Emergence of Quantum Phases in Novel Materials

Mott physics

E. Bascones

Teoría y Simulación de Materiales
Instituto de Ciencia de Materiales de Madrid

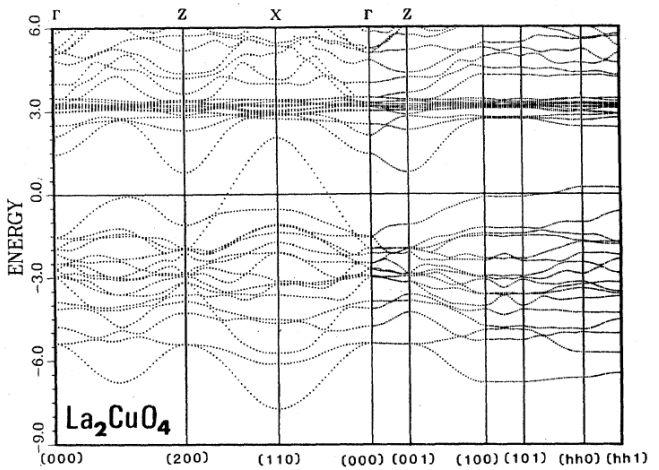
Some references

- Lecture notes on Electron Correlations and Magnetism, Patrik Fazekas, World Scientific Publishing Company
- A. Georges et al, Review Modern Physics 68, 13 (1996)
- A. Georges, arXiv:0403123

Basic concepts: metals and insulators

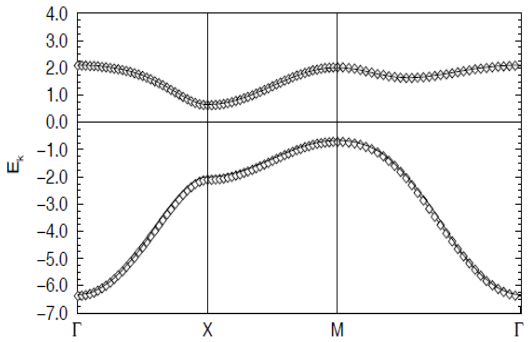
Spin degeneracy:
Each band can hold 2 electrons per unit cell

Odd number of electrons per unit cell → Metallic

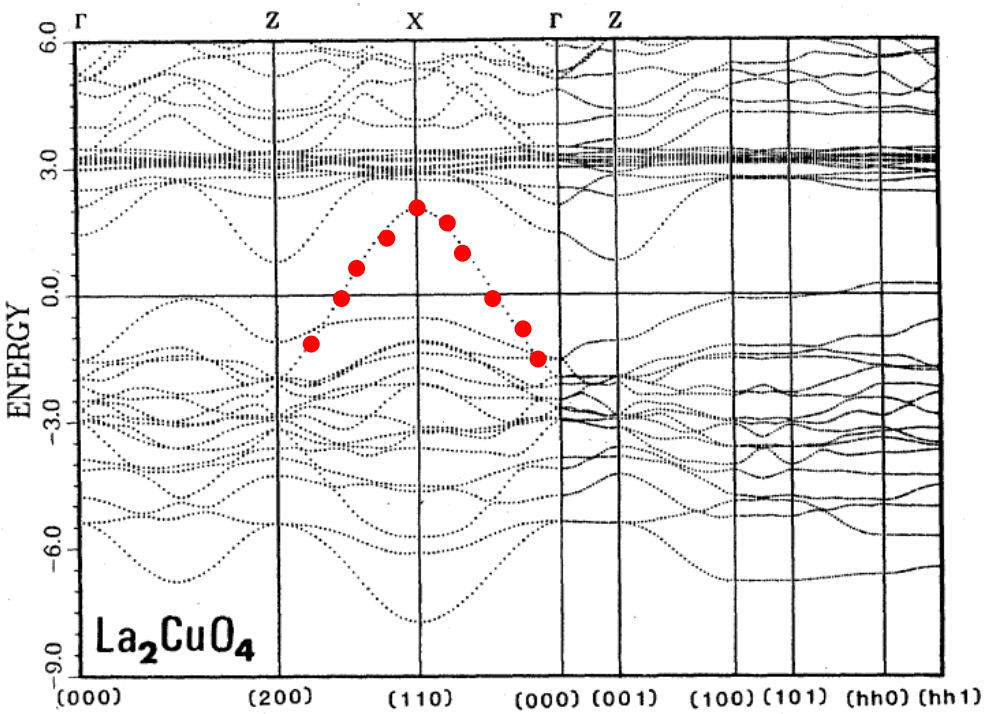


Even number of electrons per unit cell → Insulating

Even number of electrons per unit cell → Metallic (in case of band overlap)



Metals and Insulators. Mott insulators



Metallic behavior expected

Insulating behavior found

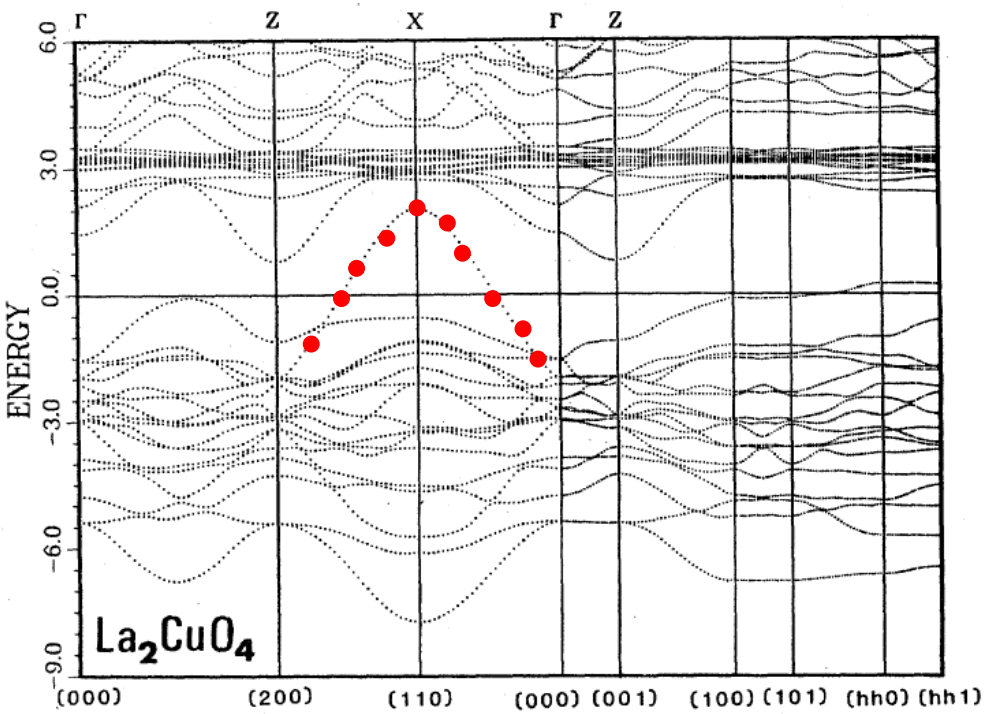
Fig: Pickett, RMP 61, 433 (1989)

Electron counting

La_2CuO_4: 2 La (57x2)+Cu (29) + 4 O (4x8)=175 electrons

Breakdown of independent electron picture

Metals and Insulators. Mott insulators



Metallic behavior expected

Insulating behavior found

Fig: Pickett, RMP 61, 433 (1989)

Mott insulator:

Insulating behavior due to electron-electron interactions: localization of electrons

Not all insulators due to electron-electron interactions are Mott insulators

Not to be confused with Anderson localization which is due to disorder

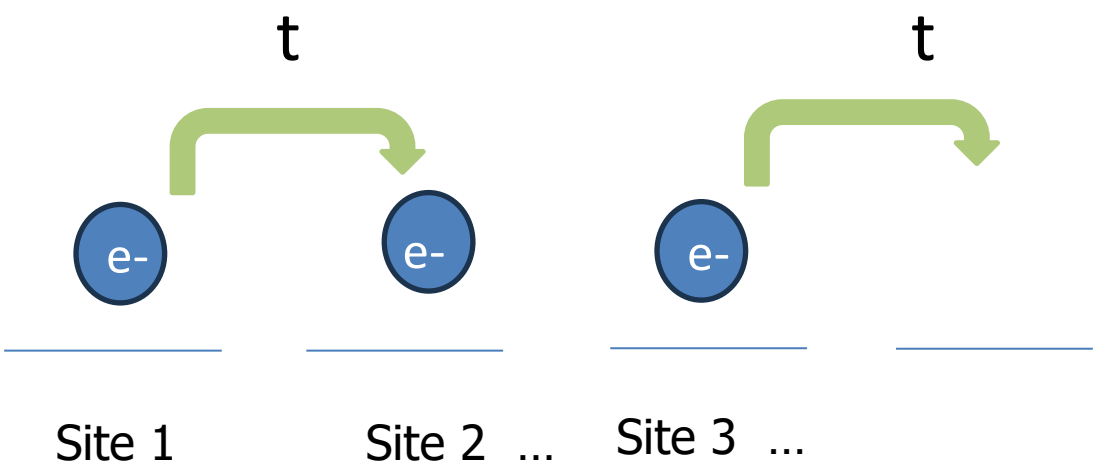
Kinetic energy. Delocalizing effect

$$\sum_{i,j,\gamma,\beta,\sigma} t_{i,j}^{\gamma,\beta} c_{i,\gamma,\sigma}^{\dagger} c_{j,\beta,\sigma} + h.c.$$

Atomic orbital

spin

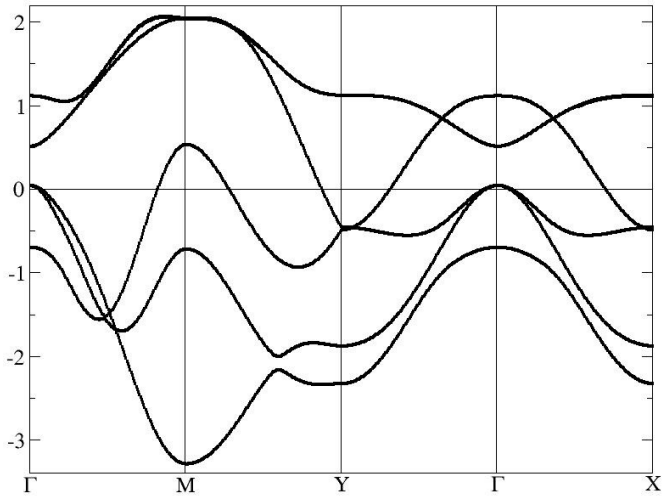
atomic site (i,j)



Kinetic Energy: going from one site to the other

Delocalizing effect

Kinetic energy only:
Bands. Rigid band shift



Kinetic and Interaction Energy. Hubbard model

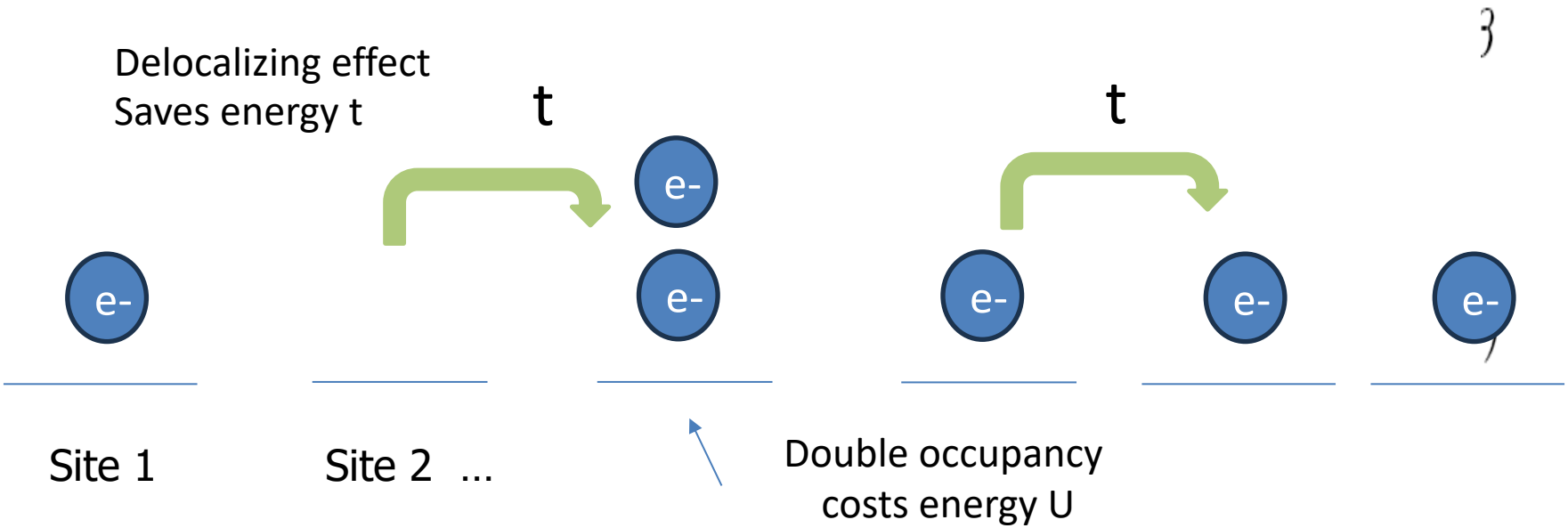
$$H = \sum_{i,j,\sigma} t_{i,j} c^{\dagger}_{i,\sigma} c_{j,\sigma} + h.c. + U \sum_{j} n_{j,\uparrow} n_{j,\downarrow}$$

Kinetic energy

Repulsion between electrons in the same site (Hubbard)

Atomic lattice with a single orbital per site and average occupancy 1 (half filling)

Delocalizing effect
Saves energy t



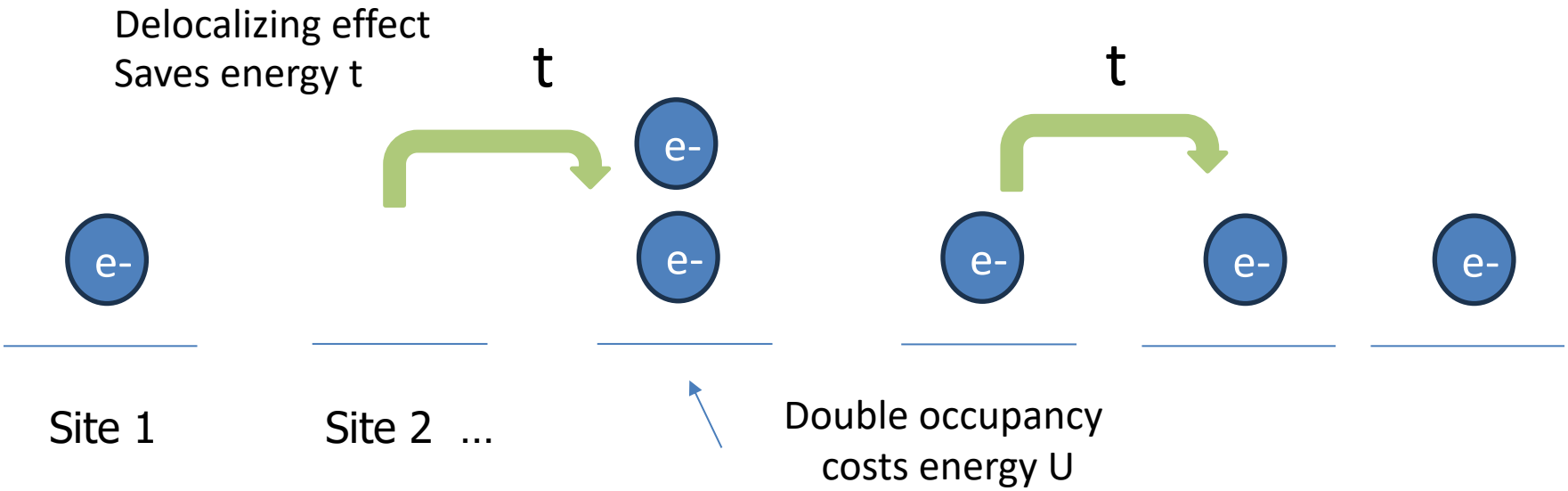
Kinetic and Interaction Energy. Hubbard model

$$H = \sum_{i,j,\sigma} t_{i,j} c^{\dagger}_{i,\sigma} c_{j,\sigma} + h.c. + U \sum_{j} n_{j,\uparrow} n_{j,\downarrow}$$

Kinetic energy

Repulsion between electrons in the same site (Hubbard)

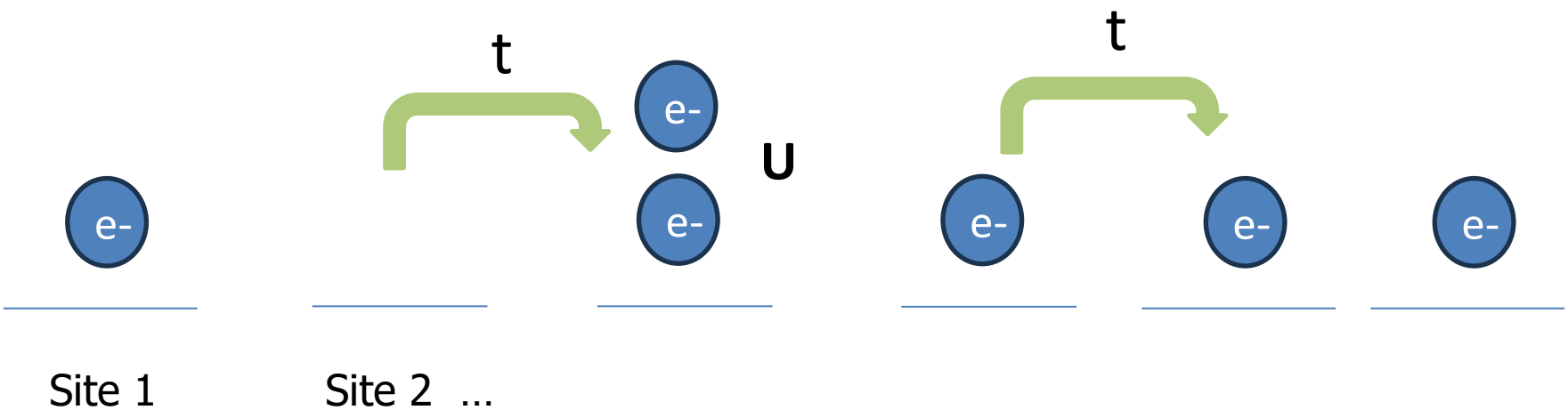
Atomic lattice with a single orbital per site and average occupancy 1 (half filling)



