Dynamical Mean Field Theory of Correlated Electron Materials

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Outline

1. Why do it?
2. What is it: analogy to density functional theory
3. The technical challenge
4. What can we do
5. Qualitative insights: orbital polarization in superlattices
6. Approaching a quantitative solution: gaps and pseudogaps in high Tc superconductors
7. New application: Quantum chemistry
8. Where next?

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Motivation

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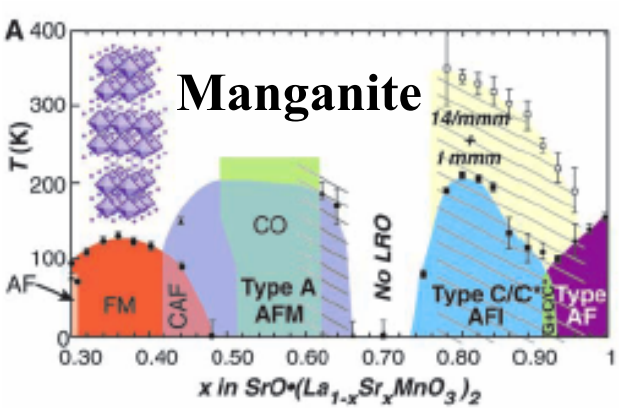
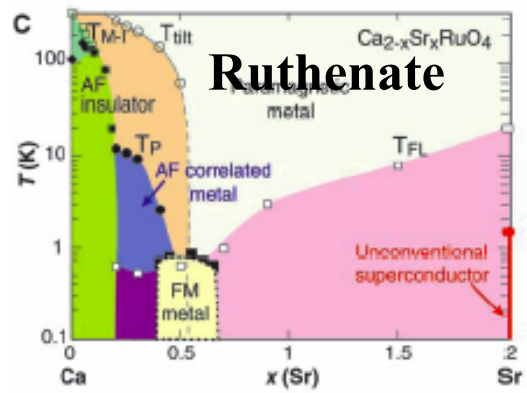
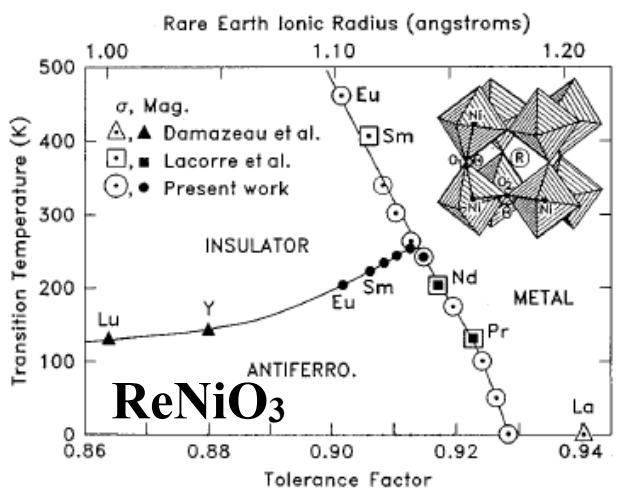
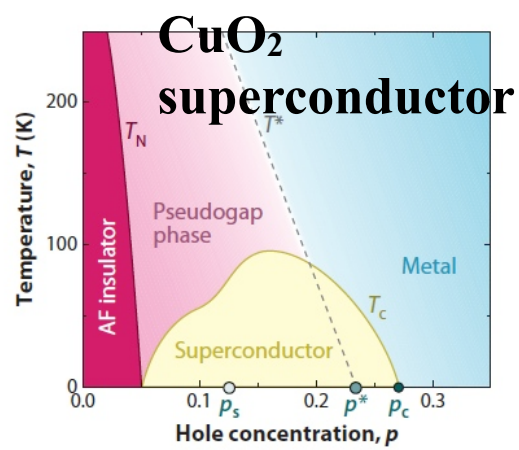
Interesting collective electronic phases









