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New States of Stable and Unstable Quantum Matter
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Electronic Structure of Strongly Correlated Electron Materials: A Dynamical Mean Field Perspective

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Electronic Structure of Strongly Correlated Electron Materials: A Dynamical Mean Field Perspective.

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Rutgers University Rutgers University

Collaborators: ***G.Kotliar, S. Savrasov, V. Oudovenko***

ICAM Symposium on Frontiers in Correlated Matter, Snowmass 2006

Overview

- Application of DMFT to real materials (Spectral density functional approach). Examples:
  - alpha to gamma transition in Ce, optics near the temperature driven Mott transition.
  - Mott transition in Americium under pressure
  - Antiferromagnetic transition in Curium
- Extensions of DMFT to clusters.

  Examples:
  - Superconducting state in t-J the model
  - Optical conductivity of the t-J model

Universality of the Mott transition

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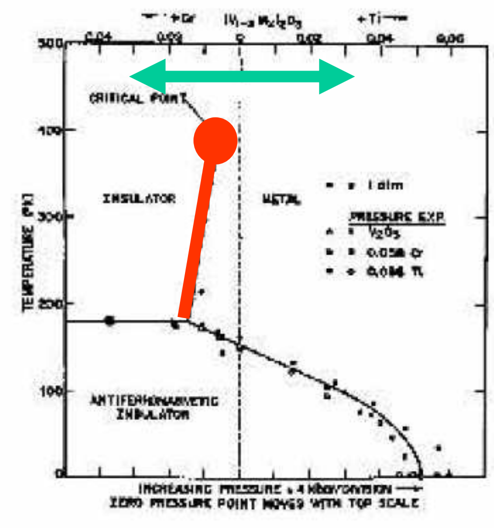
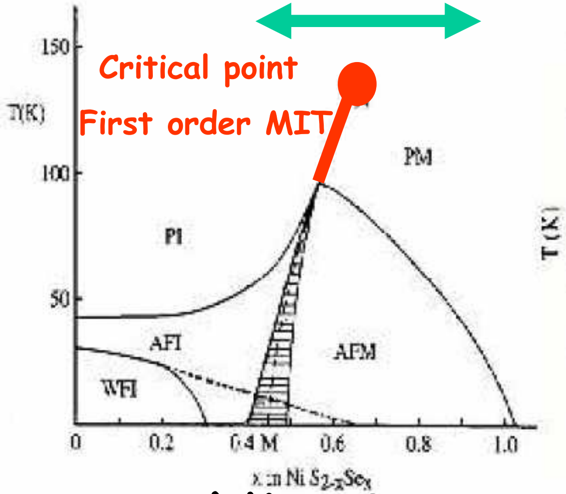
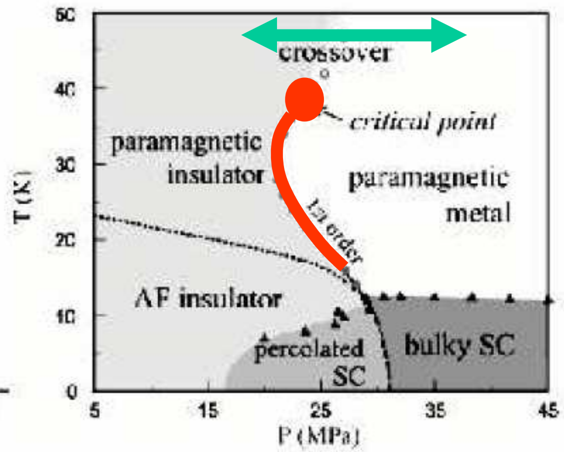
Crossover: bad insulator to bad metal



V_2O_3

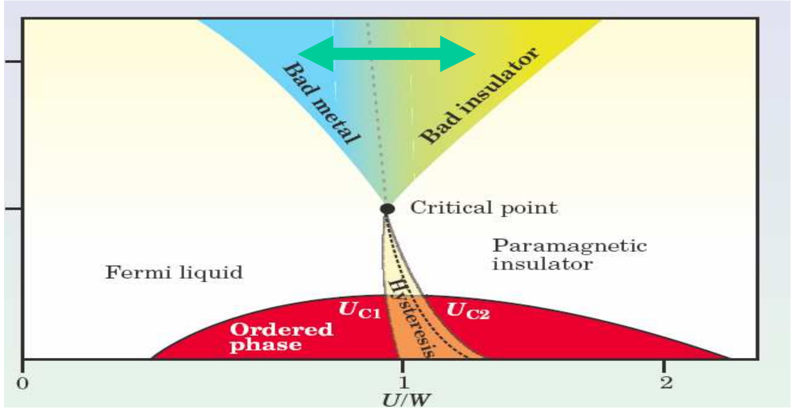


$Ni_{2-x}Se_x$



κ organics

1B HB model (DMFT):



Coherence incoherence crossover in the 1B HB model (DMFT)

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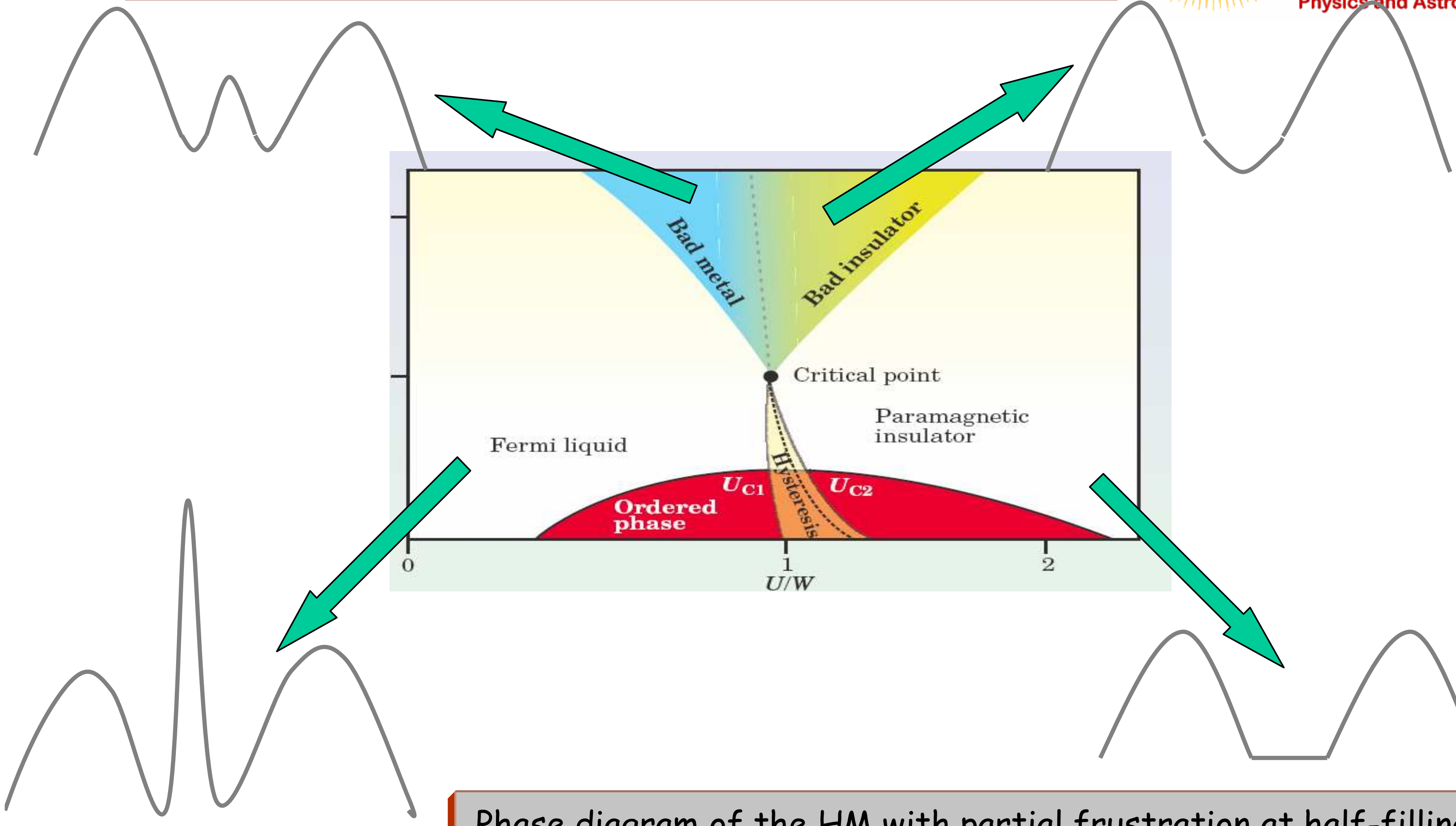


Phase diagram of the HM with partial frustration at half-filling
M. Rozenberg et.al., Phys. Rev. Lett. **75**, 105 (1995).

DMFT + electronic structure method

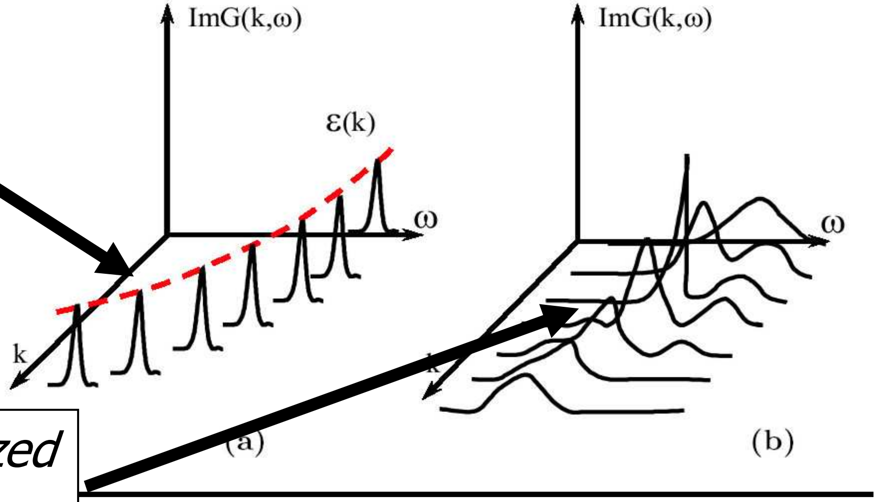
Basic idea of DMFT: reduce the *quantum many body problem* to a *one site* or a *cluster* of sites problem, *in a medium* of non interacting electrons obeying a self-consistency condition. (A. Georges et al., RMP 68, 13 (1996)).