For U >> t electrons localize: Mott insulator

The Mott transition

Hubbard model with a single spin degenerate orbital per site and average occupancy 1: Half filling



For U >> t electrons localize: Mott insulator

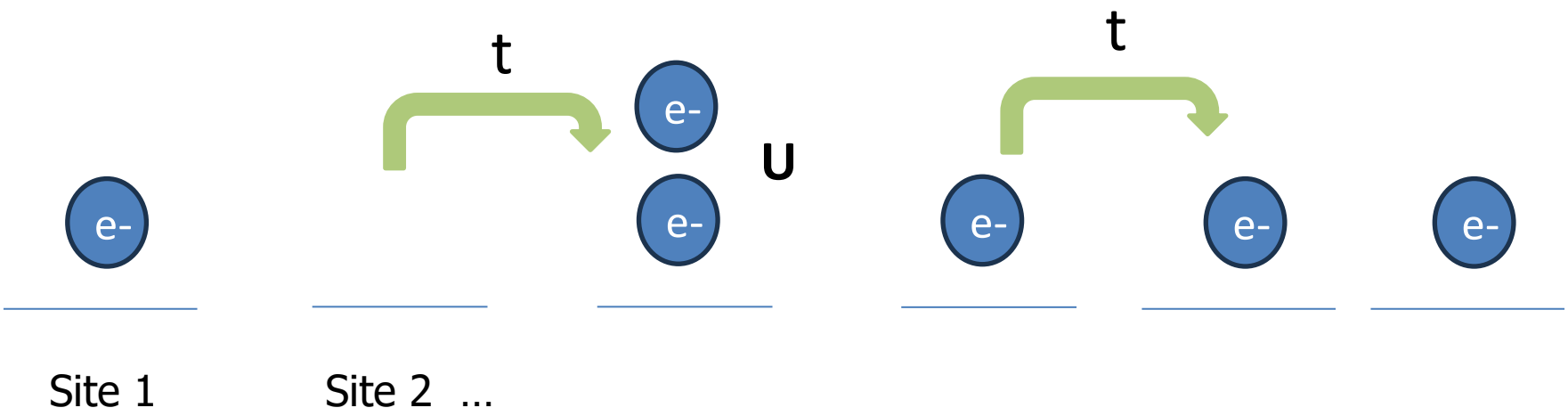
Small U/t — Increasing U/t — Large U/t

Metal → Insulator

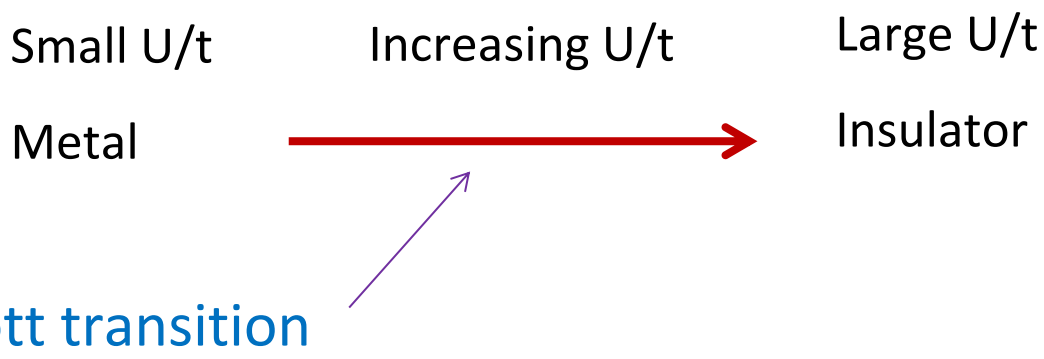
Interaction energy cost due to the hopping process > Kinetic energy saved in the hopping process

The Mott transition

Hubbard model with a single spin degenerate orbital per site and average occupancy 1: Half filling



For U >> t electrons localize: Mott insulator



The Mott transition

Hubbard model with a single spin degenerate orbital per site
and average occupancy 1: Half filling

Site 1 Site 2 ...

- Insulator with odd number of electrons per site (on spite of spin degeneracy)
- No symmetry breaking is required (but it may happen as well)
- Magnetism not used to justify the metal-insulator transition

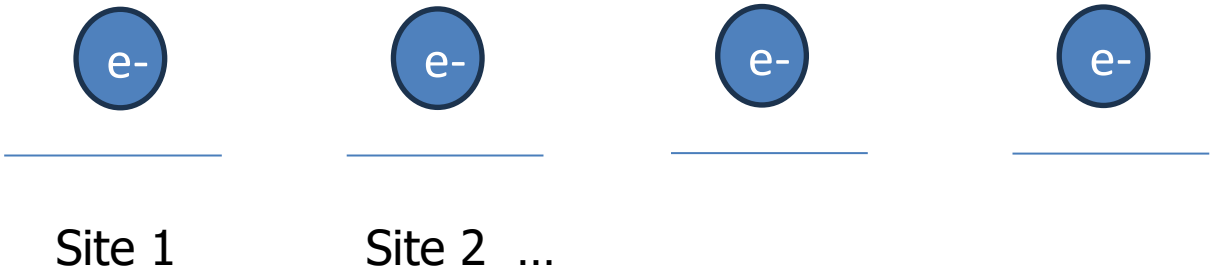
Interaction energy cost due to the hopping process	>	Kinetic energy saved in the hopping process

Electrons become localized

Doping a Mott insulator. Metallicity

Onsite repulsión U

Hopping energy t



1 electron per site/atom

U>>t Mott Insulator

Doped Mott insulator

Metallic, but correlated

Mott insulator: Charge localization

Onsite repulsión U

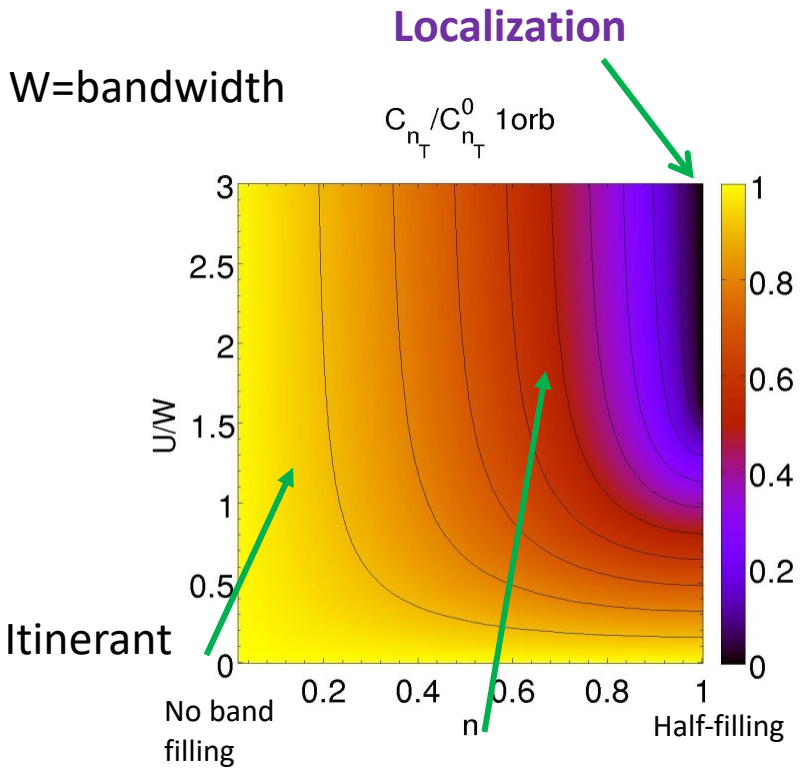
Bandwidth W

$C_T = \langle n^2 \rangle - \langle n \rangle^2 = \langle (\delta n)^2 \rangle$

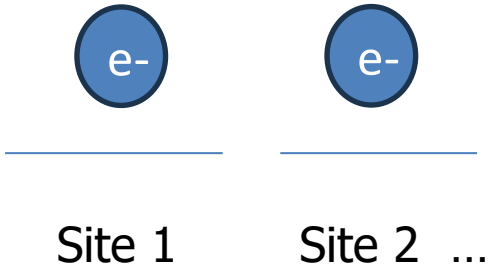
$n = \langle n \rangle + \delta n$

Fluctuations of the charge at the atom

Vanishes if electrons are localized



Charge partially localized
(correlated metal)



The electrons become progressively localized as the Mott transition is approached
(with increasing interaction or approaching half filling)

Fig: Fanfarillo & EB

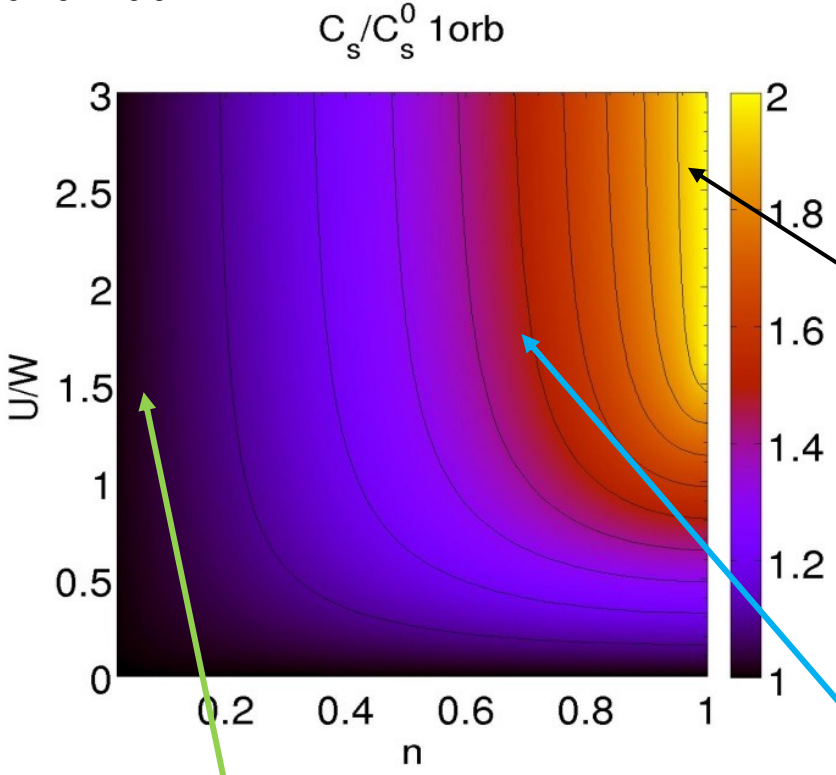
Mott insulator: Formation of local moments

Onsite repulsión U

Bandwidth W

$C_S = \langle S^2 \rangle - \langle S \rangle^2 = \langle S^2 \rangle$

W=bandwidth



C_s larger when atoms are spin polarized even if there is no long range order

At the Mott insulator each atom
Is maximally polarized
(in a paramagnetic sense
do not confuse with long range order)

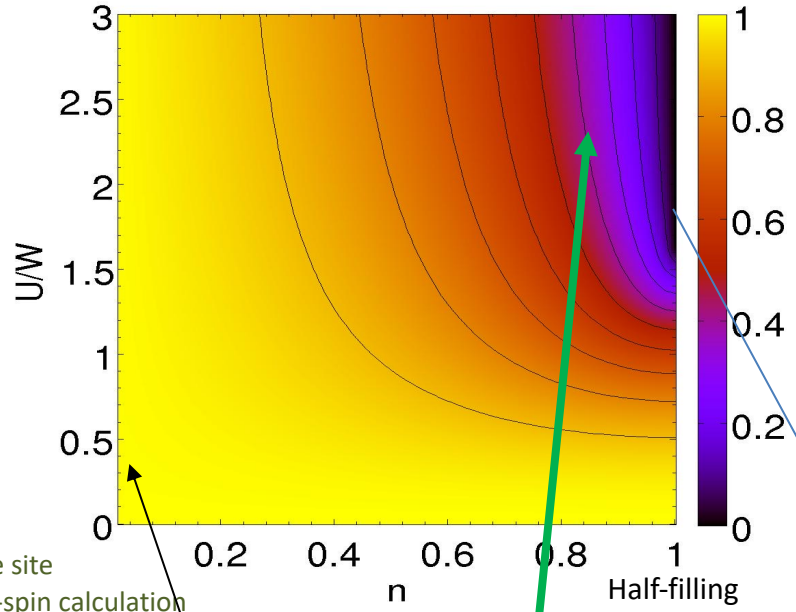
No local moment if interactions are weak or large doping away from half-filling

Magnetic moments only partially formed

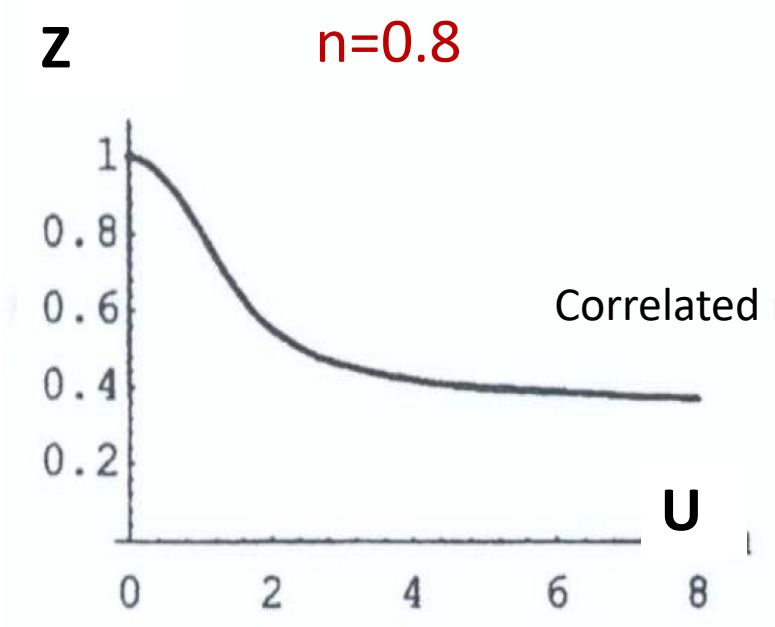
Fig: Fanfarillo & EB

Doping a Mott insulator. Correlated metal. Quasiparticle weight

Z colour plot for the Hubbard model



Single site slave-spin calculation for 2D dispersion
Fig: Fanfarillo & EB



Gutzwiller Approximation for Constant Density of States

Fig: Fazekas

Z ~1 small interaction or large doping away from half-filling

Small Z: correlated metal

Z=0 at the Mott insulator

W=bandwidth

Small quasiparticle weight Fermi liquid behavior restricted to very low temperatures and energies when Z is small

Different behavior in Fermi liquid and Mott insulator

Metal (Fermi liquid)

Resistivity increases with temperature

$$\rho \sim \rho_0 + A\,T^2$$

$$A \sim m^{*2}$$

Specific heat linear with temperature

$$C \sim \gamma\,T \qquad \gamma \sim m^*$$

Magnetic susceptibility
does not depend on temperature

$$\chi \sim \chi_0 \qquad \chi_0 \sim m^*$$

Mott insulator

Resistivity decreases with temperature

Specific heat activated like behavior

$$C \sim e^{-\Delta/KT} \quad \text{(gap)}$$

Magnetic susceptibility inversely
proportional to temperature

Paramagnetic
Curie-Weiss
behavior

$$\chi \sim \chi_0 + C'/(T+\Theta)$$

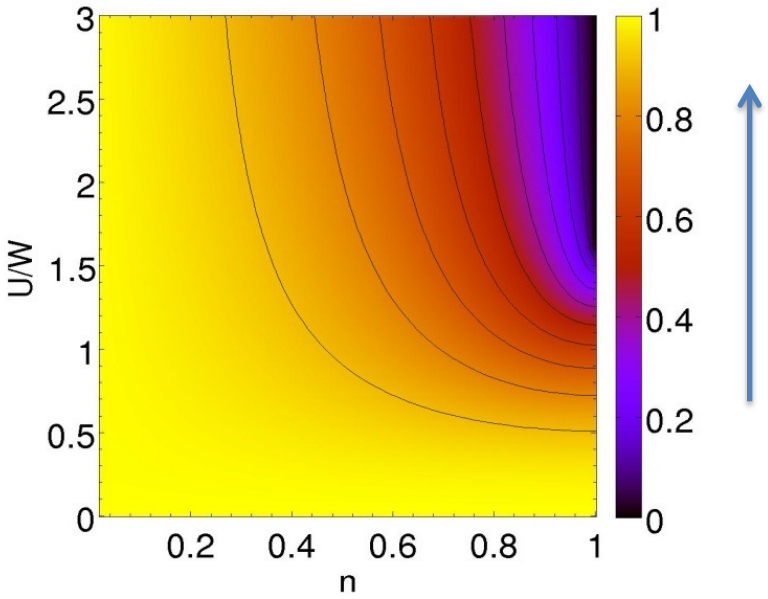
Doping a Mott insulator. Correlated metal