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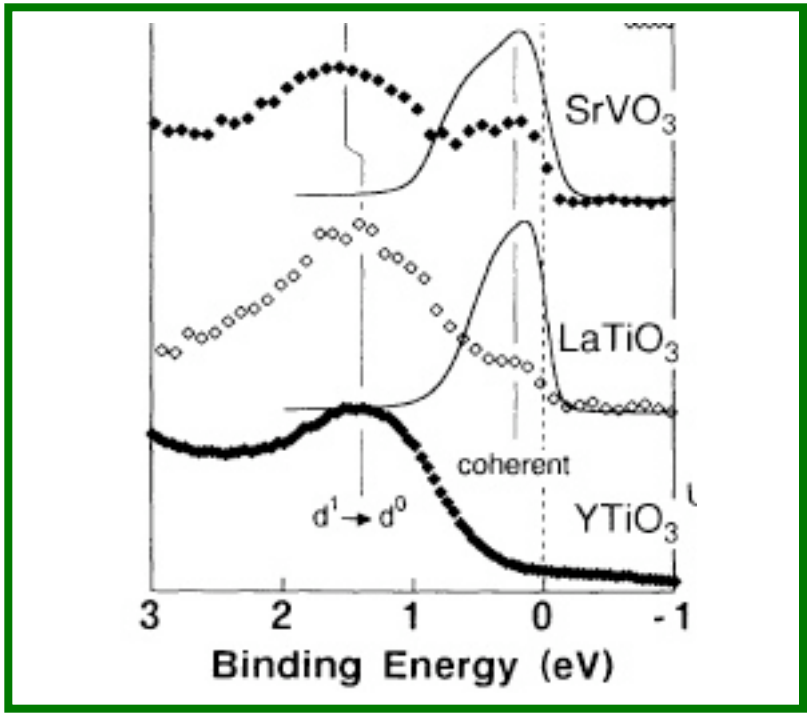
Unconventional Excitation Spectra

Photoemission
(electron removal spectrum)



A. Fujimori et al Phys. Rev. Lett. **69**, 1796 (1992)

Inconsistent with conventional electronic structure

dots: data
lines: band theory

Shakeoff (side) band present in data, absent in band theory

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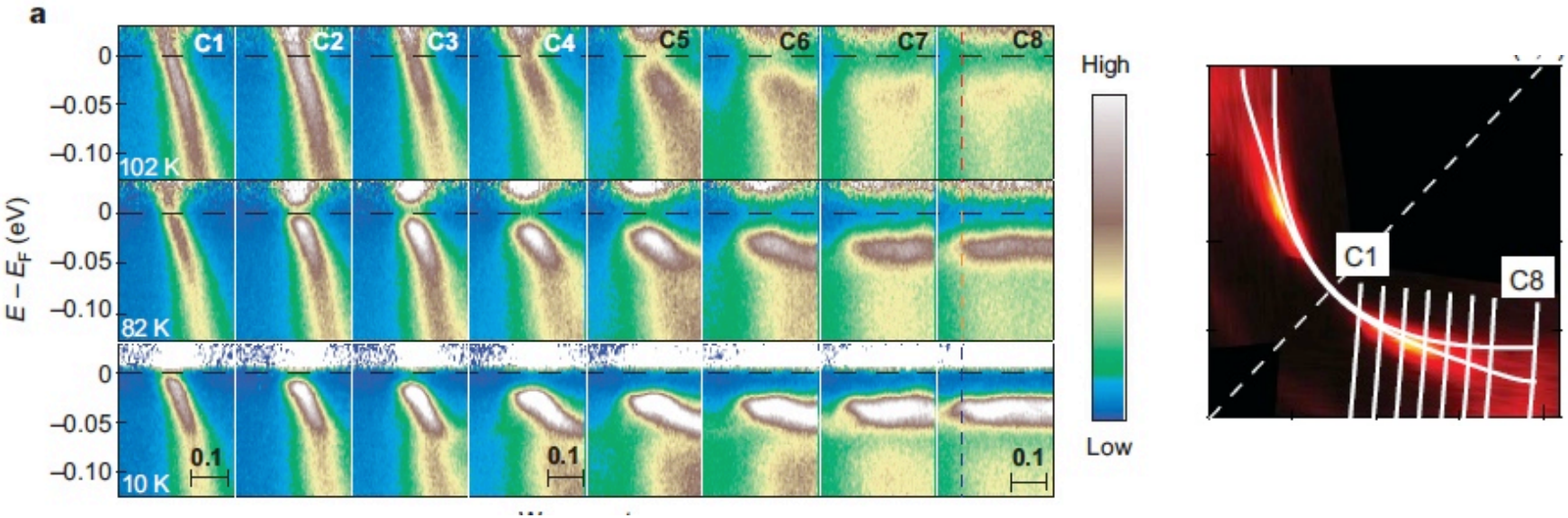
CuO_2 superconductors: `pseudogap'



S. Lee et al, Nature 450, p. 81 (2007)

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Gap without obvious long ranged order

Basic question: what is origin

Theoretical question: is this phenomenon a property of a well defined model in a well defined parameter regime?