DMFT in the language of functionals: DMFT sums up *all local diagrams* *in BK functional*

Basic idea of DMFT+electronic structure method (LDA or GW):

For less correlated bands (s,p): use LDA or GW

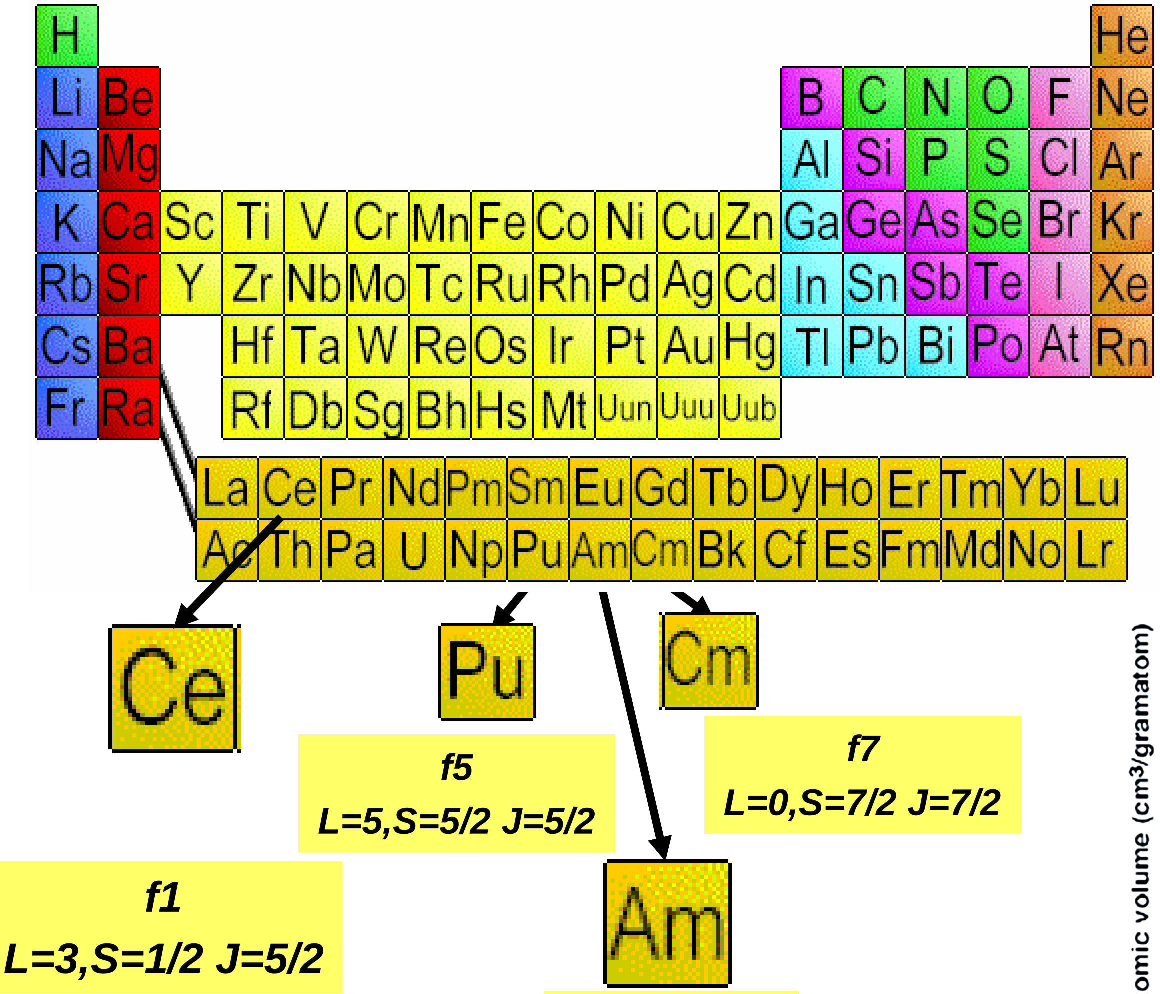
For correlated bands (f or d): *with DMFT add all local diagrams*



Effective (DFT-like) single particle Spectrum consists of delta like peaks

Spectral density usually contains renormalized quasiparticles and Hubbard bands

How good is single site DMFT for f systems?



f1

L=3,S=1/2 J=5/2

f5

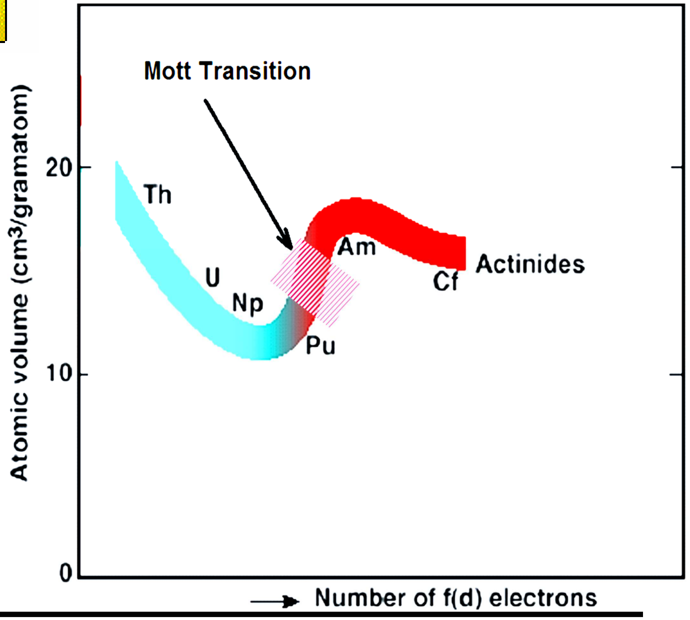
L=5,S=5/2 J=5/2

f6

L=3,S=3 J=0

f7

L=0,S=7/2 J=7/2



Cerium

Ce overview

- isostructural phase transition ends in a critical point at (T=600K, P=2GPa)

→ γ (fcc) phase

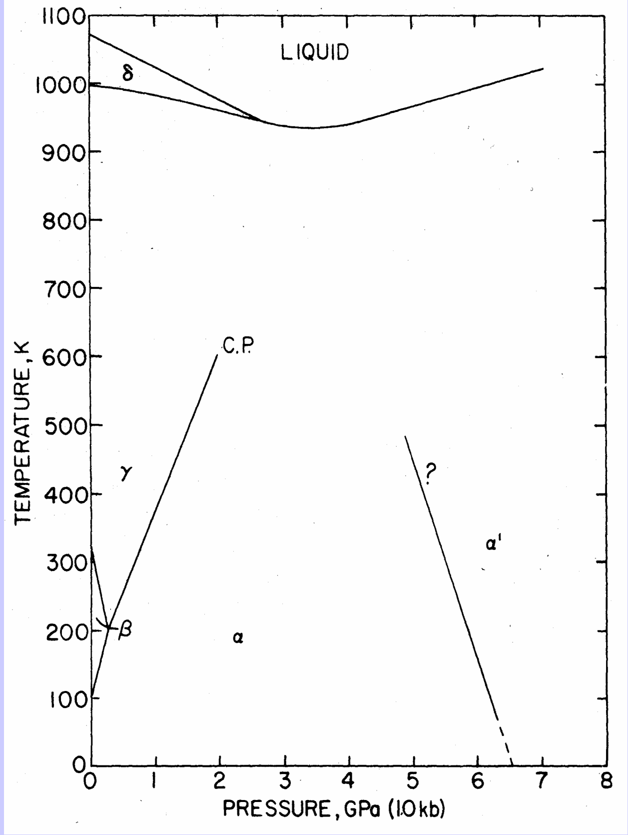
[magnetic moment (Curie-Wiess law), large volume, stable high-T, low-p]

→ α (fcc) phase

[loss of magnetic moment (Pauli-para), smaller volume, stable low-T, high-p]