Correlated metallic state. Large U

$m^* \sim m/Z$ → Mass enhancement (divergence) the Mott insulator is approached

Mass is enhanced when we approach half-filling

W=bandwidth

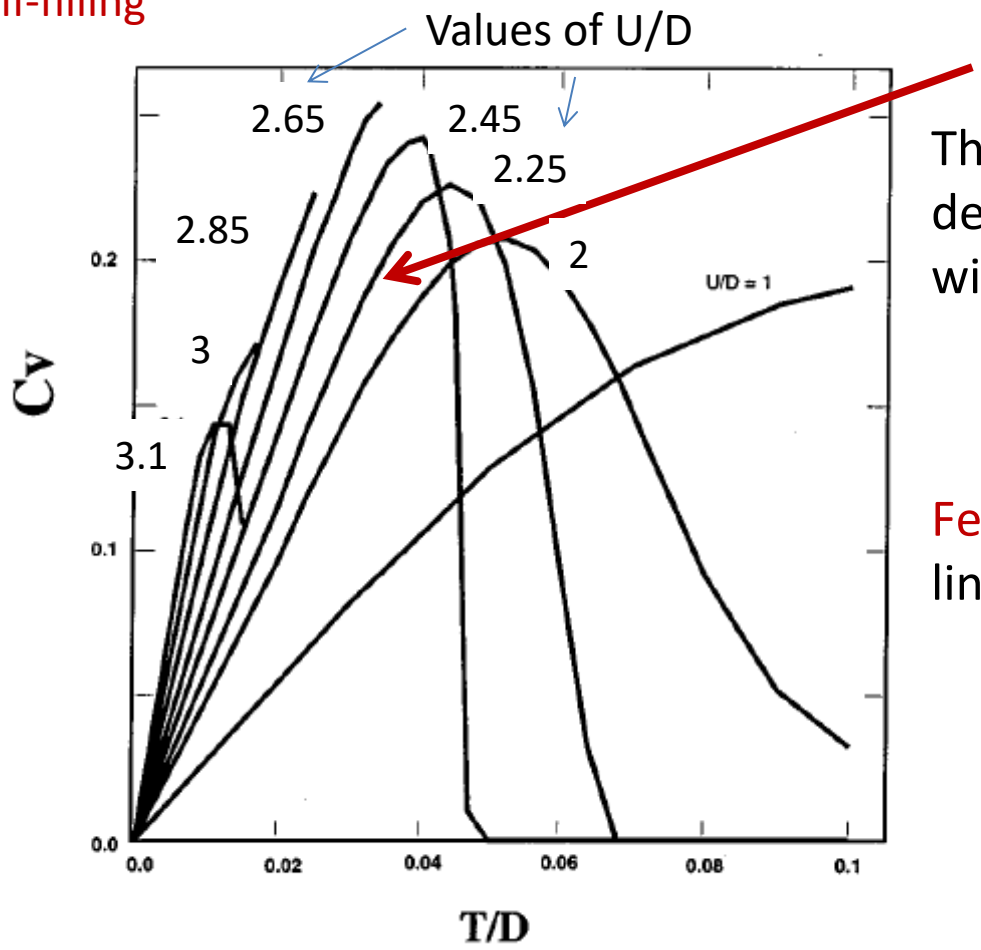


Mass is enhanced when U increases

Small quasiparticle weight → Fermi liquid behavior restricted to very low temperatures and energies when Z is small

The Mott transition. Finite temperatures

Half-filling



The slope of the linear T dependence increases with interactions

Fermi liquid: Specific heat linear with temperature

$C \sim \gamma T \qquad \gamma \sim m^*$

Mass enhanced with interactions

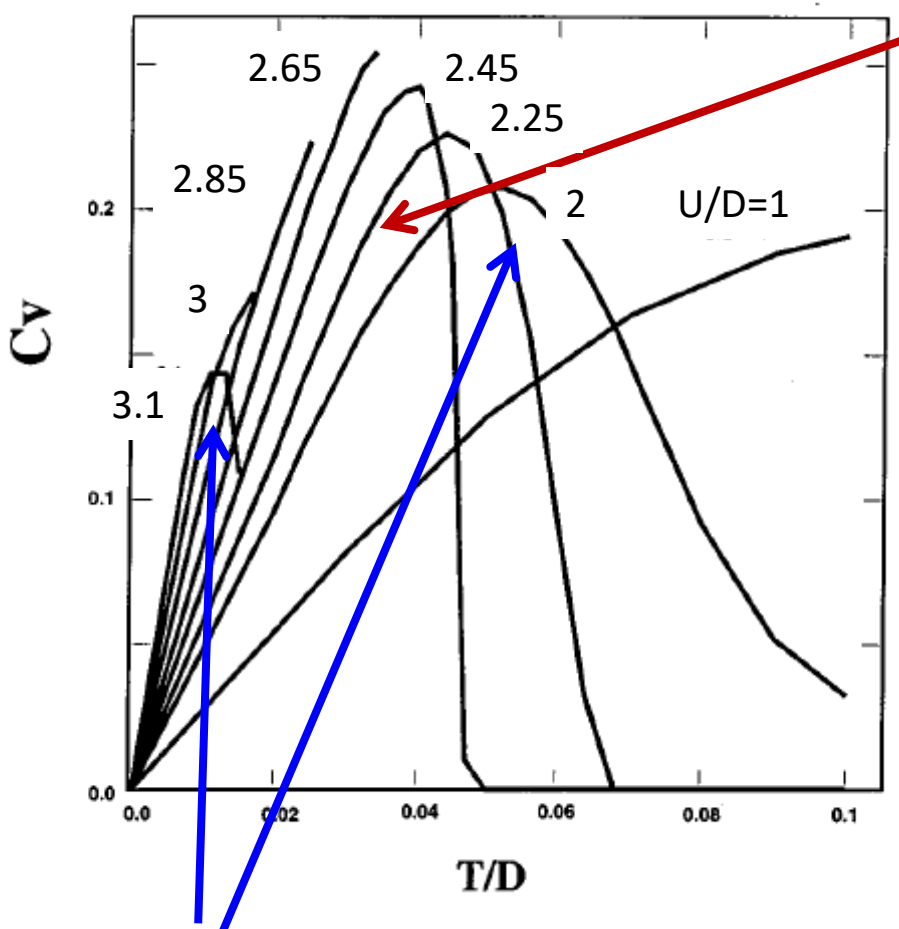
DMFT calculations for the single orbital Hubbard model at half-filling

D=half-bandwidth

Georges et al , RMP 68, 13 (1996)

The Mott transition. Finite temperatures. Single-site DMFT

Fermi liquid behavior observed at low temperatures



The slope of the linear T dependence increases with interactions

Fermi liquid: Specific heat linear with temperature

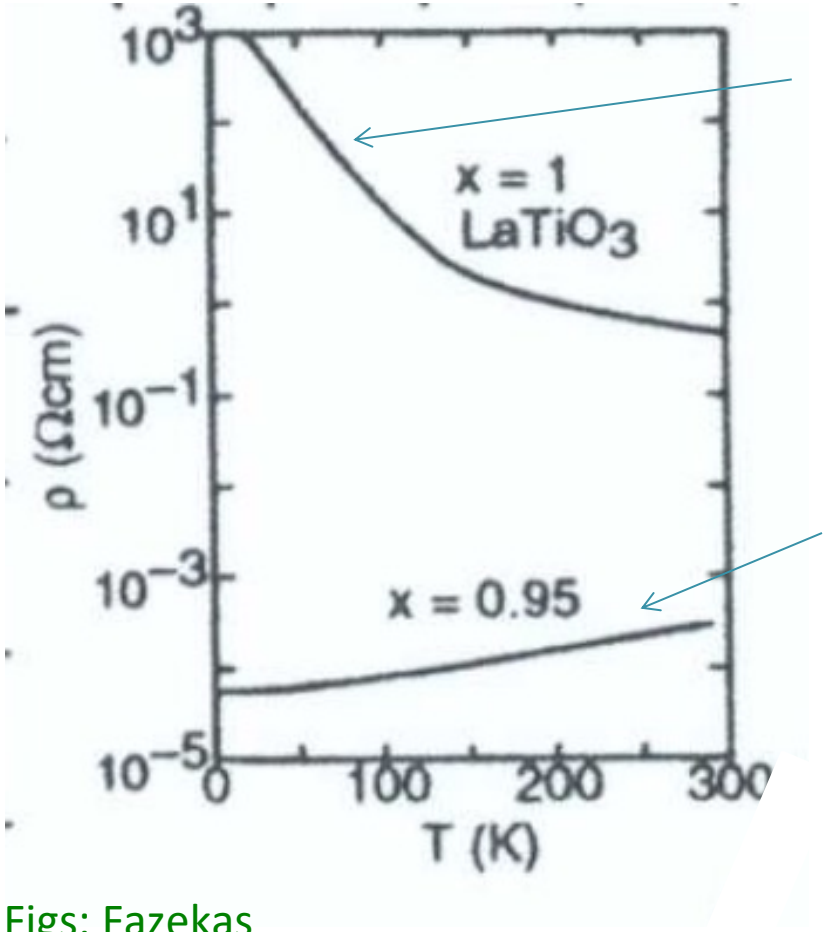
$C \sim \gamma T \qquad \gamma \sim m^*$

Mass enhanced with interactions

Linearity is lost at a temperature which decreases with increasing interactions

Georges et al , RMP 68, 13 (1996)

Doping a Mott insulator. Correlated metal



Figs: Fazekas

Undoped: Mott Insulator

Resistivity decreases with temperature

Doped: Metal

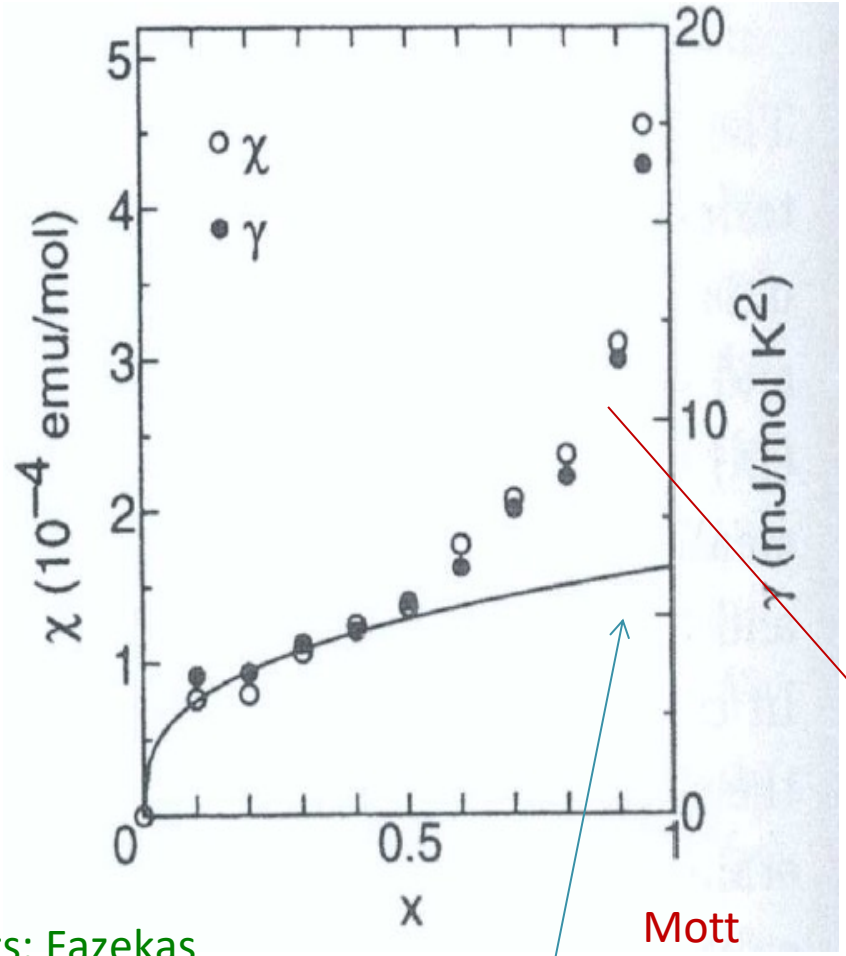
Resistivity increases with temperature

$$La_xSr_{1-x}TiO_3$$

Insulator to Metal transition with doping

This example corresponds to a Mott insulator but not to a single orbital Hubbard model

Doping a Mott insulator. Correlated metal



Figs: Fazekas

$La_xSr_{1-x}TiO_3$

Metal (Fermi liquid)

Specific heat linear with temperature

$C \sim \gamma T \qquad \gamma \sim m^*$

Magnetic susceptibility
does not depend on temperature

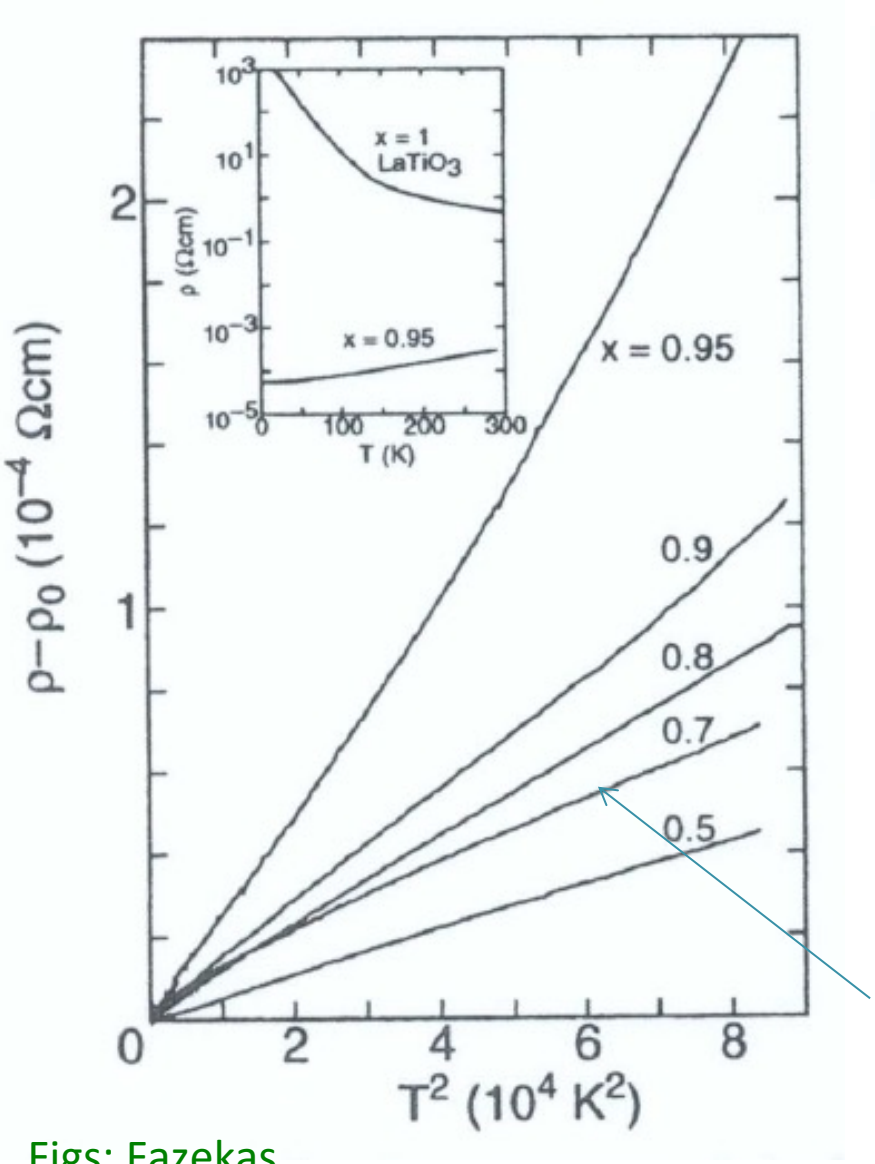
$\chi \sim \chi_0 \qquad \chi_0 \sim m^*$

Mott

Enhancement of the mass
when approaching half-filling

Expected for
independent electrons

Doping a Mott insulator. Correlated metal



Figs: Fazekas

$La_xSr_{1-x}TiO_3$

Metal (Fermi liquid)

Resistivity increases with temperature

$\rho \sim \rho_0 + A\,T^2$

$A \sim m^{*2}$

The coefficient of the T^2 law increases as we approach half-filling (enhancement of the mass)

This example corresponds to a Mott insulator but not to a single orbital Hubbard model

Quasiparticle weight, charge & spin fluctuations in the Hubbard model

Strength of correlations

Quasiparticle weight Z:

$0 \leq Z \leq 1$

Charge Fluctuations

$C_T = \langle n^2 \rangle - \langle n \rangle^2 = \langle (\delta n)^2 \rangle$

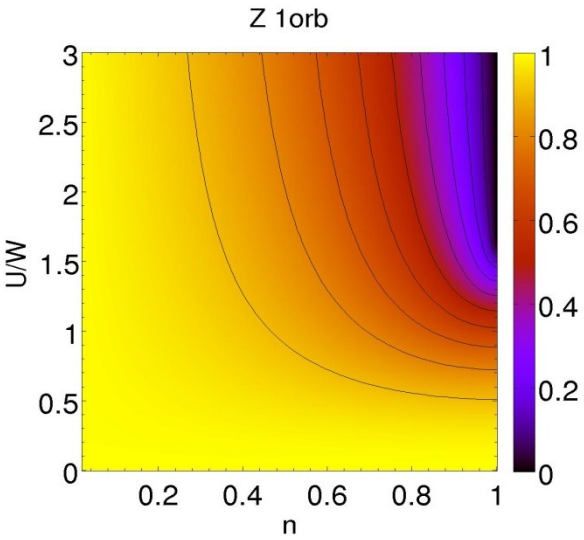
$n = \langle n \rangle + \delta n$

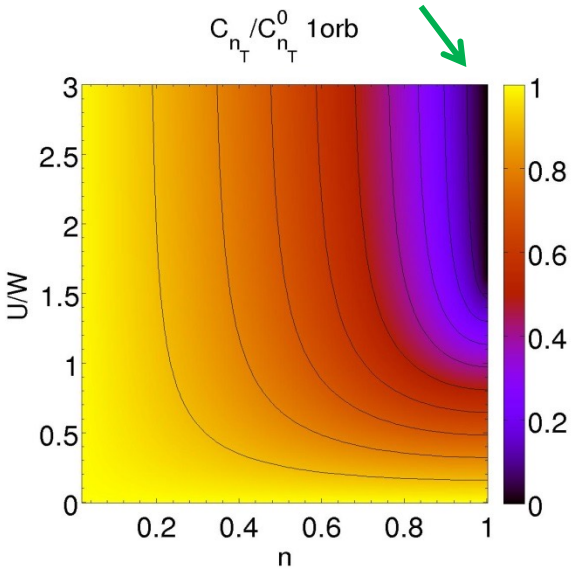
Localization

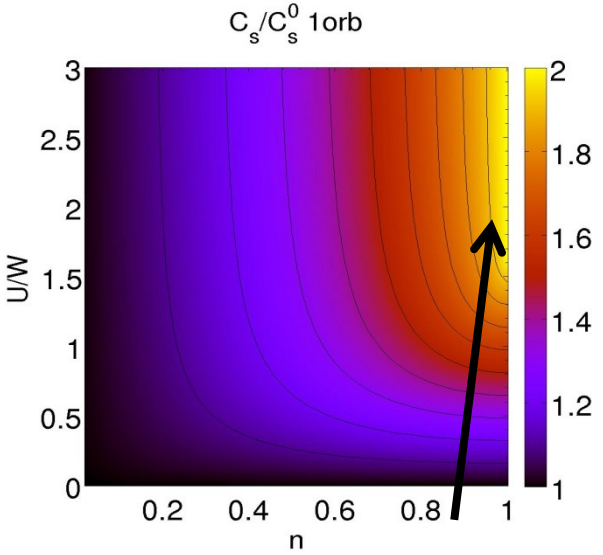
Moment Formation

$C_S = \langle S^2 \rangle - \langle S \rangle^2 = \langle S^2 \rangle$

C_s larger when atoms are spin polarized even if there is no long range order







Polarized atom

In the single orbital Hubbard model, the emergence of correlations is concomitant with the localization of the charge and with the formation of magnetic moments.