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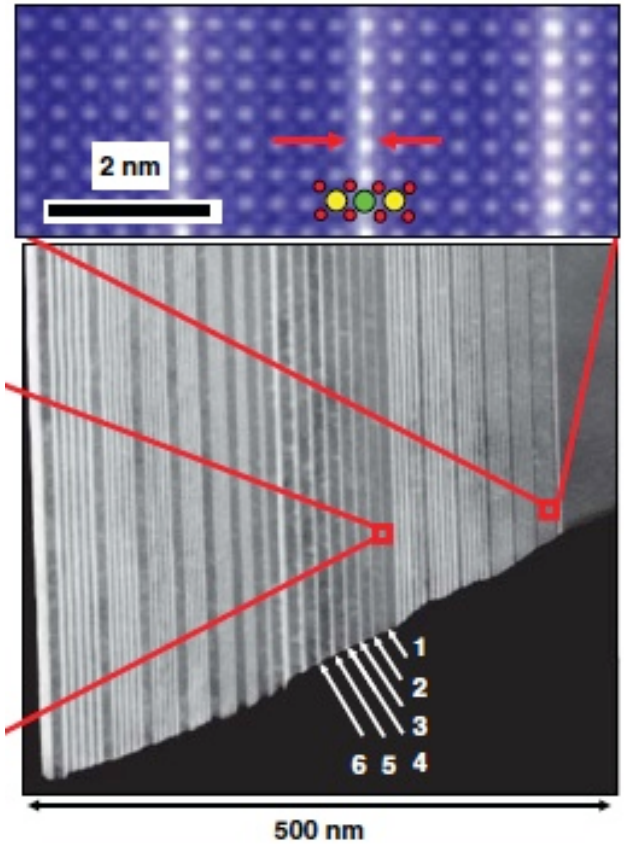
Oxide superlattices: materials by design

$(SrTiO_3)_m(LaTiO_3)_n$



$SrTiO_3$:
d^0 "band" insulator

$LaTiO_3$:
d^1 "Mott" insulator

'any' desired (n,m) can be synthesized

Ohtomo, Muller, Grazul and Hwang, Nature **419** p. 378 (2002)

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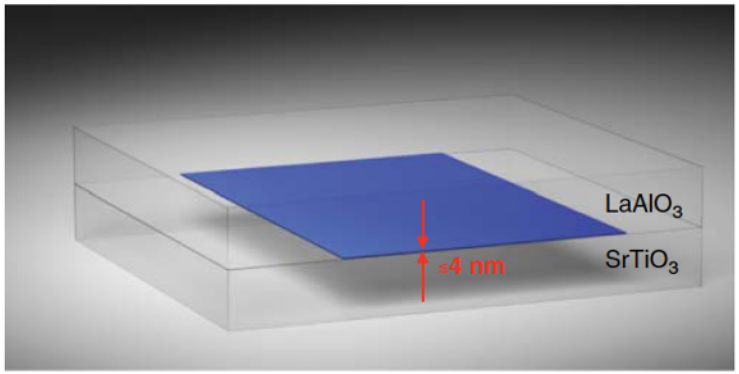
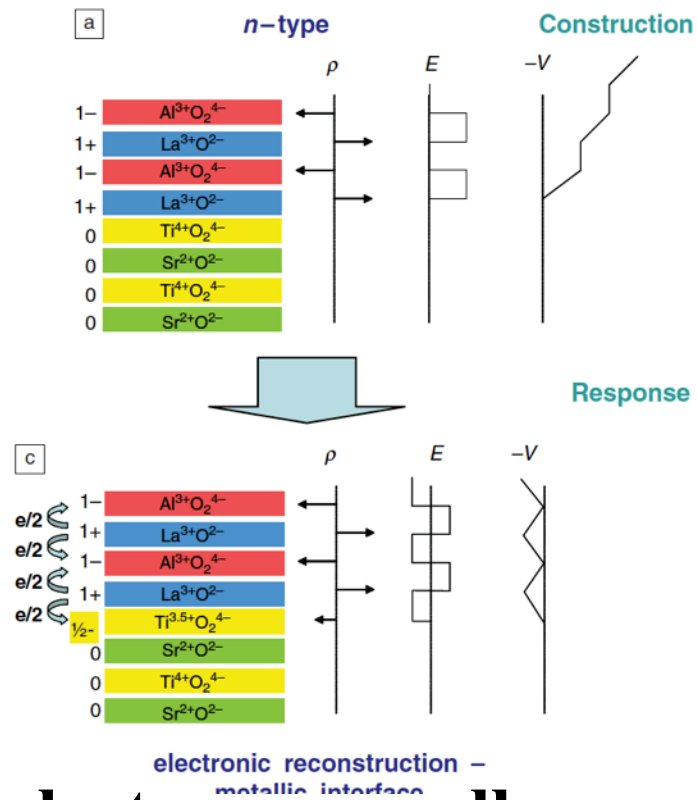
polar interface=>high doping





Mannhart, Blank, Hwang, Millis, Triscone, Bull MRS 33 (1027)