with large volume collapse

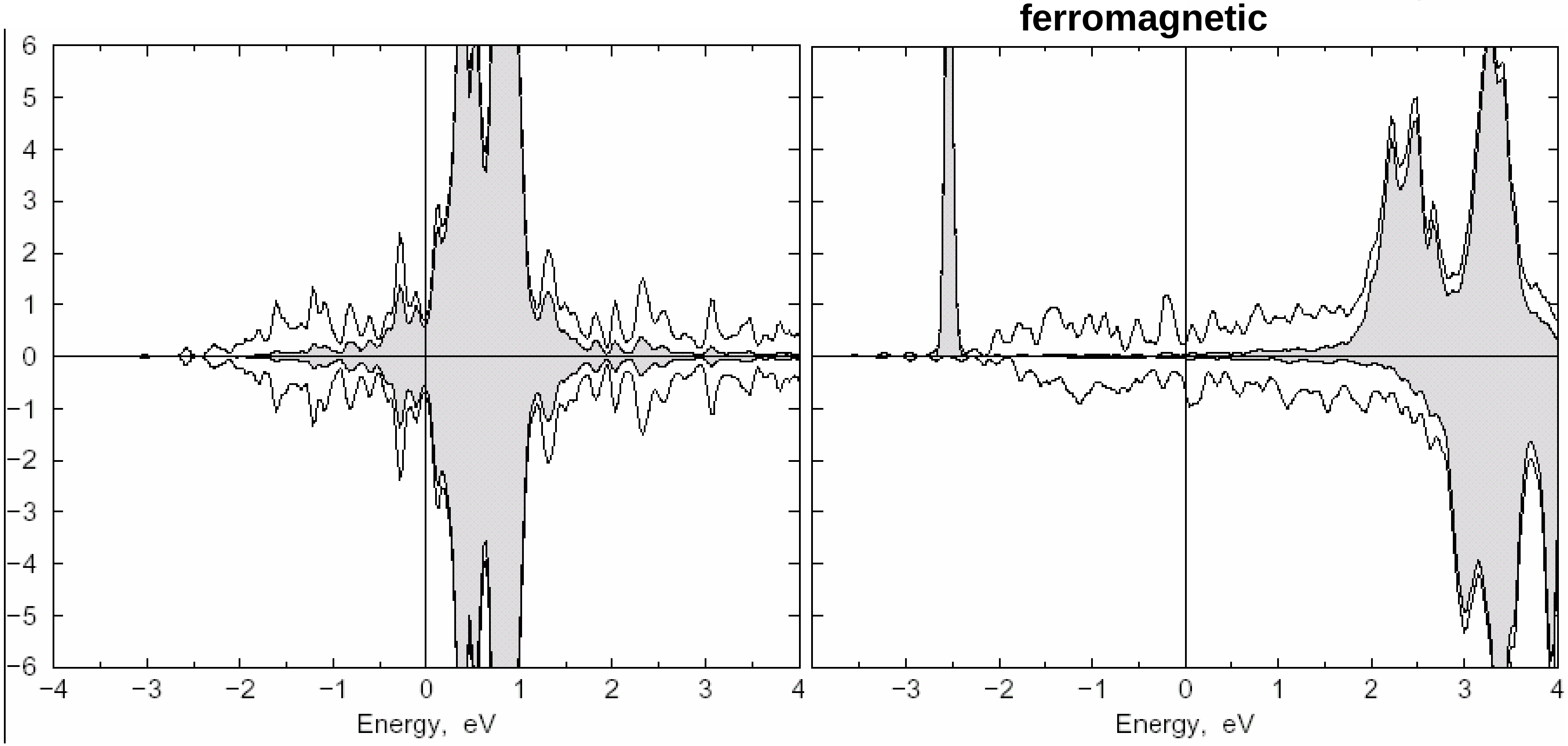
$\Delta v/v \approx 15\%$



volumes	exp.	LDA	LDA+U
α	28Å^3	24.7Å^3	
γ	34.4Å^3		35.2Å^3

- *Transition is 1.order*
- *ends with CP*

LDA and LDA+U

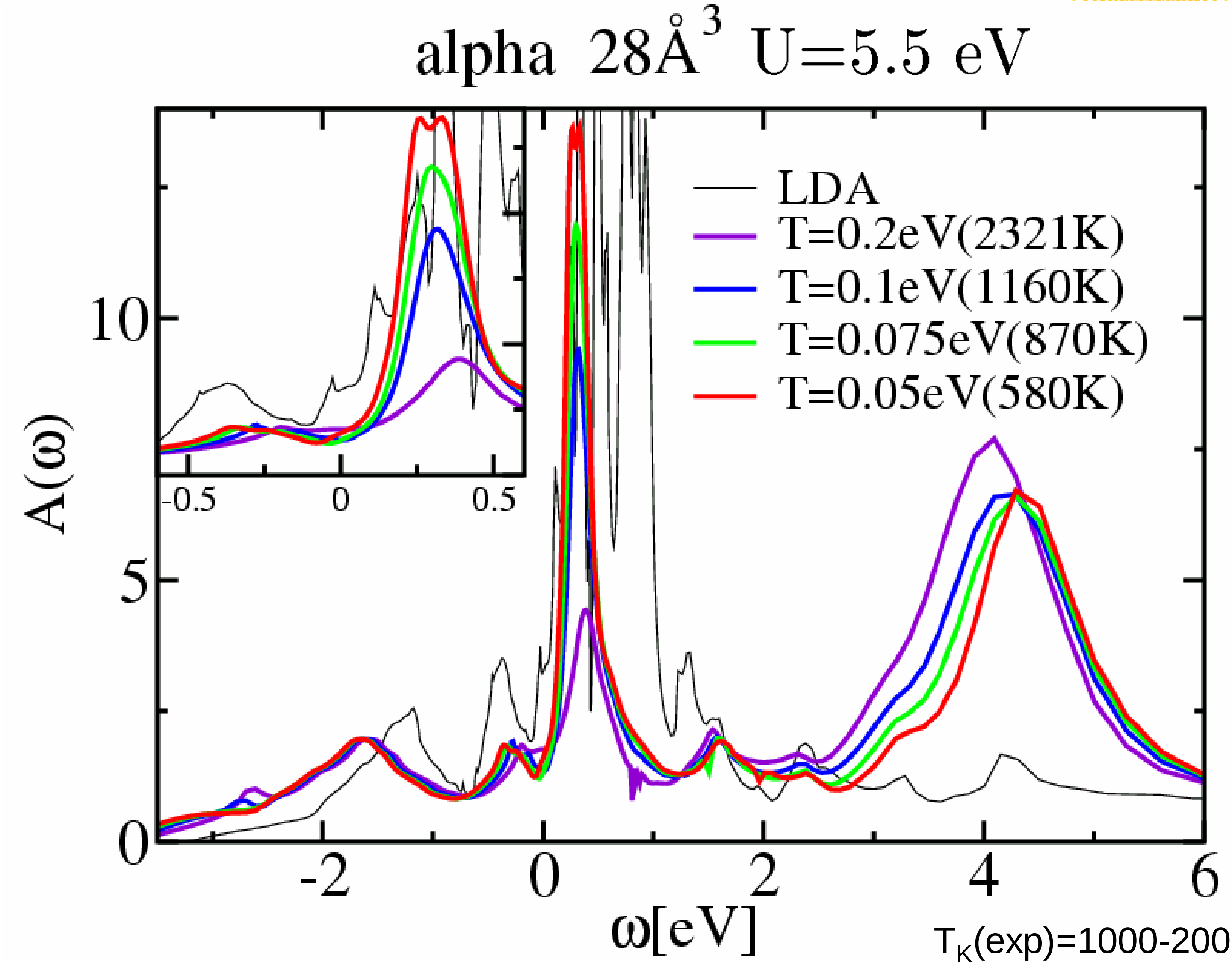


▨ f DOS

☐ total DOS

volumes	exp.	LDA	LDA+U
α	28Å^3	24.7Å^3	
γ	34.4Å^3		35.2Å^3

LDA+DMFT alpha DOS



T_K(exp)=1000-2000K

LDA+DMFT gamma DOS

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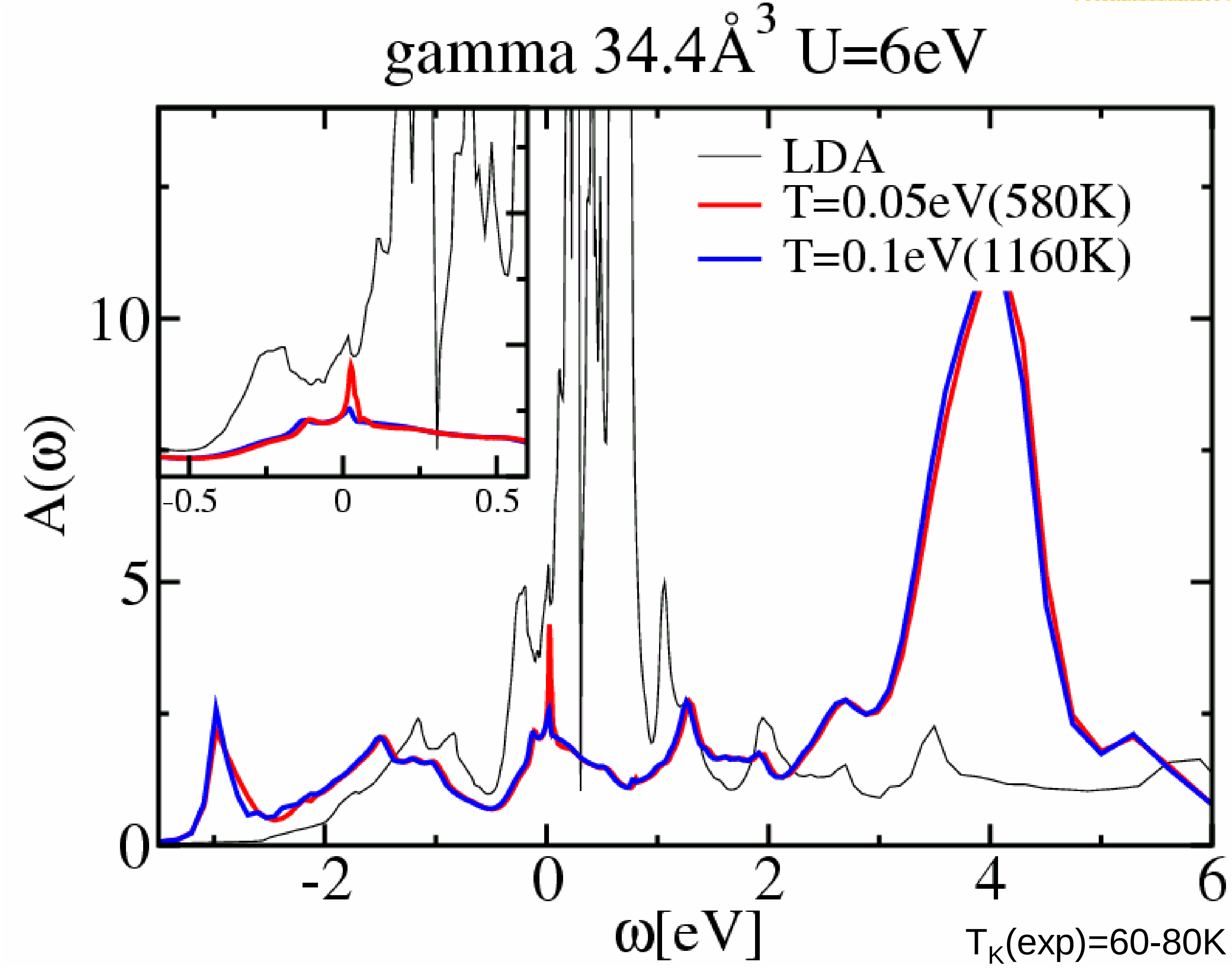
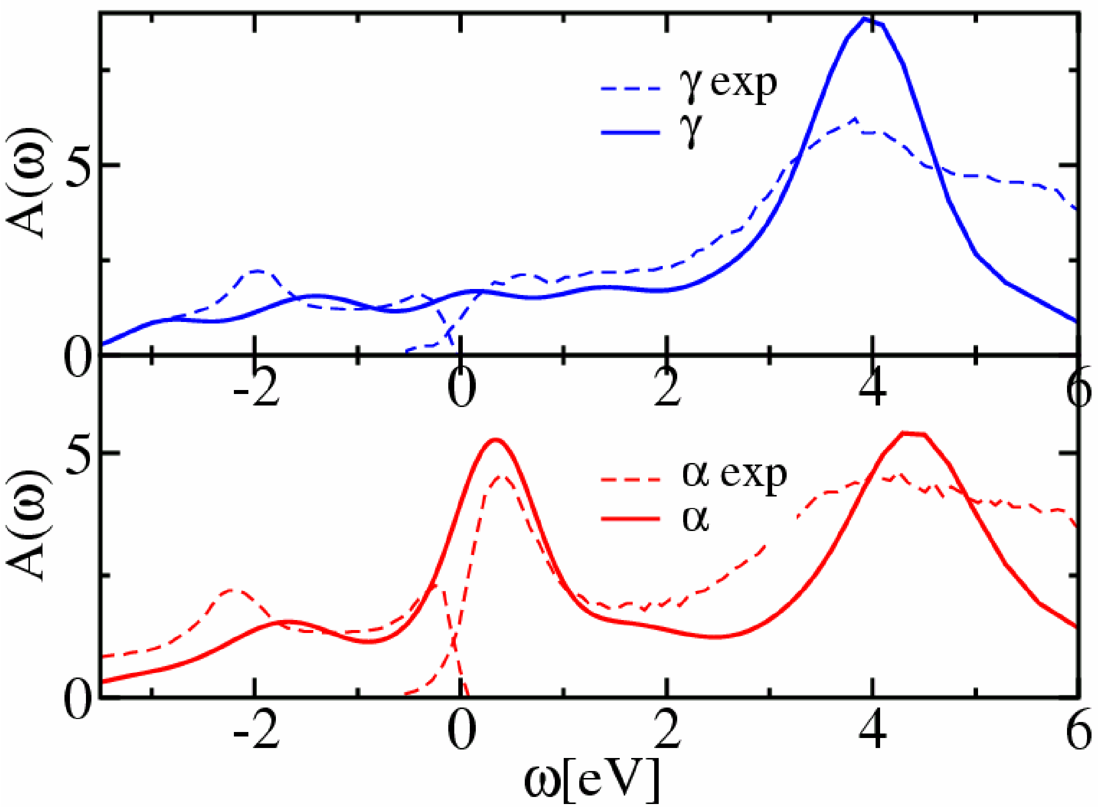


T_K(exp)=60-80K

Photoemission&experiment



- A. Mc Mahan K Held and R. Scalettar (2002)
- K. Haule V. Udovenko and GK. (2003)

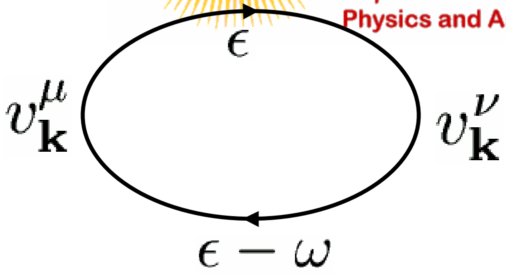
Fenomenological approach describes well the transition