Fanfarillo & EB

Doping a Mott insulator. Correlated metal

Correlated metallic state. Large U

Small/Large filling

Metal

Z=1

Changing the filling
Mott transition

0 <Z <1

Half-filling

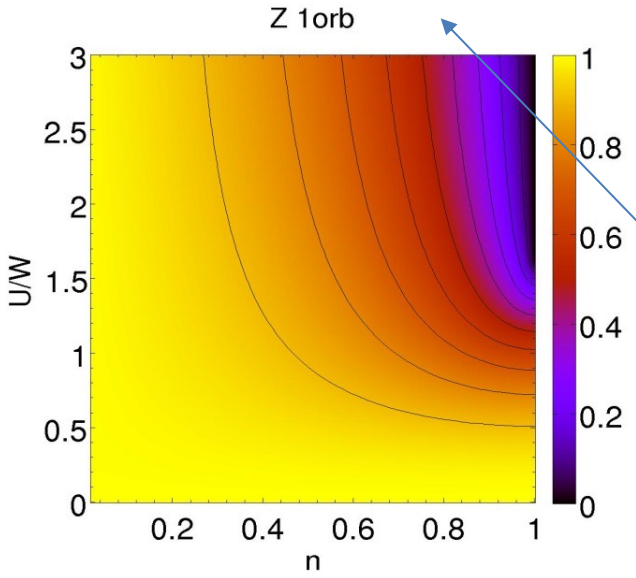
Mott Insulator

Z=0

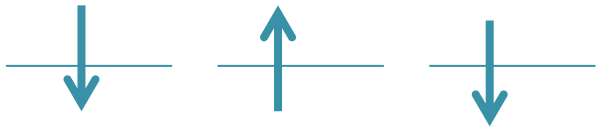
Description in terms of electronic bands.

Fermi surface physics

Color plot Quasiparticle weight Z



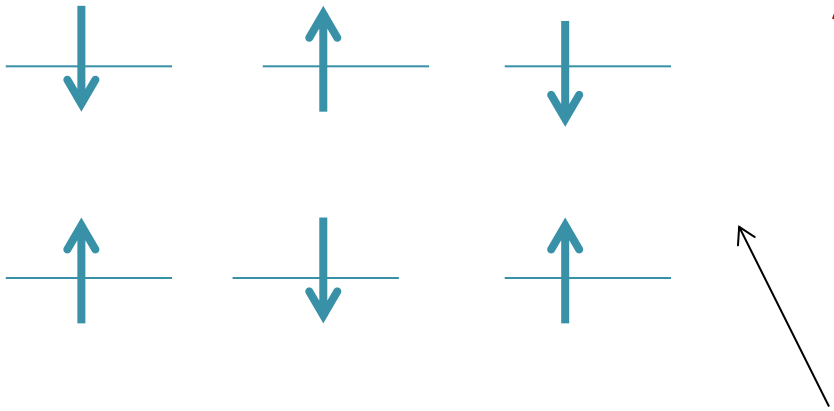
Description as localized spins



The mass diverges

In between better description
In terms of simple effective models is less obvious

Spin models. Tendency towards antiferromagnetism



Antiferromagnetic interactions
between the localized spins
(not always ordering)

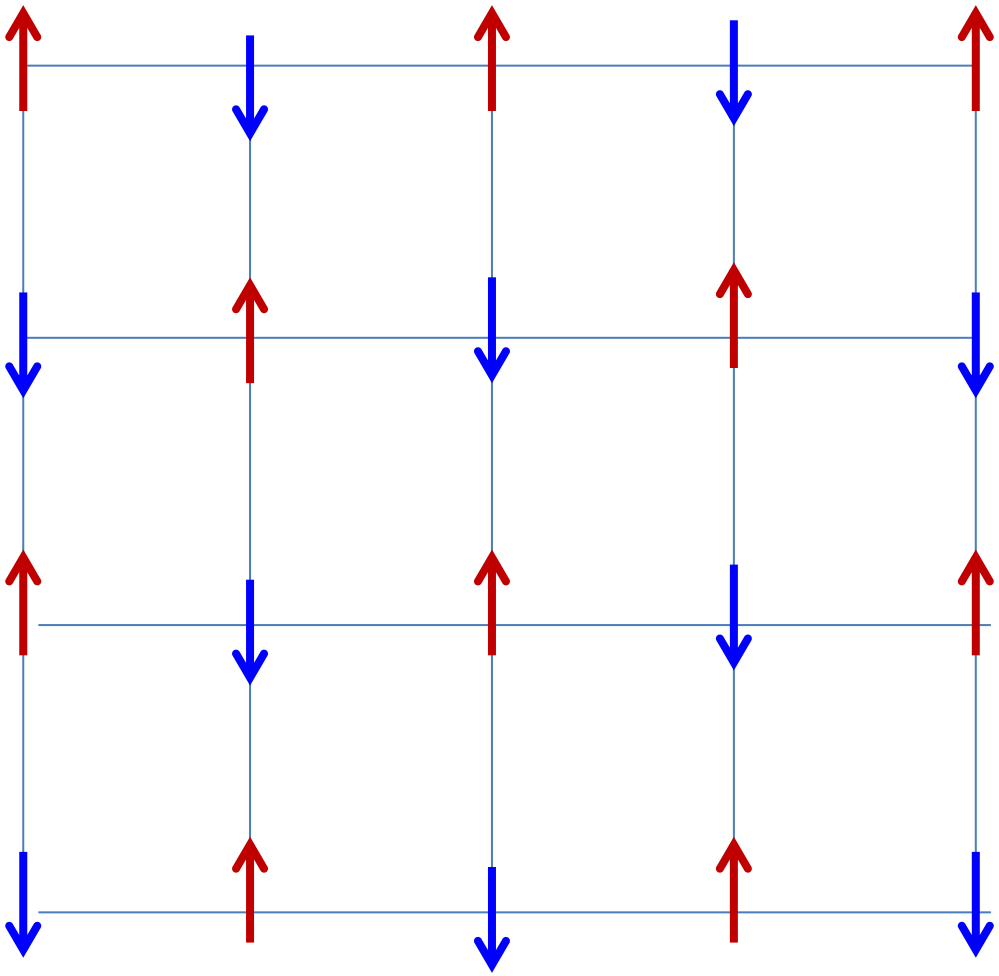
Effective exchange interactions
(second order perturbation theory)

$J \sim t^2/U$ $\quad\quad$ $J \ll U$

$J \rightarrow 0$ in the pure flat band limit

Antiferromagnetic **correlations/ordering** can reduce the energy
of the localized spins. Depends on the degree of frustation of the lattice

Tendency towards antiferromagnetism

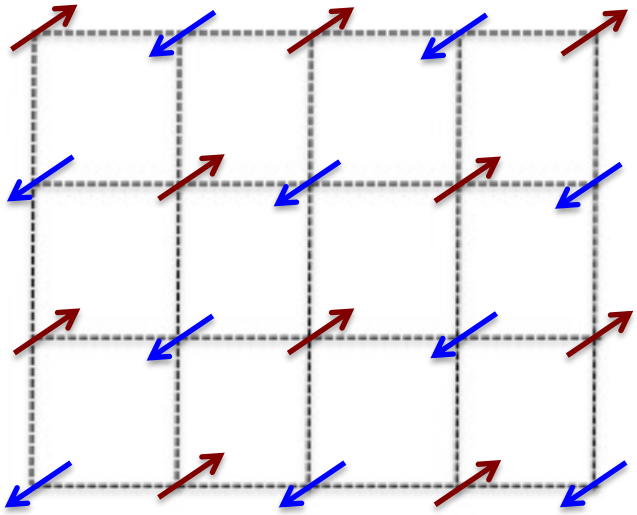


Interaction between
Local spins

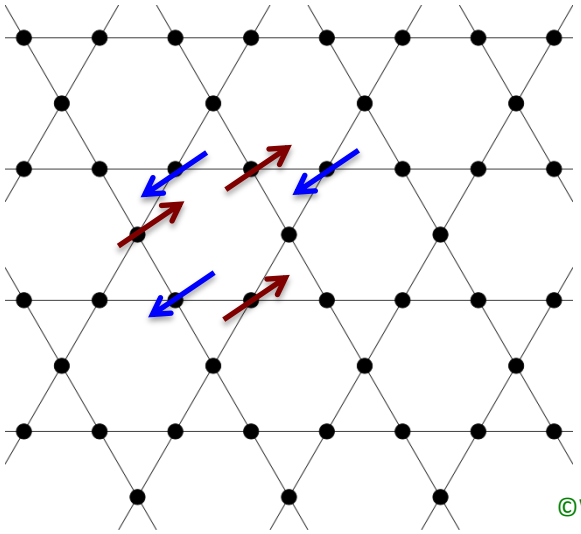
Antiferromagnetic order
in a bipartite lattice

Tendency towards antiferromagnetism

Bipartite lattice



Frustrated lattice



©WilliamSix

Triangular lattice: hopping restricted to first neighbors

Hubbard model in triangular lattice with hopping to many neighbors

120º-AF

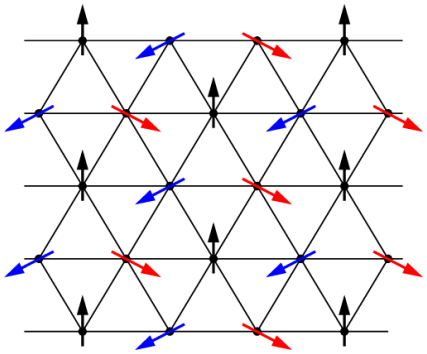


Fig: Chernishev, Zhitomirsky, PRB 79, 144416 (2009)

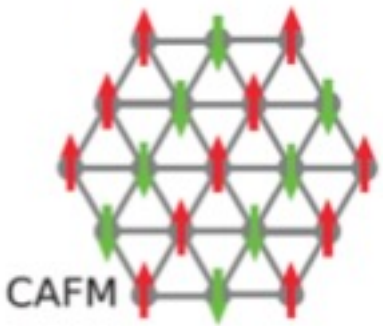
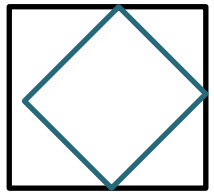


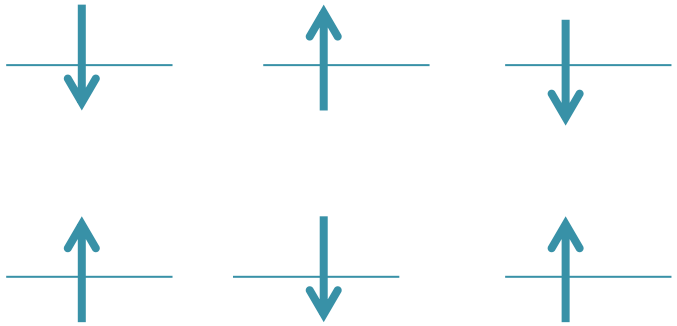
Fig: arXiv: 2007.06086

Slater vs Mott insulators

The shape of the Fermi surface can lead to an antiferromagnetic instability



Antiferromagnetism doubles the unit cell



1 electron per site

↓

2 electrons per unit cell
(even number of electrons/unit cell)

Slater insulators: Insulating behavior associated to unit cell doubling (Antiferromagnetism)

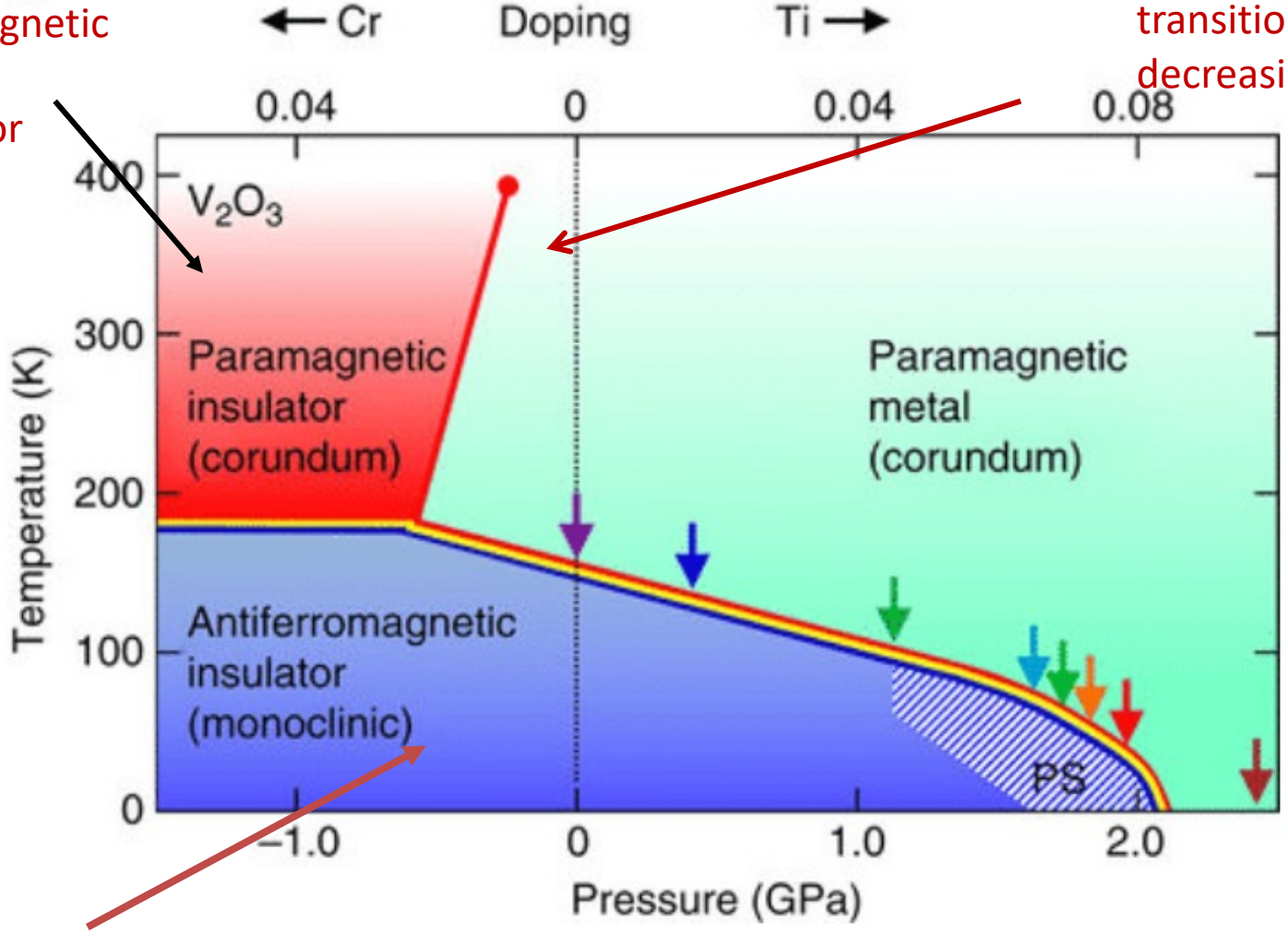
Mott insulators: Insulating behavior does not require AF,
(but the local spins have a strong tendency to order antiferromagnetically)

There are ferromagnetic Mott insulators!