Polar discontinuity=>1/2 electron per cell (areal density 3 x 10^{14} cm^{-2})

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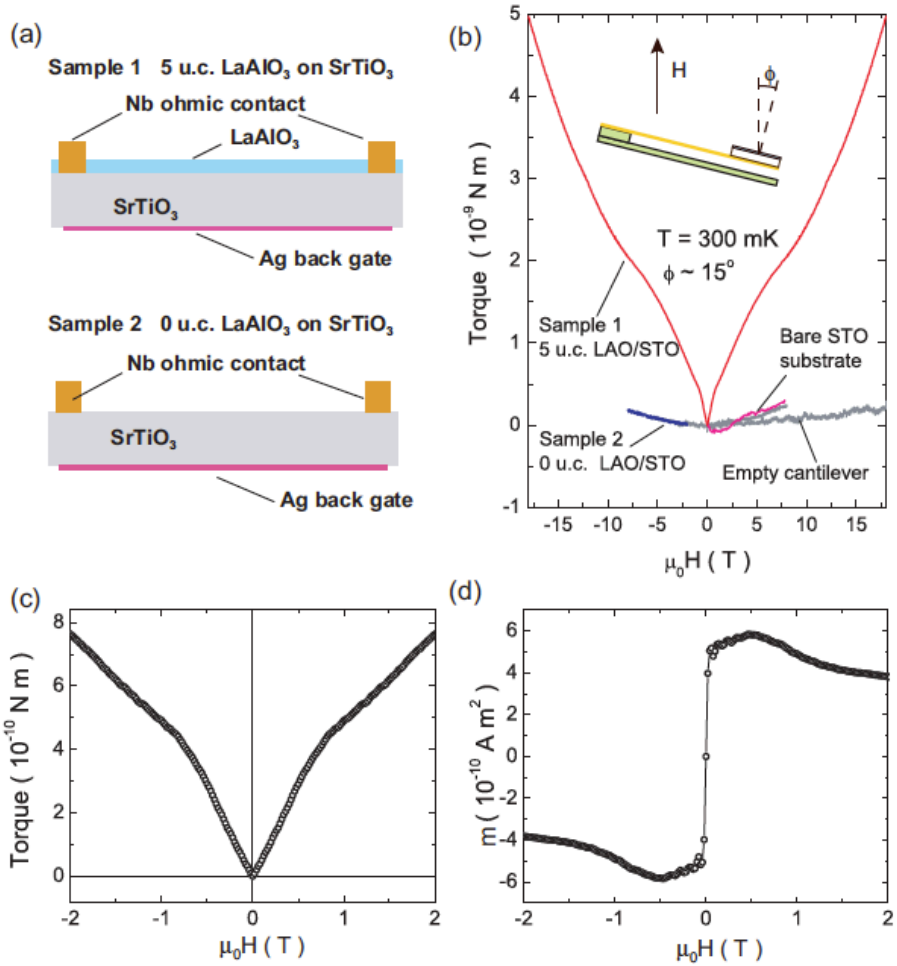
new claim: doped gas is magnetic



Lu Li, R. Ashoori et al
arXiv:1105.0235
(Nature Physics in press)

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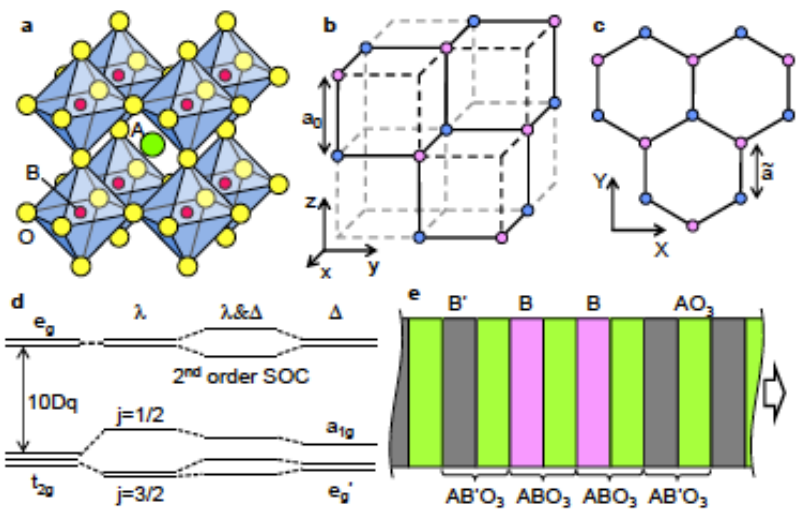
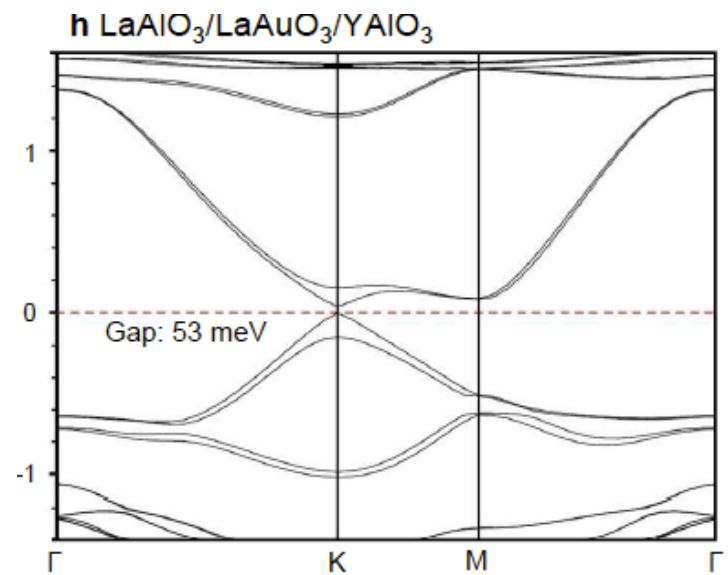
More exotic behavior possible