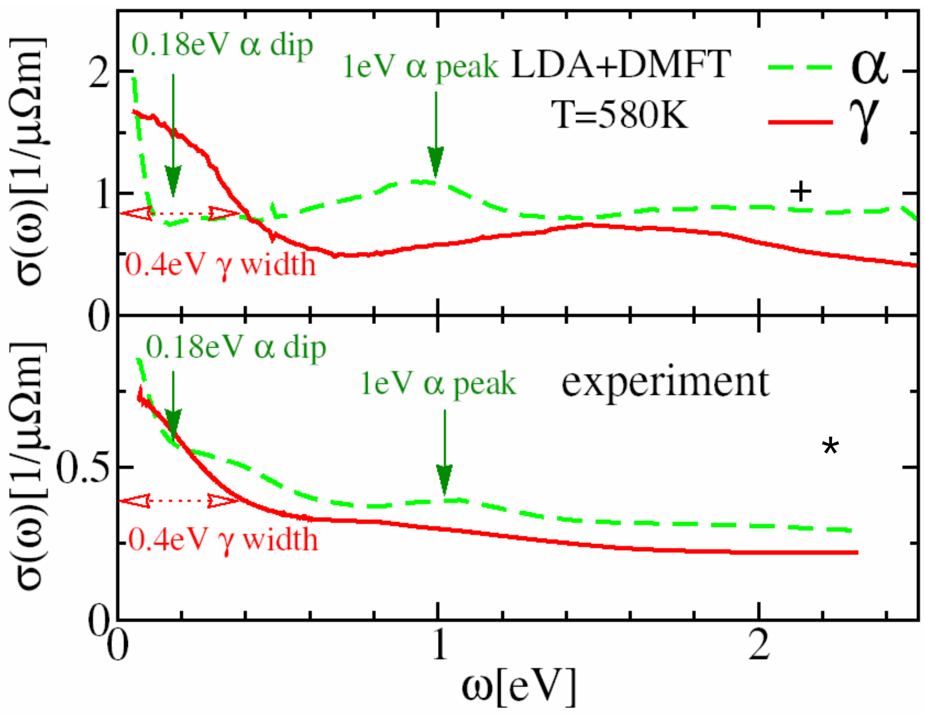
Kondo volume colapse (J.W. Allen, R.M. Martin, 1982)

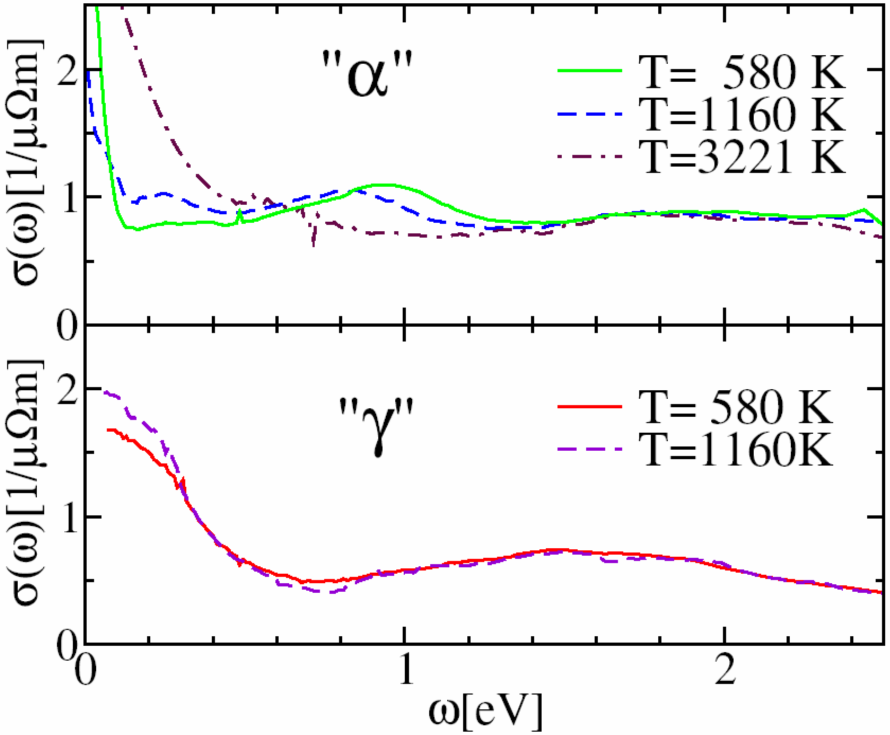
Optical conductivity

$$\sigma^{\mu\nu}(\omega) = \int w(\epsilon,\omega) \sum_{\mathbf{k}} \mathrm{Tr}\left(v^{\mu}_{\mathbf{k}} \rho_{\mathbf{k}}(\epsilon) v^{\nu}_{\mathbf{k}} \rho_{\mathbf{k}}(\epsilon-\omega)\right)$$

$$w(\epsilon,\omega) = \frac{f(\epsilon-\omega) - f(\epsilon)}{\omega}$$



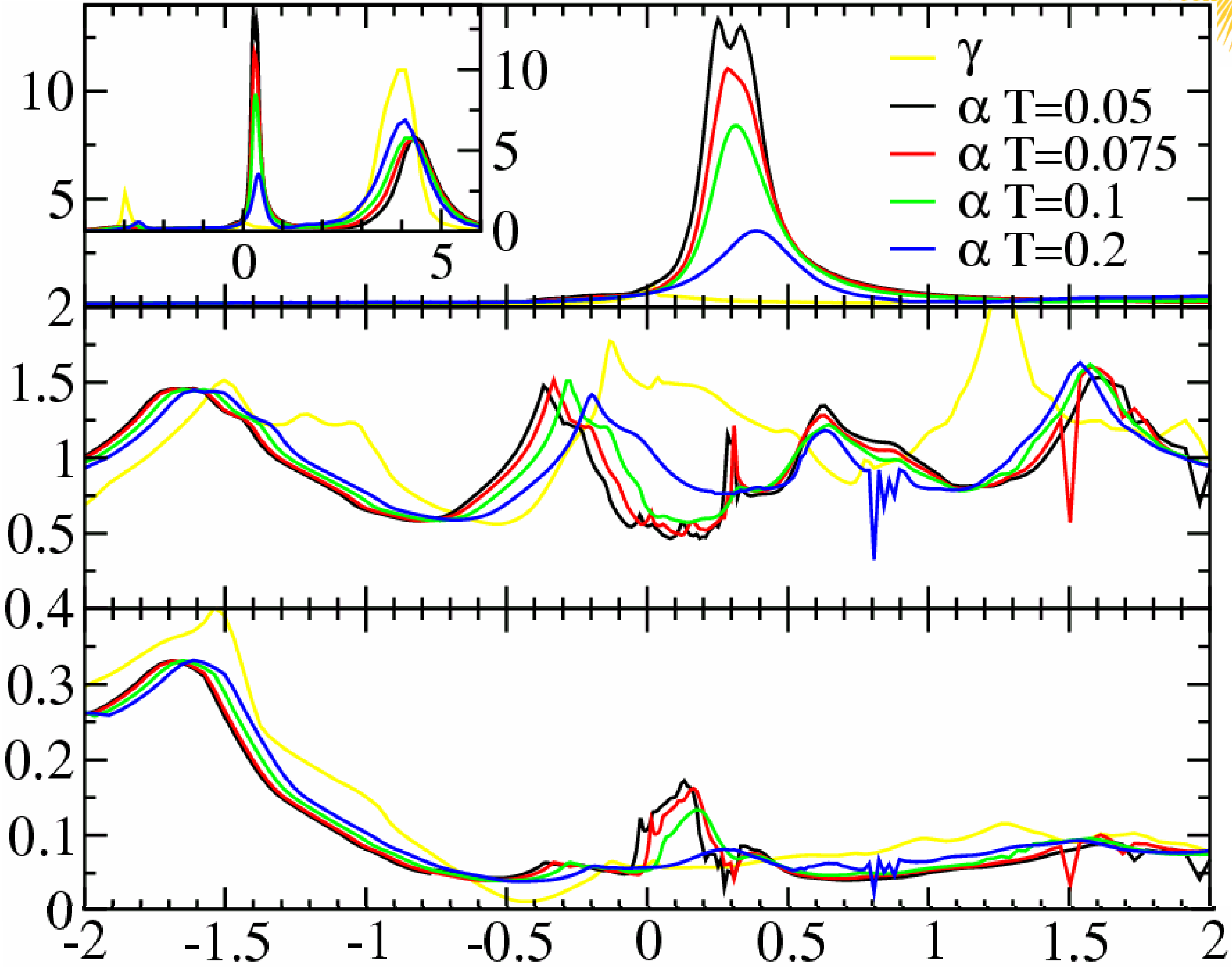




+ K. Haule, et.al.,
Phys. Rev. Lett. **94**, 036401 (2005)

* J.W. van der Eb, A.B. Ku'zmenko, and D. van der Marel,
Phys. Rev. Lett. **86**, 3407 (2001)

Partial DOS



4f Z=0.33

5d

6s

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Americium

Americium

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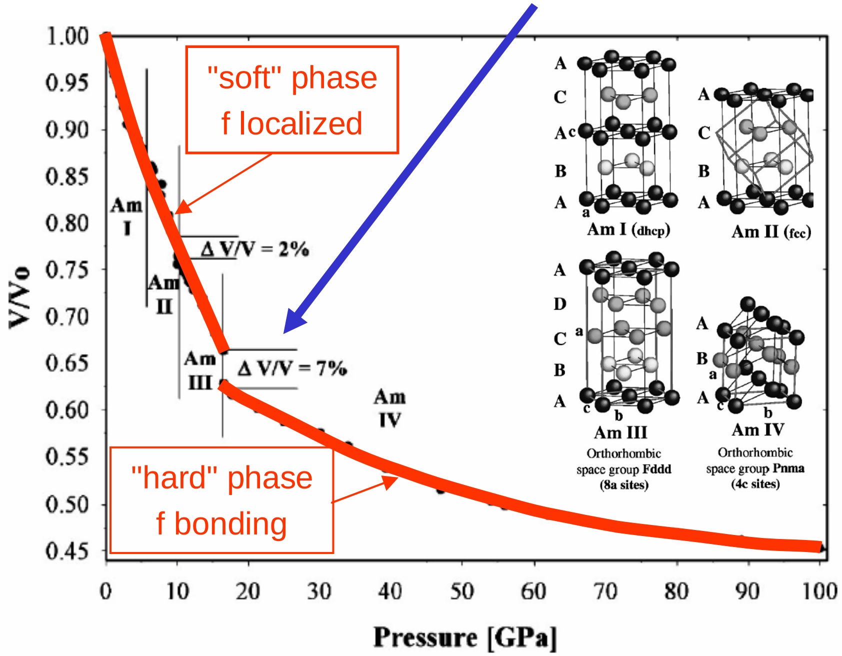
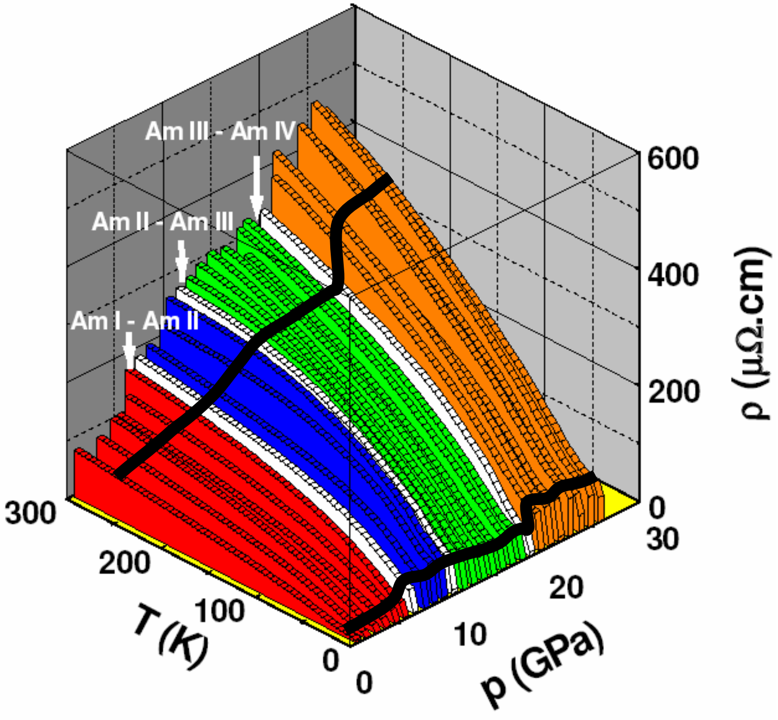
lanthanons	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	53 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
actinons	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

