Multiorbital Mott systems: new results from continuous-time Quantum Monte Carlo calculations

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Support:
DOE ER-46169,
NSF-DMR-0705847

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References:

Phys. Rev. Lett. **97**, 056802 (2006).
Phys. Rev. **B74**, 155107 (2006).
Phys. Rev. **B** in press (cond-mat/0609438)
Phys. Rev. **B** in press (cond-mat/07040057)

(see also Phys. Rev. **B75**, 085108 (2006) and cond-mat/0701730 (PRL in press))

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Outline

- **Physics problem=>theoretical problem**
- **New "CT-QMC" solver**
- **Results: 2 and 3 orbital models--**
  - **classifying Mott insulators**
  - **crystal field splitting and orbital selectivity**
- **Summary and prospects**

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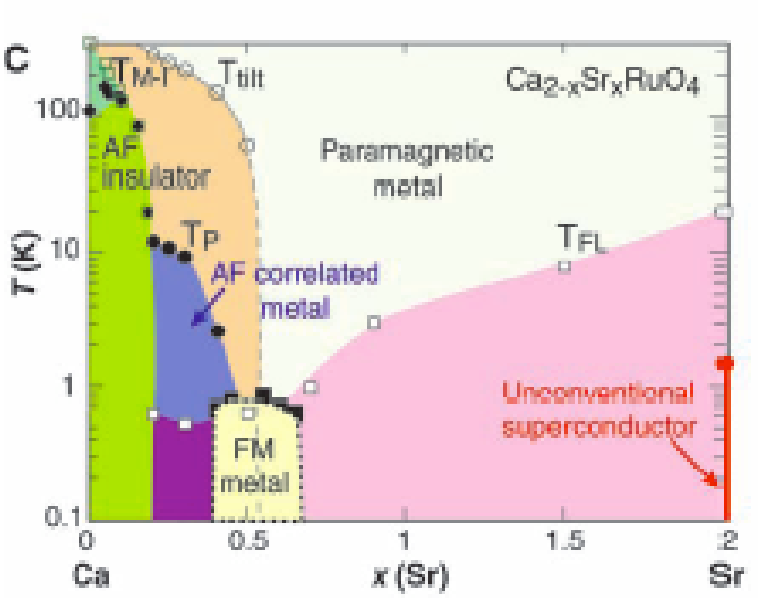
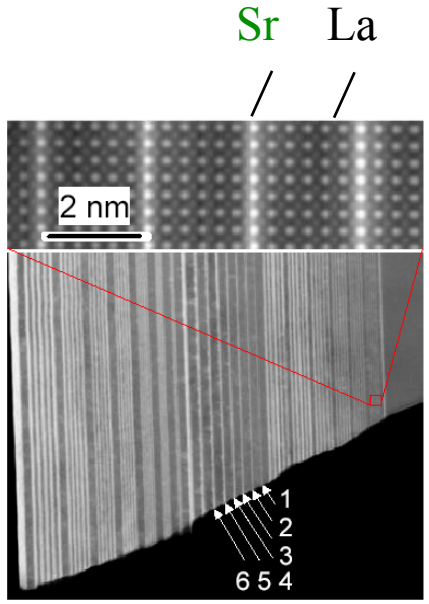
Transition metal oxides with partly filled d-shells: many challenges

Ca/SrRuO:
(Maeno/MacKenzie/....



Ru: t_{2g} d-orbitals (3x degenerate)
$SrRuO_3$: Ferromagnet
Ca_2RuO_4: Mott insulator

$LaTiO_3/SrTiO_3$ heterostructure
(Ohtomo et al Nature 2002)



Ti: t_{2g} d-orbitals (3x degenerate)
$LaTiO_3$: Mott insulator
$SrTiO_3$: Band insulator

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Work-horse of Theoretical Materials Science: Density Functional Theory

Basic Theorem (Hohenberg and Kohn): $\exists$ functional Φ of electron density n(r): minimized at physical density; value at minimum gives ground state energy

$$\Phi[\{n(r)\}] = \Phi_{univ}[\{n(r)\}] + \int (dr) V_{lattice}(r) n(r)$$

Useful because:

***Have uncontrolled (but apparently good) approximations to Φ**

***Have efficient way to carry out minimization**

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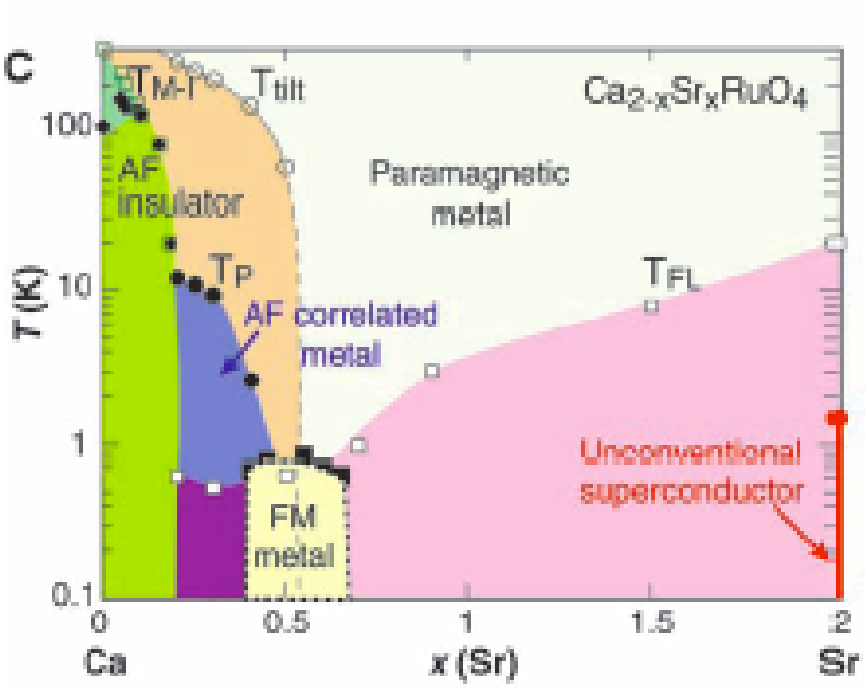
Density Functional Theory: Difficulties

$$\Phi[\{n(r)\}] = \Phi_{univ}[\{n(r)\}] + \int (dr) V_{lattice}(r) n(r)$$

Density is not the optimal variable: phases with quite different physical properties have almost the same density



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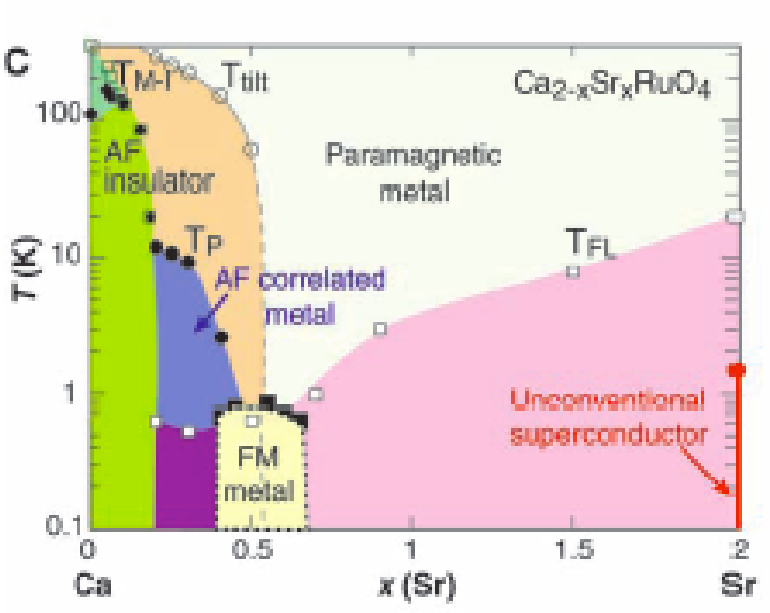
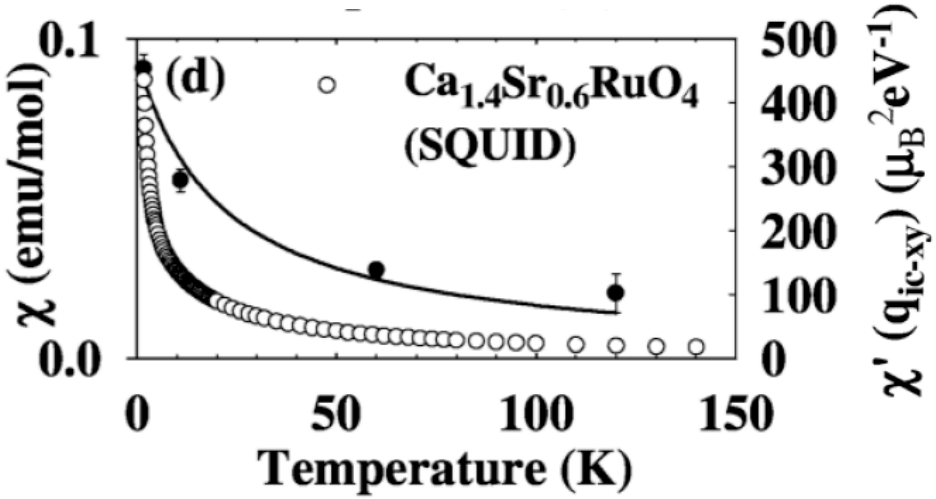
Density Functional Theory: Difficulties

Ground state is not the only interest: different phases at different temperatures: need theory with local moments, entropic effects





Fermi liquid; low T. ?Local moment? at higher T?

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Density Functional Theory: Difficulties

Density functional theory: focus is on ground state but excitations are important

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Example: photoemission from Sr/LaTiO3 heterostructure:

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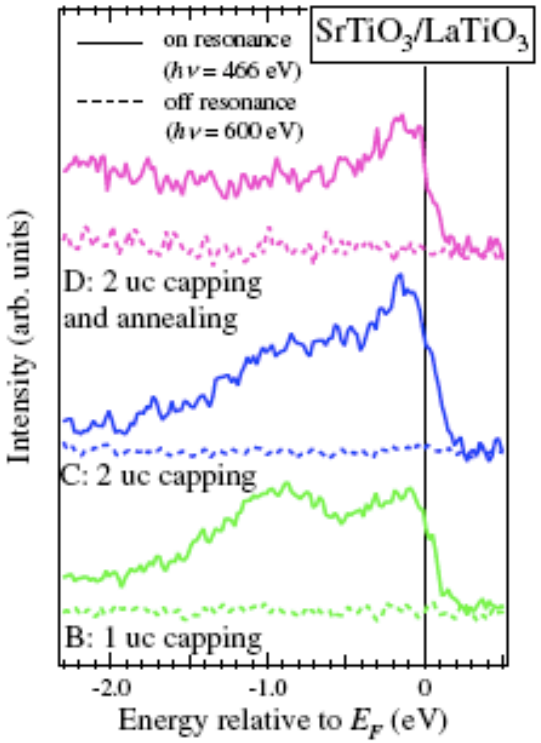

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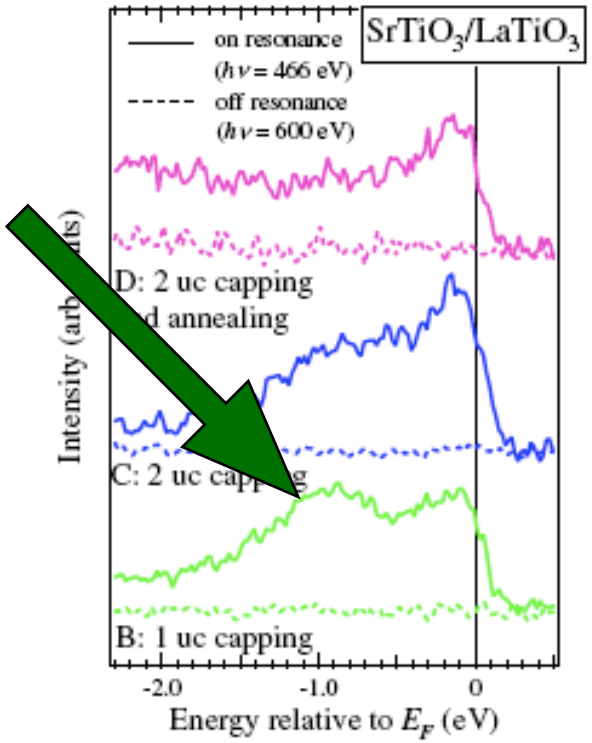
Density Functional Theory: Difficulties

Density functional theory: focus is on ground state but excitations are important

Example: photoemission from Sr/LaTiO3 heterostructure:
"shakeoff band": is this incipient "lower hubbard band"



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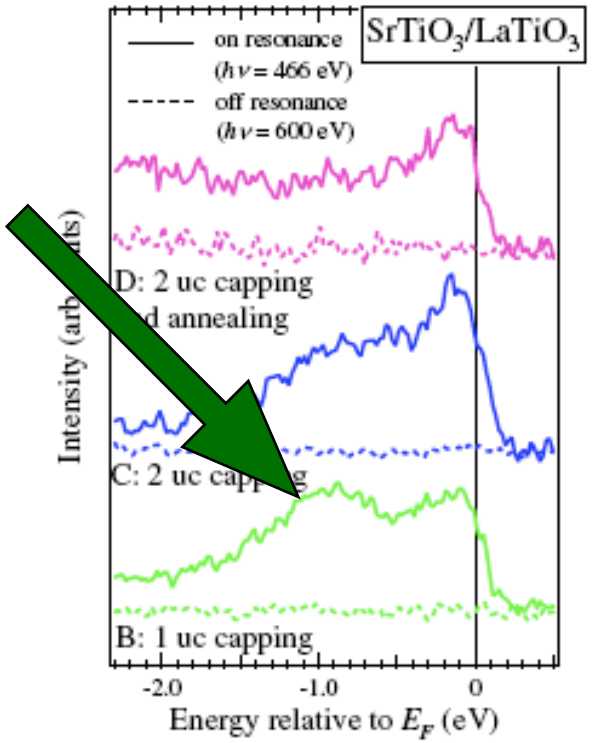

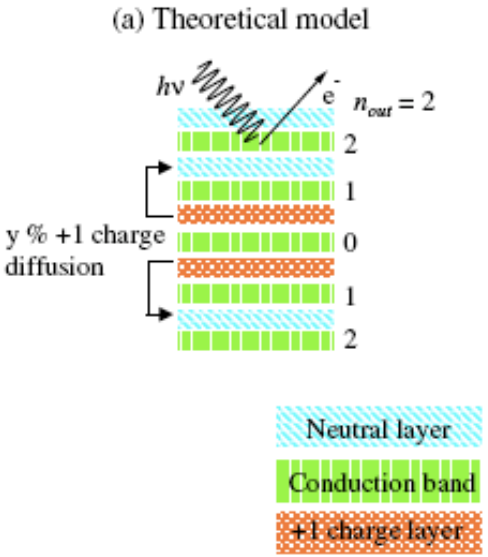

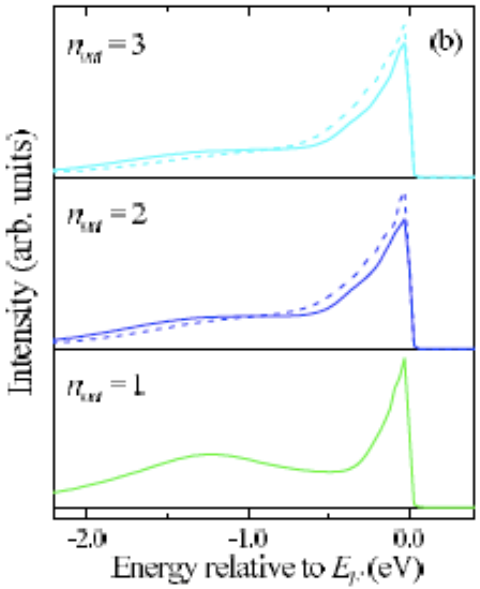

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Dynamical Mean Field Method

Metzner/Vollhardt; Mueller Hartmann **KOTLIAR/GEORGES**

- **Standard many-body theory=>exists a functional of self energy**

$$F[\{\Sigma(p,\omega\}] = F_{univ}[\{\Sigma(p,\omega\}] + Tr\left[ln\left(G_0^{-1}(p,\omega) - \Sigma(p,\omega)\right)\right]$$

extremized at correct self energy and from which ALL RESPONSE FUNCTIONS can be extracted.