Ex: use (111) superlattice to make material with intersting 'topological insulator' properties:

$LaAlO_3$/$AlAuO_3$/$YAlO_3$





Xiao...Okamoto arXiv:1106:4296

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’materials by design’

Appropriate choice of heterostructure

=>devices with controlalbe many-body physics effects

--superconductivity
--magnetism
--metal-insulator transitions

<u>May</u> be possible

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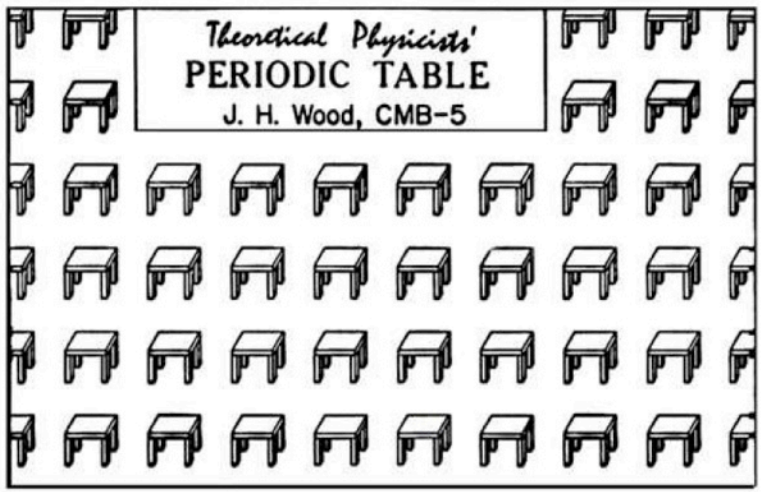
Stereotypical theorist's view of condensed matter physics



Periodic table from J H Wood;

quoted in Z Fisk 2010 KITP talk

Quantum Field Theory

$$\Rightarrow \quad \frac{\kappa}{2}\epsilon^{\mu\nu\lambda}\mathbf{A}_\mu\partial_\nu\mathbf{A}_\lambda + \ldots$$

Experimental side of the field has advanced:
We (theorists) need to do better!

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Thus:

If experimentalists can make 'anything' --what would one want to make?

How do we connect crystal structure/atomic properties to interesting electronic behavior?

=>Dynamical electronic structure problem

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Why Dynamical Mean Field Theory:

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Quantum many-body problem is 'hard'

We know the equation: $\mathbf{H}\Psi_n = -i\partial_t \Psi_n$

$$\mathbf{H} = \sum_i \frac{-\nabla_i^2}{2m_e} + \sum_i V_{ext}(r_i) + \frac{1}{2}\sum_{i\neq j} \frac{e^2}{|r_i - r_j|}$$

BUT:

need more than ground state.

interaction 'entangles' coordinates: $\mathbf{\Psi}(\vec{r}_1, \ldots . \vec{r}_{1000} \ldots)$

Even worse: Ψ is fully antisymmetric function of spins and coordinates

=>``Fermion sign problem"

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Stochastic (Monte-Carlo) exploration of configuration space

To estimate expectation value of some quantity A

$$\langle A \rangle_p = \frac{1}{Z} \int_{\mathcal{C}} d\mathbf{x} \mathcal{A}(\mathbf{x}) p(\mathbf{x})$$

Select M points x_i with probability $p(x_i)$ and compute

$$\langle A \rangle_p \approx \langle A \rangle_{MC} \equiv \frac{1}{M} \sum_{i=1}^{M} \mathcal{A}(\mathbf{x}_i)$$

