac{D^2}{4}Z$$



$$Z = 1 - \left(\frac{U}{3D}\right)^2$$

Recent algorithmic breakthroughs

entering a new age for DMFT approaches (and extensions) …

Continuous-time quantum Monte Carlo (CT-QMC)

*Rubtsov 2005 Interaction expansion(CT-INT)

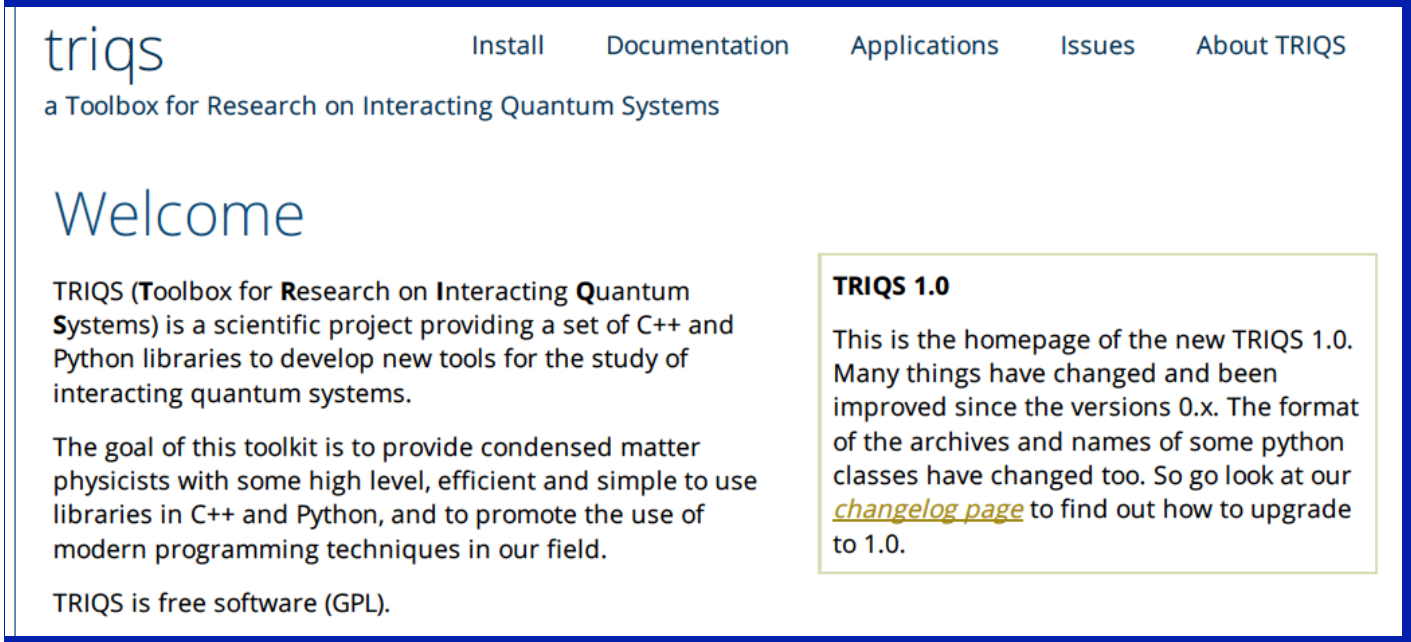
*P. Werner, M.Troyer, A.Millis et al 2006
Hybridization expansion(CT-HYB)

*Gull/Parcollet 2008 Auxiliary field (CT-AUX)

See lecture by F.Assaad
Recent review: Gull et al. Rev Mod Phys 83, 349 (2011)

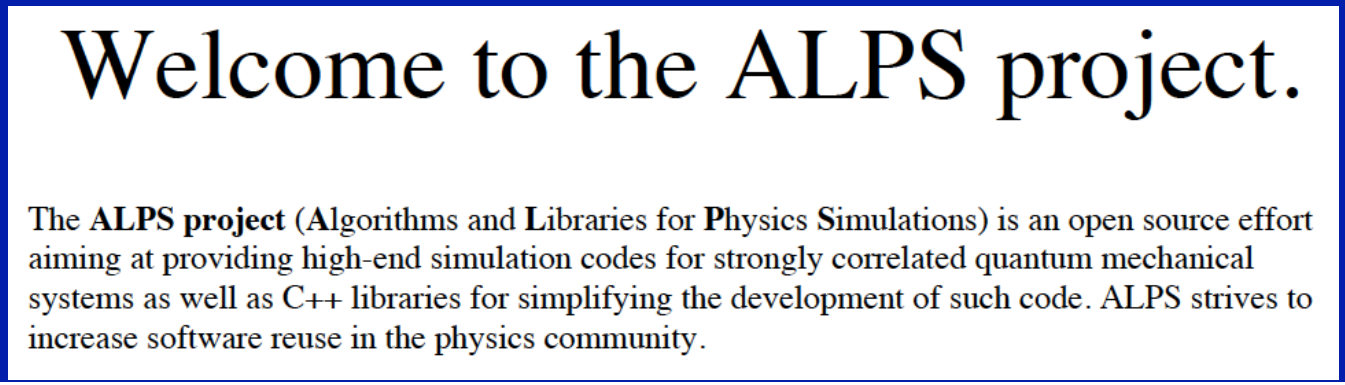
Need for efficient development and sharing of code libraries

The TRIQS project (O.Parcollet, M.Ferrero et al.)

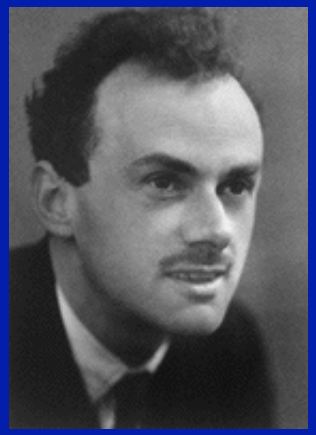


ipht.cea.fr/triqs

alps.comp-phys.org



In simplest cases (e.g. single-orbital),
the DMFT construction avoids the
fermion minus-sign problem
(absent for simplest quantum impurity problems,
effectively 1+1-dimensional)



`` It therefore becomes desirable
that approximate practical methods
of applying quantum mechanics
should be developed,
which can lead
to an explanation of the main features
of complex atomic systems
without too much computation"