f^6 -> L=3, S=3, J=0

Mott Transition?



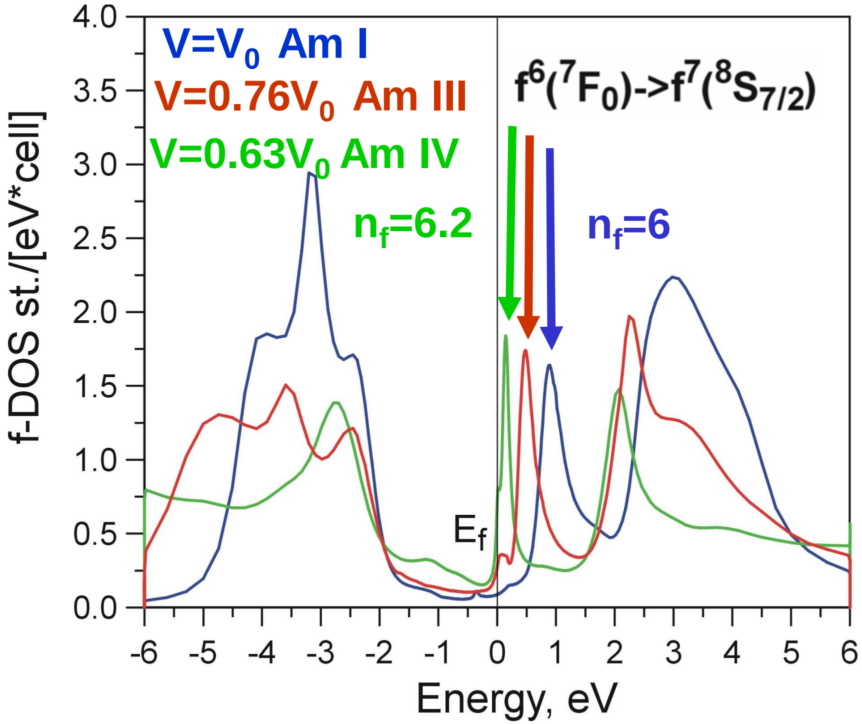


A.Lindbaum, S. Heathman, K. Litfin, and Y. Méresse,
Phys. Rev. B **63**, 214101 (2001)

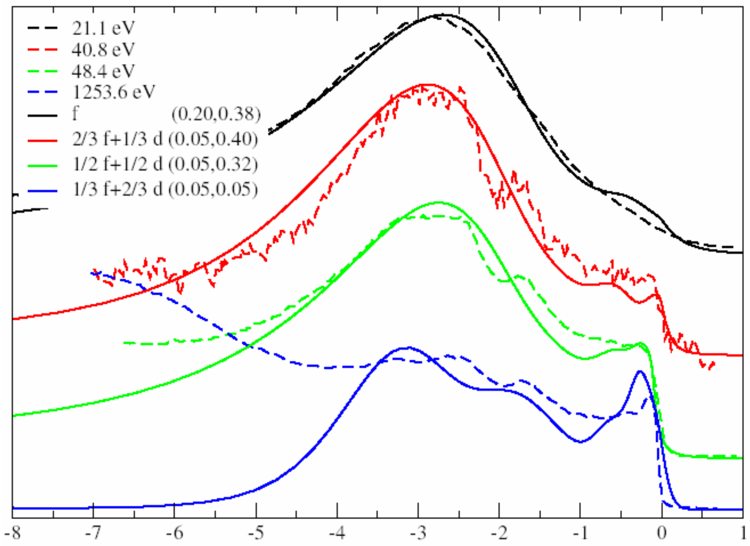
J.-C. Griveau, J. Rebizant, G. H. Lander, and G.Kotliar
Phys. Rev. Lett. **94**, 097002 (2005)

Am within LDA+DMFT

from J=0 to J=7/2



Comparisson with experiment



Exp: J. R. Naegele, L. Manes, J. C. Spirlet, and W. Müller
Phys. Rev. Lett. **52**, 1834-1837 (1984)

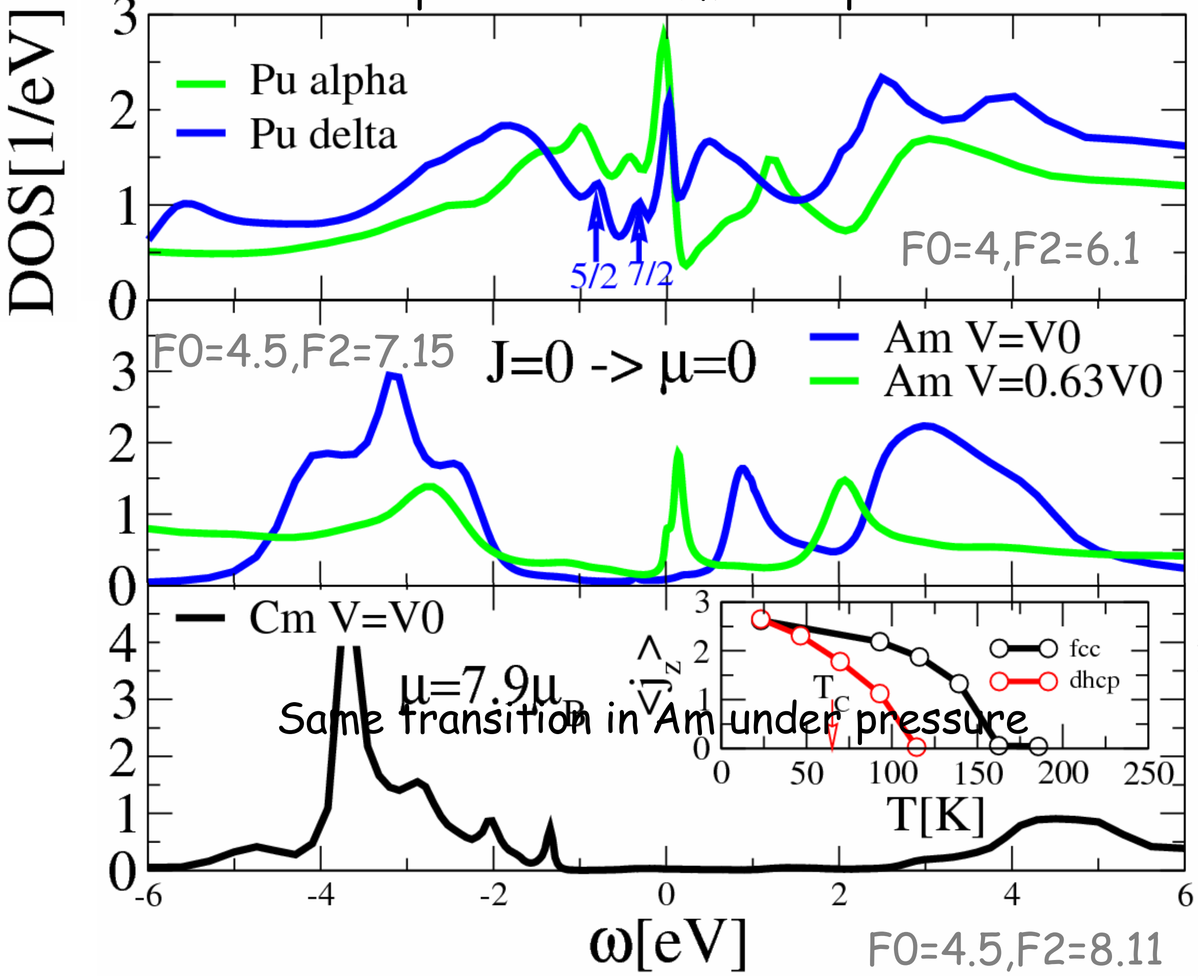
Theory: S. Y. Savrasov, K. Haule, and G. Kotliar
Phys. Rev. Lett. **96**, 036404 (2006)

- *"Soft" phase very different from γ Ce*
  not in local moment regime since J=0 (no entropy)
- *"Hard" phase similar to α Ce,*
  Kondo physics due to hybridization, however,
  nf still far from Kondo regime

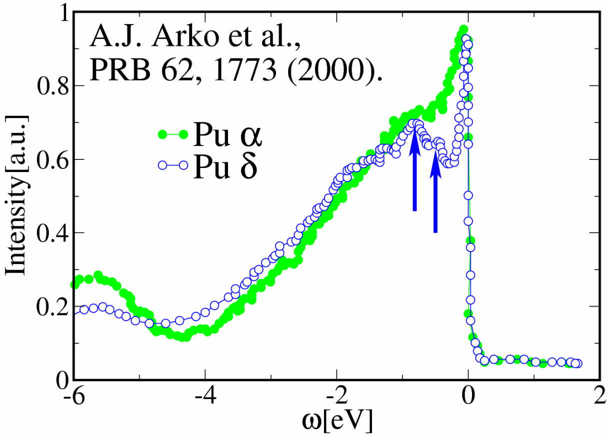
Different from Sm!

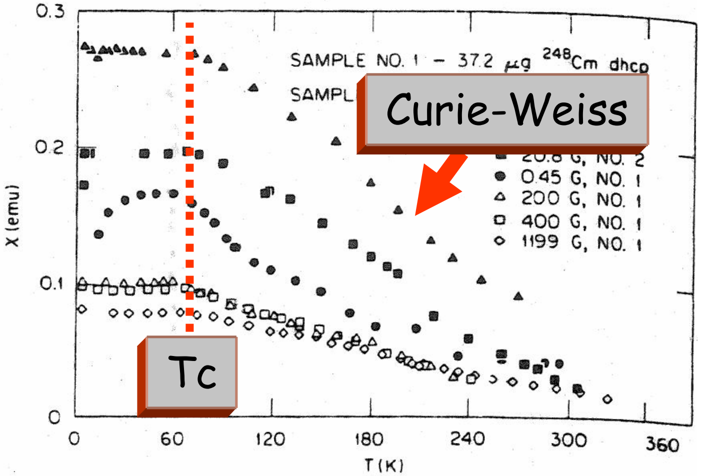
Trends in Actinides

alpa->delta volume collapse transition



Same transition in Am under pressure





Curium has large magnetic moment and orders antif.