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BUT for fermions

Sign problem: antisymmetry of wave function means that different configurations come with different signs. p(x) not always positive

Sample $$\langle A \rangle_p = \frac{1}{Z} \int_{\mathcal{C}} d\mathbf{x} \mathcal{A}(\mathbf{x}) p(\mathbf{x})$$

using $\rho(x) = |p(x)|$

$$\langle \mathbf{A} \rangle_{\mathbf{p}} = \frac{\langle \mathbf{A}\ \mathbf{sign}[\mathbf{p}] \rangle_\rho}{\langle \mathbf{sign}[\mathbf{p}] \rangle_\rho} = \frac{\frac{1}{\mathbf{Z}} \int_{\mathbf{C}} \mathbf{dx} \mathbf{A}(\mathbf{x})\ \mathbf{sign}[\mathbf{p}(\mathbf{x})] |\mathbf{p}(\mathbf{x})|}{\frac{1}{\mathbf{Z}} \int_{\mathbf{C}} \mathbf{dx}\ \mathbf{sign}[\mathbf{p}(\mathbf{x})] |\mathbf{p}(\mathbf{x})|}$$

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Problem (Ceperly, 1996; Assaad, 1991....)

$$\langle sign \rangle = Z = e^{-\frac{F_{fermion}}{T}}$$

Partition function of fermion system

|p(x)|: weight of configuration of ``boson" system

$$|p(x)| \sim Exp[-\frac{F_{\text{“boson''}}}{T}]$$

with $\mathbf{F_{\text{“boson''}}} < \mathbf{F_{fermion}}$

$$\boxed{\langle \mathbf{sign} \rangle = \frac{\mathbf{Z}}{\mathbf{Z}_\rho} = \mathbf{Exp}\left[-\left(\frac{\mathbf{F_{fermion}} - \mathbf{F_{boson}}}{\mathbf{T}}\right)\right]}$$

Vanishes exponentially as T->0 or system size-> infinity

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Theoretical approach:
Reduce size of 'active' Hilbert space

Single particle basis $|\phi_a(\tilde{r})\rangle$

Hamiltonian $H = T^{ab} + I^{abcd}$

$$T^{ab} = \langle \phi_a | \frac{-\nabla^2}{2m} + V(r) | \phi_b \rangle$$

$$I^{abcd} = \langle \phi_a(r_1)\phi_b(r_2) | \frac{e^2}{r_1 - r_2} | \phi_c(r_1)\phi_d(r_2) \rangle$$

Choose relevant subset of states $a_1, a_2, \ldots$
keep exact matrix elements within subspace; use simple approximation for remainder

=> ``Model System"

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How to solve model system?

N basis states=> Hilbert space dimension 4^N

Direct approaches encounter difficulties

- **Diagonalization fails before you can reach interesting sizes**
- **'DMRG': best for 1d systems and for ground state properties**
- **direct Quantum Monte Carlo: sign problem**

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Because direct approach inadequate,

Indirect approach: express (some aspects of) solution of physical problem in terms of solution of auxiliary problem

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Best Known Example: Density Functional Theory

Hohenberg and Kohn: ∃ functional Φ of electron density n(r) minimized at physical n(r) value at minimum gives ground state energy

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

Kohn and Sham:

Minimize by solving auxiliary (band theory) problem with self-consistently determined potential V_{XC}

different choices for V_{XC} =>different 'flavors' of density functional theory (LDA, GGA, B3LYP....)

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Dynamical Mean Field Theory

Metzner/Vollhardt; Mueller Hartmann KOTLIAR/GEORGES

Luttinger and Ward: $\exists$ functional F of electron self-energy $\Sigma(\mathbf{r},\omega)$ minimized at physical $\Sigma(\mathbf{r},\omega)$ value at minimum gives ground state energy and implies all response functions

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

Kotliar and Georges:

Extremize by solving auxiliary (quantum impurity) problem with self-consistently determined hybridization

different choices =>different 'flavors' of DMFT (DCA, CDMFT..)