Slater vs Mott insulators

Paramagnetic Mott Insulator

Metal-Insulator transition with decreasing pressure



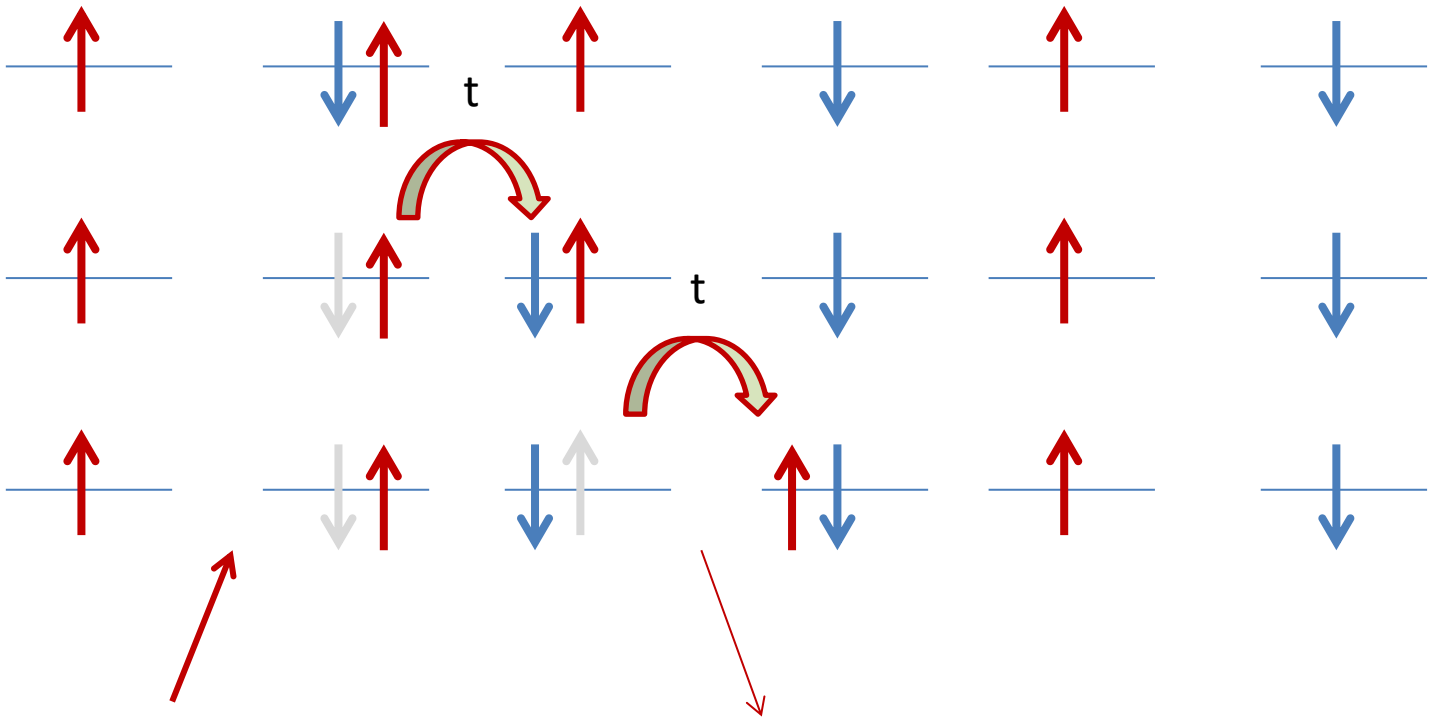
Antiferromagnetism

Increasing Pressure: decreasing U/W

McWhan et al, PRB 7, 1920 (1973)

Doping a Mott insulator. AF state. Disappearance of AF

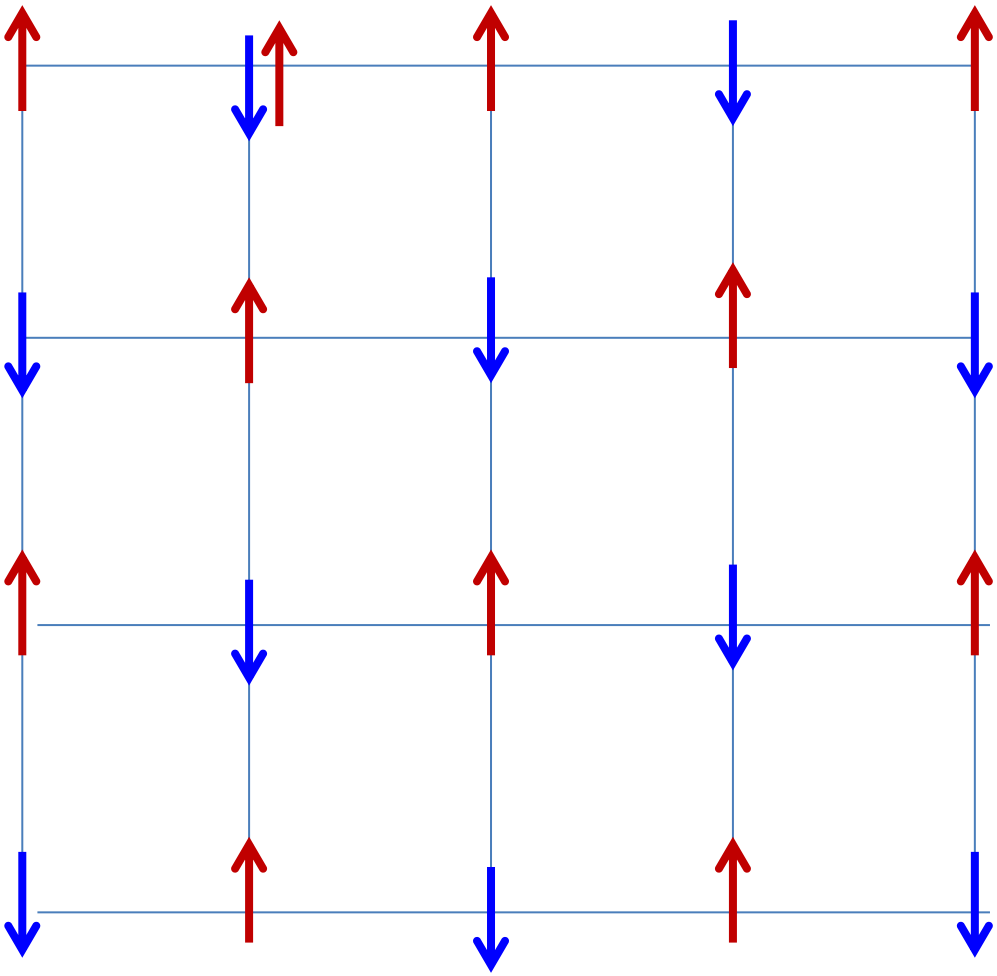
Doping a Mott insulator. AF state. Disappearance of AF



Antiferromagnetic background is distorted & eventually destroyed

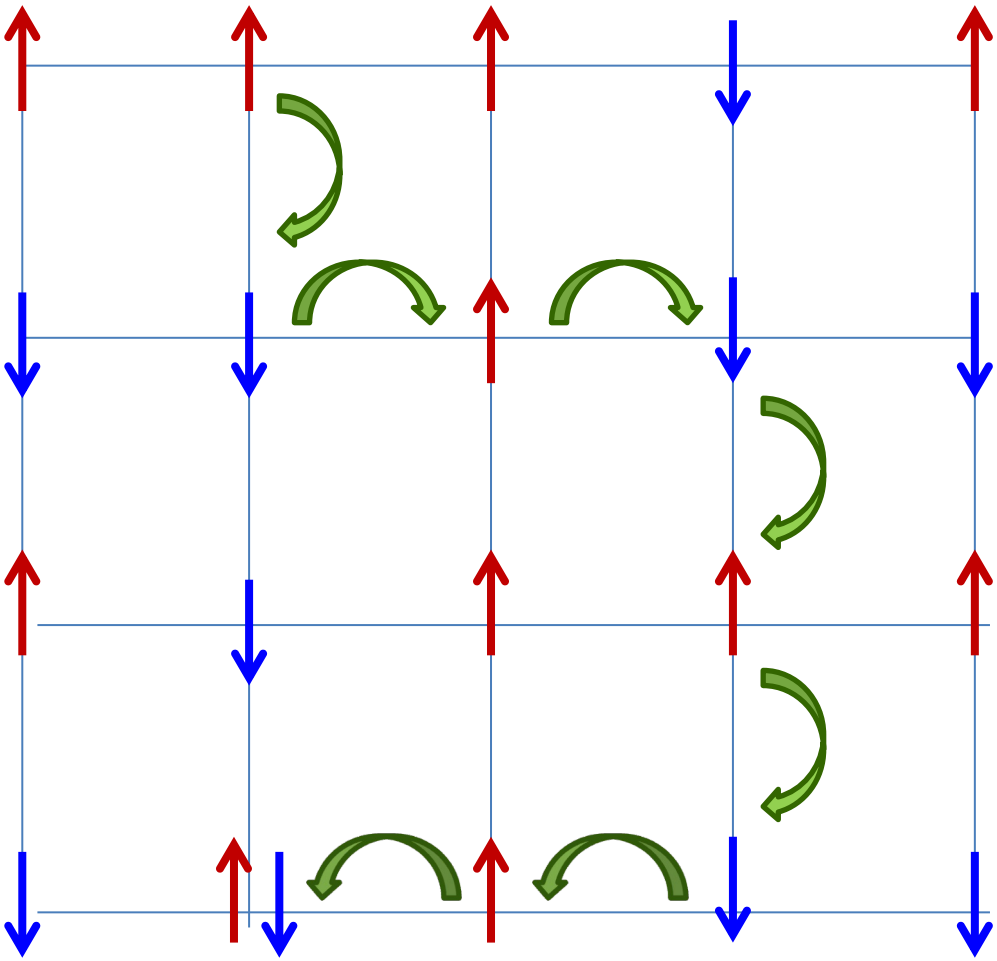
Mass enhanced by AF correlations
AF background tends to localize the electron (Spin bags)

Doping a Mott insulator. AF state. Disappearance of AF



LOCAL
Magnetism

Doping a Mott insulator. AF state. Disappearance of AF



Mass enhanced
by AF correlation
Tendency to
Localization due to AF
(Spin bags)

Antiferromagnetic ordering is destroyed by doping

Spectrum in the atomic limit

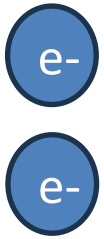
Consider 1 atom with a single orbital $U \sum_j n_{j\uparrow} n_{j\downarrow}$

E =0

Atom (site)

1 electron
(two possible states up or down)

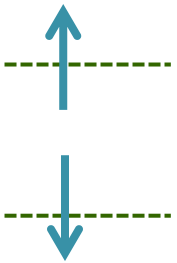
2 electron



E =U

Atom (site)

To add a second electron to single filled orbital costs energy U
Energy depends on the occupancy
Non rigid band shift

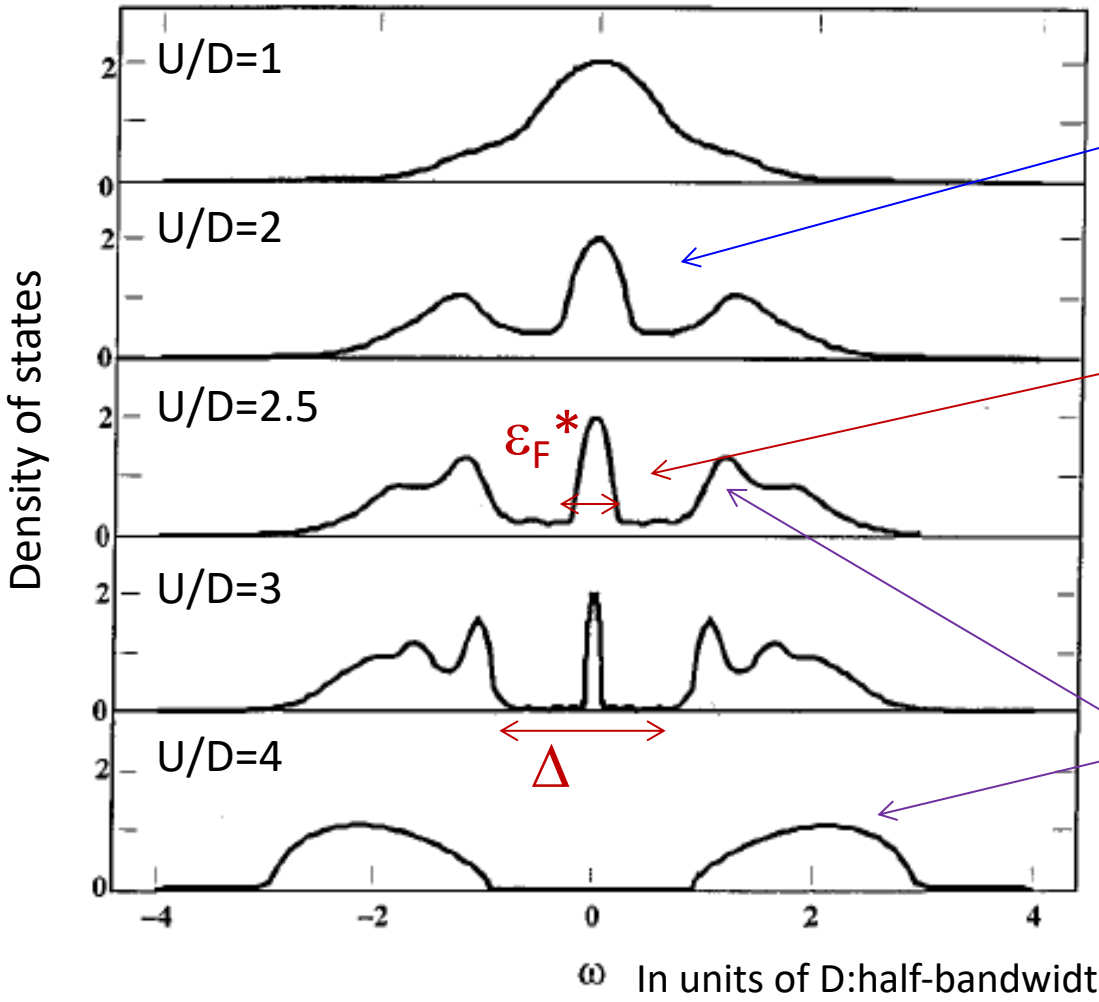


Mott transition. Non magnetic state. (single site) DMFT picture

Georges et al , RMP 68, 13 (1996)

Infinite dimensions

Numerical technique



Three peak structure

Heavy quasiparticles (coherent)

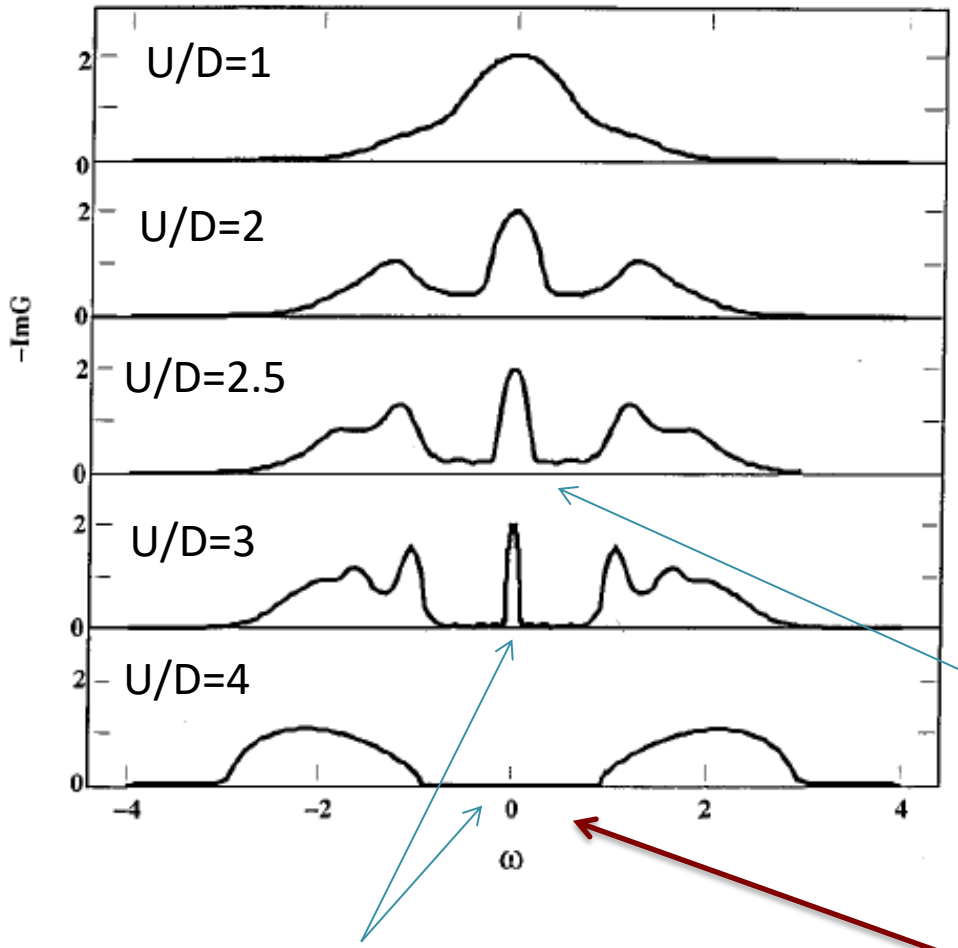
Hubbard bands (incoherent or continuum)

Two energy scales: ε_F^* and the gap Δ between the Hubbard bands (gap of Mott insulator)

ε_F^* control the energy range in which we expect to find Fermi liquid behavior

Mott transition. Paramagnetic state. DMFT picture

Infinite dimensions



Transfer of spectral weight from the quasiparticle peak to the Hubbard bands with increasing interactions