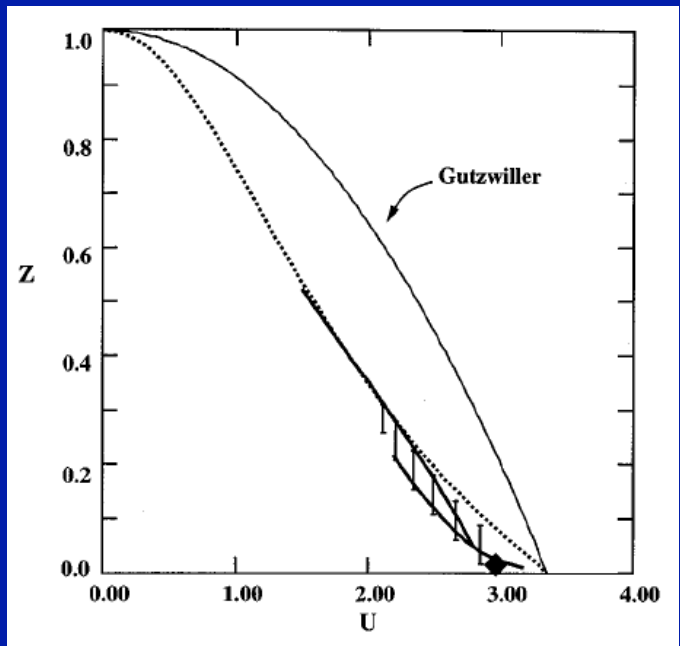
Nature of the metallic phase

- At (possibly very) low T,ω: a Fermi liquid

$$\mathrm{Re}\Sigma(\omega + i0^+) = U/2 + (1 - 1/Z)\omega + O(\omega^3),$$
$$\mathrm{Im}\Sigma(\omega + i0^+) = -B\omega^2 + O(\omega^4).$$

- At U_{c2} transition: Z→0 (~ Brinkman-Rice)
- Heavy quasiparticles:

m*/m=1/Z
(divergence reflects
large entropy of insulator,
see below)



LaTiO3: AF Mott insulator

AF persists up to ~ 5% hole-doping

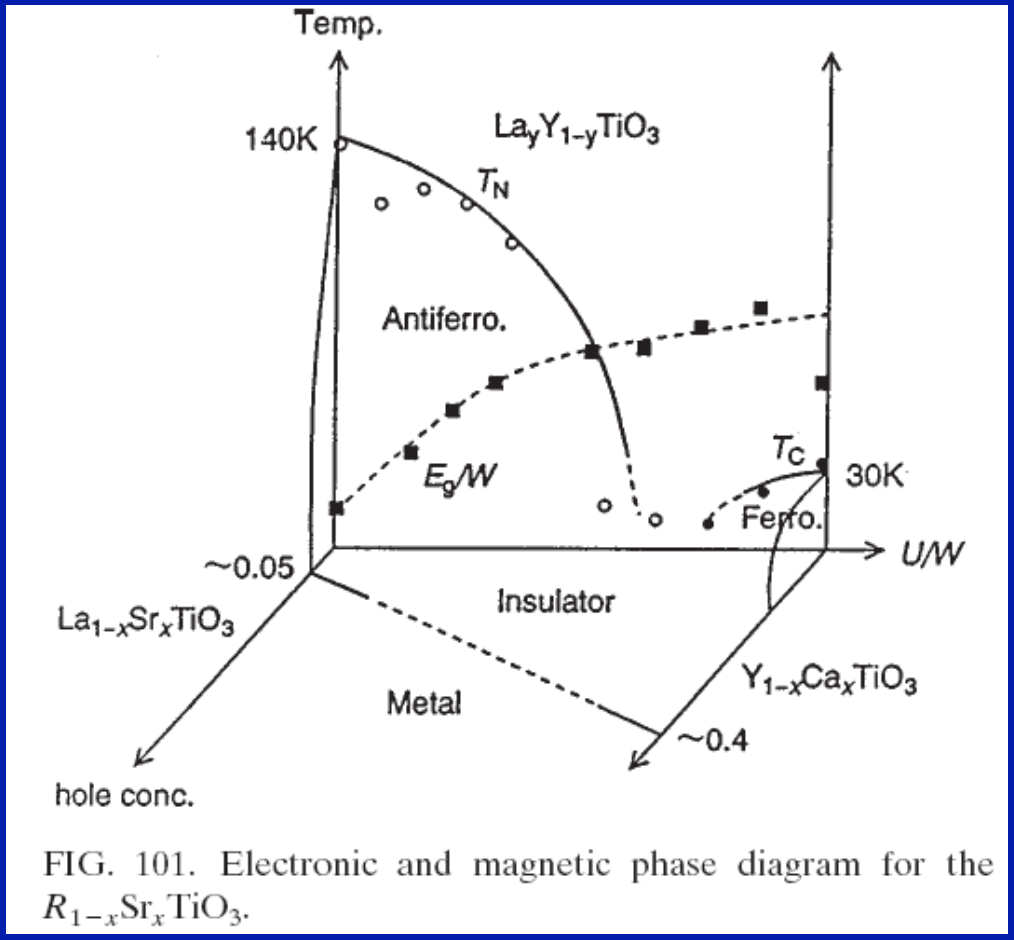
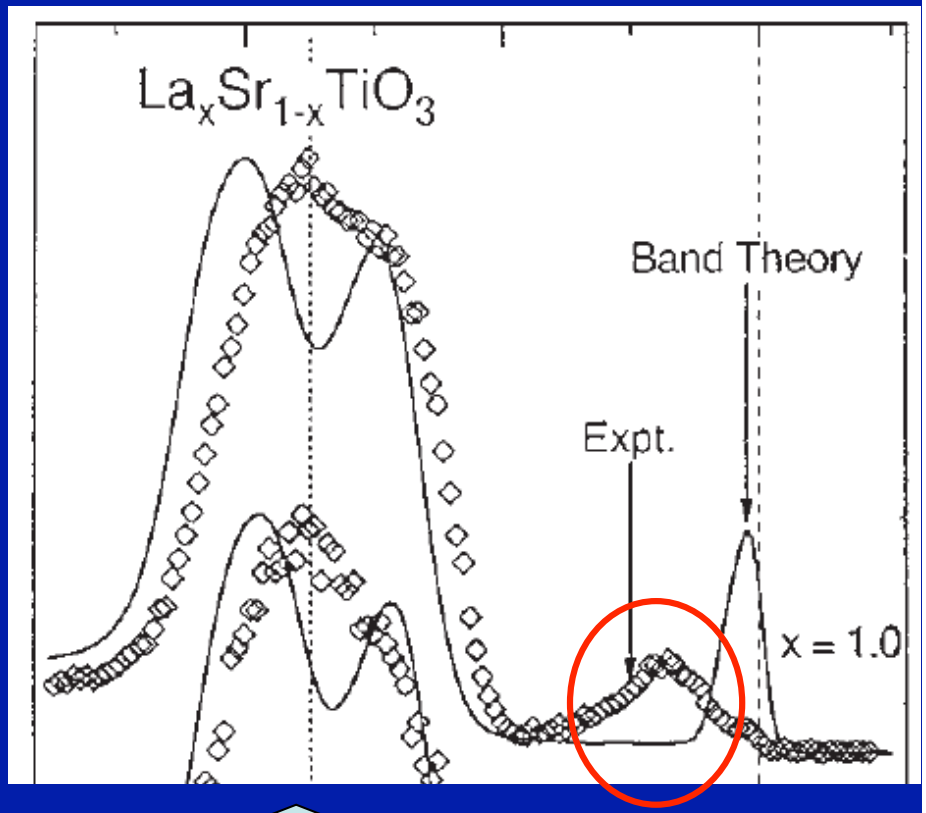