- **But: F_{univ} only known perturbatively=> general formalism not useful. (Kotliar/Georges): there is an ?accurate? approximation**

$$\Sigma_p(\omega) \rightarrow \Sigma_p^{approx}(\omega) = \sum_a \phi_a(p)\Sigma_a(\omega)$$

and 'convenient' procedure for doing minimization over restricted sub-space of approximate self energies

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Dynamical Mean Field II

$$\Sigma_p(\omega) \rightarrow \Sigma_p^{approx}(\omega) = \sum_a \phi_a(p)\Sigma_a(\omega)$$

Different choices of basis function $\phi_a(p)$=> different "flavors" of DMFT (1-site, DCA, CDMFT....).

This talk: single-site $\phi_a(p) \rightarrow \phi_a = 1$ **"a": local orbitals**

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Dynamical Mean Field: III

$$F[\{\Sigma_p(\omega\}] \rightarrow F_{approx} =$$

$$F_{univ}\left[\{\sum_a \phi_a(p)\Sigma_a(\omega)\}\right] + Trln\left[G_0^{-1} - \sum_a \phi_a(p)\Sigma_a(\omega)\right]$$

- **F_{approx}: functional of a small number of functions of frequency <=> "quantum impurity model", i.e. a (0+1) dimensional quantum field theory**
- **Stationarity w.r.t variations in self energy:**

$$\frac{\delta F_{univ}}{\delta \Sigma_a(\omega)} \equiv G_a^{QI}(\omega) = \int (dp) \frac{\phi_a(p)}{\omega - \varepsilon_p - \Sigma_p^{approx}(\omega)}$$

fixes form of quantum impurity model
(n.b.: issues remain with choosing interaction term)

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Dynamical Mean Field IV

Challenge: accurate solution of quantum impurity model

<=>find local (d-d) green functions of

$$H_{QI} = H_{loc}[\{d_a^\dagger, d_a\} + \sum_{p,a} \left(V_{pa} d_a^\dagger c_{pa} + H.c\right) . + H_{bath}[\{c_{pa}^\dagger c_{pa}\}]$$

ex: Slater-Kanamori d-multiplet interactions

$$H_{e-e}^{(i)} = U \sum_a n_{ia\uparrow} n_{ia\downarrow} + (U' - J) \sum_{a>b,\sigma} n_{ia\sigma} n_{ib\sigma} + U' \sum_{a\neq b} n_{ia\uparrow} n_{ib\downarrow}$$

$$+ J \sum_{a\neq b} d_{ia\uparrow}^\dagger d_{ib\uparrow} d_{ib\downarrow}^\dagger d_{ia\downarrow}. \qquad (4)$$

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What can we do?

DMFT work-horse: "Hirsch-Fye" quantum Monte Carlo

Write model as (imaginary time) path integral Discretize time axis. At time step i, use discrete Hubbard-Stratonovich transformation

$$e^{\Delta\tau U(n_\uparrow - n_\downarrow)^2} = \frac{1}{2}\left(e^{\lambda_i(n_\uparrow - n_\downarrow)} + e^{-\lambda_i(n_\uparrow - n_\downarrow)}\right)$$

=>problem reduces to solving non-interacting electron problem in some time dependent configuration of "magnetic" fields, then summing over all field configurations

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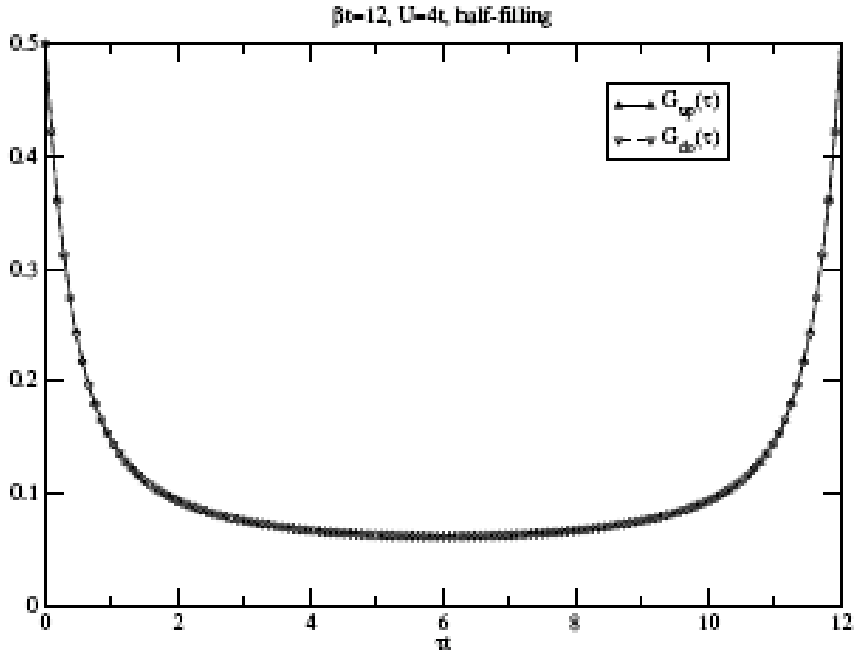
Issues with Hirsh-Fye

- **time discretization: need fixed time grid, but main contribution to the energy is in the details of the initial drop of G=> need many time slices**



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Issues with Hirsh-Fye II

- **partitioning of phase space--at strong coupling, simulation has trouble equilibrating**

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Plot: probability of average value of auxiliary field

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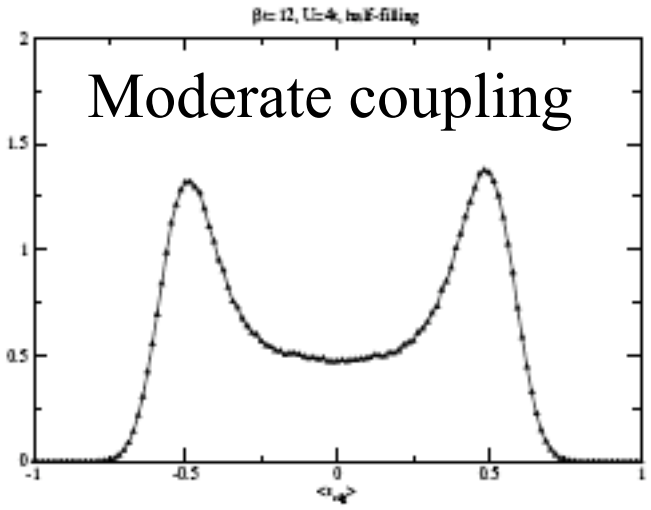

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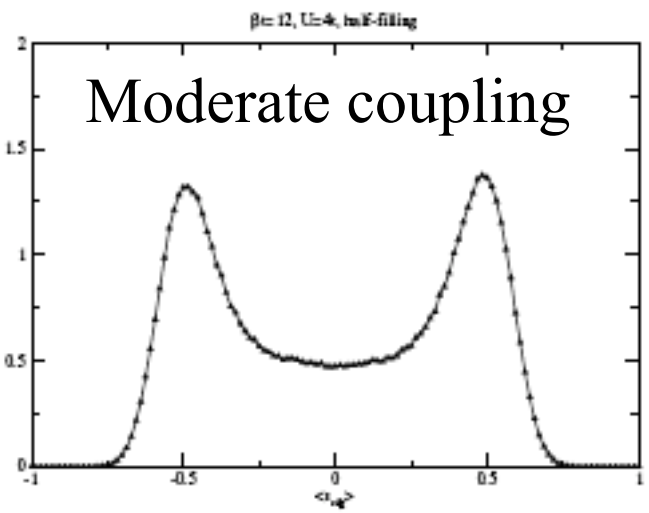
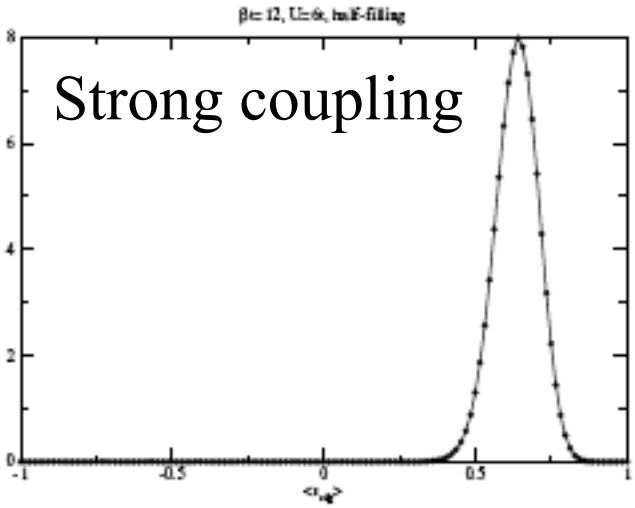
Plot: probability of average value of auxiliary field





At strong coupling, simulation trapped in wrong minimum

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Issues with Hirsh-Fye III

- **For "Slater-Kanamori" multiplet interactions, no good decoupling exists**

$$H_{e-e}^{(i)} = U\sum_{a} n_{ia\uparrow}n_{ia\downarrow} + (U' - J)\sum_{a>b,\sigma} n_{ia\sigma}n_{ib\sigma} + U'\sum_{a\neq b} n_{ia\uparrow}n_{ib\downarrow}$$
$$+ J\sum_{a\neq b} d_{ia\uparrow}^{\dagger}d_{ib\uparrow}d_{ib\downarrow}^{\dagger}d_{ia\downarrow}. \qquad (4)$$

*Too many terms
*Rotational invariance hard to implement.

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Much important work optimizing Hirsh-Fye methods

- **N. Bluemer (Ph.D. thesis and recend cond-mat)--minimizing time discretization errors**
- **T. Sakai--Hubbard-Stratonovich for Slater-Kanamori interactions (many auxiliary fields!)**

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Needed: other methods

- **"Exact diagonalization" (Caffarel/Krauth; Capone; Liebsch)**

$$H_{QI} = H_{loc}[\{d_a^\dagger, d_a\} + \sum_{p,a} \left(V_{pa} d_a^\dagger c_{pa} + H.c\right) . + H_{bath}[\{c_{pa}^\dagger c_{pa}\}]$$

  - **approximate continuous bath by small number (typically 6-9) of appropriately chosen states**

- "CT-QMC" (Rubtsov; Werner)
  - Expand in H_{loc} (Rubtsov) or in V/T (Werner)

This talk: Expand in V/T

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Basic idea:

$$H_{QI} = H_{loc}[\{d_a^\dagger, d_a\} + \sum_{p,a} \left(V_{pa} d_a^\dagger c_{pa} + H.c\right). + H_{bath}[\{c_{pa}^\dagger c_{pa}\}]$$

- **interaction representation with respect to H_{loc}, H_{band}**

$$Z = Tr\left[T_\tau e^{\sum_{p,a}\left(V_{pa}^I d_a^\dagger(\tau) c_{pa}(\tau) + H.c.\right)}\right]$$

- **formal expansion in V**

$$= \sum_k \frac{1}{k!} \int_0^\beta d\tau_1 ... d\tau_k Tr\left[T_\tau \hat{\mathbf{V}}^{\mathbf{I}}(\tau_1) ... \hat{\mathbf{V}}^{\mathbf{I}}(\tau_k)\right]$$

- **sample series stochastically: add/remove V; accept or reject by usual importance sampling**

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Technical issues

$$Tr\left[T_\tau \hat{\mathbf{V}}^{\mathbf{I}}(\tau_1)...\hat{\mathbf{V}}^{\mathbf{I}}(\tau_k)\right] = Tr\left[T_\tau d^\dagger d d^\dagger ...\right] * Tr\left[T_\tau c^\dagger_{pa} c_{pa} ...\right]$$

- **Tr[c...]--determinant=> sum all contractions at once. Essential to do this, or face serious sign problem. No sign problem found so far in calculations Tr[d...]--product of matrices in Hilbert space of H_{loc}**
- **n orbitals=>4^n dimensional matrices. As yet--no "fast update" for trace or quick way to know if Tr of product vanishes. Important limitation. 3 orbitals or 4-site cluster possible. Bigger system=>use tricks**

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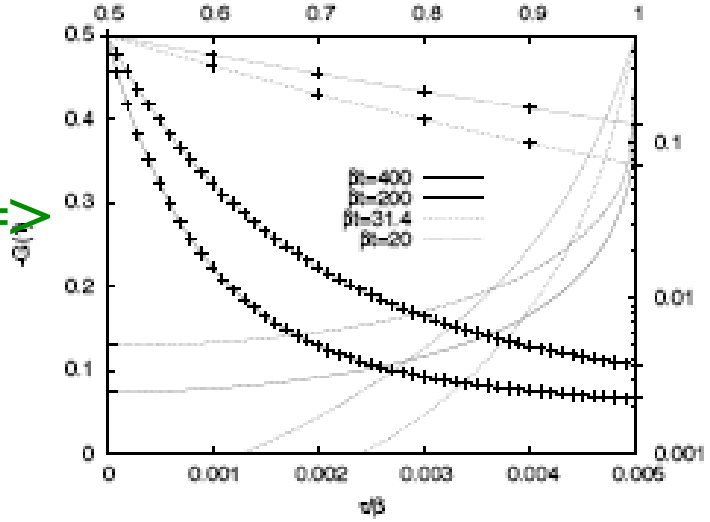
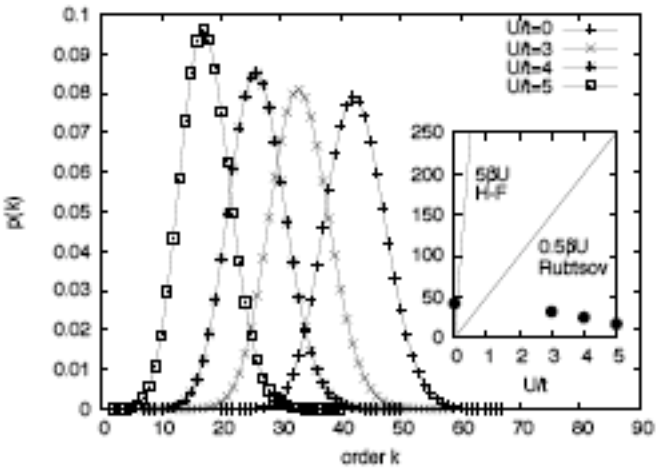
Advantages:

- **Continuous time=> more points where G varies fast=> Much better energies**



- **Mean perturbation order lower at strong coupling**



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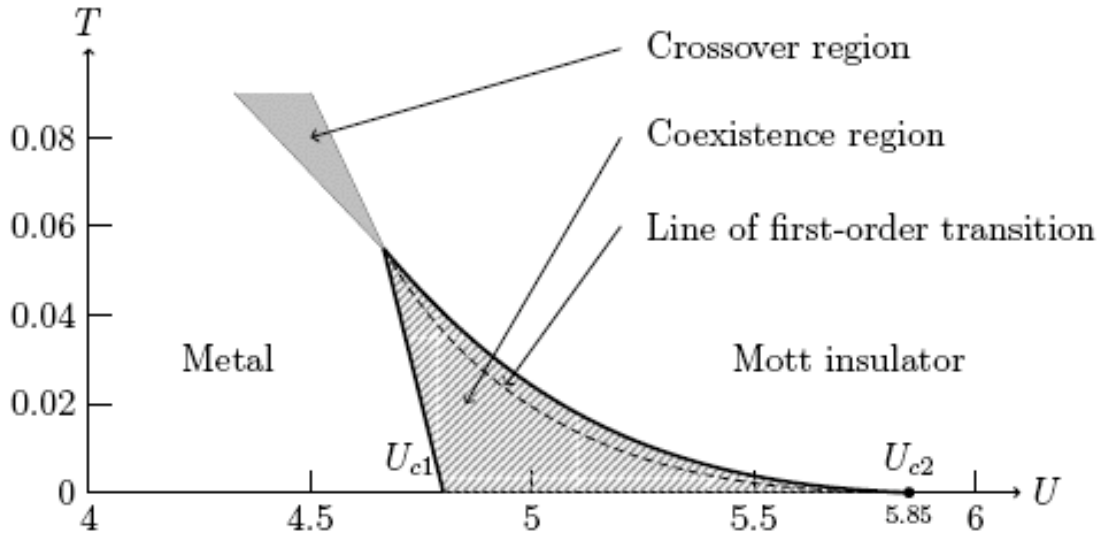
1 orbital Hubbard model

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

Single-site DMFT phase diagram:

Bethe lattice.
bandwidth =4



$U<U_{c2}$ paramagnetic metal (if no AF order or correlations)
$U>U_{c2}$: Mott insulator.