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Analogy

Density functional <=> ‘Luttinger Ward functional

Kohn-Sham equations <=> quantum impurity model

Particle density <=> electron Green function

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Theoretical structure

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

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

=>

$$\mathbf{F}_{approx} \rightarrow \tilde{F}\left[\{\Sigma_a(\omega)\}\right] + Tr\left[ln\left(G_0^{-1}(p,\omega) - \sum_a \phi_a(p)\Sigma_a(\omega)\right)\right]$$

$\tilde{\mathbf{F}} = F_{univ}\left[\{\Sigma_p^{approx}\}\right]$ **is a functional of a small number of functions of frequency–so can be represented by a quantum impurity model ($0+1$ dim. field theory)**

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stationarity

$$\mathbf{F}_{approx} \rightarrow \tilde{F}\left[\{\Sigma_a(\omega)\}\right] + Tr\left[ln\left(G_0^{-1}(p,\omega) - \sum_a \phi_a(p)\Sigma_a(\omega)\right)\right]$$

$$\frac{\delta F}{\delta \Sigma(p,\omega)} = 0$$ **=> self-consistency condition**

$$\frac{\delta \tilde{F}}{\delta \Sigma_a(\omega)} = G_{QI}(\omega) = Tr\left[\phi_a(p)\left(G_0(p,\omega) - \sum_a \phi_a(p)\Sigma_a(\omega)\right)^{-1}\right]$$

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Nontrivial questions

- Does F^{approx} exist?
- How to construct the theory which gives G^{QI}
- What basis functions are acceptable
- Do the solutions make any sense at all, given the brutality of the approximation involved
- what is the best way to generalize to (approximations to) functionals of two particle correlators

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But before dealing with these: note advantages

*'Moving part' $Tr_p\left[\phi_a(p) G_{lattice}(\Sigma^{approx})\right]$

spatial average over electron spectral function
--but still a function of frequency
--not a quasiparticle and not a density.

=>releases many-body physics from twin tyrannies of
--focus on coherent quasiparticles/expansion about well understood broken symmetry state
--emphasis on particle density and ground state properties

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Ways to approximate self energy

X: generalized spatial coordinate

$$\mathbf{\Sigma_X}(\omega) \rightarrow \mathbf{\Sigma_X^{approx}}(\omega) = \sum_{\mathbf{a}} \phi_{\mathbf{a}}(\mathbf{X})\mathbf{\Sigma_a}(\omega)$$

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

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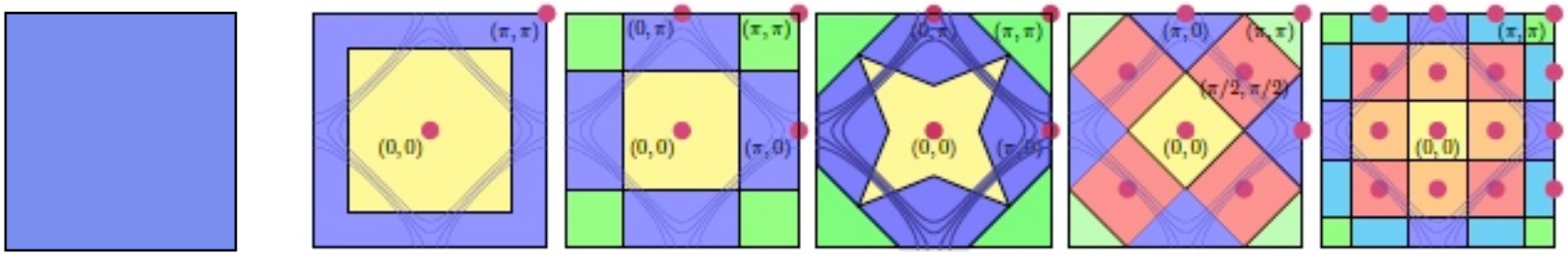
Most of this talk: 'DCA'

M. H. Hettler, M. Mukherjee, M. Jarrell, and H. R. Krishnamurthy
Phys. Rev. B **61**, 12739 (2000)

Momentum space approach: tile Brillouin zone
Choose N momenta K_a, draw an equal area patch around each one => N site impurity problem



1 **2** **4** **4*** **8** **16**

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

$\phi_a(p) = 1$ **if** p **is in the patch containing** K_a **and is 0 othewise**

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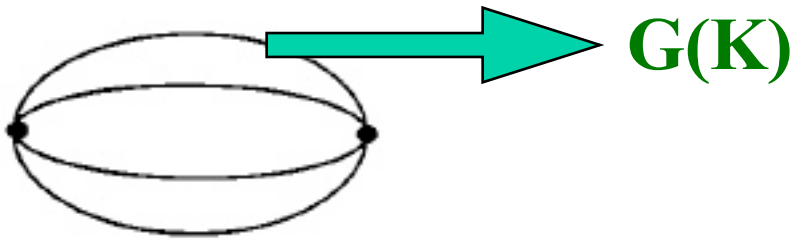
Approx Luttinger Ward functional: coarse-grain interaction in k space

$$\mathbf{V(k_1, k_2, k_3, k_4) \rightarrow V(K_1, K_2, K_3, K_4)}$$

$$\left(V(K_a ... = \int_{patch\ a} (dk) V(k, ...) \right)$$

Interpret G-lines in diagram as discrete set G(K) of QI model G's



The diagrams are the vacuum to vacuum diagrams for a quantum impurity model with interactions given by coarse-grained versions of the interactions of the original model