FIG. 101. Electronic and magnetic phase diagram for the $R_{1-x}Sr_xTiO_3$.

Photoemission spectrum: definitely a Mott insulator

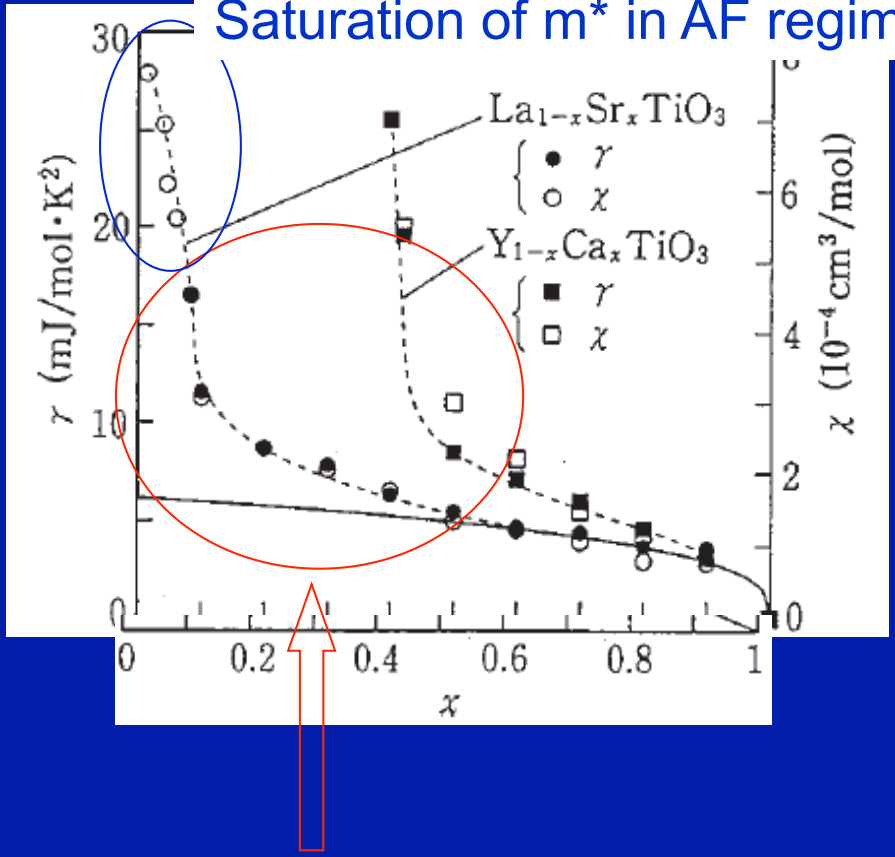


Oxygen states

Lower Hubbard band d1→d0

Approach to the Mott state in titanates

Saturation of m* in AF regime



Increase of effective mass

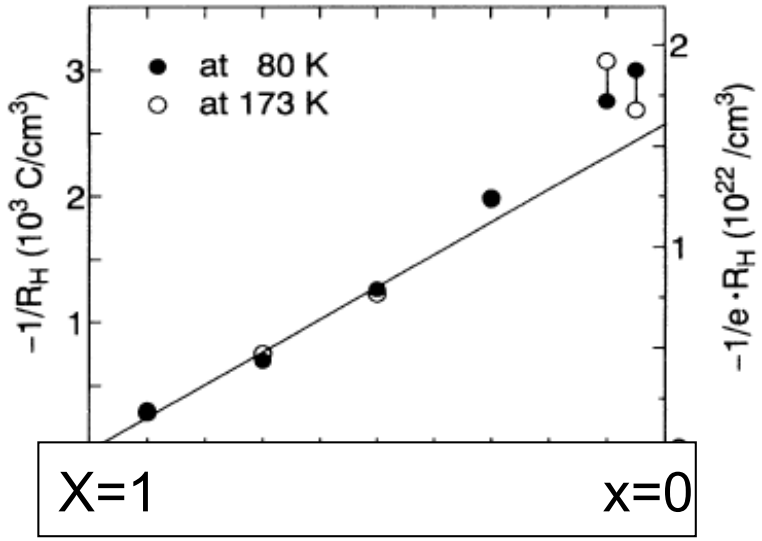
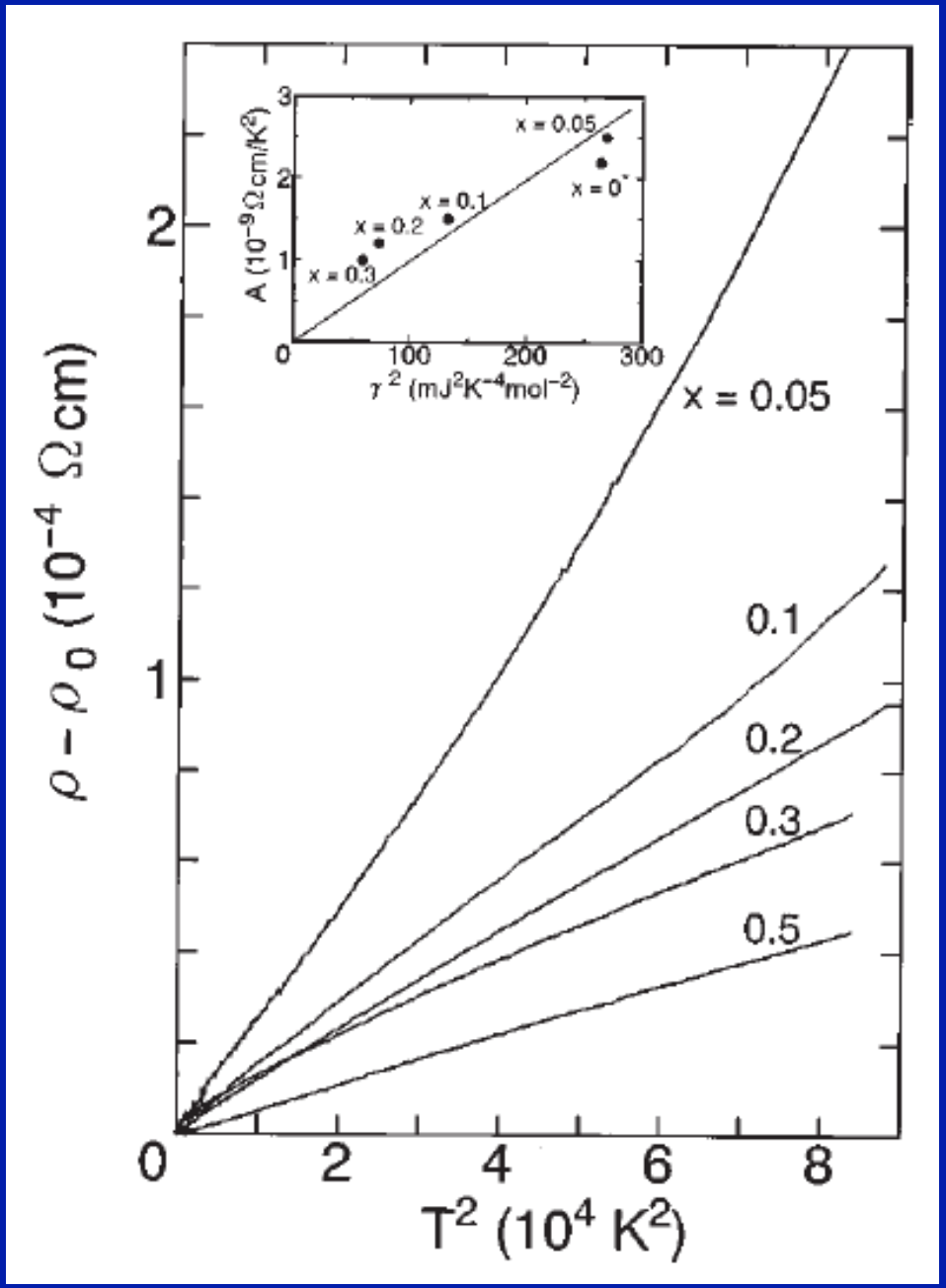