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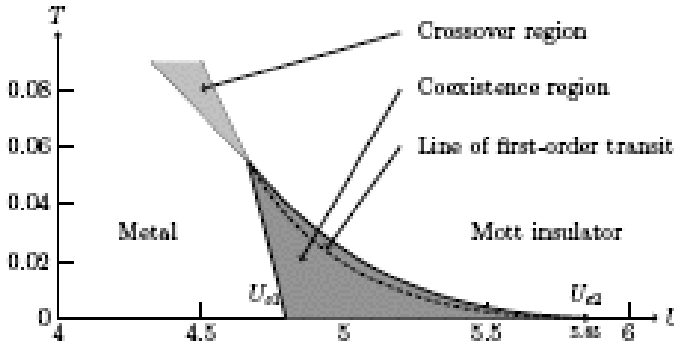
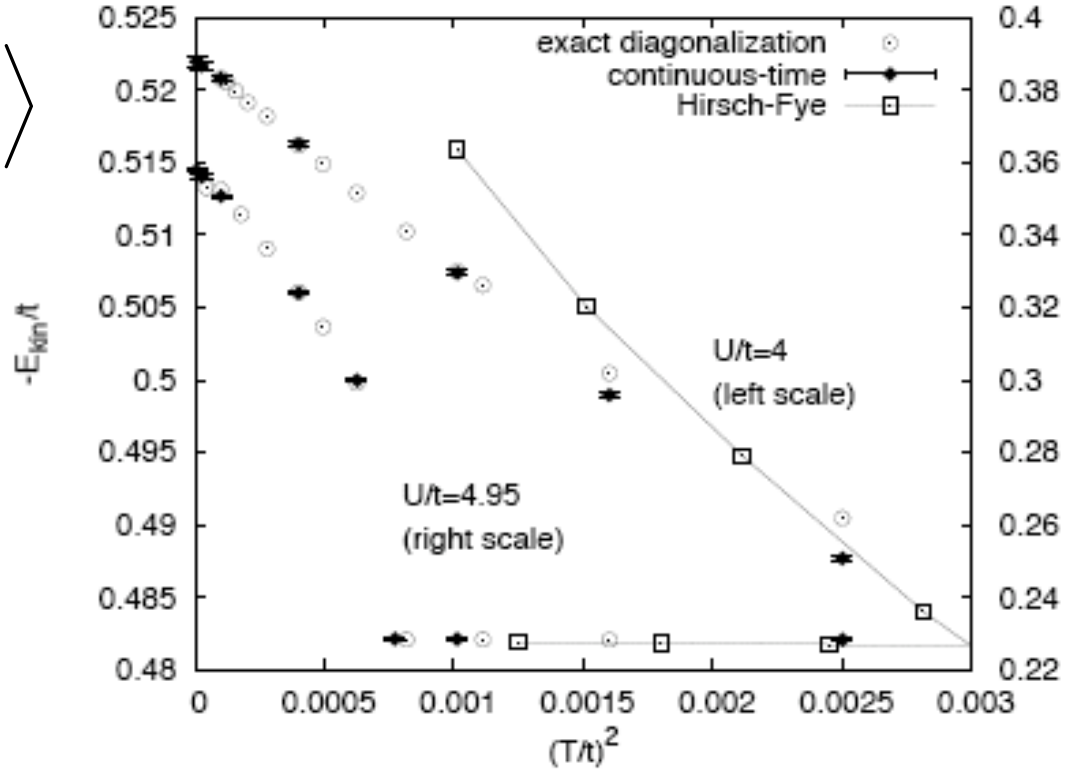
1 orbital Hubbard model

Compare: Hirsh-Fye, CT-QMC, T-dep ED

$$K = \sum_{ij,\sigma} \left\langle t_{ij} c^{\dagger}_{i,\sigma} c_{j,\sigma} + H.c. \right\rangle$$





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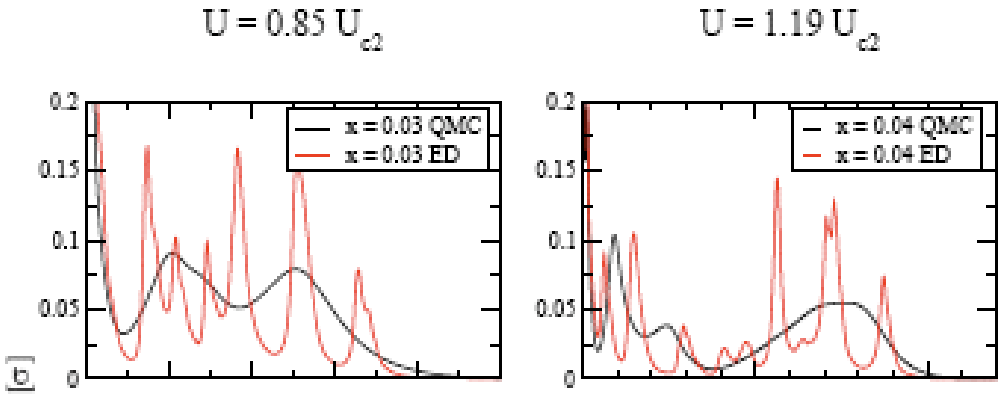
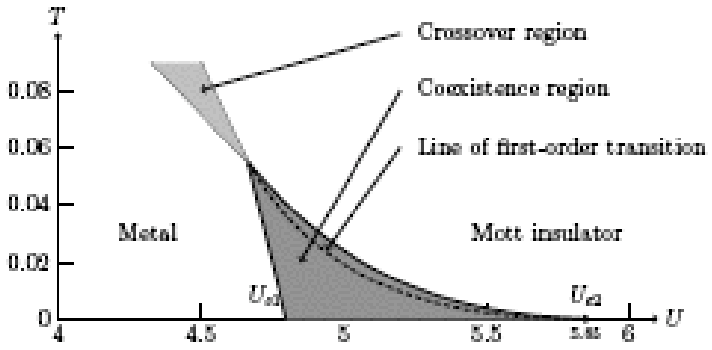
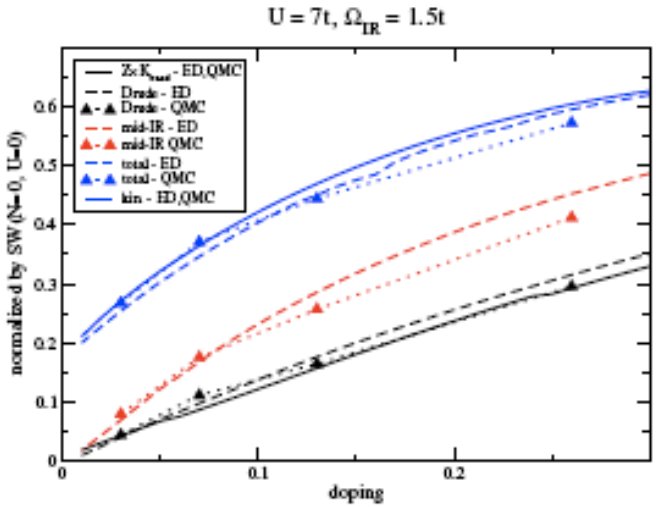
1 orbital Hubbard model

Compare: CT-QMC+analytical continuation, ED

$$K(\Omega) = \int_0^{\Omega} \frac{2d\omega}{\pi}\sigma(\omega)$$







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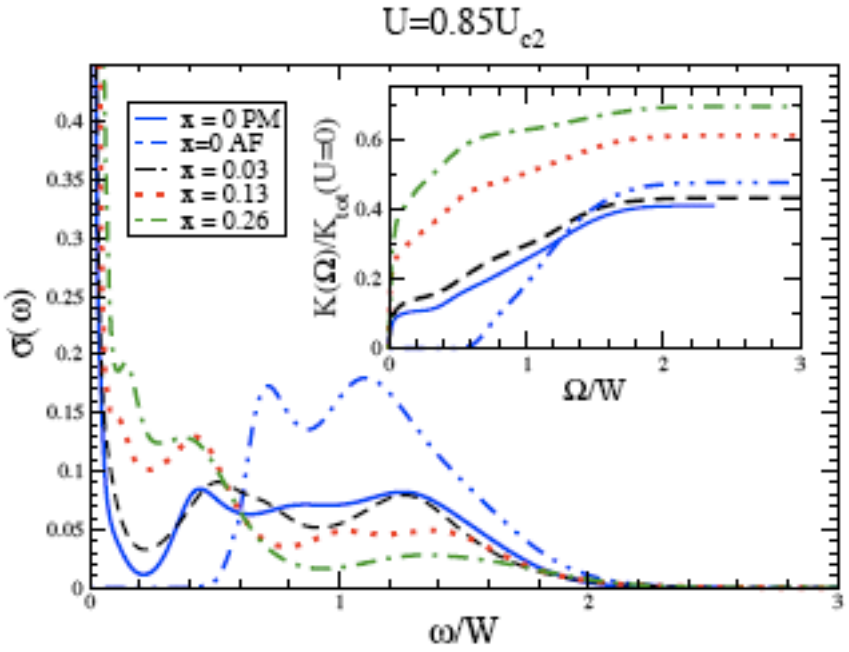
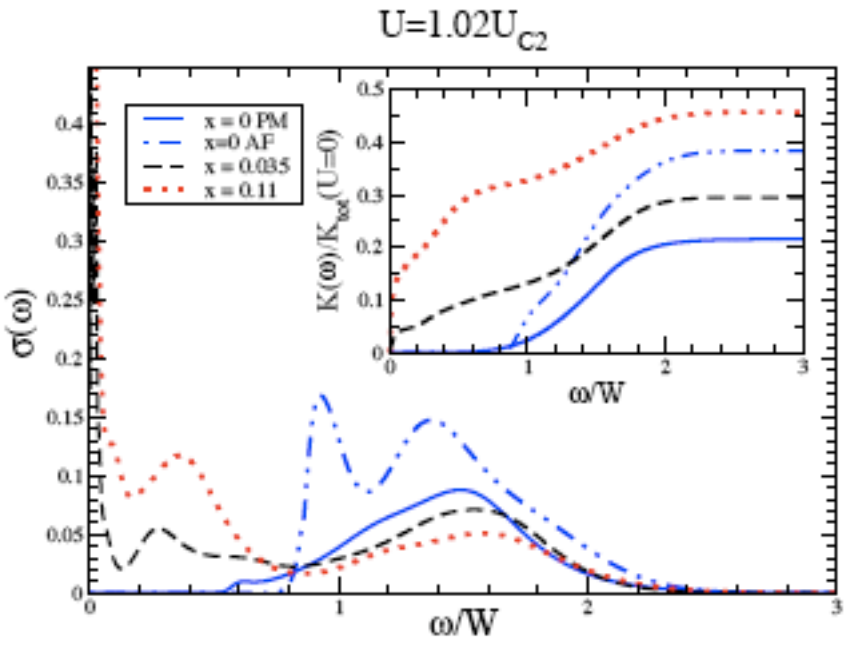
Conductivity of Hubbard model
PM and AF phases

W=bandwidth





U>U$_{c2}$, paramagnetic (Mott) phase: only very small fraction of spectral weight near gap edge

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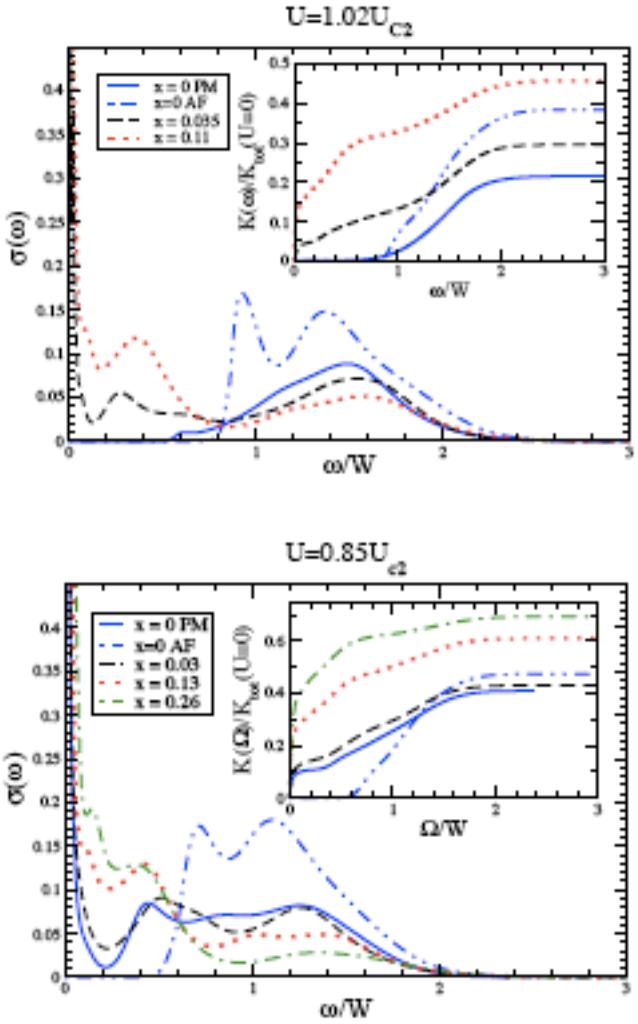
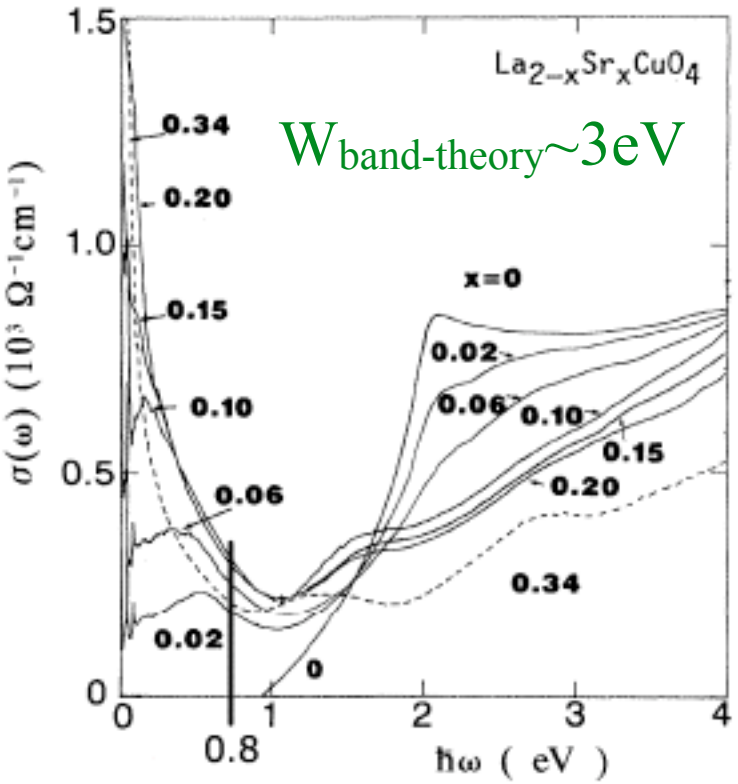
Conductivity of Hubbard model Compare to Data





$K(W)=0.2K_{band}$

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Query

Should the cuprates be regarded as Mott insulators if $U<U_{c2}$ and antiferromagnetism (or at least correlations) is needed to stabilize insulating state???