What is captured by single site DMFT?

- Captures volume collapse transition (first order Mott-like transition)
- Predicts well photoemission spectra, optics spectra, total energy at the Mott boundary
- Antiferromagnetic ordering of magnetic moments, magnetism at finite temperature
- Qualitative explanation of mysterious phenomena, such as the anomalous raise in resistivity as one applies pressure in Am,..

Beyond single site DMFT

What is missing in DMFT?

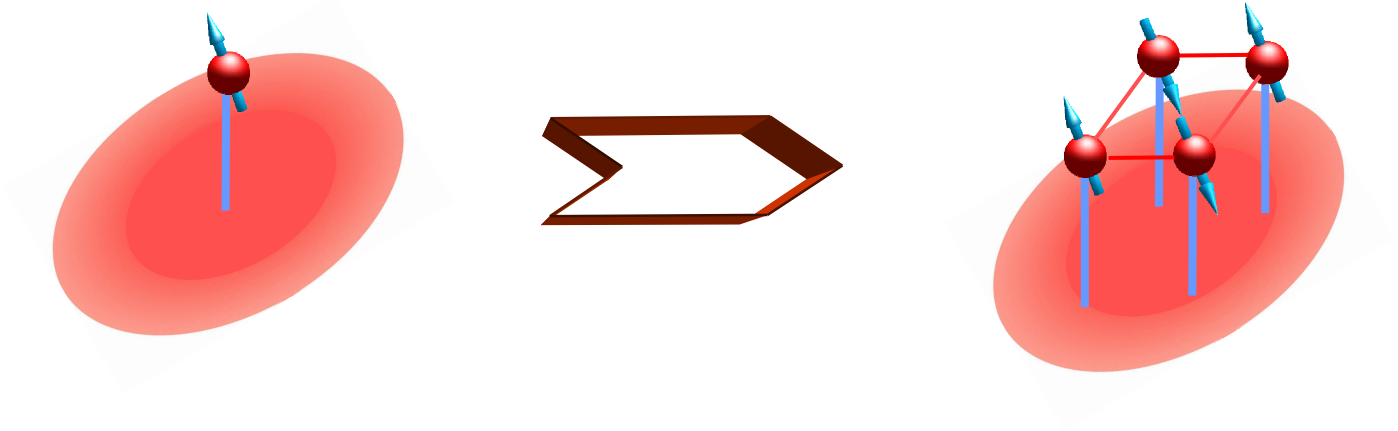
- Momentum dependence of the self-energy $m^*/m=1/Z$
- Various orders: d-waveSC,…
- Variation of Z, m^*,τ on the Fermi surface
- Non trivial insulator (frustrated magnets)
- Non-local interactions (spin-spin, long range Columb,correlated hopping..)

Present in DMFT:

- *Quantum time fluctuations*

Present in cluster DMFT:

- *Quantum time fluctuations*
- *Spatially short range quantum fluctuations*



The simplest model of high Tc's

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$$H = -\sum_{ij\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + \frac{1}{2}\sum_{ij} J_{ij} \mathbf{S}_i \mathbf{S}_j$$

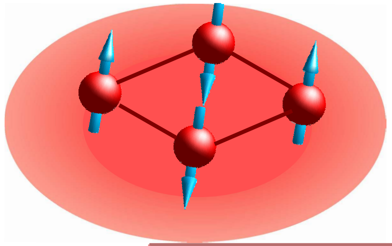
t-J, PW Anderson

Hubbard-Stratonovich ->(to keep some out-of-cluster quantum fluctuations)

$$S = \int_0^\beta d\tau \left\{ \sum_{\mathbf{k}\sigma} c^\dagger_{\mathbf{k}\sigma}(\tau)\left(\frac{\partial}{\partial\tau} - \mu + \epsilon_{\mathbf{k}}\right) c_{\mathbf{k}\sigma}(\tau) + \sum_i U n_{i\uparrow}(\tau) n_{i\downarrow}(\tau) + \sum_{\mathbf{q}} \left[\mathbf{\Phi}^\dagger{}_{\mathbf{q}}(\tau) \frac{2g^2}{J_{\mathbf{q}}} \mathbf{\Phi}_{\mathbf{q}}(\tau) + ig\, \mathbf{S}_{\mathbf{q}} \left(\mathbf{\Phi}^\dagger_{\mathbf{q}}(\tau) + \mathbf{\Phi}_{-\mathbf{q}}(\tau) \right) \right] \right\}$$

$$\begin{aligned}\Gamma[\mathcal{G},\mathcal{D}] &= -\mathrm{Tr}\log(G_0^{-1} - \Sigma) - \mathrm{Tr}[\mathcal{G}\Sigma] \\ &+ \frac{1}{2}\mathrm{Tr}\log(\mathcal{D}_0^{-1} - \Pi) + \frac{1}{2}\mathrm{Tr}[\mathcal{D}\Pi] + \boxed{\Phi[\mathcal{G},\mathcal{D}]}\end{aligned}$$

BK Functional, Exact



cluster in k space

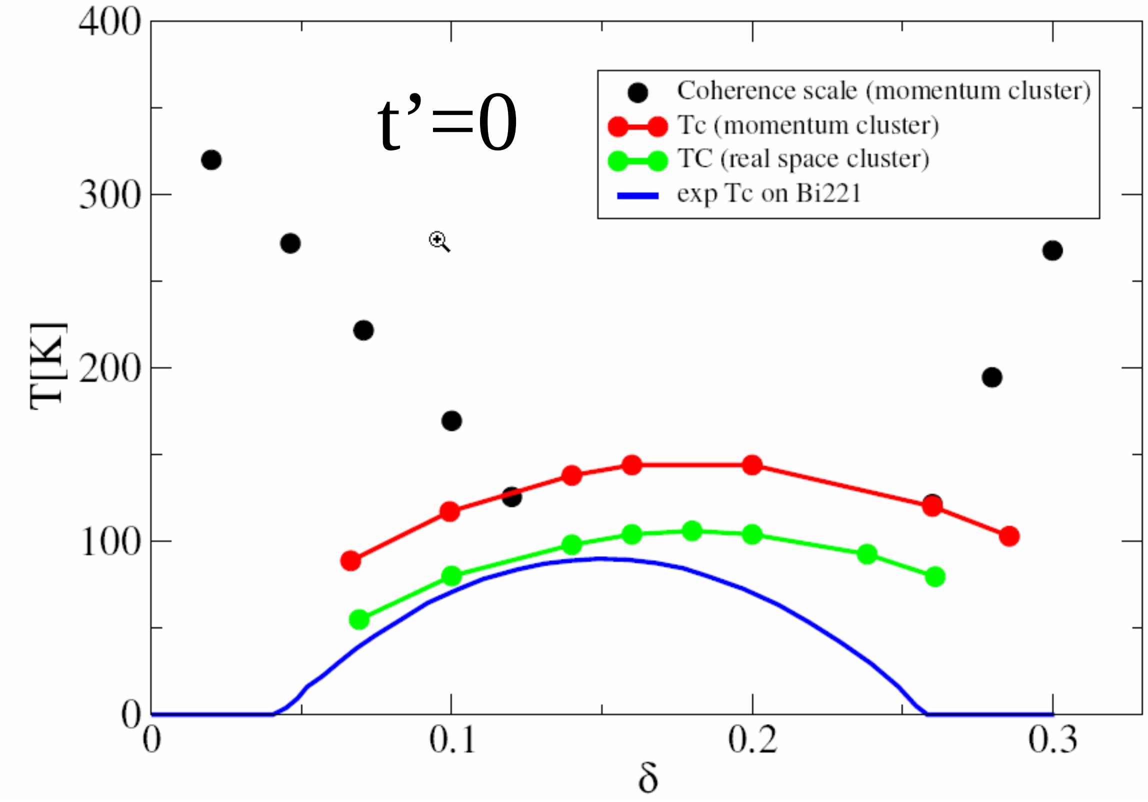
$$\mathcal{G}_{\mathbf{K}}(\omega) \rightarrow \sum_{\mathbf{k}\in\mathbf{K}} \mathcal{G}_{\mathbf{k}}$$

cluster in real space

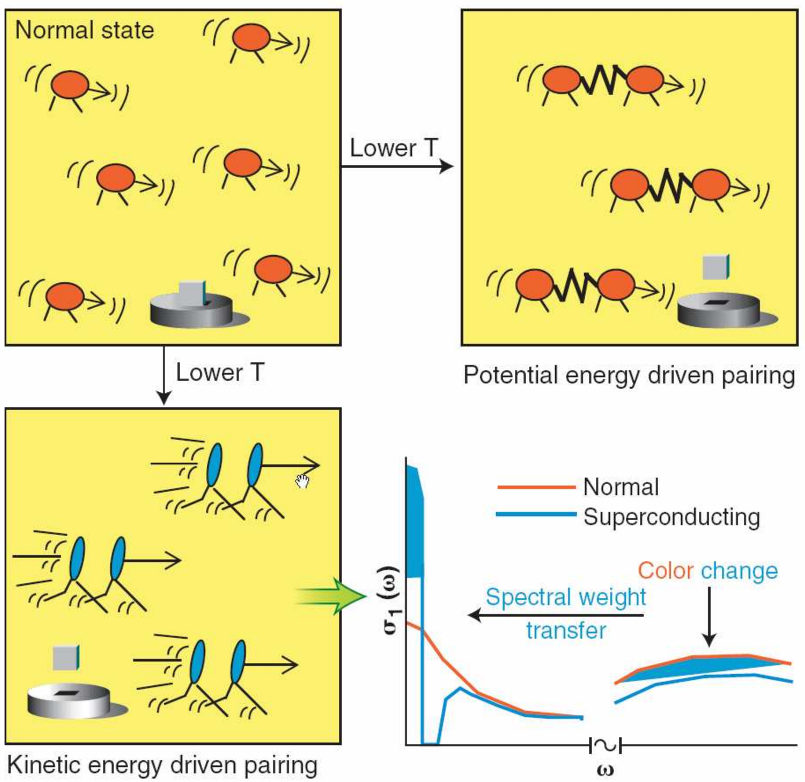
$$\mathcal{G}_{\mathbf{R}_i-\mathbf{R}_j}(\omega) \rightarrow \begin{cases} \mathcal{G}_{\mathbf{R}_i-\mathbf{R}_j}(\omega) & \mathbf{R}_i - \mathbf{R}_j \textit{ inside the cluster} \\ 0 & \textit{otherwise} \end{cases}$$

What can we learn from "small" Cluster-DMFT?

Phase diagram



Insights into superconducting state (BCS/non-BCS)?