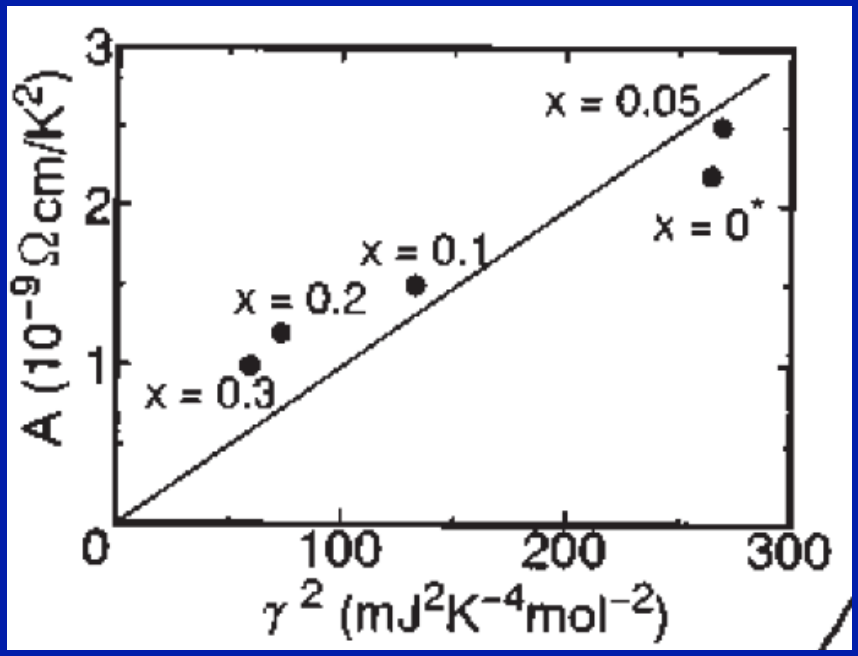


FIG. 2. The filling (x) dependence of the inverse of Hall coefficient (R_H^{-1}) in $Sr_{1-x}La_xTiO_3$. Open and closed circles represent the values measured at 80 K and 173 K, respectively. A solid line indicates the calculated one based on the assumption that each substitution of a Sr^{2+} site with La^{3+} supplies the compound with one electron-type carrier per Ti site.