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Multiorbital systems:
Hunds coupling and response to crystal field

- **Many transition metal oxides: partly filled orbitally degenerate d-shells**
- **Okamoto/AJM ($(Sr/Ca)_2RuO_4$) Bierman/ Georges ($BaVaO_3$): nontrivial interplay between Mott threshold, Hunds coupling, crystal field.**
- **Anisimov/Rice,Liebsch, Georges,...Orbitally selective Mott transition**

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Multiorbital systems:
"Two-orbital" model

$$H_{\rm loc} = -\sum_{\alpha=1,2}\sum_{\sigma}\mu n_{\alpha,\sigma} + \sum_{\sigma}\Delta(n_{1,\sigma} - n_{2,\sigma})$$
$$+ \sum_{\alpha=1,2} U n_{\alpha,\uparrow} n_{\alpha,\downarrow} + \sum_{\sigma} U' n_{1,\sigma} n_{2,-\sigma}$$
$$+ \sum_{\sigma}(U' - J) n_{1,\sigma} n_{2,\sigma}$$
$$- J(\psi^{\dagger}_{1,\downarrow}\psi^{\dagger}_{2,\uparrow}\psi_{2,\downarrow}\psi_{1,\uparrow} + \psi^{\dagger}_{2,\uparrow}\psi^{\dagger}_{2,\downarrow}\psi_{1,\uparrow}\psi_{1,\downarrow} + h.c.)$$

U'=U-2J

Temp=0.005W

Issue: interplay between J
and crystal field splitting Δ

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Two-orbital model: orbital filling vs crystal field
2 electrons per site; plot filling/spin

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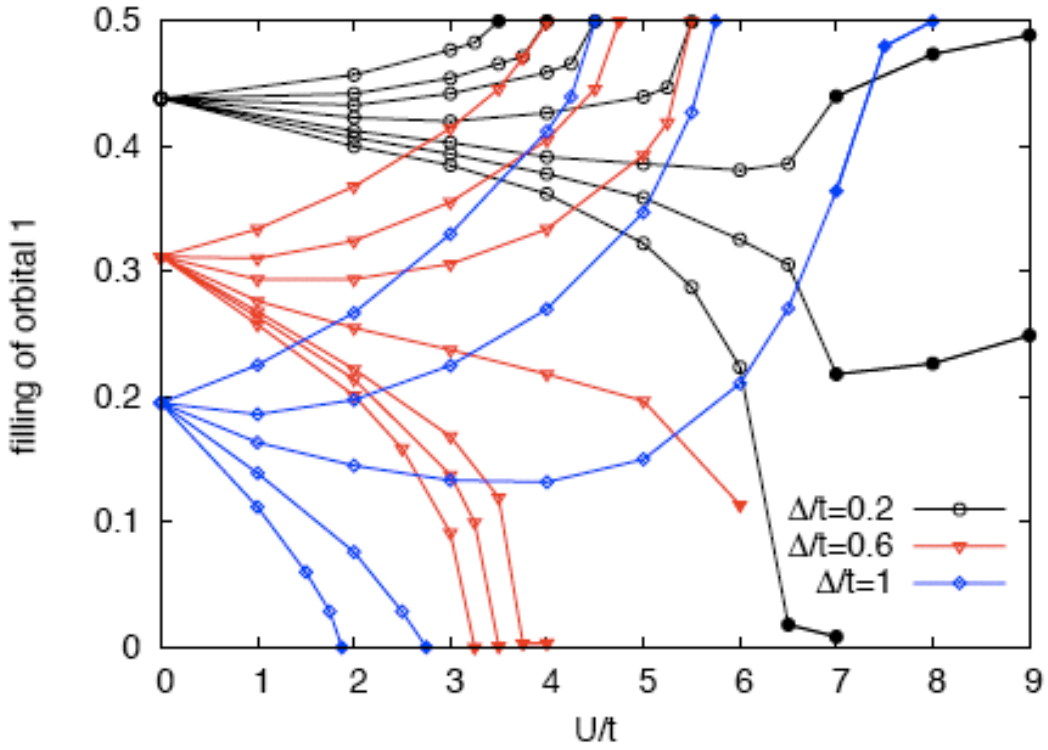
Two-orbital model: orbital filling vs crystal field
2 electrons per site; plot filling/spin



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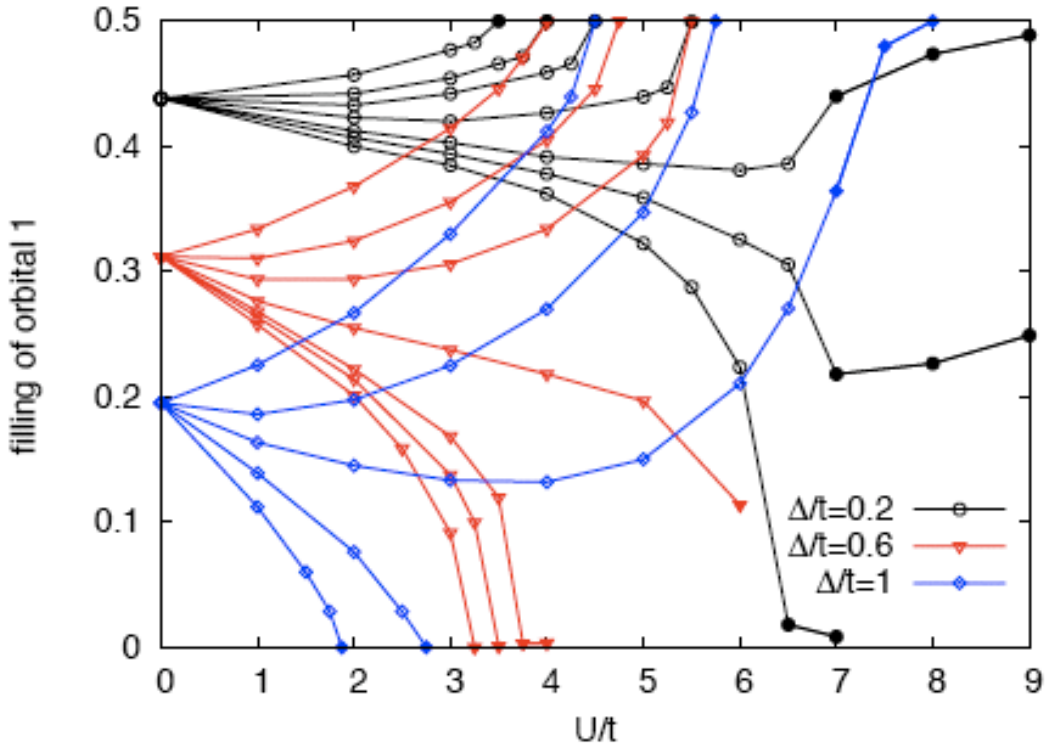
Two-orbital model: orbital filling vs crystal field
2 electrons per site; plot filling/spin



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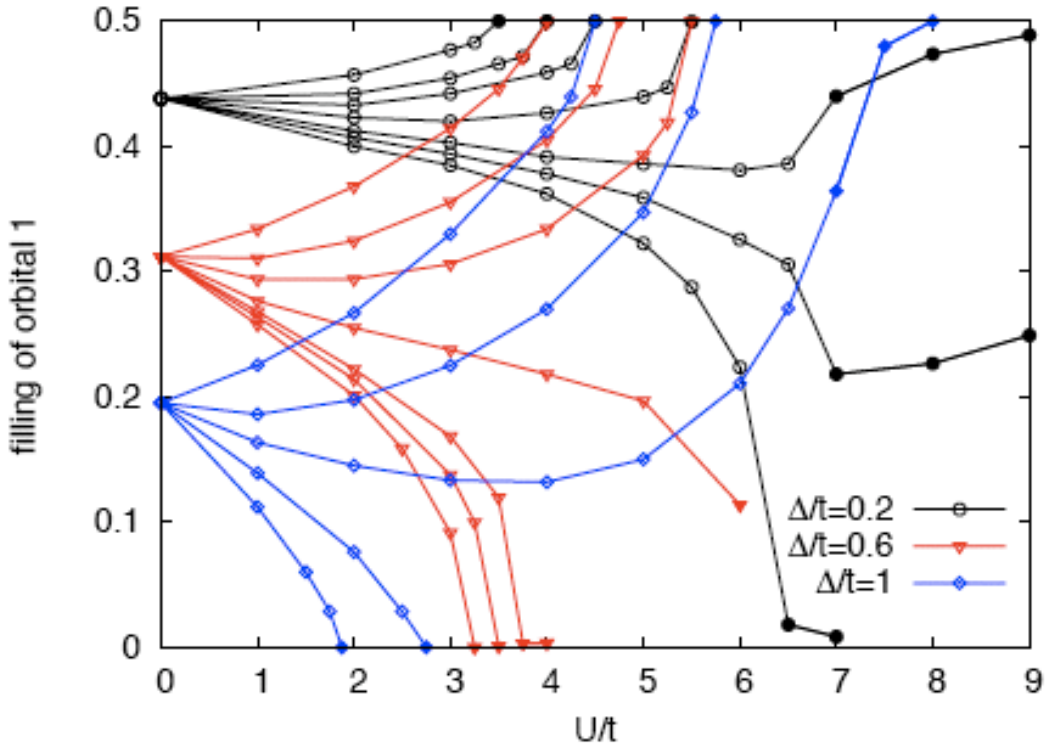
Two-orbital model: orbital filling vs crystal field
2 electrons per site; plot filling/spin

Large J/U:
"orbital stability"
(Okamoto/AJM)



J/U=
0.25
0.15
0.10
0.05
(0.02)
(0.01)

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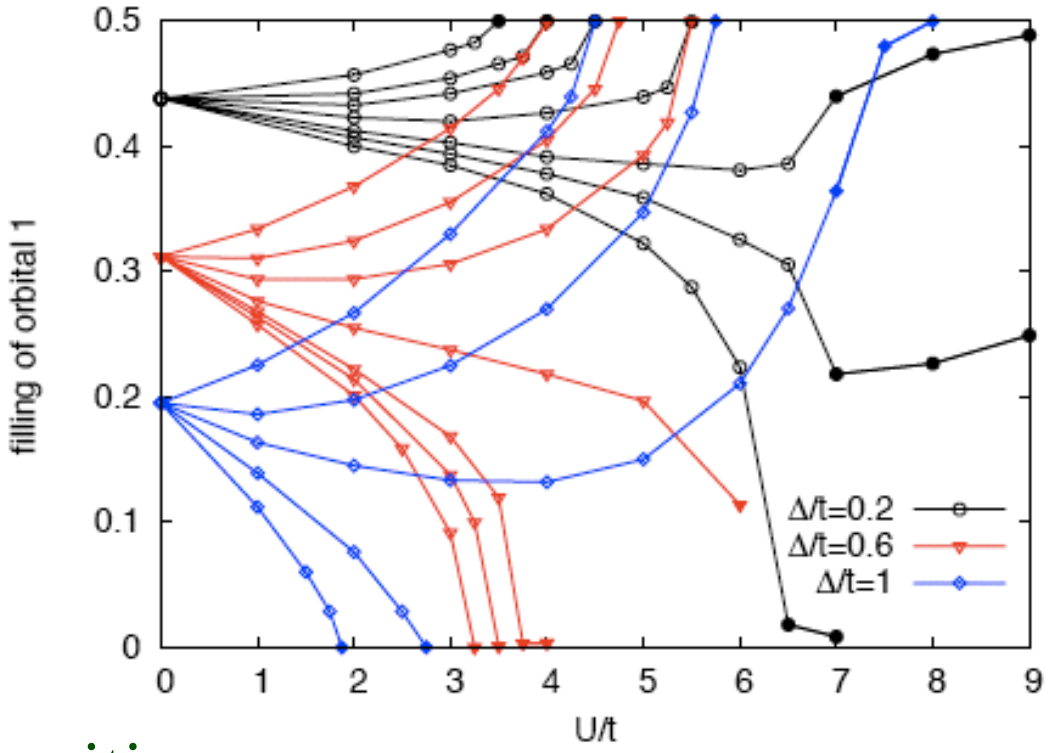

J/U=
0.25
0.15
0.10
0.05
(0.02)
(0.01)

3 metal-insulator transitions:
Band filling (n_1->0). Symmetric Mott (n_1->0.5)
Asymmetric Mott (n_1 intermediate)

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Two-orbital model phase diagram: U vs crystal field; 2 electrons.

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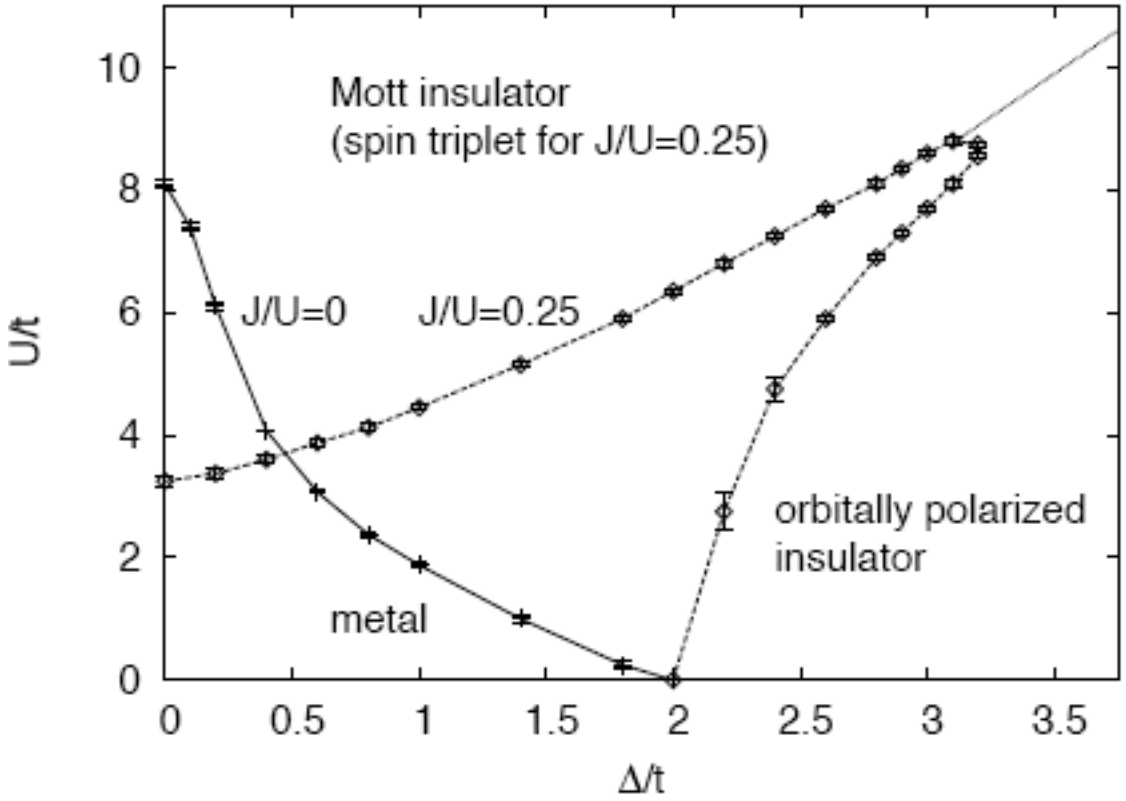