BCS: upon pairing potential energy of electrons decreases, kinetic energy increases

(cooper pairs propagate slower)

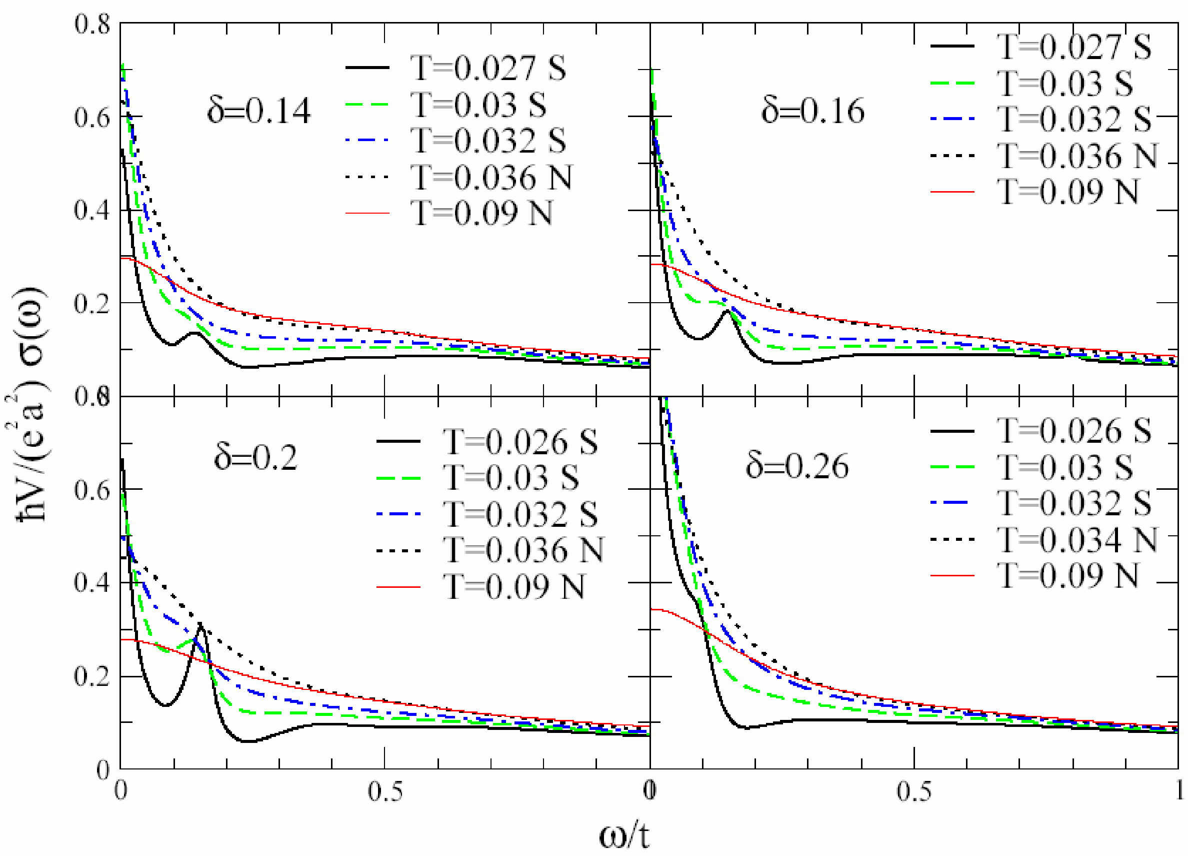
Condensation energy is the difference

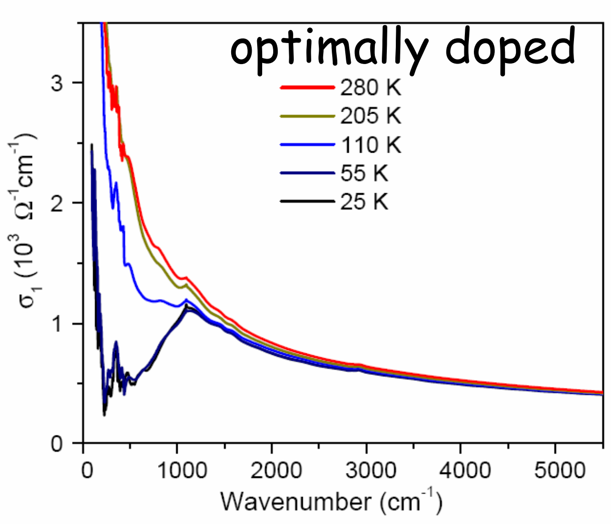
non-BCS: kinetic energy decreases upon pairing

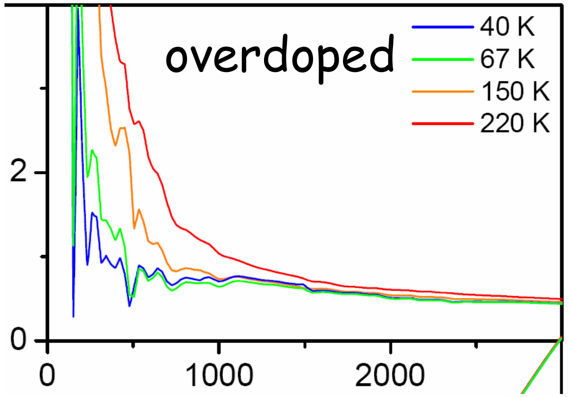
(holes propagate easier in superconductor)

J. E. Hirsch, *Science*, **295**, 5563 (2226)

Optical conductivity







cond-mat/0601478

D van der Marel, Nature 425, 271-274 (2003)

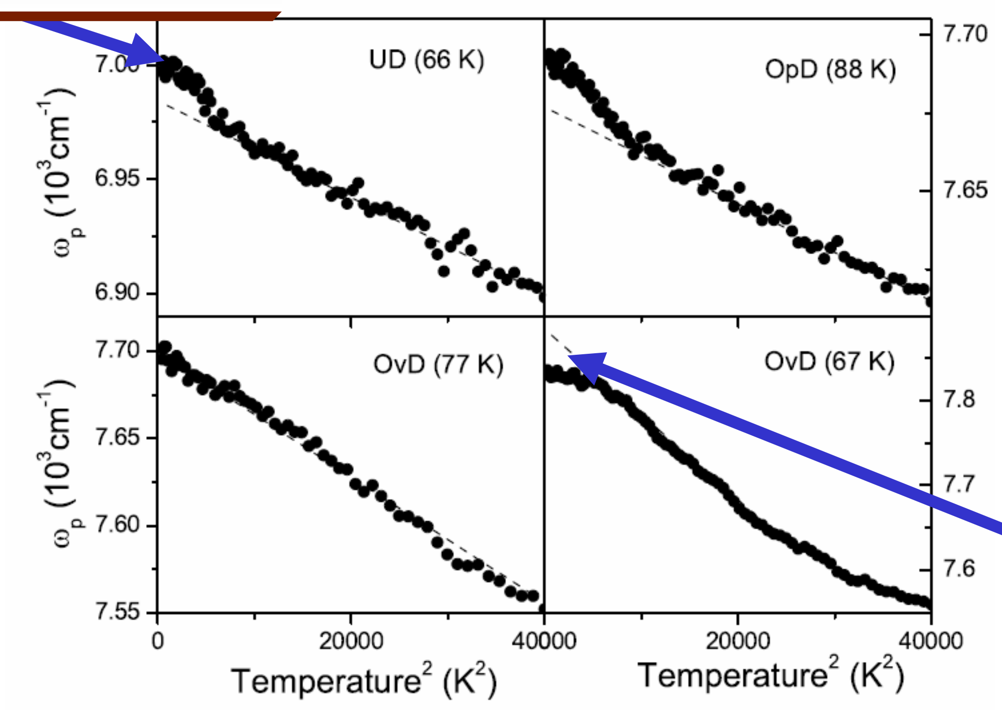
Optical weight, plasma frequency

$$W(\Omega_c, T) \equiv \int_0^{\Omega_c} \sigma_1(\omega, T) d\omega = \frac{\pi e^2 a^2}{2\hbar^2 V} < -\hat{K} >$$

Ω_c ~1eV

Weight bigger in SC,
K decreases (**non-BCS**)

Bi2212



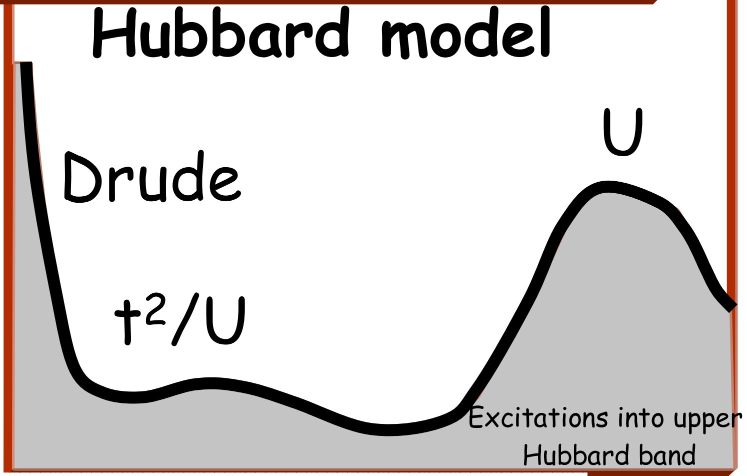
Weight smaller in SC,
K increases (**BCS-like**)

D. van der Marel et.al., in preparation

Hubbard versus t-J model

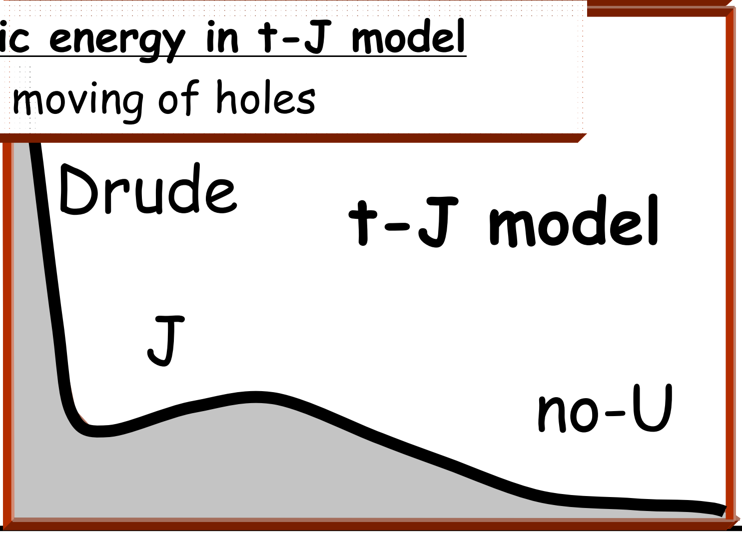
Kinetic energy in Hubbard model:

- Moving of holes
- Excitations between Hubbard bands

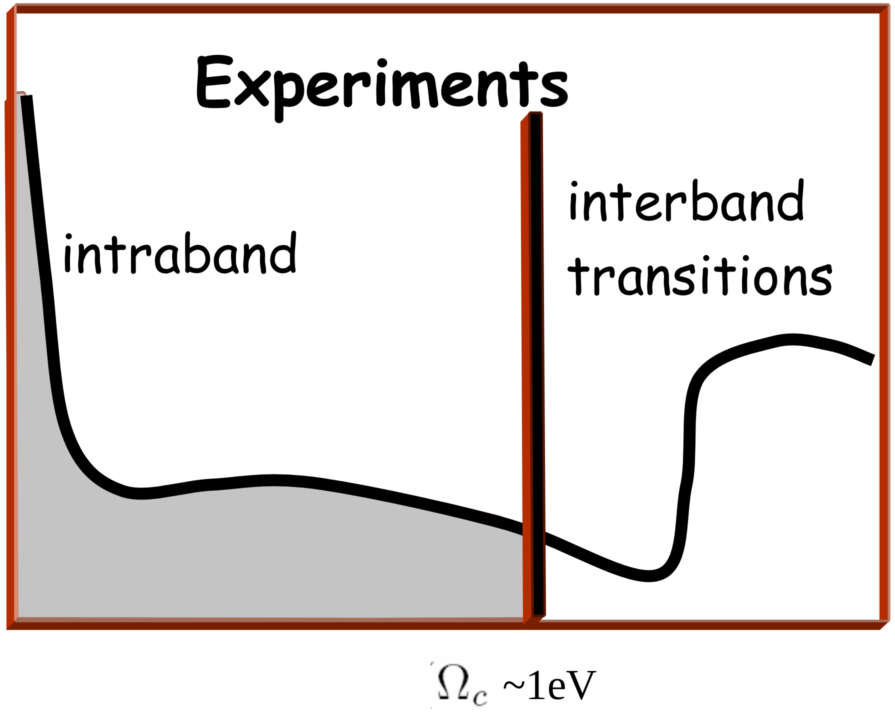


Kinetic energy in t-J model

- Only moving of holes



$$W(\Omega_c,T) \equiv \int_0^{\Omega_c} \sigma_1(\omega,T)\,d\omega = \frac{\pi e^2 a^2}{2\hbar^2 V} < -\hat{K} >$$



Kinetic energy change

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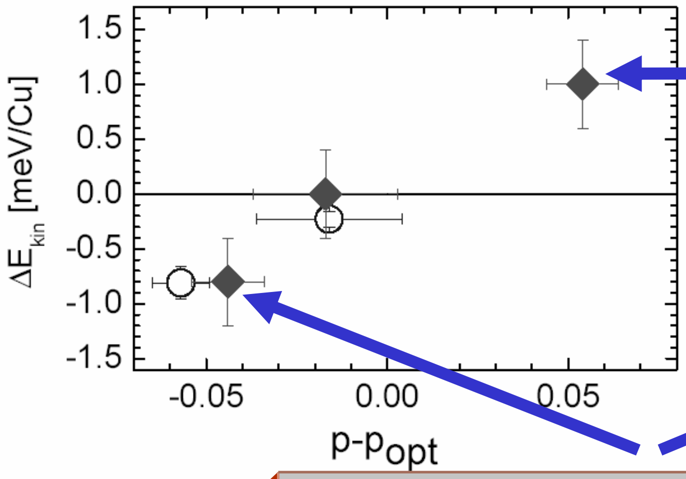
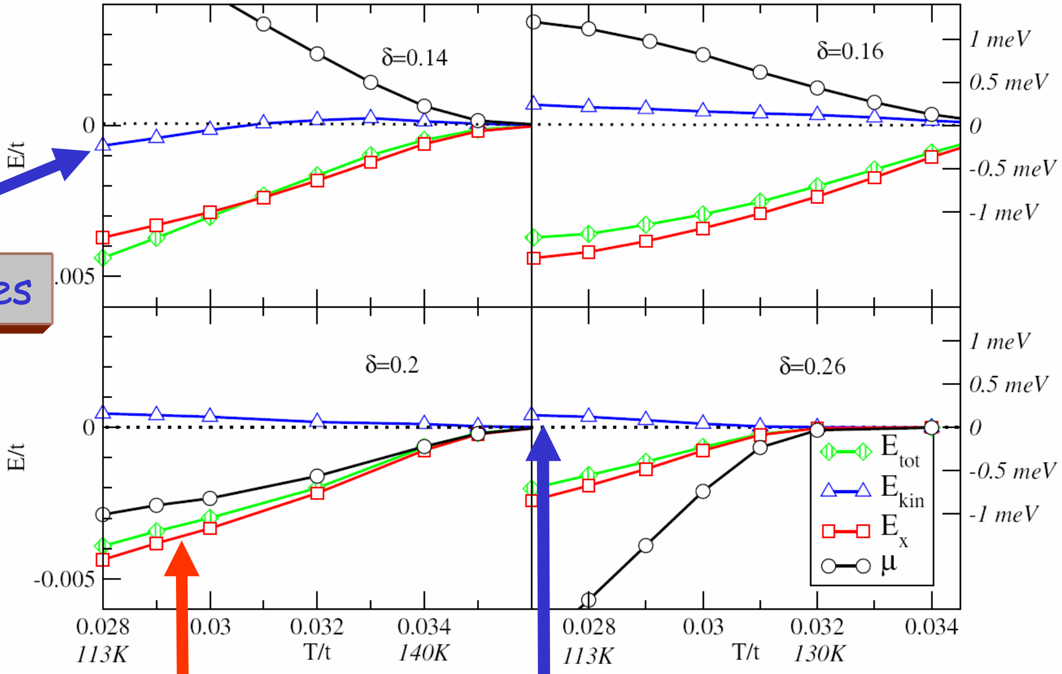


Kinetic energy increases

cluster-DMFT, cond-mat/0601478

Kinetic energy decreases

FIG. 2: Change ΔE_{kin} of the kinetic energy, in meV per copper site, calculated from equations (1) and (3), versus the charge p per copper with respect to p_{opt} (Eq.6). Full diamonds: data from Ref.[3], high frequency cut-off 1 eV. Open circles: data from Ref.[1], high frequency cut-off 1.25 eV. Error bars: vertical, uncertainties due to the extrapolation of the temperature dependence of the normal state spectral weight down to zero temperature; horizontal, uncertainties resulting from $T_c/T_{c,max}$ through Eq.6 (see text). We have taken $T_{c,max} = (83 \pm 2)$ K for films and (91 ± 2) K for crystals.



Kinetic energy increases

Guy Deutscher[1], Andrés Felipe Santander-Syro[2]
and Nicole Bontemps[3]

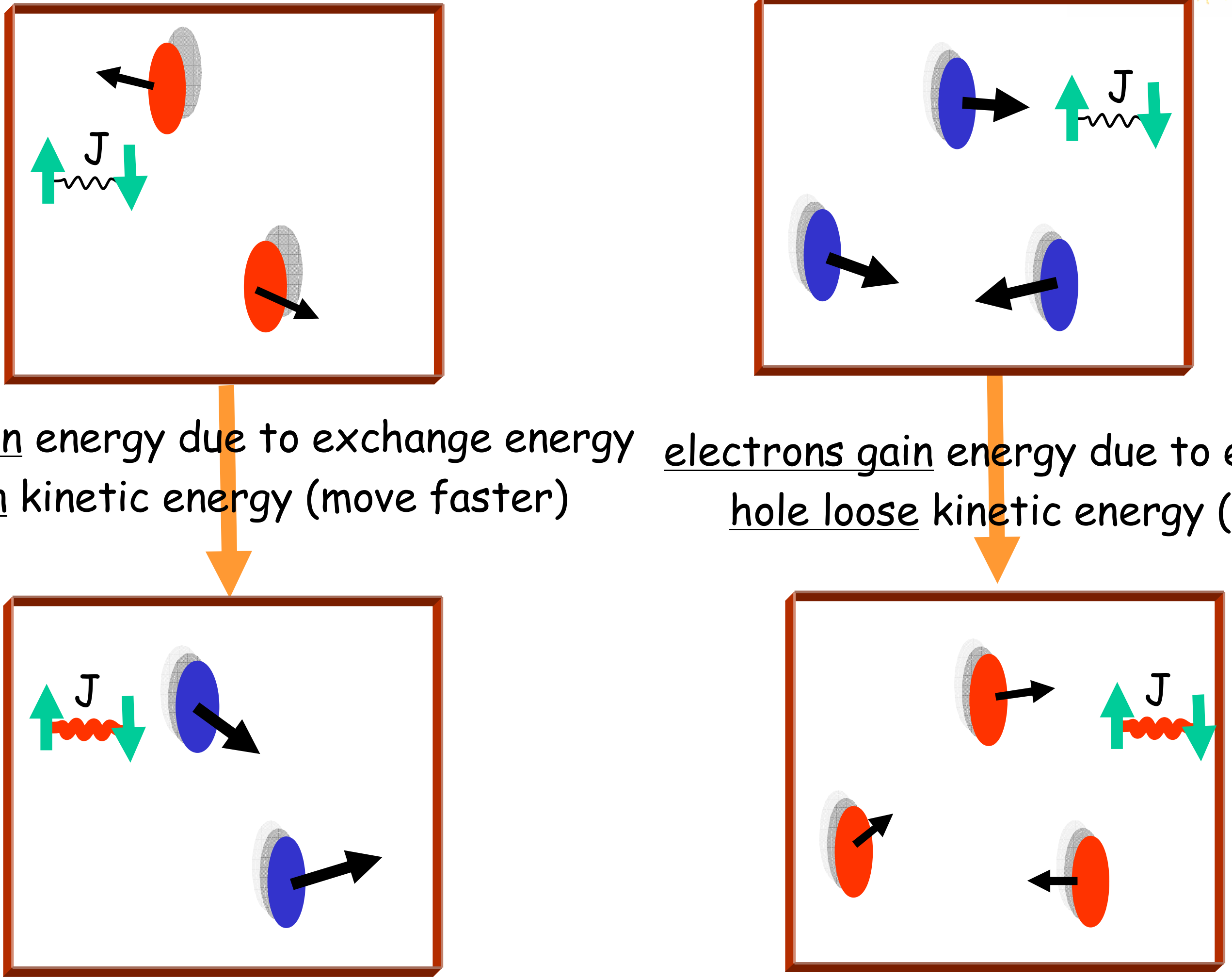
cond-mat/0503073

Phys Rev. B **72**, 092504 (2005)

Exchange energy decreases and gives

largest contribution to condensation energy

Kinetic energy upon condensation



underdoped

electrons gain energy due to exchange energy
holes gain kinetic energy (move faster)

overdoped

electrons gain energy due to exchange energy
hole loose kinetic energy (move slower)

same as RVB (see P.W. Anderson Physica C, 341, 9 (2000),
or slave boson mean field (P. Lee, Physica C, 317, 194 (1999)

BCS like

Conclusions

- LDA+DMFT can describe interplay of lattice and electronic structure near Mott transition. Gives physical connection between spectra, lattice structure, optics,....
  - Allows to study the Mott transition in open and closed shell cases.
  - In both Ce and Am single site LDA+DMFT gives the zeroth order picture
  - Am: Rich physics, mixed valence under pressure.
  - Describes magnetism of Curium
- 2D models of high-Tc require cluster of sites. Some aspects of optimally doped, overdoped and slightly underdoped regime can be described with cluster DMFT on plaquette:
  - Evolution from kinetic energy saving to BCS kinetic energy cost mechanism