R_H reported as ~ T-independent and consistent w/ large Fermi surface

Tokura et al.
PRL, 1993





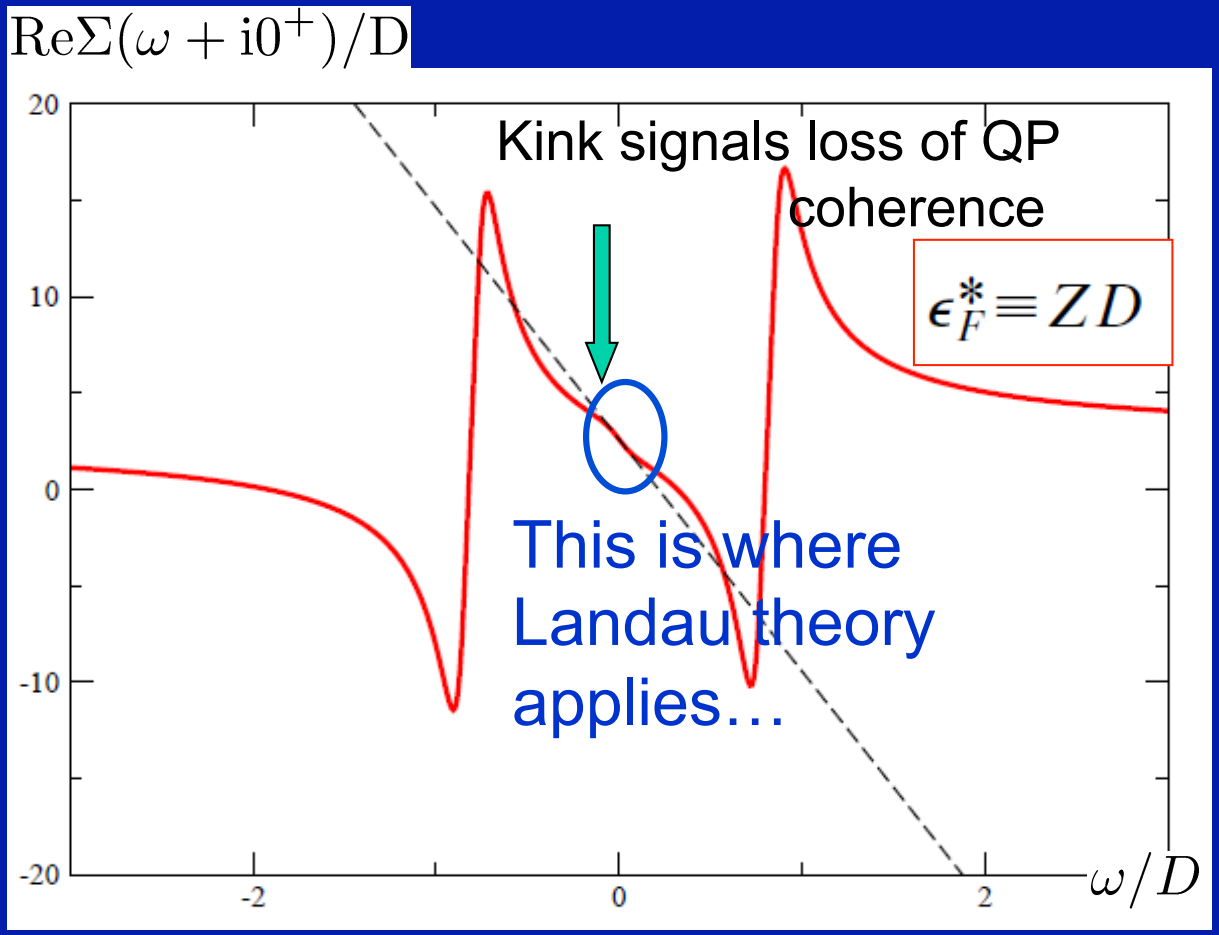
Titanates/transport:

$$\rho_{dc} = AT^2 + \cdots$$

$$A/\gamma^2 \sim \text{const.}$$

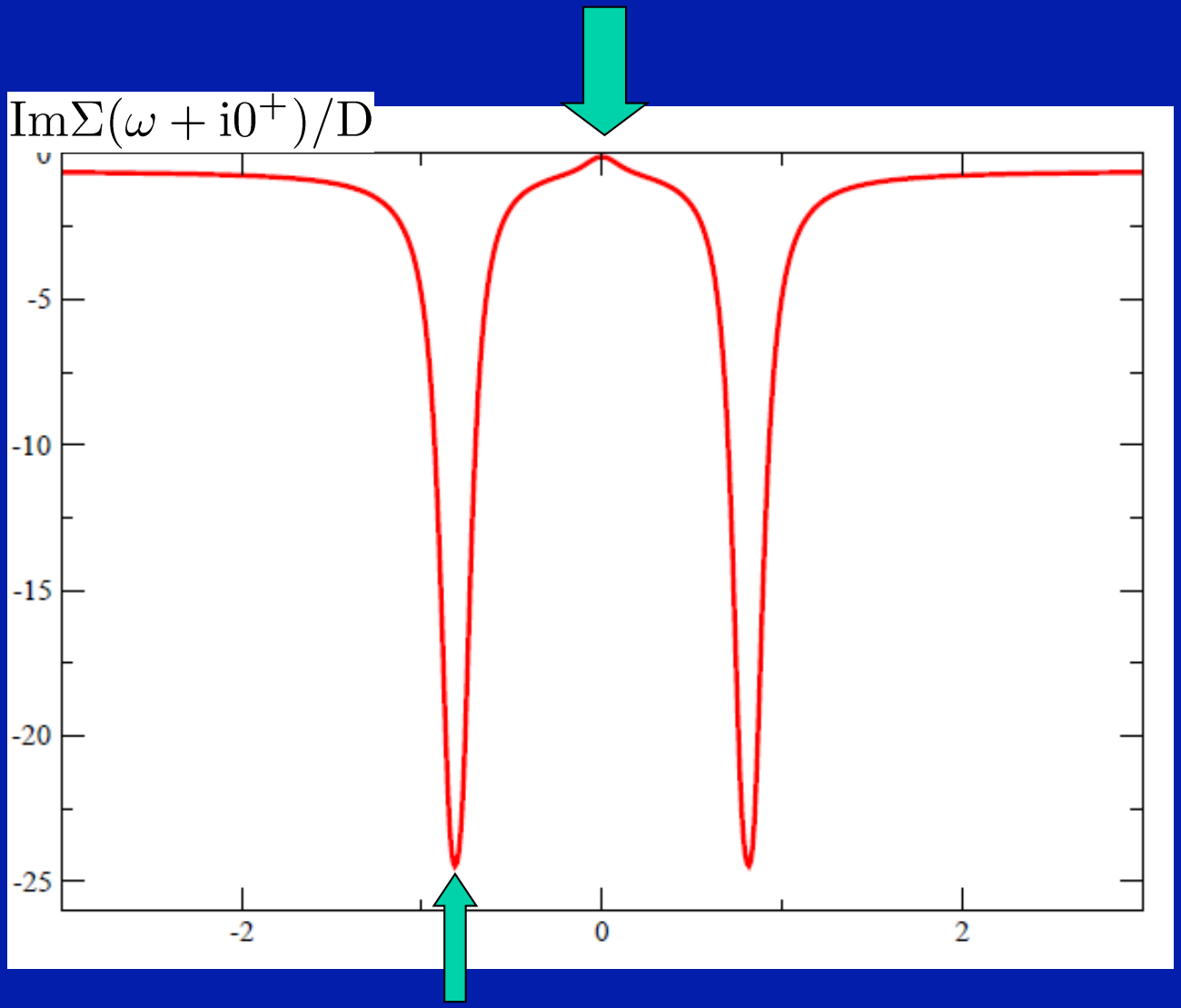
Fermi liquid behavior observed
Below ~ 100K @ 5% doping