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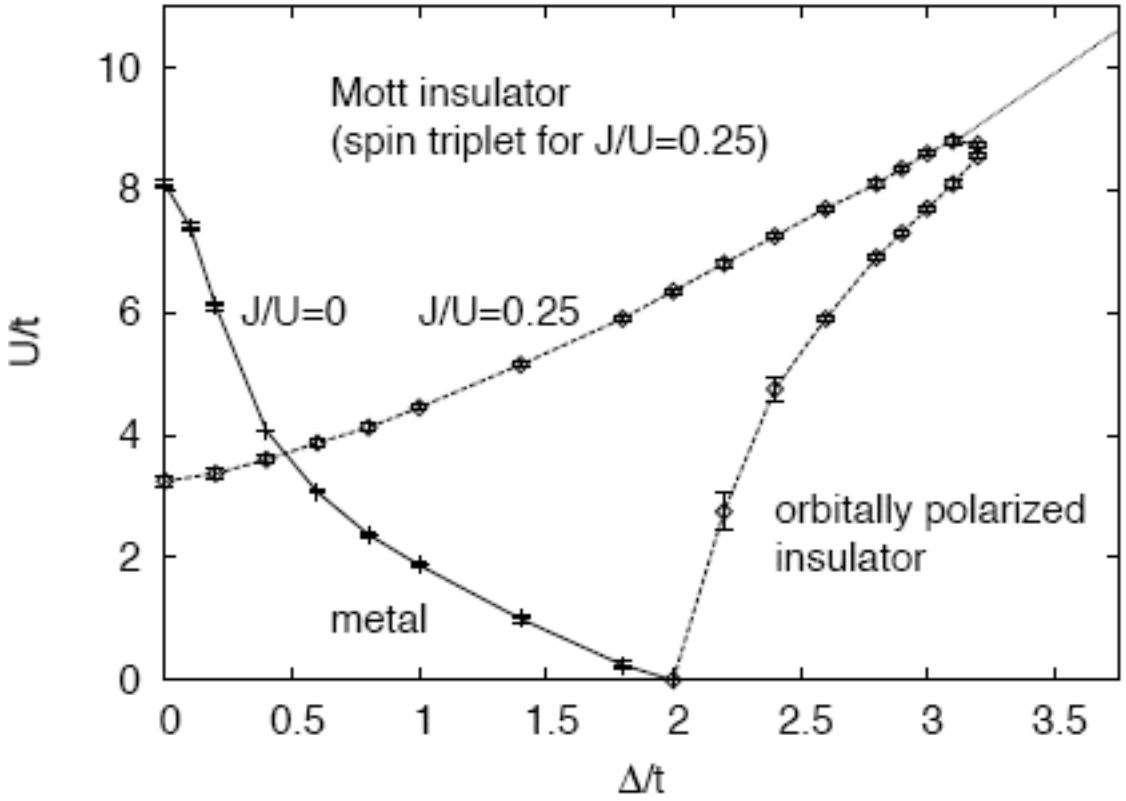

Δ=0: increasing J drastically reduces Mott critical value

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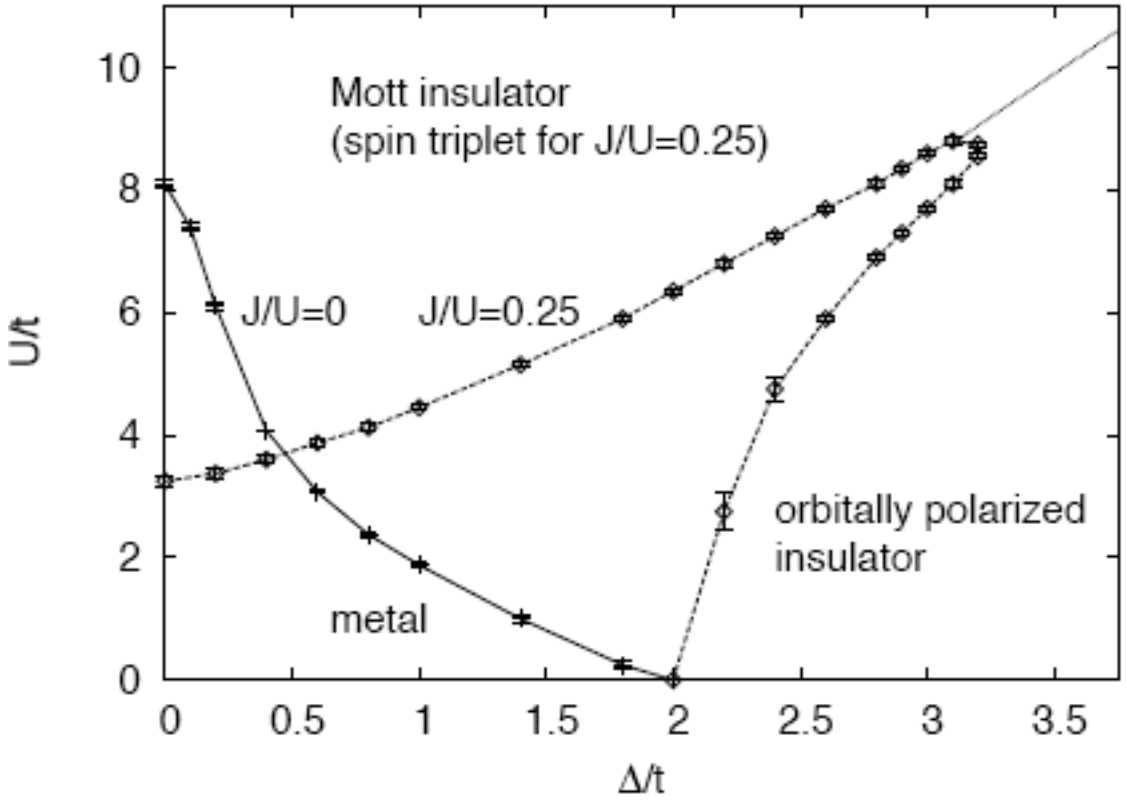

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J>0: much wider metallic regime.

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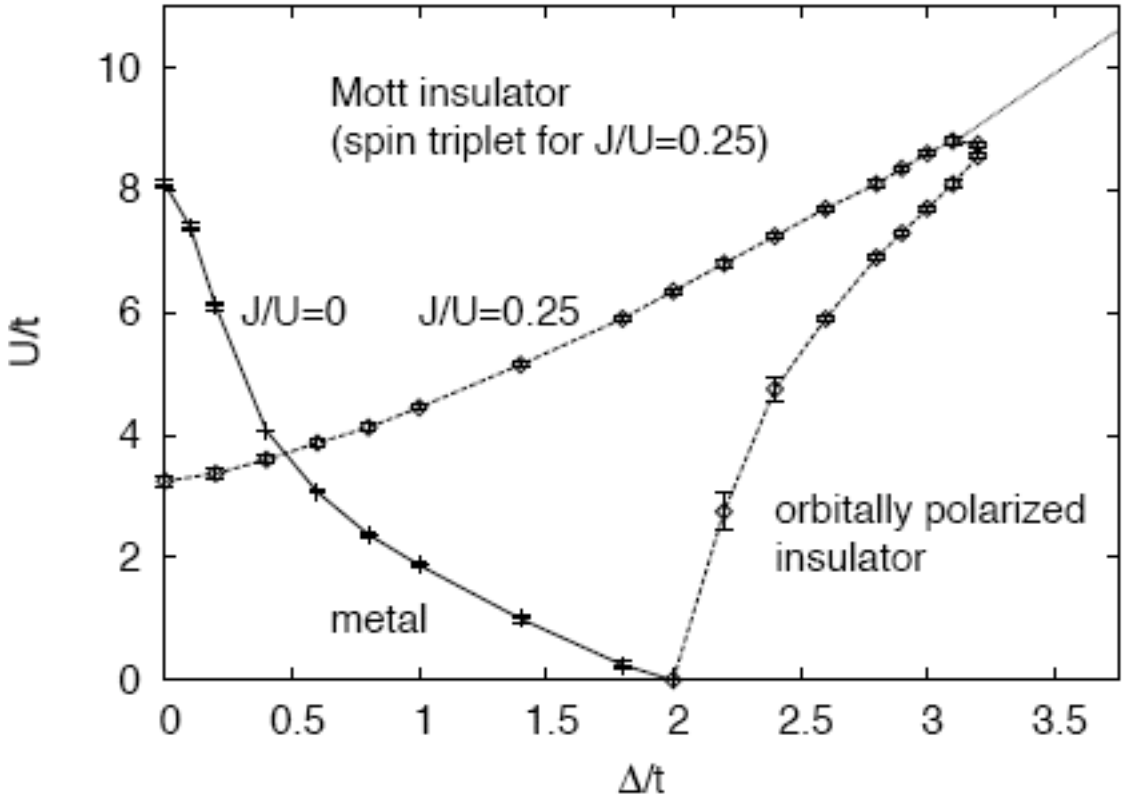

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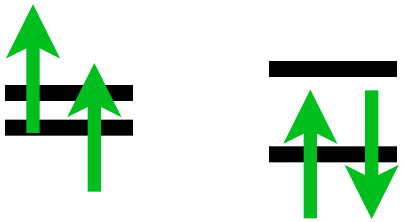
Physics: triplet vs singlet



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Two-orbital model phase diagram: Characterize insulator: small J; U=2.25W

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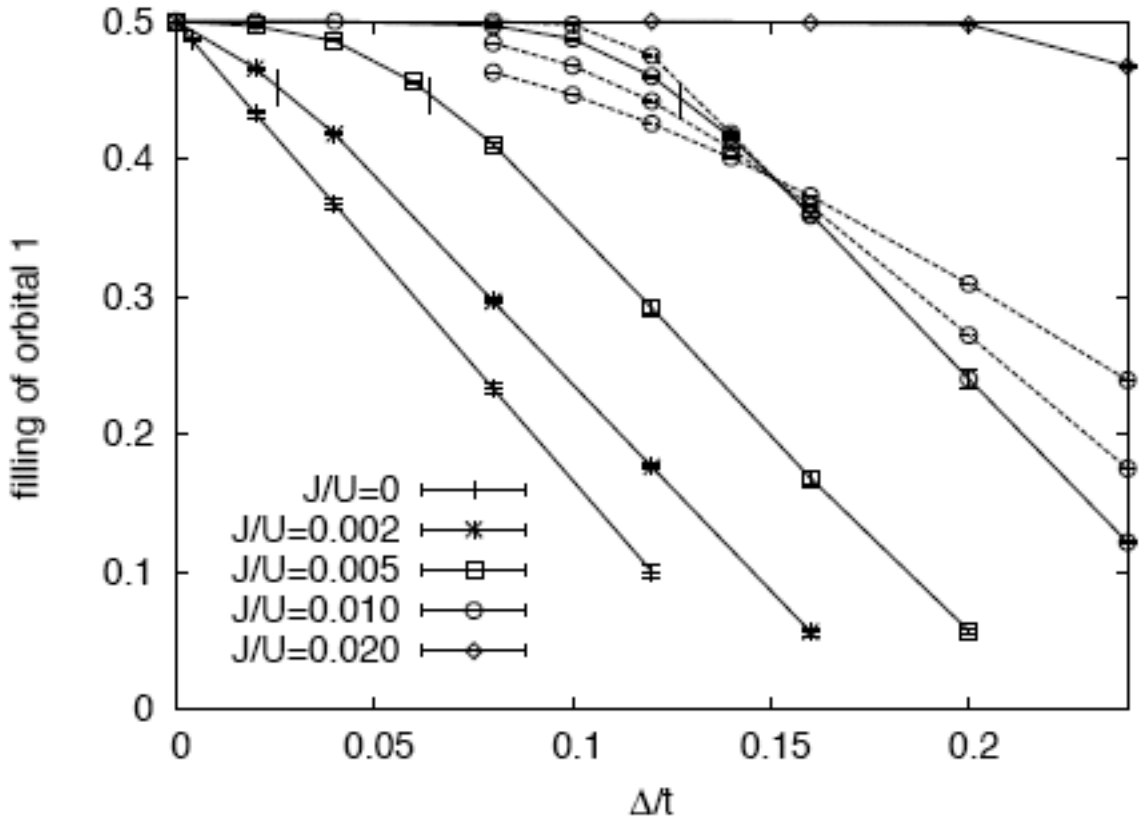

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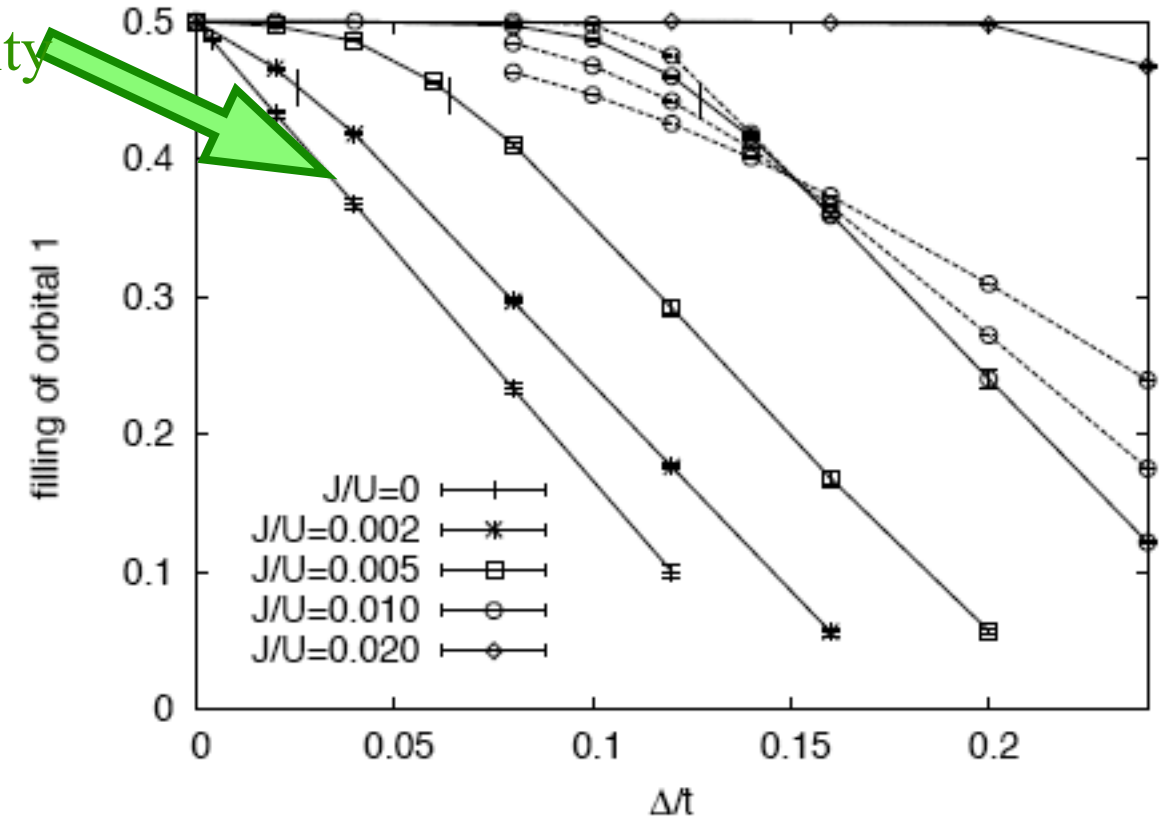
J=0: Kugel-Khomskii orbital susceptibility ~t^2/U