The gap between the Hubbard bands opens in the metallic state
Correlated metal

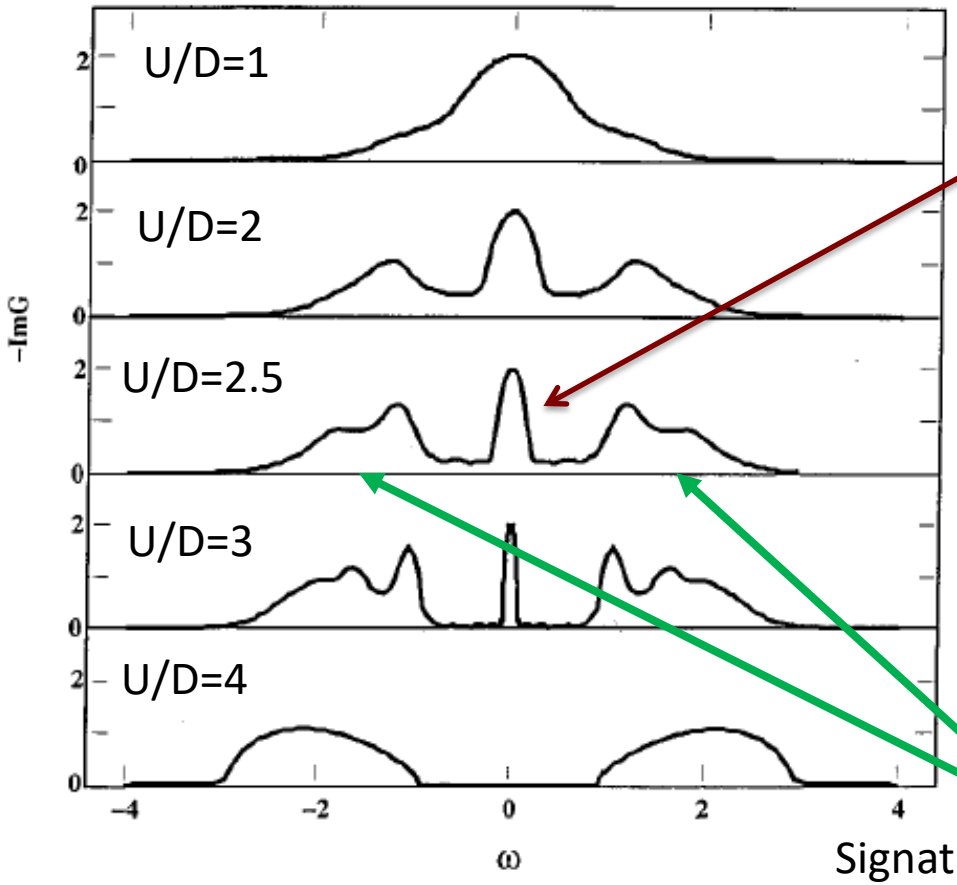
Quasiparticles disappear at the Mott transition

Gap between Hubbard bands ~W, U
Large gap

Georges et al , RMP 68, 13 (1996)

Mott transition. Paramagnetic state. DMFT picture

Infinite dimensions



Georges et al , RMP 68, 13 (1996)

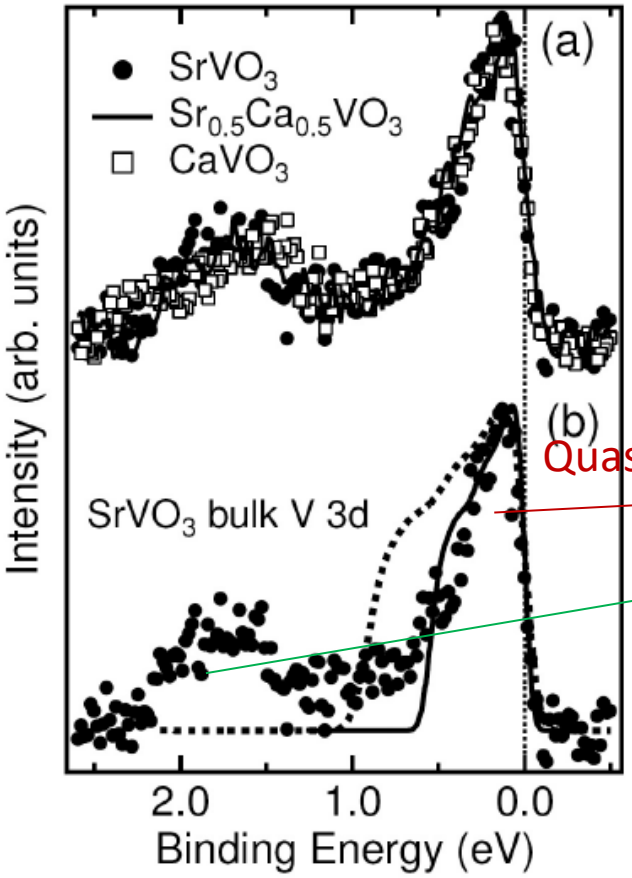
Correlated metal

Quasiparticle peak
Signature of metal
(at very low T and energy Fermi liquid behavior)

Hubbard bands
Signature of localized electrons (local moments)
(at large T and energy non-Fermi liquid)

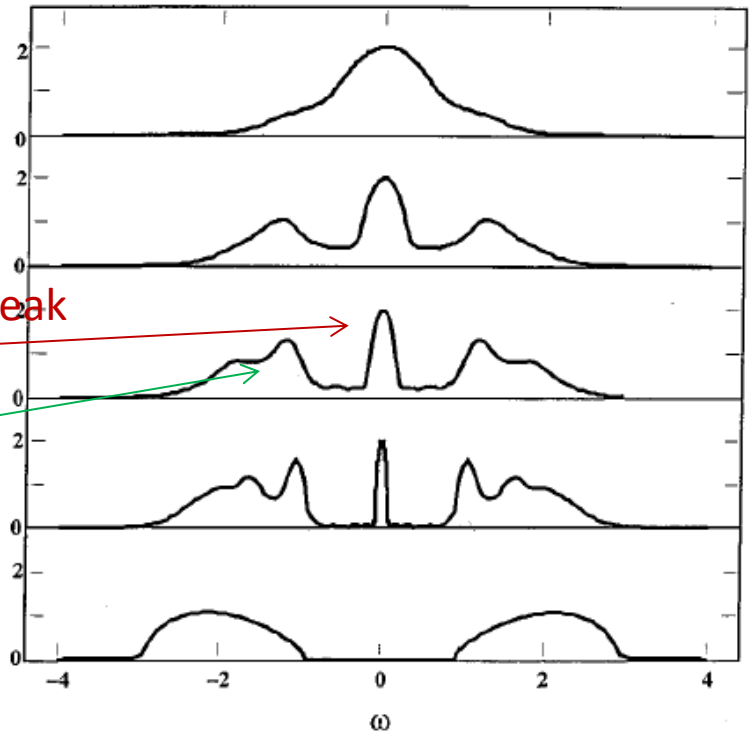
Moreover the spectrum is going to be modified
If the temperatura increases

Mott transition. Paramagnetic state. DMFT picture



Quasiparticle peak

Hubbard band



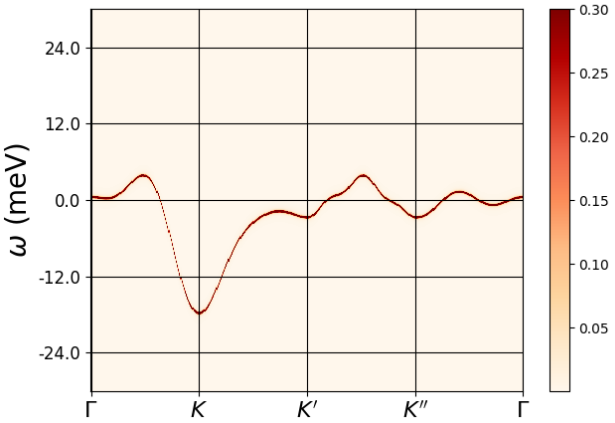
Photoemission

Sekiyama et al, arXiv:0206471; Georges, arXiv:0403123

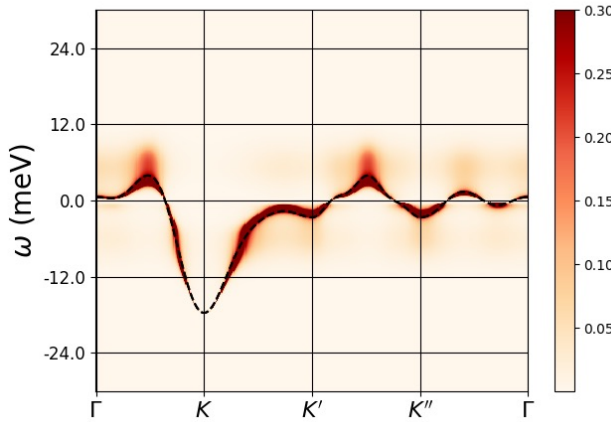
Correlated bands. Non magnetic states

Uc: Interaction at which the Mott transition takes place

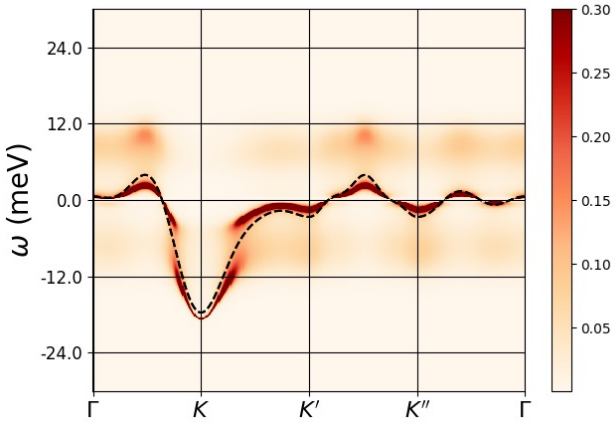
Non-interacting band n=2



U= 0.18 Uc

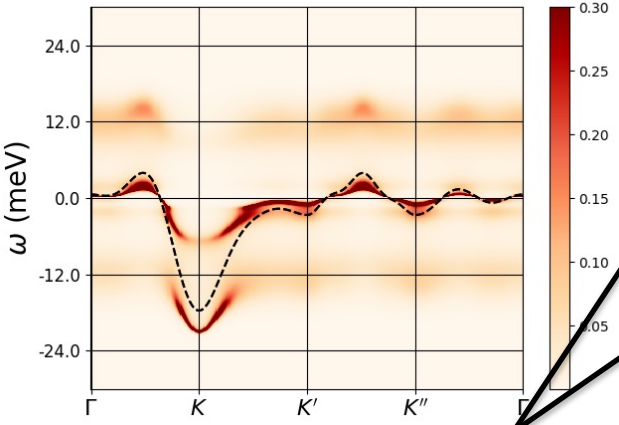


U= 0.36 Uc

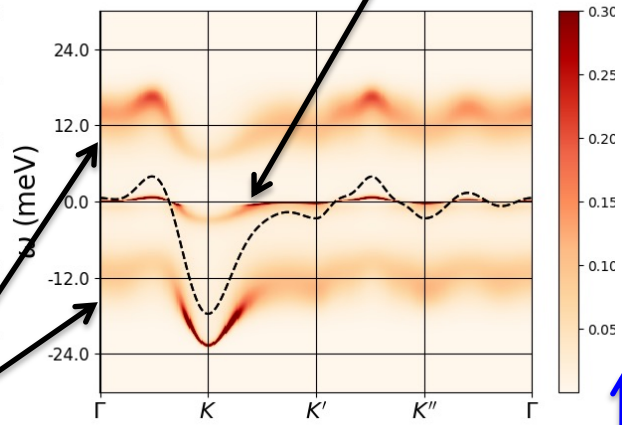


Strongly renormalized quasiparticle band

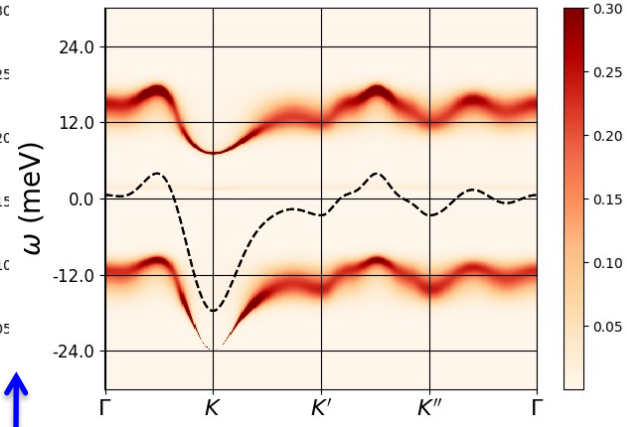
U= $U_c/2$



U= 0.82 Uc



U=1.07 Uc



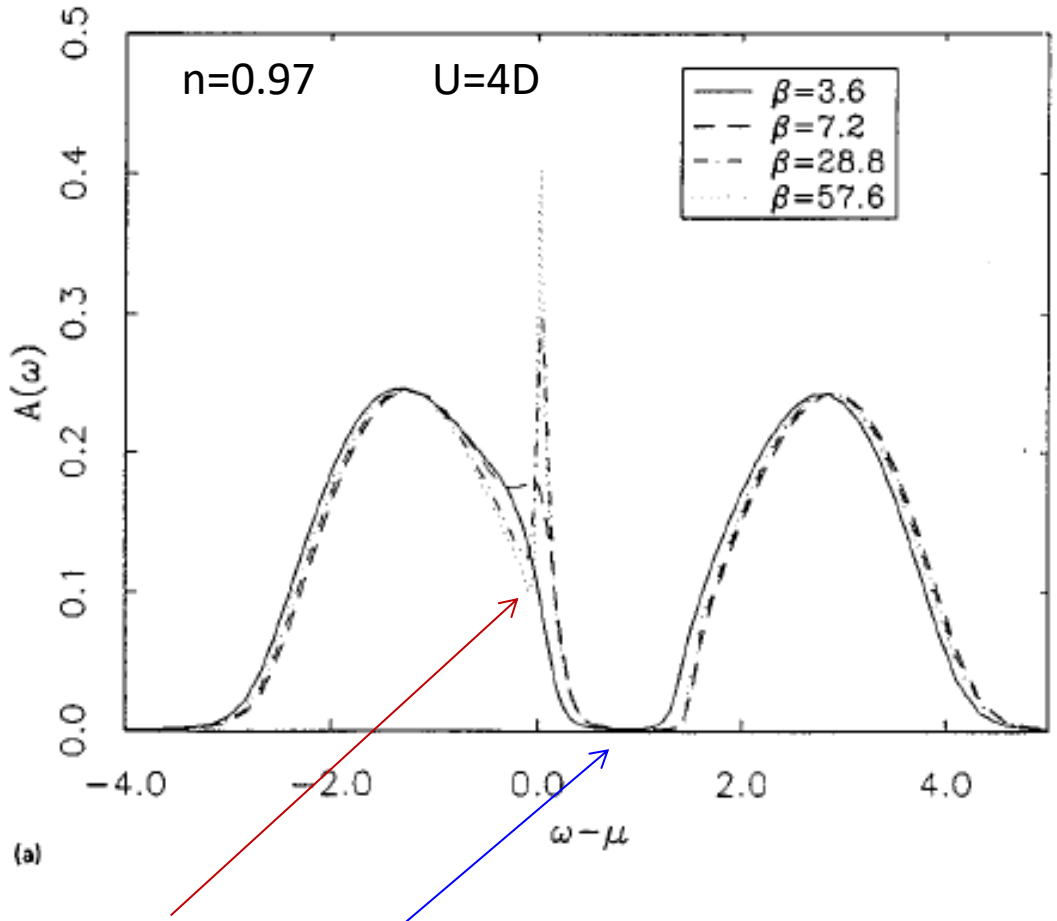
Hubbard bands (incoherent)

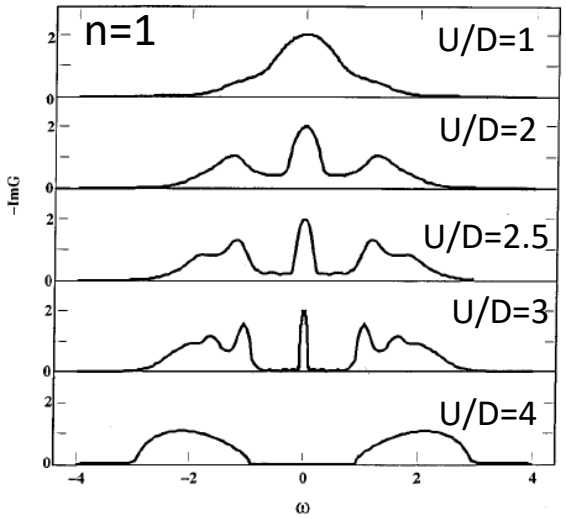
U_{Mott}

This is not just a simple gap opening

Two-orbital Hubbard model at half-filling Calderón, Camjayi & EB, (2023)

Doping a Mott insulator. Correlated metal

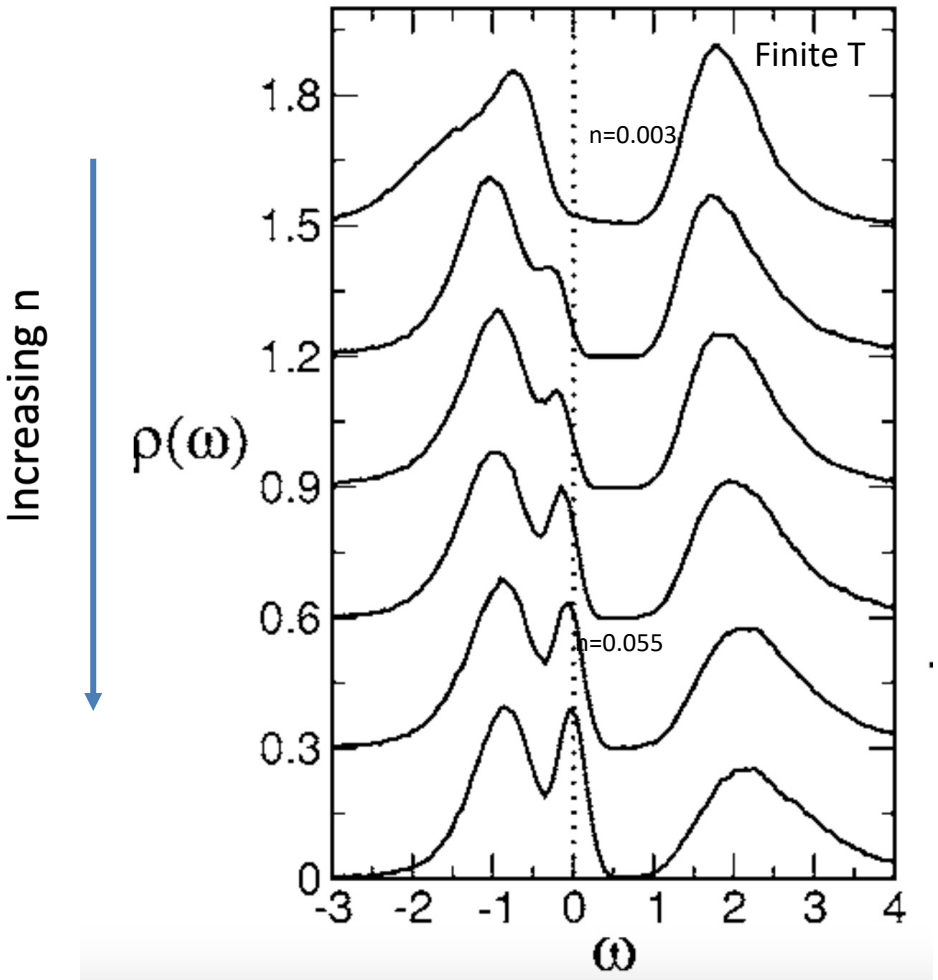




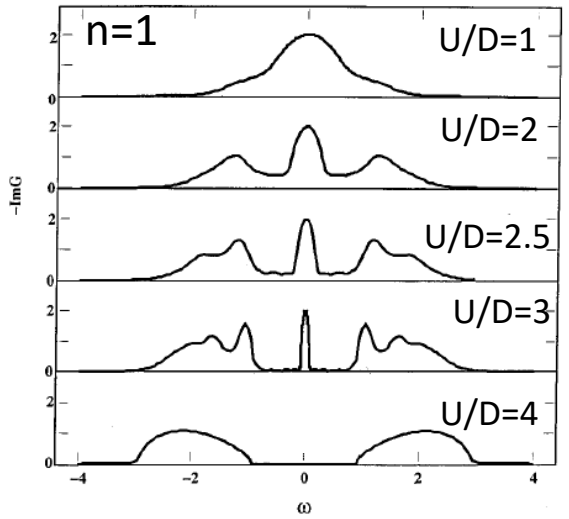
Georges et al , RMP 68, 13 (1996)