But… there is (plenty of) life beyond the Fermi-liquid regime



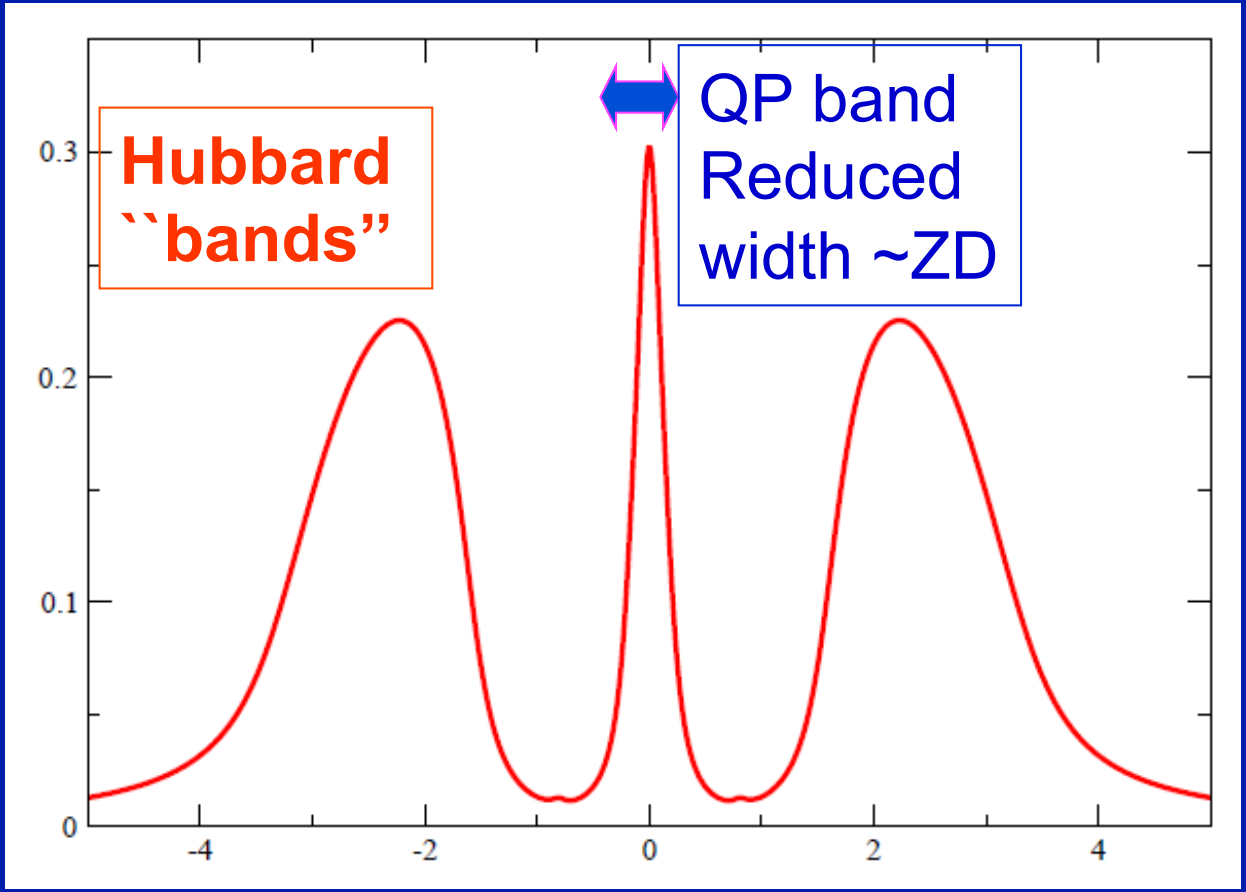
CTQMC+Analytical continuation (Pade),
courtesy M.Ferrero, compares perfectly to NRG

$B\omega^2$ applies only below coherence scale
B-coefficient is enhanced ~ $1/Z^2$



These 2 peaks will coalesce into a pole at ω=0
as insulator is reached

k-integrated spectral function (total d.o.s) :

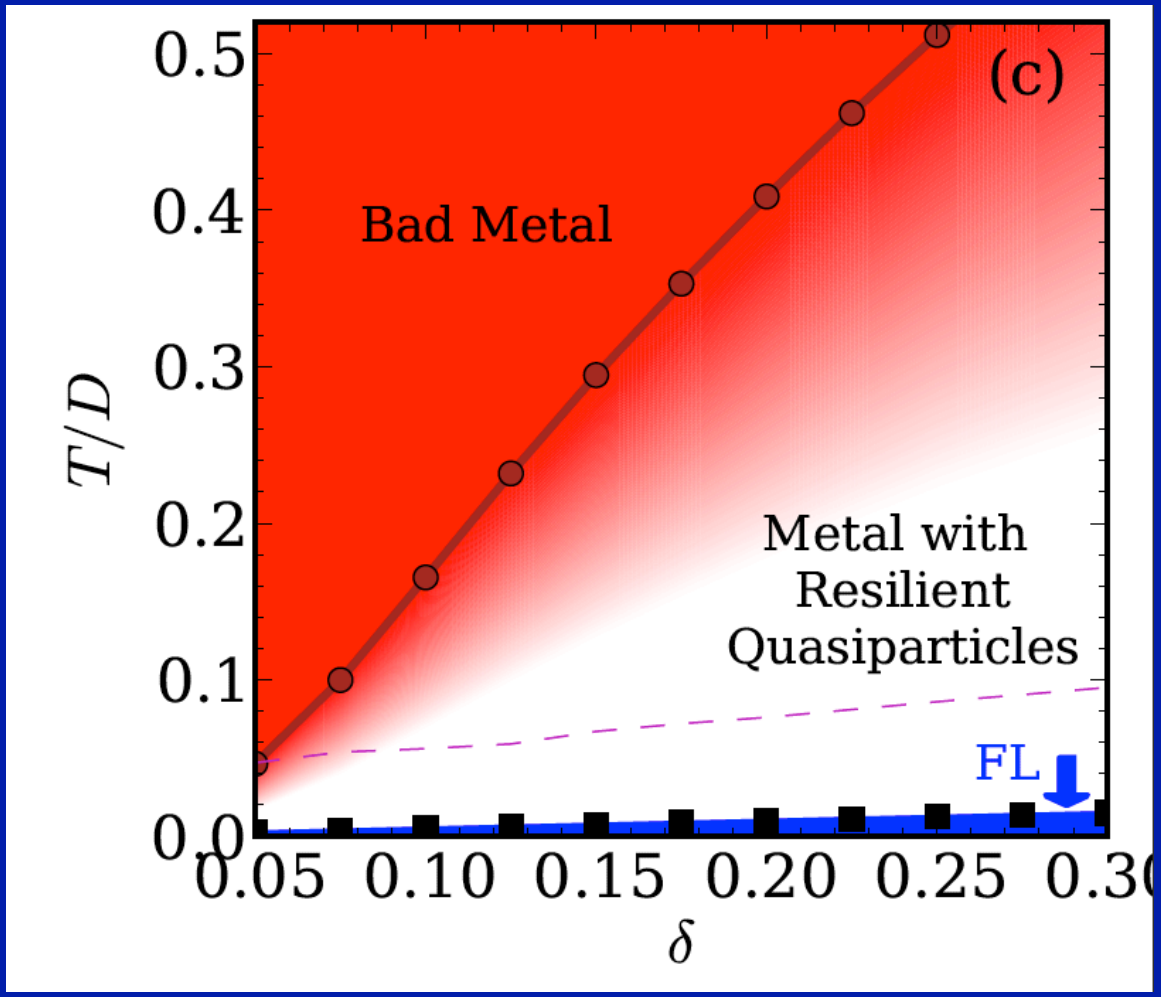


Value of A(ω=0) is pinned at U=0 value due to Luttinger theorem

→ Low-energy quasiparticles and incoherent Hubbard bands Coexist in one-particle spectrum of correlated metal