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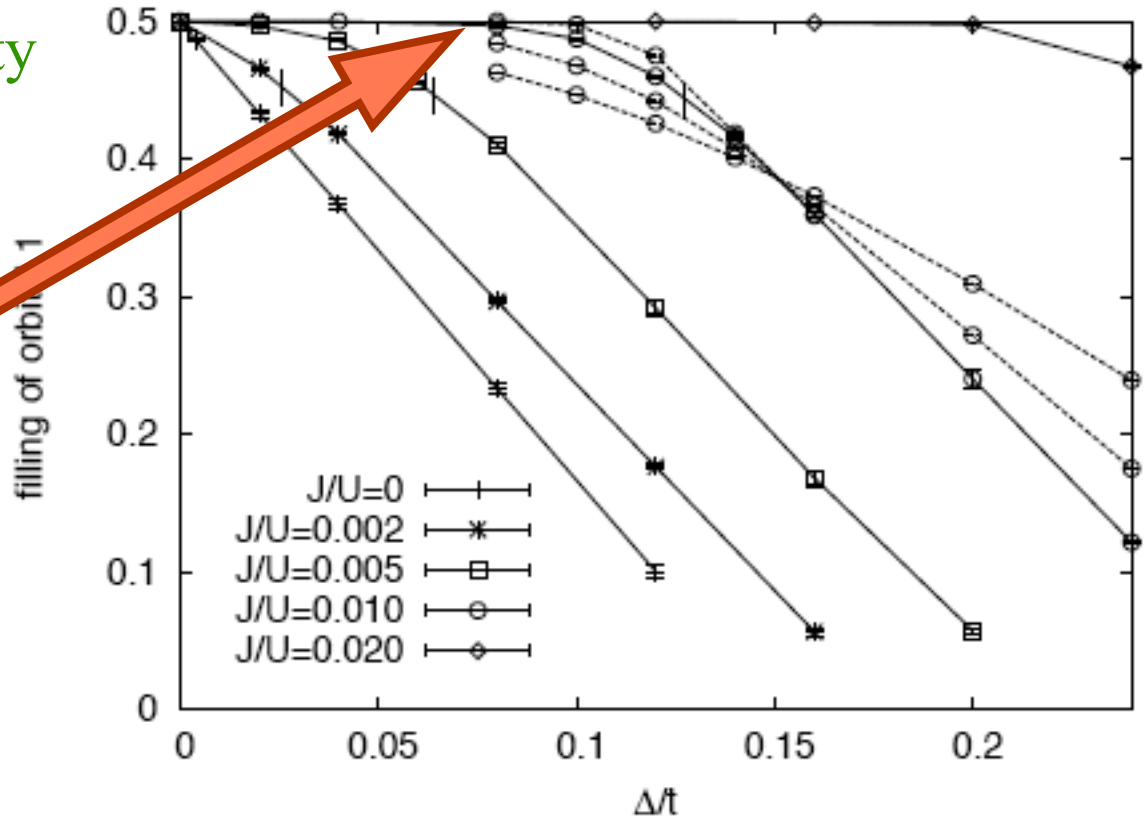

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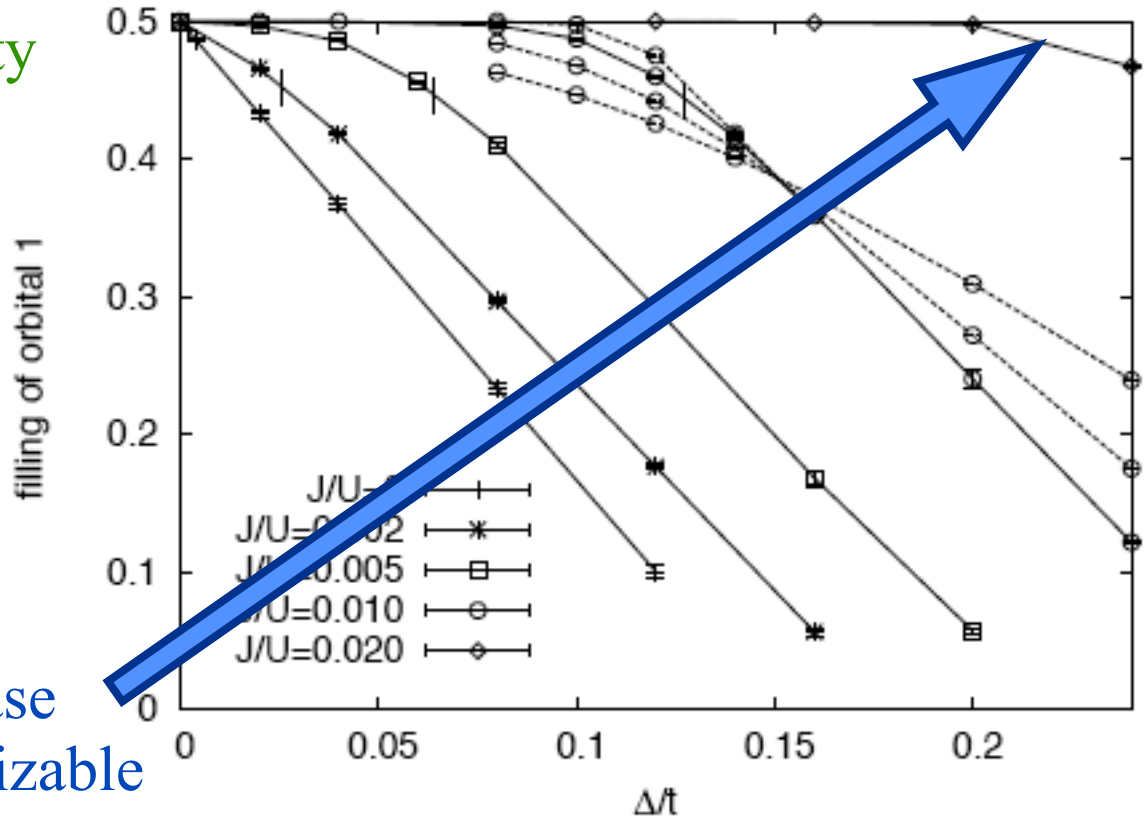

Larger Δ:T=0 phase transition to polarizable phase

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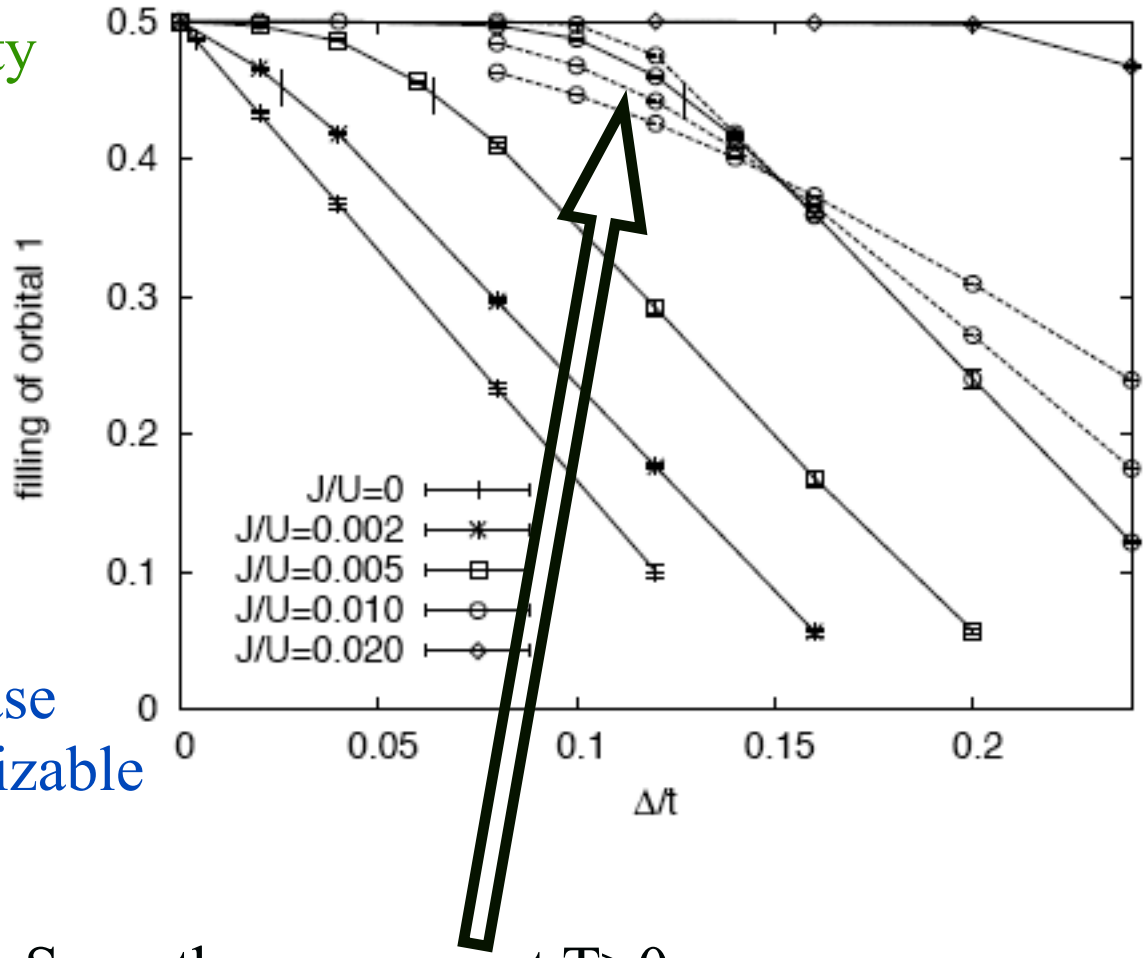

Larger ΔT=0 phase transition to polarizable phase

Smooth crossover at T>0

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Two-orbital model phase diagram: Characterize transitions: small J; U=1.5W

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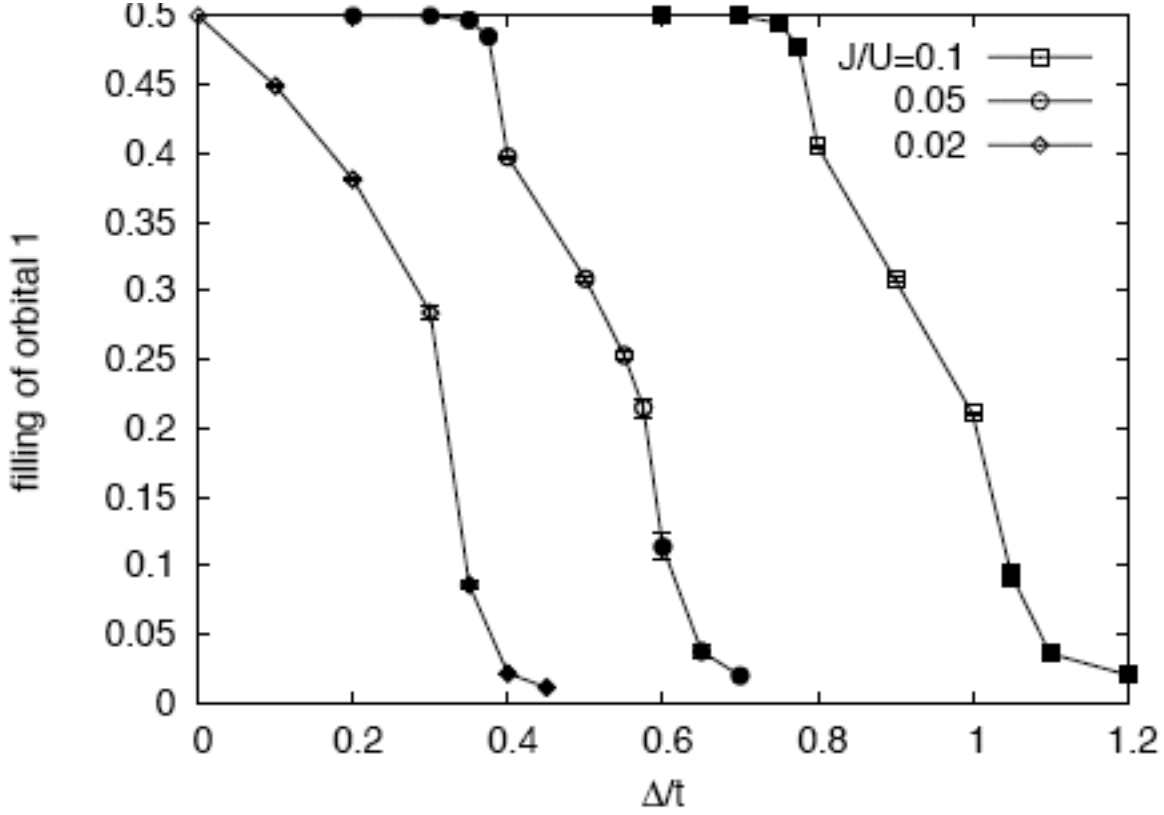

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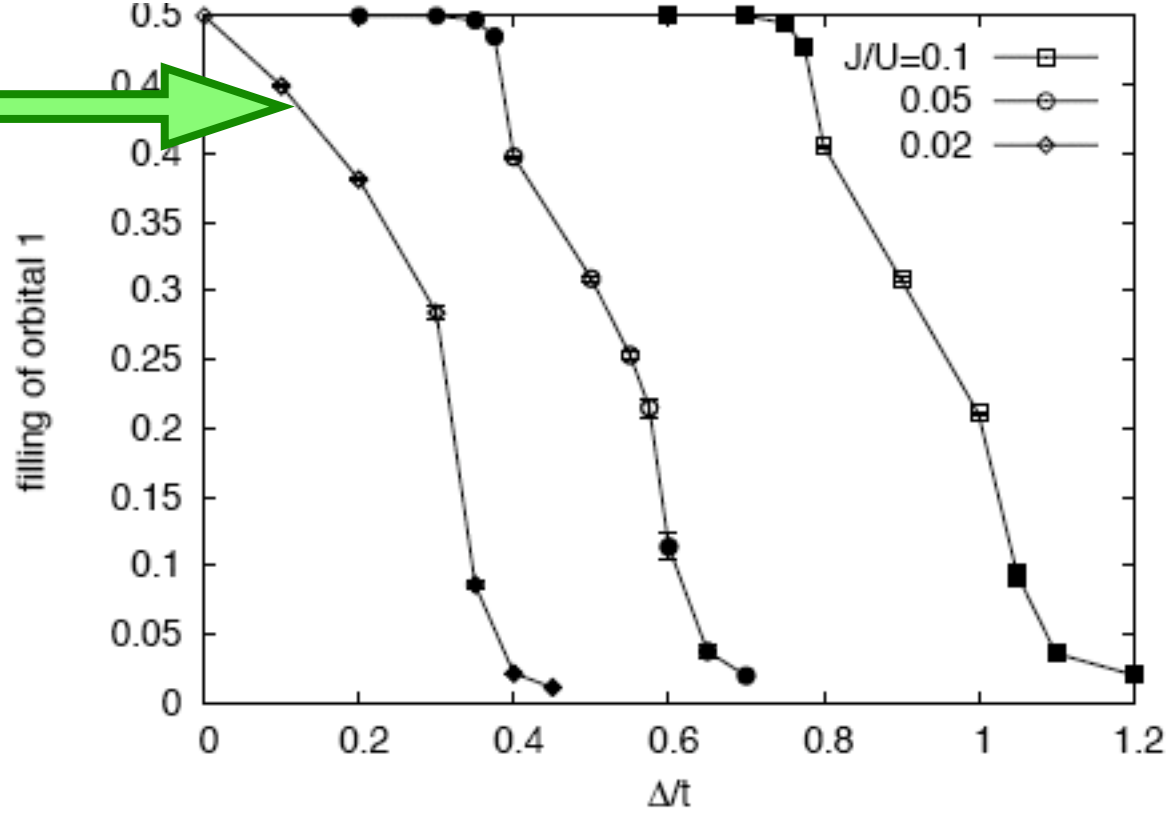
Two-orbital model phase diagram: Characterize transitions: small J; U=1.5W

J=0.02: Δ=0 state metallic w/ large orbital suscept.