Finite density of states at the Fermi level
but correlation features remain

Doping a Mott insulator. Correlated metal



Camjayi et al, PRB (2007)



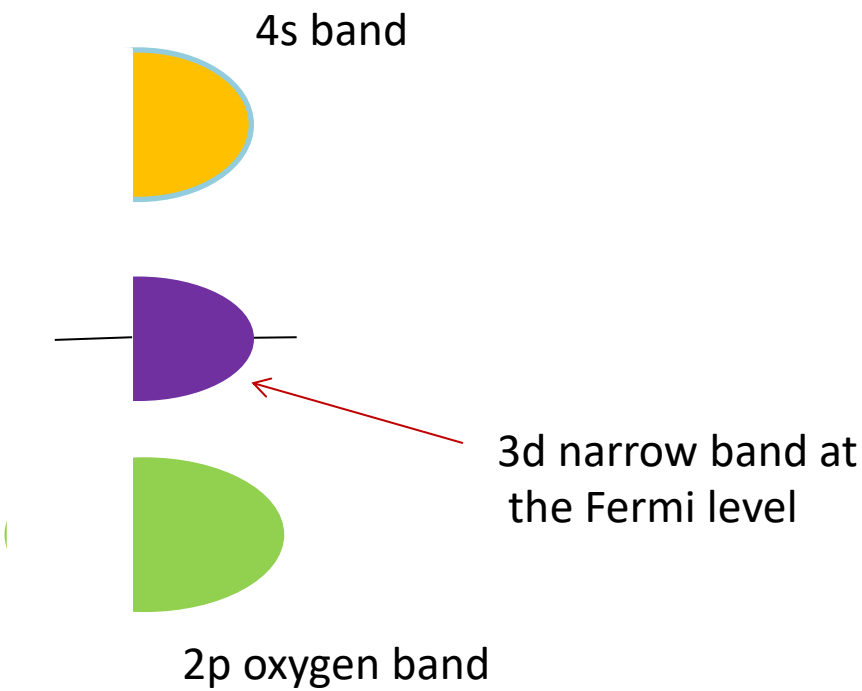
Georges et al , RMP 68, 13 (1996)

Non-rigid band shift

Mott vs charge transfer insulators

3d oxides

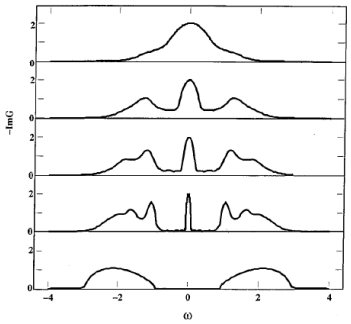
U=0



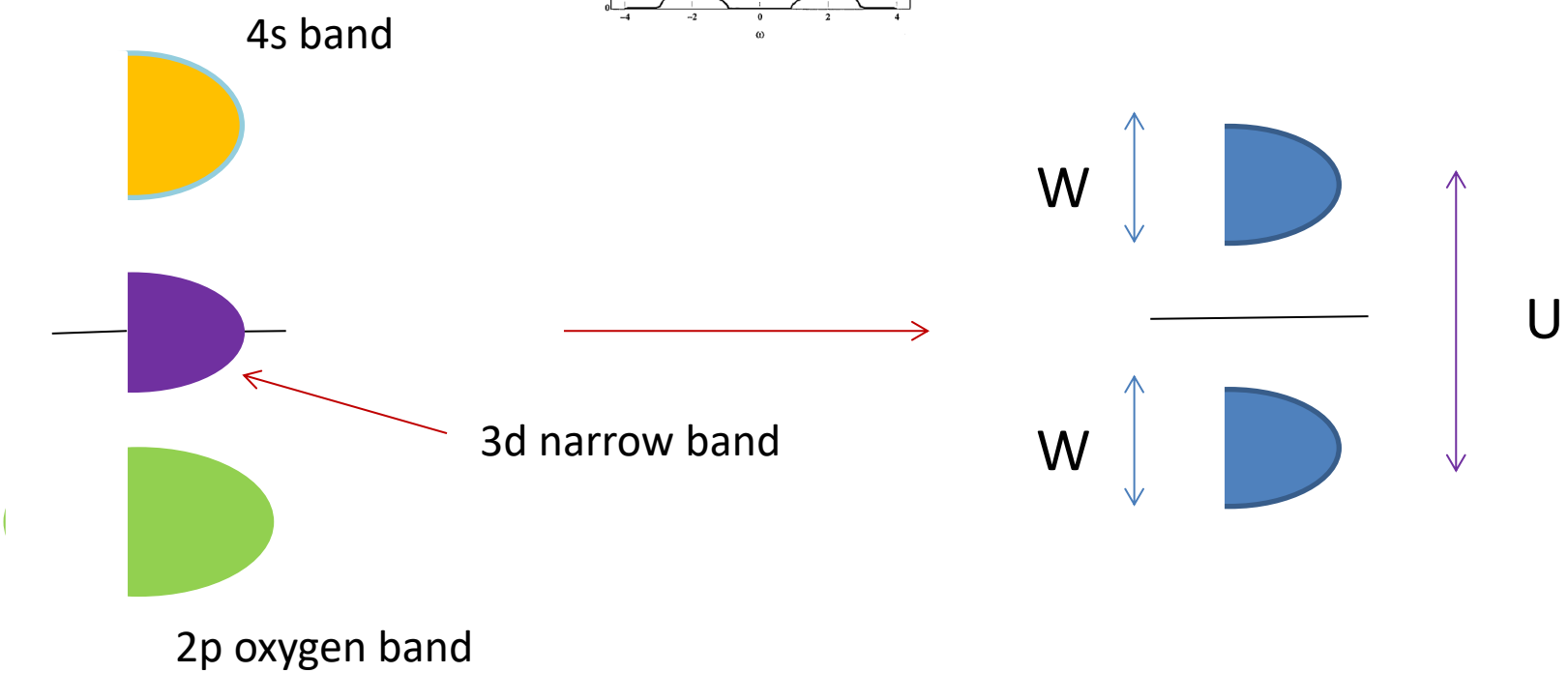
Mott vs charge transfer insulators

3d oxides

U=0



Mott gap opens in the 3d narrow band

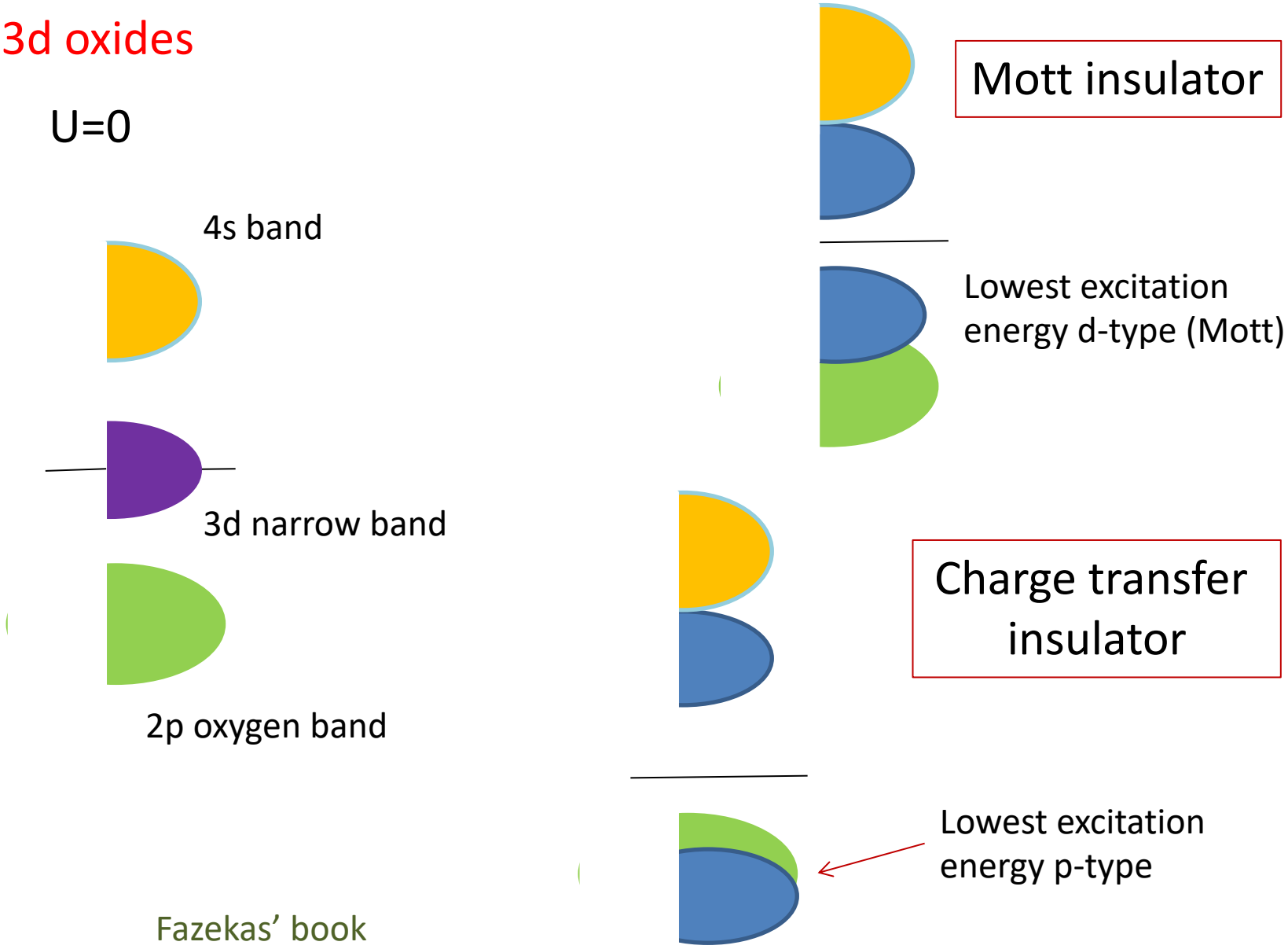


Fazekas' book

Mott vs charge transfer insulators

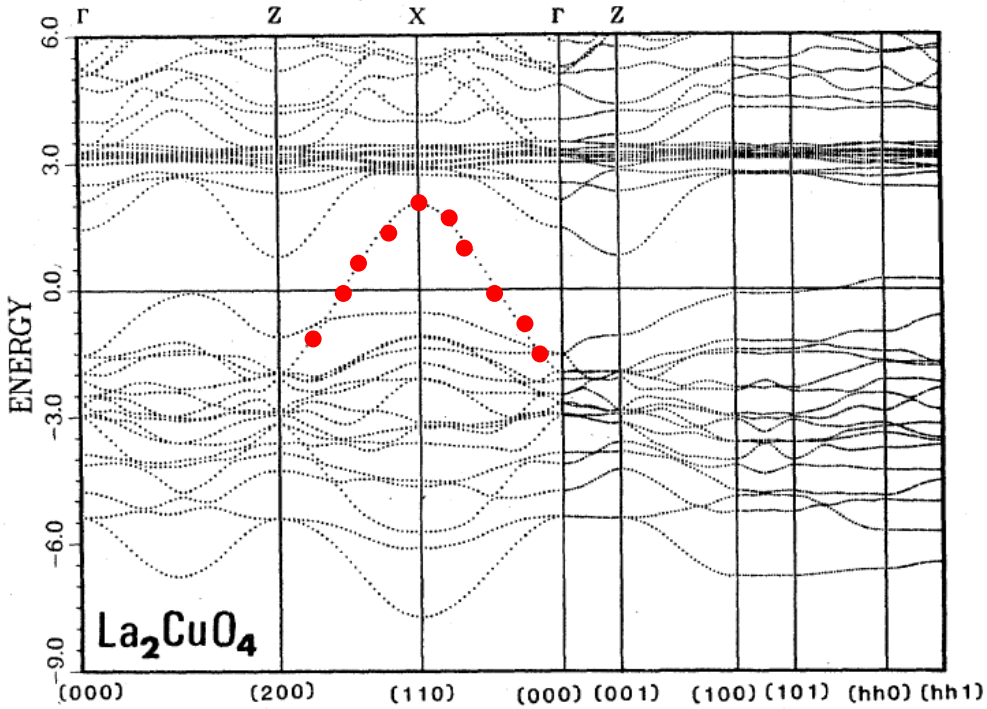
3d oxides

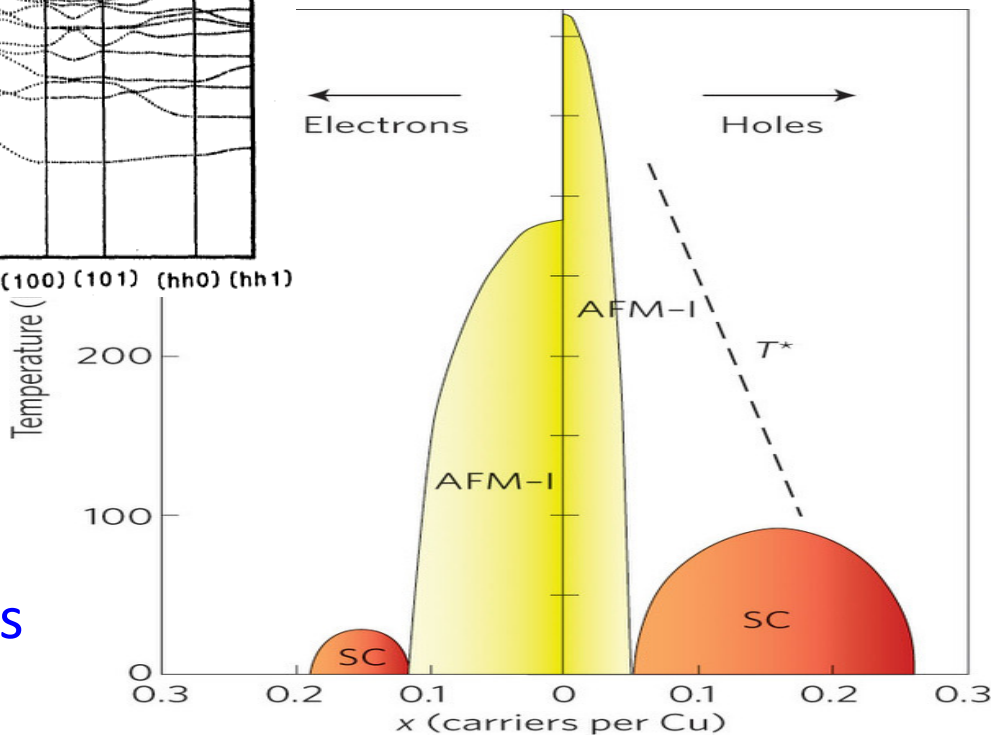
U=0



Fazekas' book

Mott vs charge transfer insulators

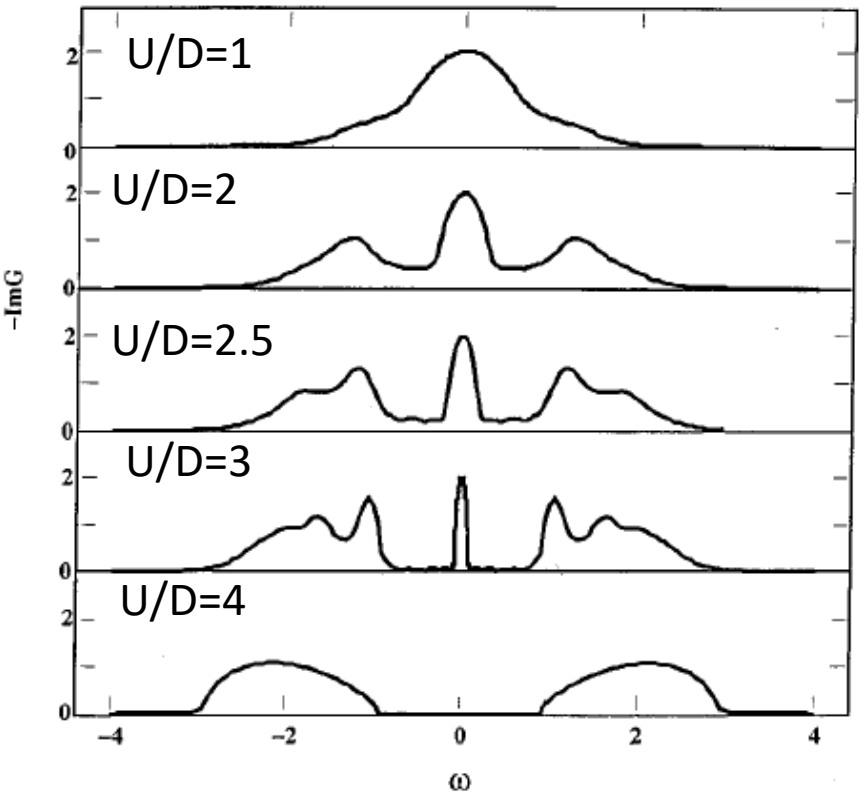




Cuprates are
charge transfer insulators

Correlated metallic state at finite temperatures

Single-site DMFT



At finite temperature:

- the incoherent background (continuum) is explored

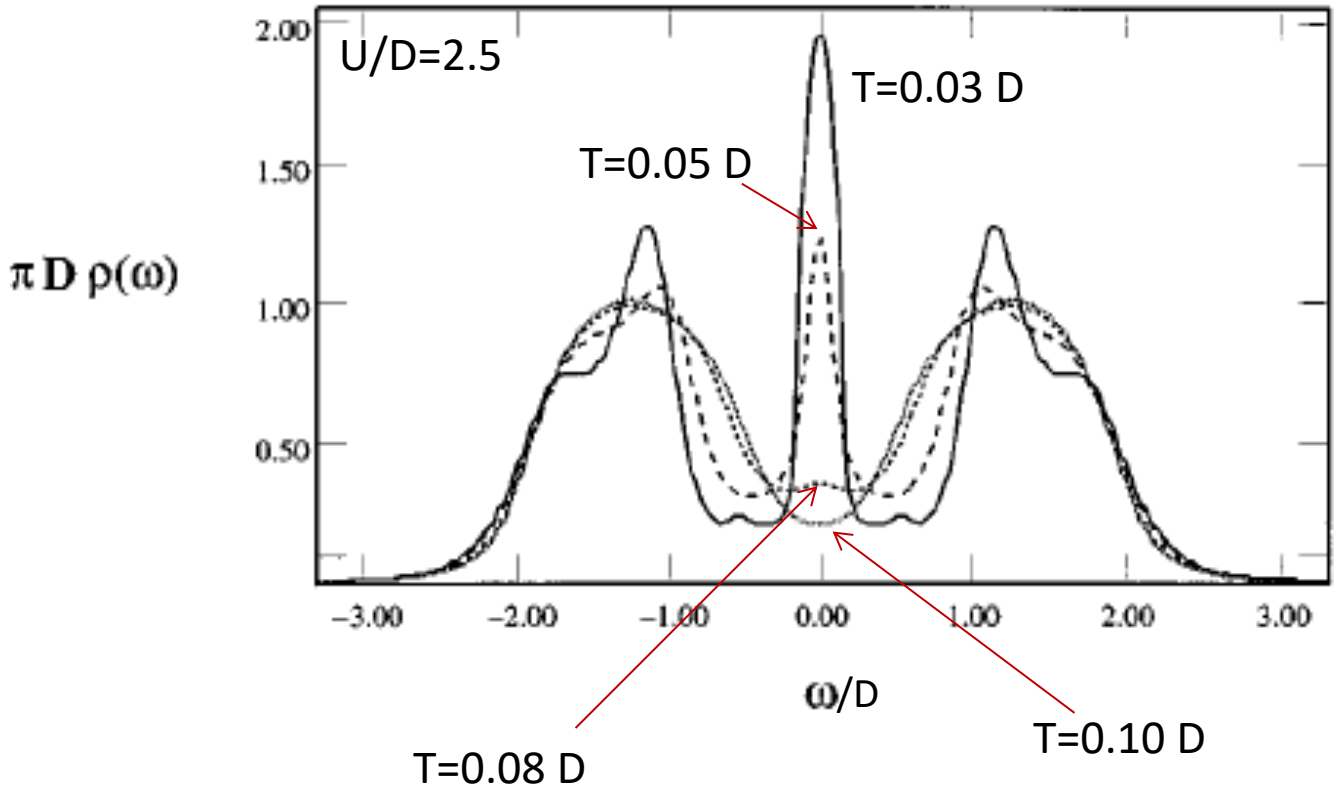
- The quasiparticle is badly defined

Finite temperature promotes more incoherent behavior

(opposite to transitions which involve symmetry breaking)

At the metallic side of the Mott transition. Finite temperatures

Single-site DMFT

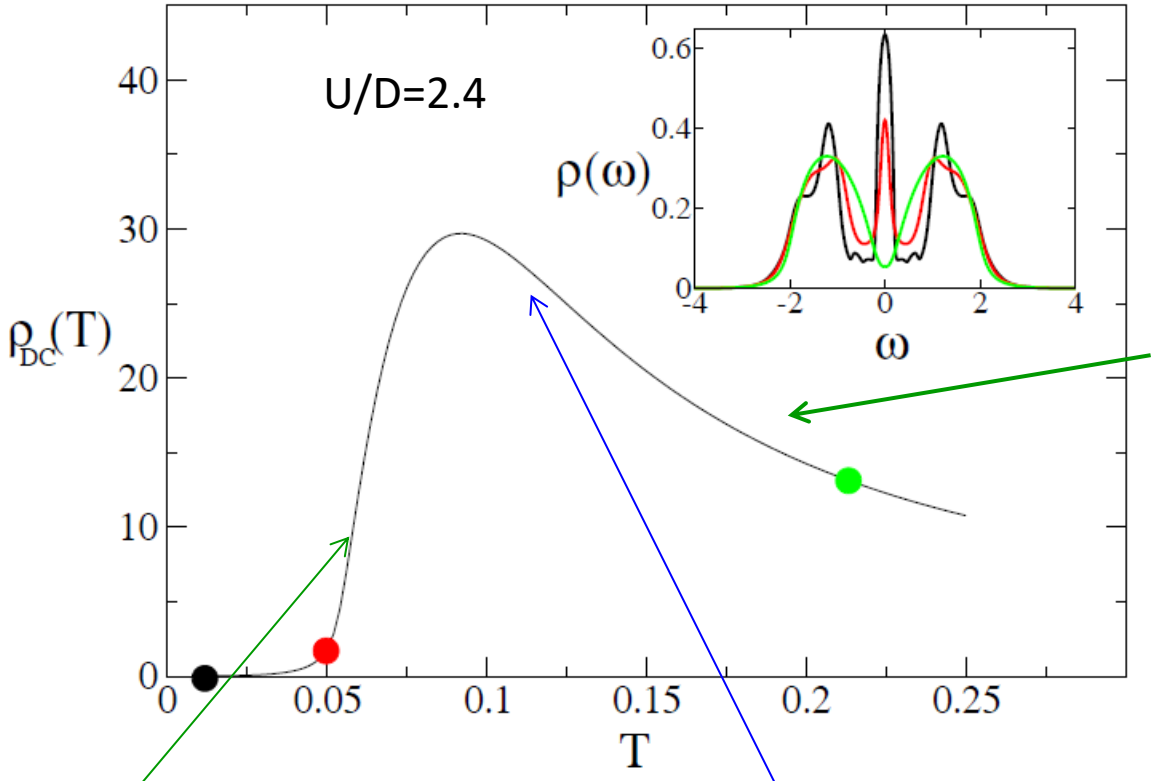


The quasiparticle weight Z decreases with increasing temperature

Finite temperature
promotes more incoherent behavior

At the metallic side of the Mott transition. Finite temperatures

Single-site DMFT



Resistivity decreases with temperature (looks like insulator)

Resistivity increases with temperature (metal)

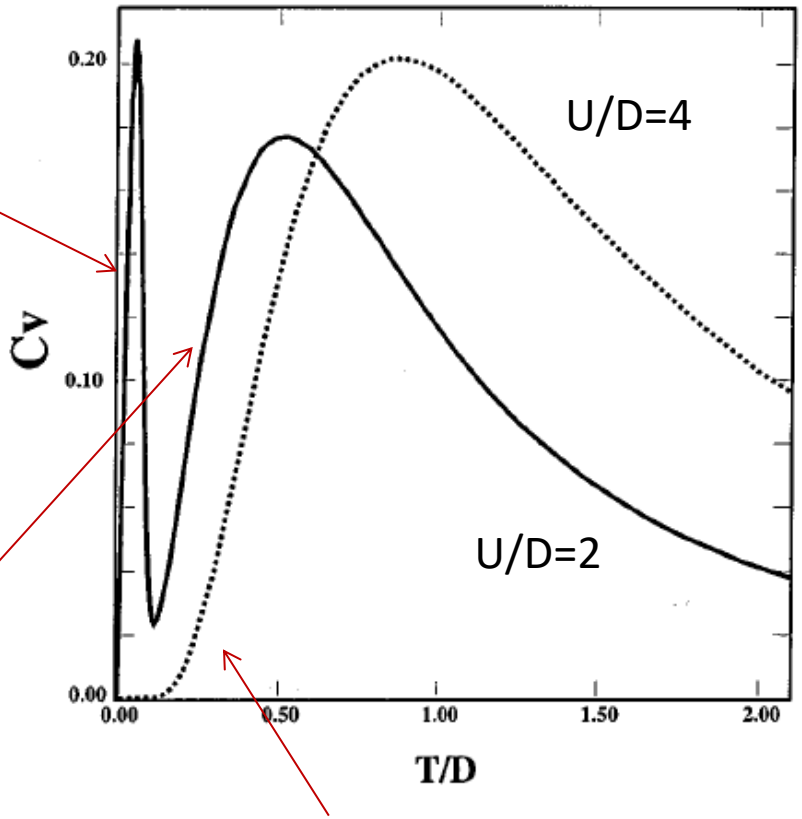
Change from metallic to insulating like behavior at a given temperature

Georges et al, J. de Physique IV 114, 165 (2004), arXiv:0311520

The Mott transition. Finite temperatures. Single-site DMFT

Fermi liquid behavior observed only at low temperatures

T-linear dependence at low temperatures (Metallic)



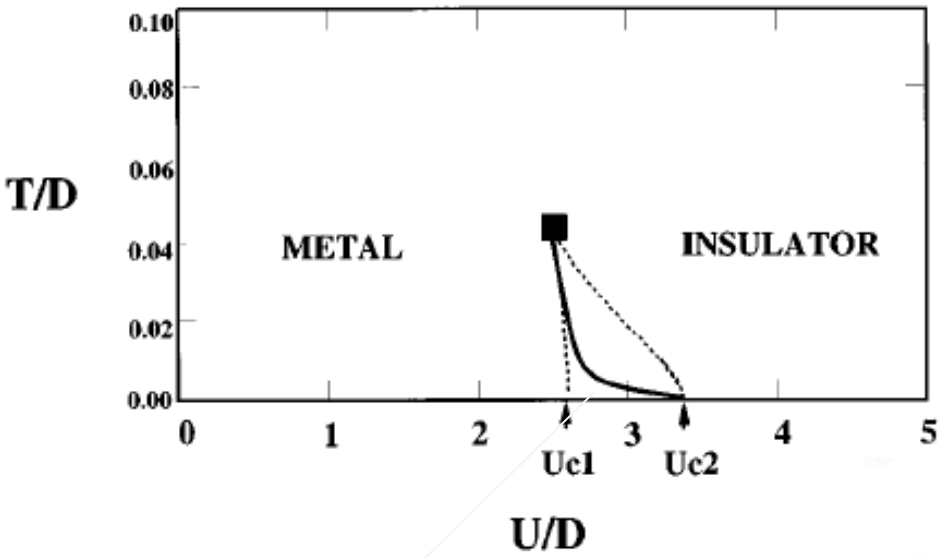
Change to insulating like behavior at high temperatures

Activated behavior at low temperatures (Insulating)

DMFT

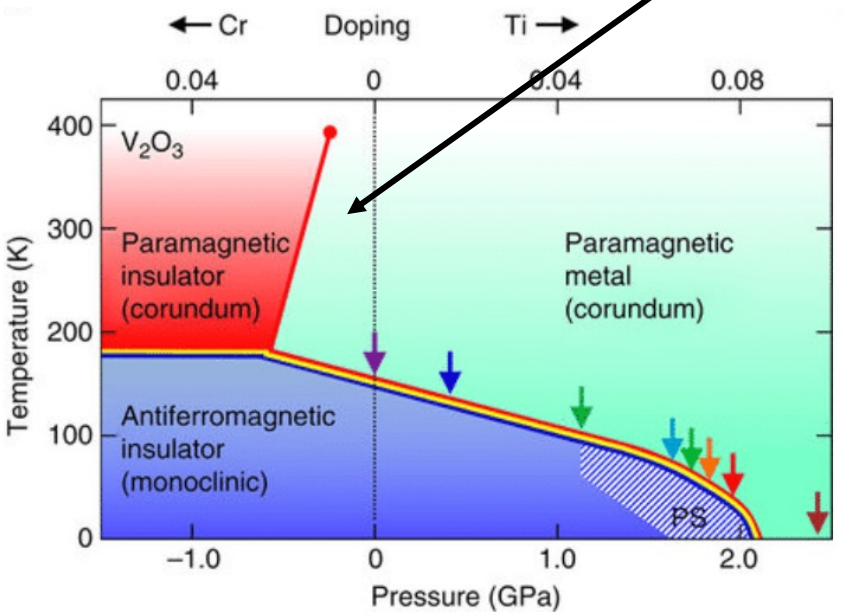
Georges et al , RMP 68, 13 (1996)

The Mott transition. Finite temperatures. Single site DMFT



The system becomes insulating with increasing temperature

Slope



The insulating state remaims even if the antiferromagnetism disappears

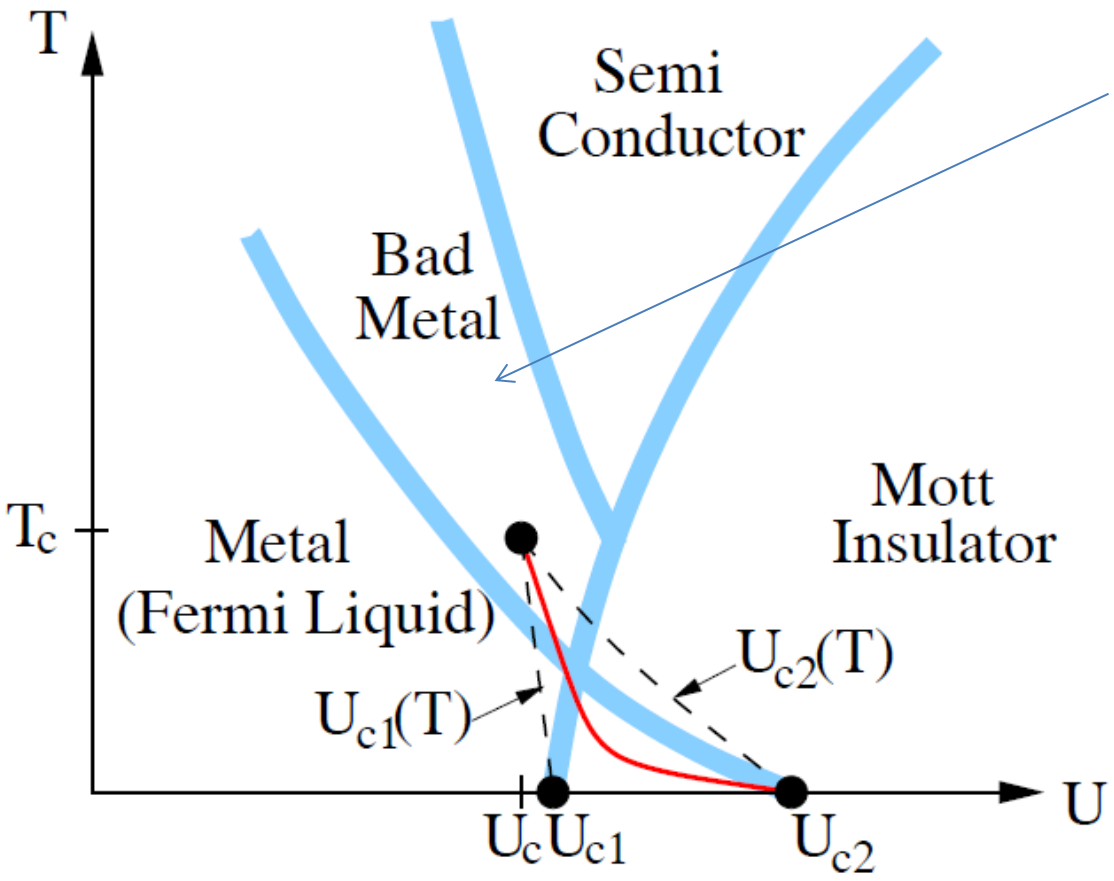
With increasing temperature the insulating behavior can show up

Georges et al , RMP 68, 13 (1996)

McWhan et al, PRB 7, 1920 (1973)

The Mott transition. Finite temperatures. Single site DMFT

Not so clear distinction between a metal and an insulator at finite temperatures



Bad metal: the electronic scattering mean free path is smaller than the lattice constant

Mott-Ioffe-Regel limit

$\rho > \rho_{MIR}$

Georges et al, J. de Physique IV 114, 165 (2004), arXiv:0311520

Mott physics

- Hubbard model at half-filling. Mott transition with increasing U
- Insulator with odd number of electrons per site (on spite of spin degeneracy)
- No symmetry breaking is required (but it may happen as well)
- Magnetism not used to justify the metal-insulator transition
- Charge localization. Local moments are formed (but not necessarily ordered).
- Description in terms of spin models. Tendency towards antiferromagnetism in Hubbard model at half-filling (if band is not perfectly flat. Finite hopping t). $J \sim t^2/U$.
- Spectrum strongly modified in both the insulator and the correlated metal (Hubbard bands and correlated metals). Anomalous “normal” state.
- Doping induces metallicity and suppresses the Antiferromagnetism.
- Incoherence increases with increasing temperature
- Charge transfer insulators versus Mott.