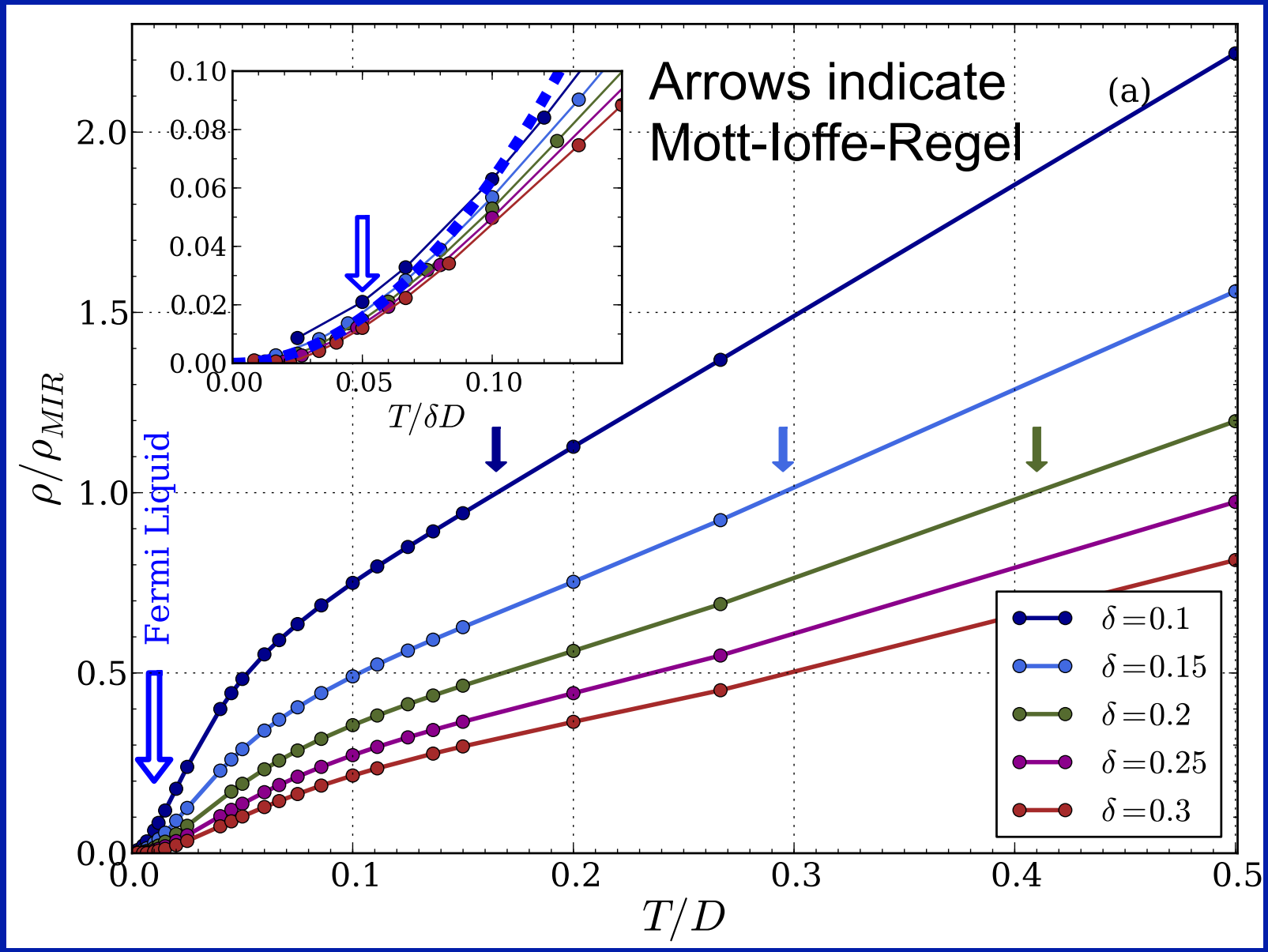
Recent insights into an old problem: "How bad metals become good"