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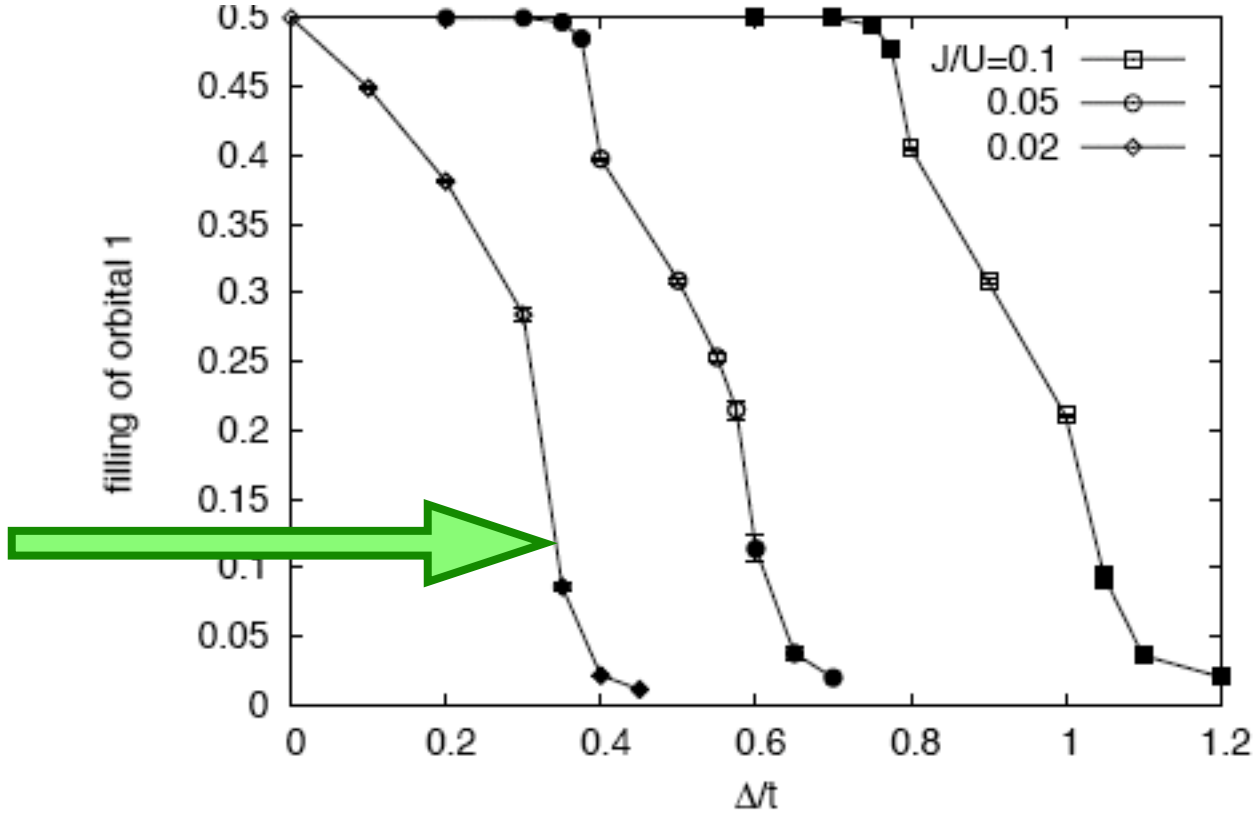
1st order metal-insulator transition to orbitally polarized insulator



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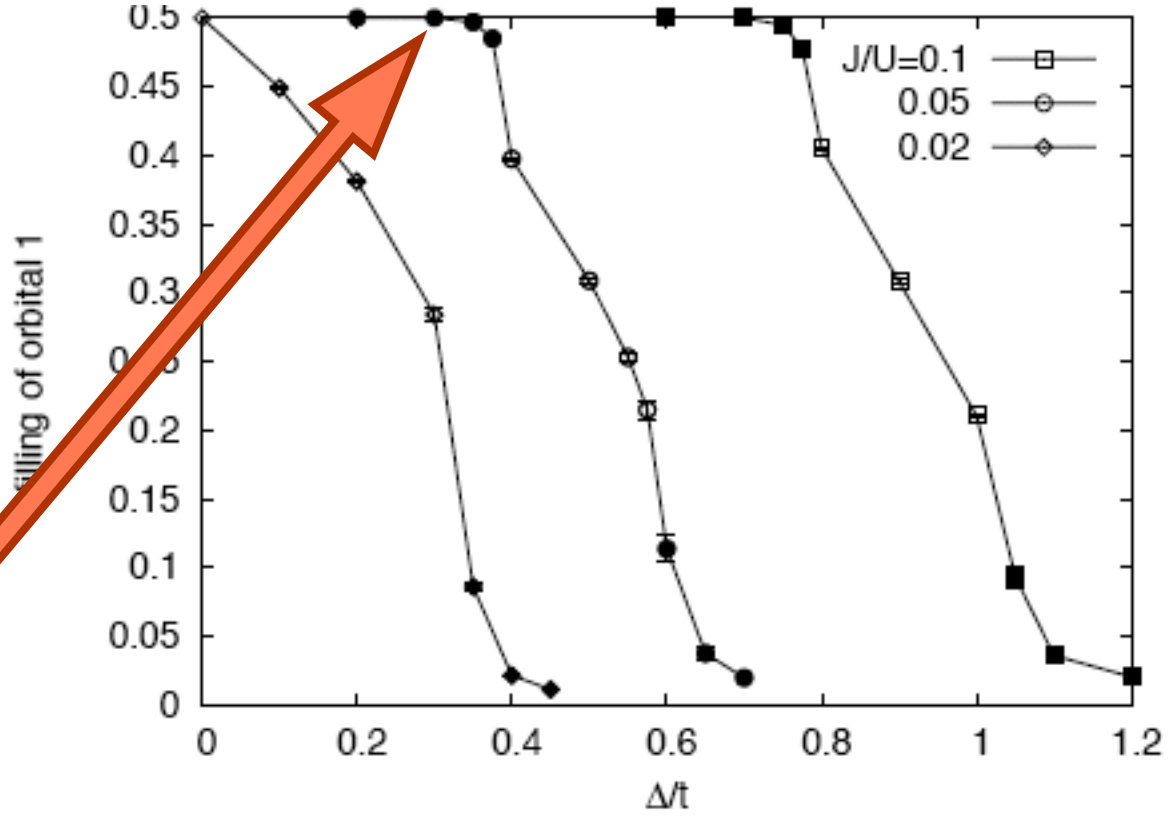
1st order metal-insulator transition to orbitally polarized insulator

J=0.05,0.1:Δ=0 state, orbital suscep vanishes.



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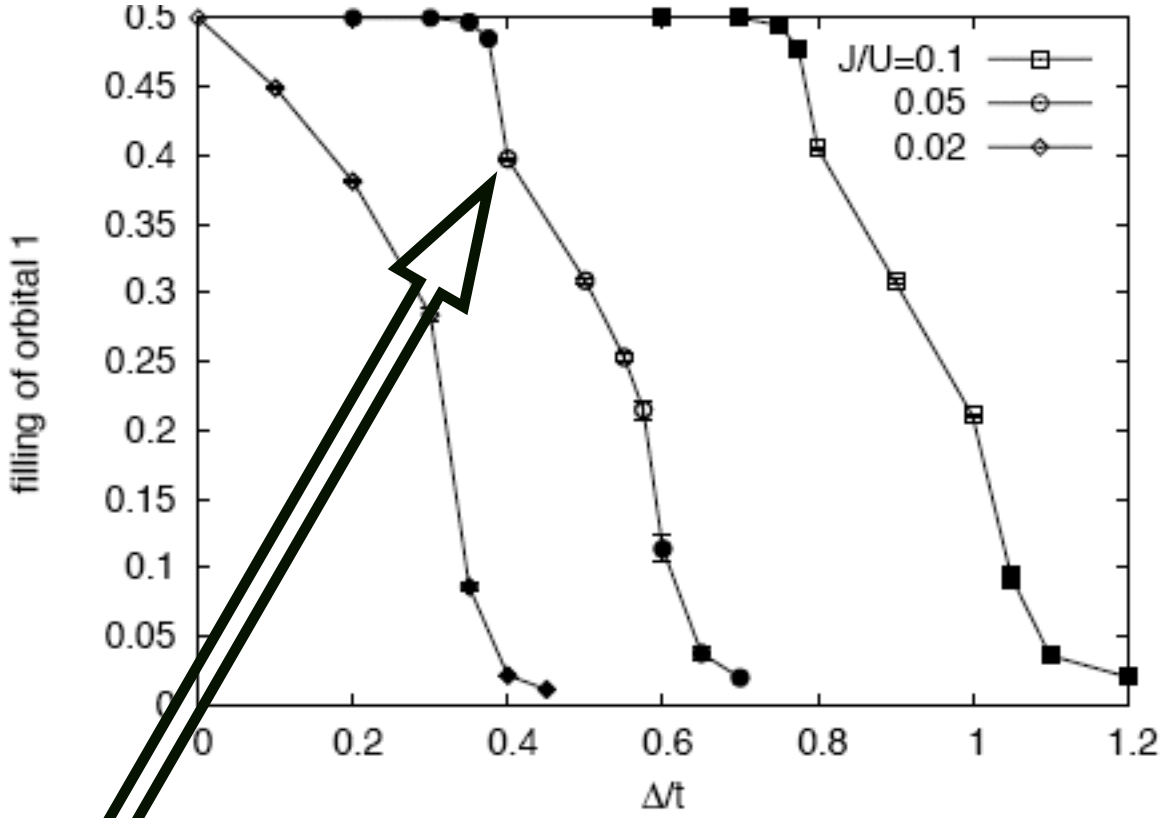

Larger Δ:1st order transitions to metallic phase

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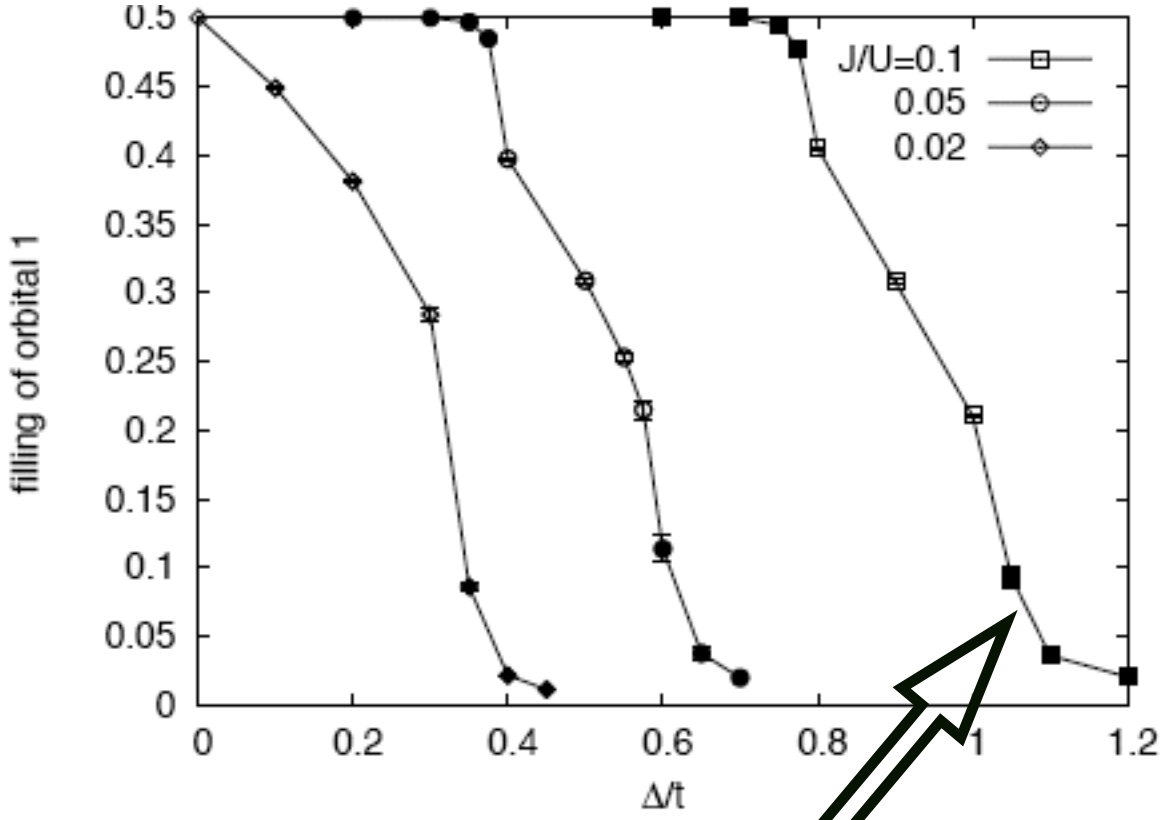

Larger Δ1st order transitions to metallic phase, then to pol. insulator T=0 phase transition to polarizable phase

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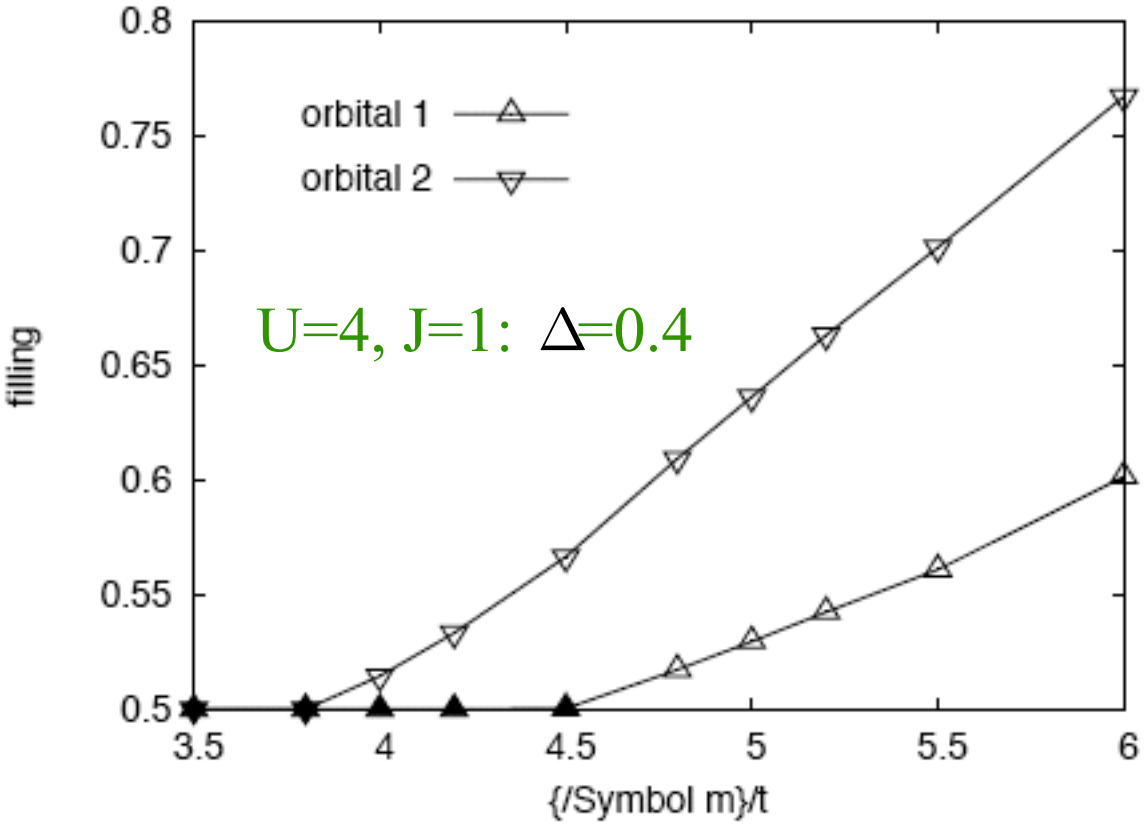
Two-orbital model phase diagram: Doping with $\Delta \neq 0$.

Doping driven Mott transition is "orbitally selective" (not surprising)



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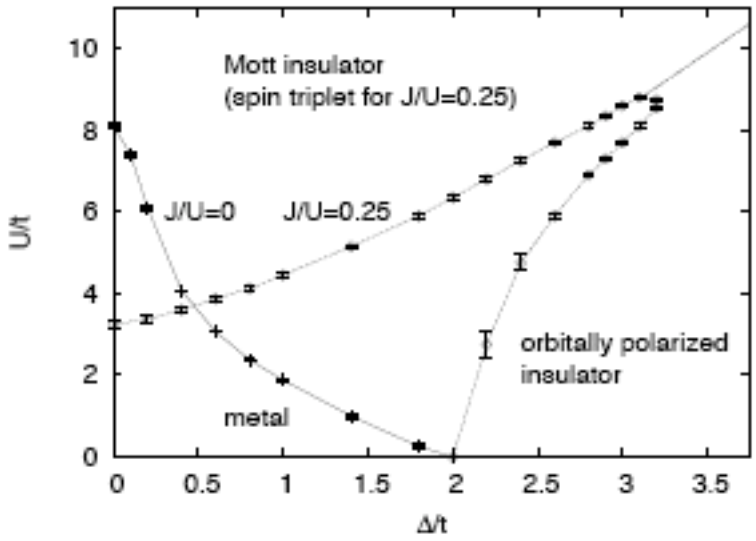
Summary: 2 orbital half-filled model

- **Intricate interplay: J,Δ**
- **"new" Mott phase-- vanishing** $\chi_{orbital}$
- **=>? additional gauge symmetry**
- **sharp transition between Mott phases at T=0 only**



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Work in progress: 3 orbitals (Ruthenates; C-60)

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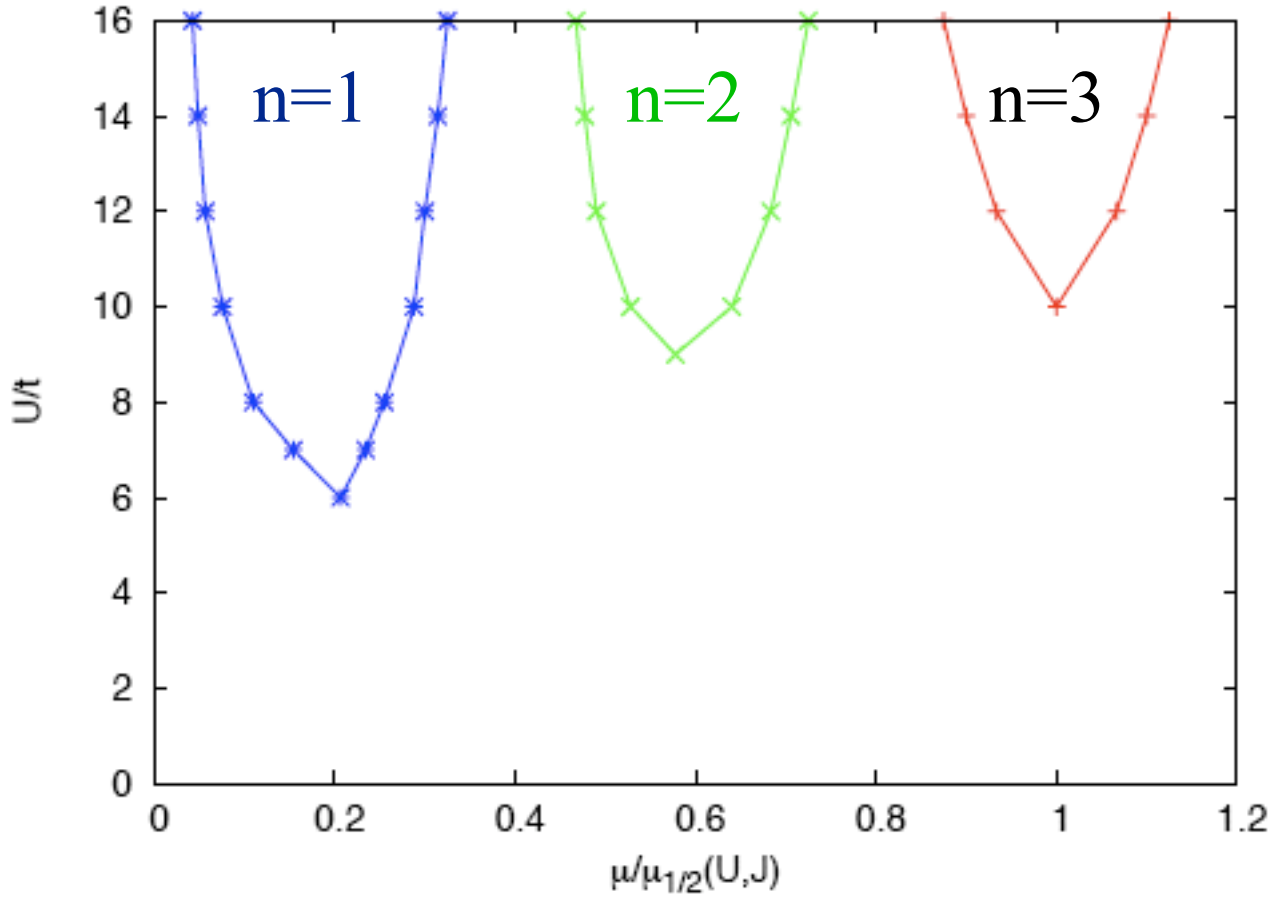
Mott phase boundary J=0 Δ=0



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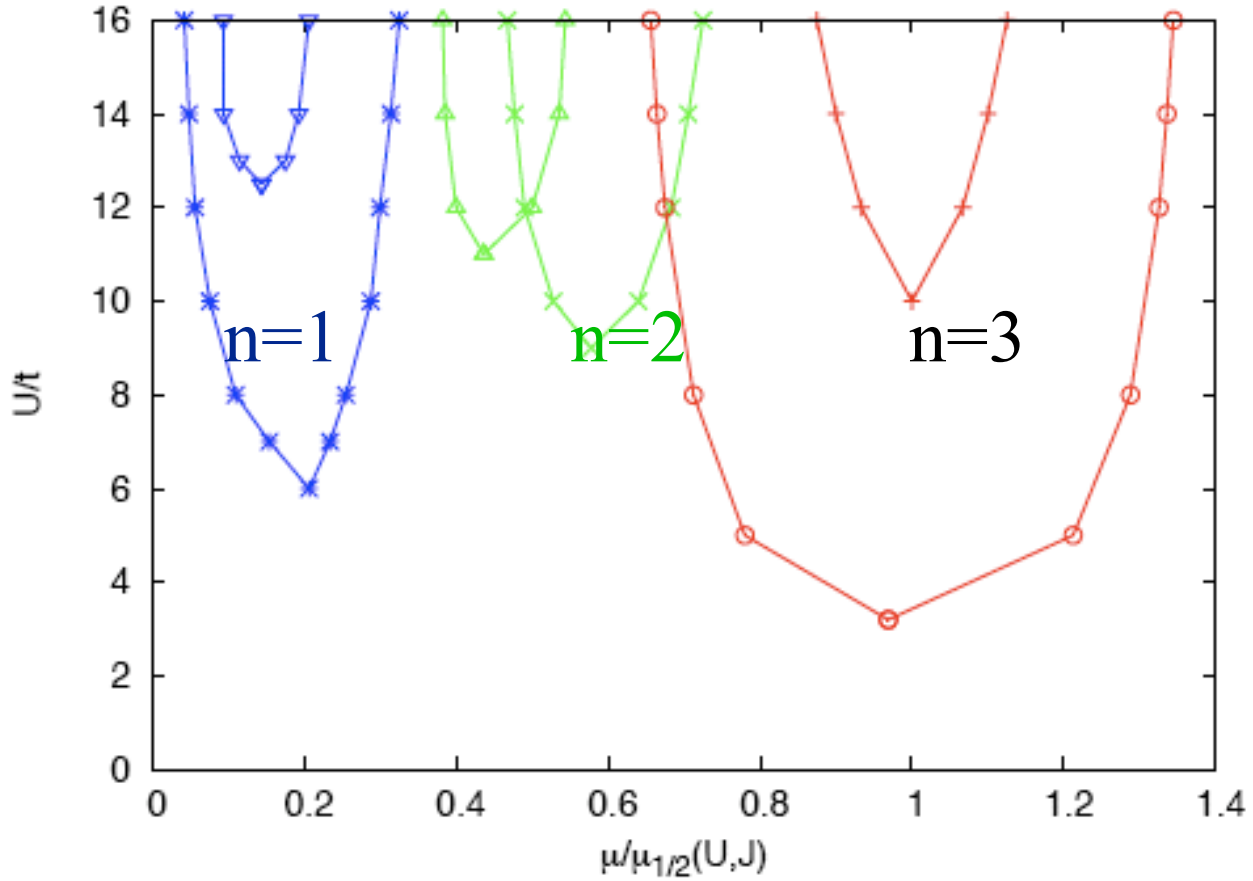
Work in progress: 3 orbitals (Ruthenates; C-60)

Mott phase boundary J=U/6 Δ =0



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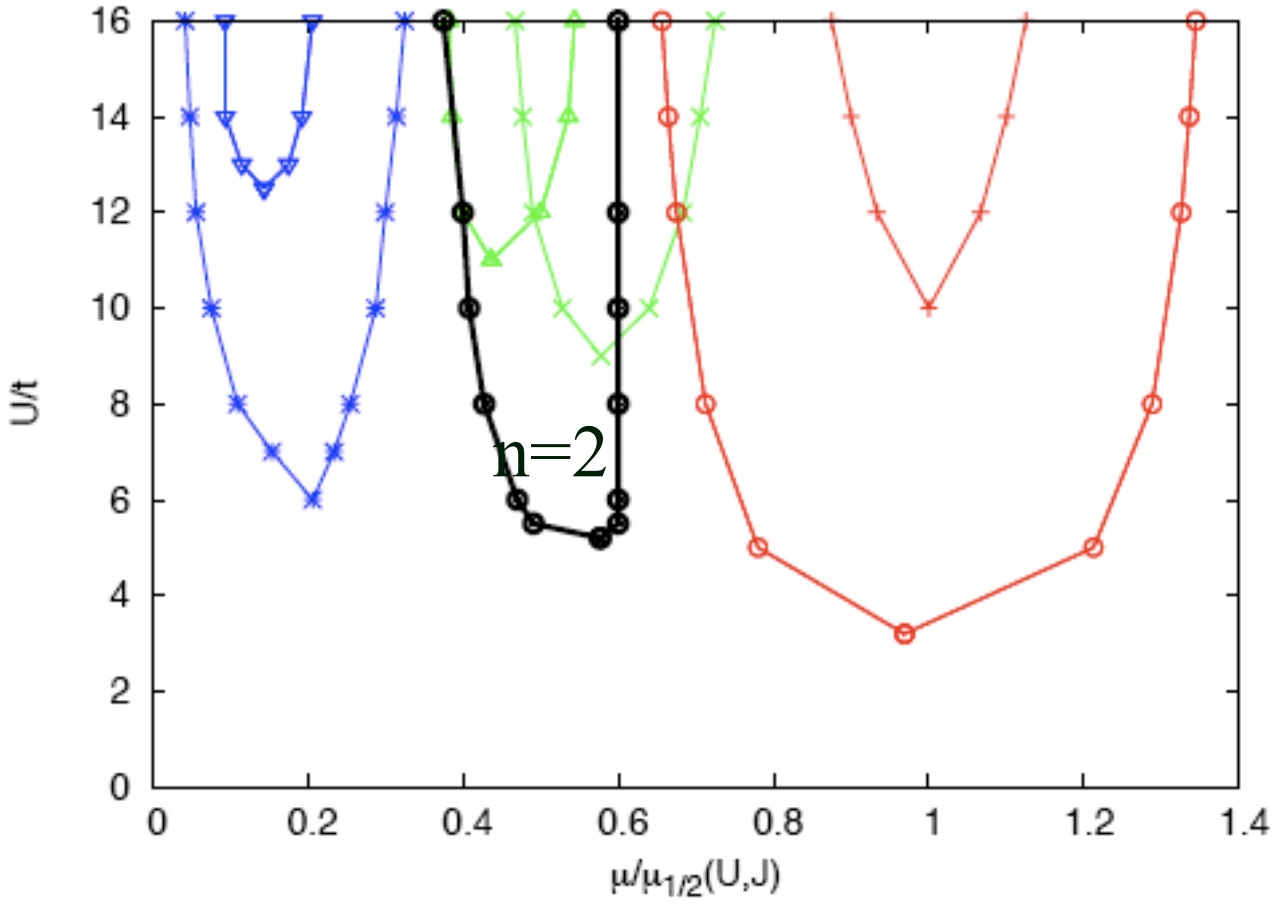
Work in progress: 3 orbitals (Ruthenates; C-60)

Mott phase boundary J=U/6 Δ=W/4 (n=2 only)



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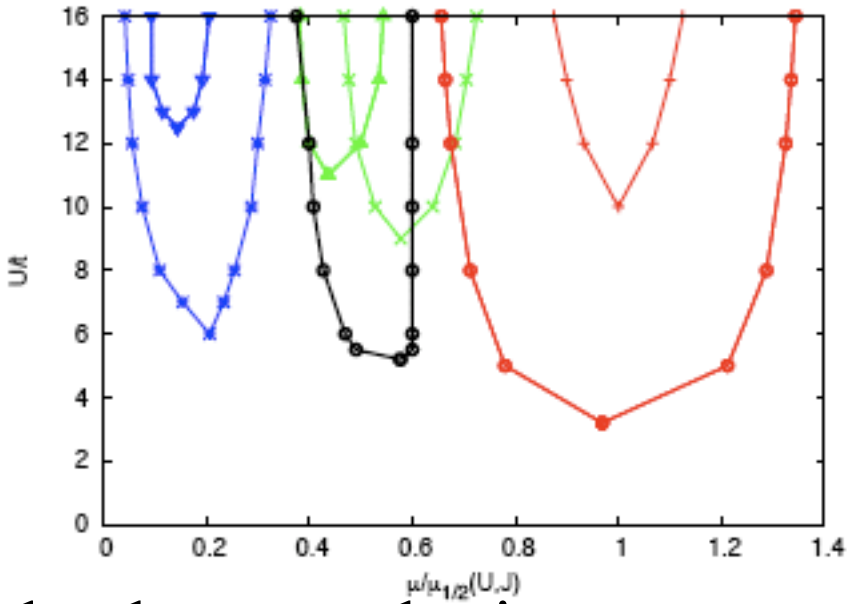
3 orbitals--summary

- **J helps insulator, n=3, hurts insulator, n=1,2**
- **Δ helps insulator; n=2**
- **Shifts are very large**



Important general issue: interplay between lattice distortions, spontaneous orbital order and Mott behavior

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Directions

- **Real materials**
- **Larger systems: reduce size of Hilbert space.**
  - **Effective low energy theories**
  - **Simple truncation (Haule)**
- **Nonequilibrium problems (Rabani)**

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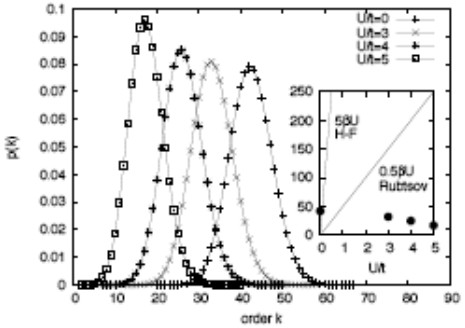
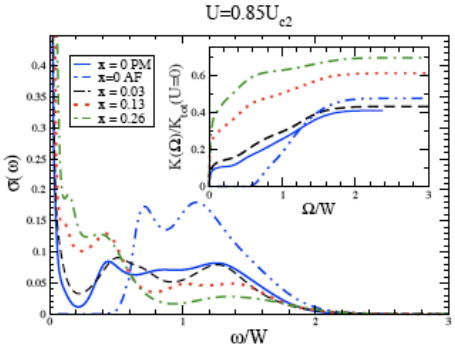
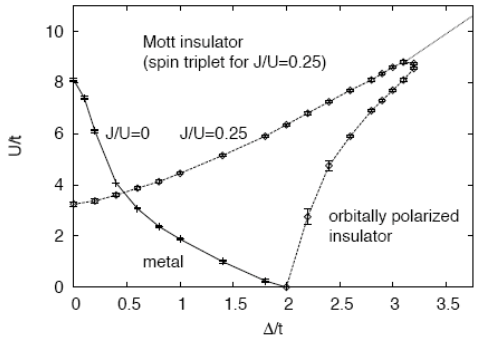
Conclusions







- **New method--seems very useful**
- **Cuprates: are the "Mott" insulators? what is a Mott insulator anyway**
- **Orbital degeneracy, Hunds coupling and crystal fields: new phases and new transitions**

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