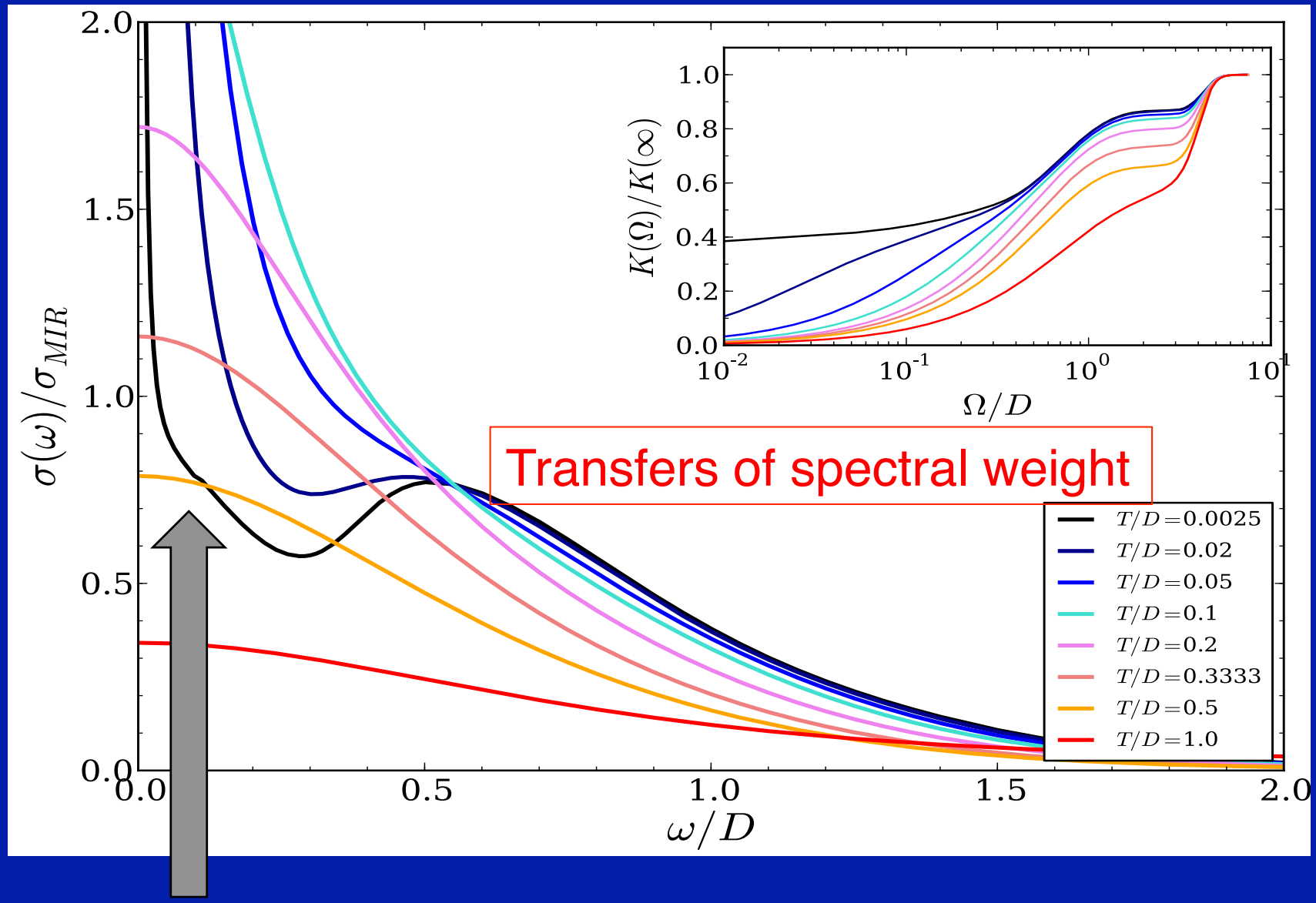
`Resilient' *quasiparticles* *beyond Landau Theory*



Deng et al.
PRL 110 (2013)
086401

Overview of calculated resistivity vs. T





This non-Drude ``foot'' is actually the signature of Landau's Fermi liquid in the optical spectrum !

Signature of the two crossovers (FL, MIR) in optical spectroscopy:

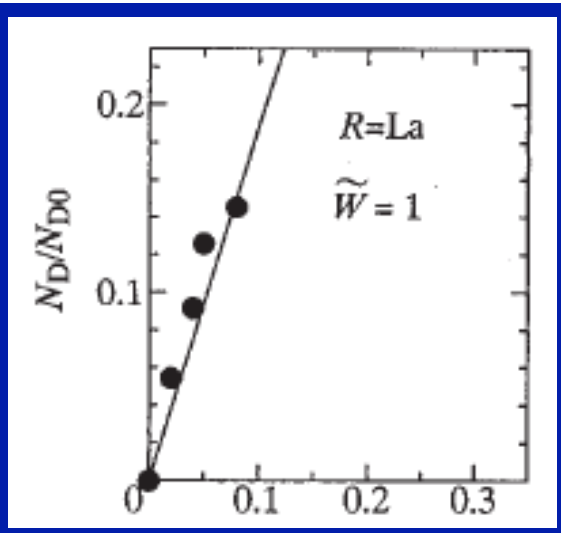
1. Merging of Drude peak and mid-infrared into broad peak at T_{FL}
2. Merging of QP band and LHB at T_{MIR}
3. Redistribution of sp.weight over very high energies at MIR, but involving only Drude+mid-infrared below T_{MIR}

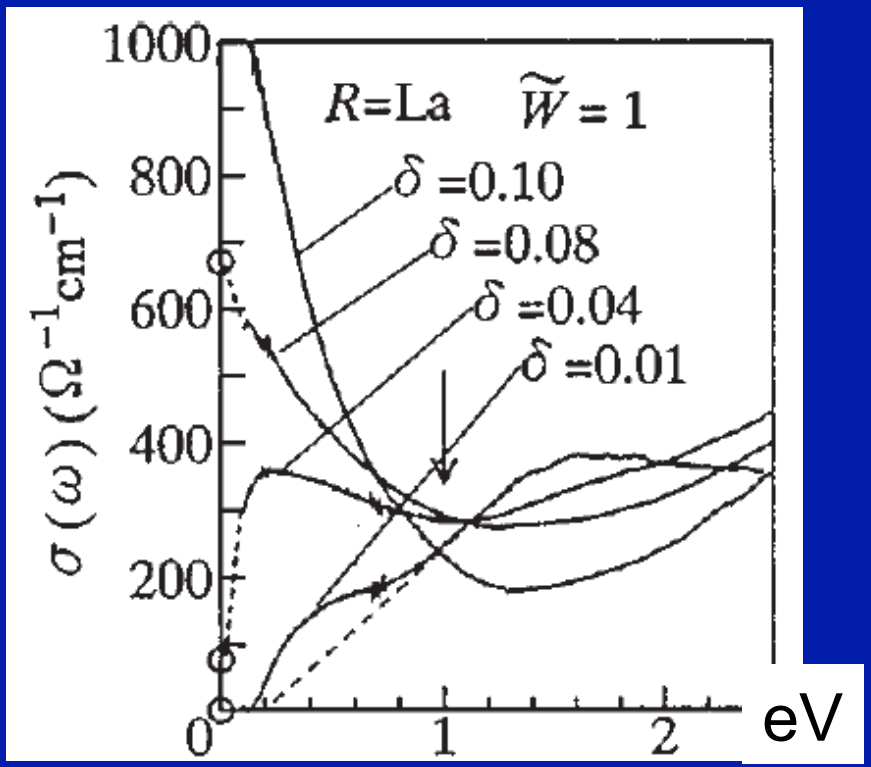
cf. Hussey, Takenaka et al. LSCO PRB 2003
Hussey, Phil Mag
Gunnarsson RMP

Optical conductivity

Drude weight ~ doping

FIG. 108. N_D to N_{D0} as a function of δ (Katsufuji, Okimoto, and Tokura, 1995) for $La_{1-x}Sr_xTiO_3$.





Large transfers of spectral weight

FIG. 107. Optical conductivity spectra in $R_{1-x}Sr_xTiO_{3+y}$ or $R_{1-x}Ca_xTiO_{3+y}$ (R=La, Nd, Sm, and Y). From Katsufuji, Okimoto, and Tokura, 1995.