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Extremum condition

$$\mathbf{G^{QI}(K_a) = \int_a (dk)\,(\omega - \varepsilon_k - \Sigma(K))^{-1}}$$

Advantages:
--translation invariance preserved
--off site interactions can be treated (in some approximation)

Disadvantages:
--piecewise constant self energy
--translation invariance required

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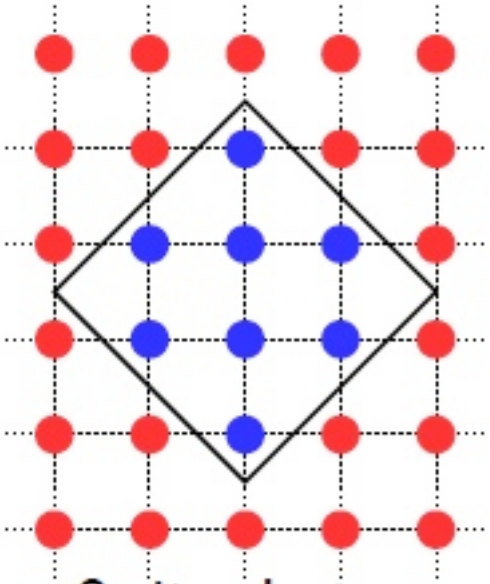
Useful to view auxiliary problem as 'quantum impurity model' (cluster of sites coupled to noninteracting bath)



8-site cluster

Quantum impurity model is in principle nothing more than a machine for generating self energies (as Kohn-Sham eigenstates are artifice for generating electron density)

As with Kohn Sham eigenstates, it is tempting (and maybe reasonable) to ascribe physical signficance to it

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The Technical Challenge:

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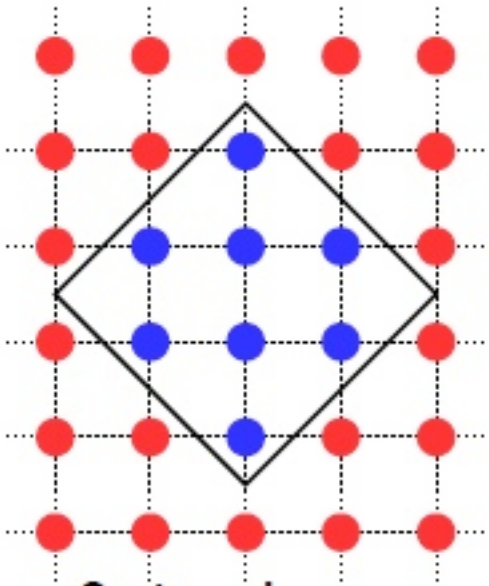
‘Impurity Solver’

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

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



8-site cluster

0 (space)+1(time) dimensional quantum field theory (with N sites x M orbitals x 2 spins discrete quantum numbers)

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Until recently, impurity model solvable for simplest models or in uncontrolled approximations

Breakthrough: continuous-time quantum Monte Carlo (CT-QMC)

***Rubtsov 05 Interaction expansion(CT-INT)**
***Werner/AJM 06 Hybridization expansion(CT-HYB)**
***Gull/Parcollet 08 Auxiliary field (CT-AUX))**

Review: RMP 83 349 (2011).

Goal now: controlled solutions =>non qualitative results

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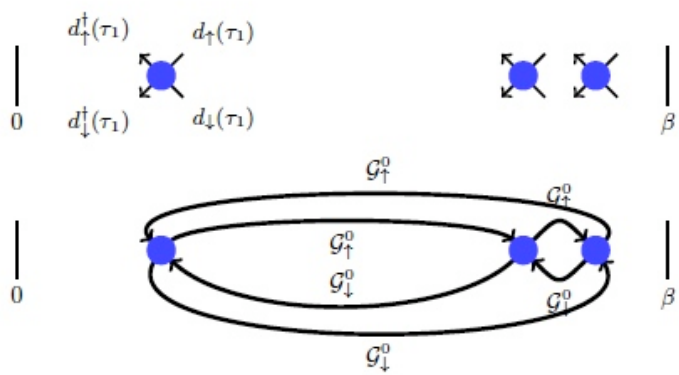
CT-QMC: stochastic sampling of diagram series

$$\mathbf{H_{QI} = E_0 + H_{int}^{loc} + \sum_{p,a} \left(V_{pa} d_a^\dagger c_{pa} + H.c \right) + \ldots}$$

CT-INT:
Expand in interaction



Large systems, but only density-density interaction

CT-HYB: Interaction rep; formal expansion in V

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

General interactions but small systems

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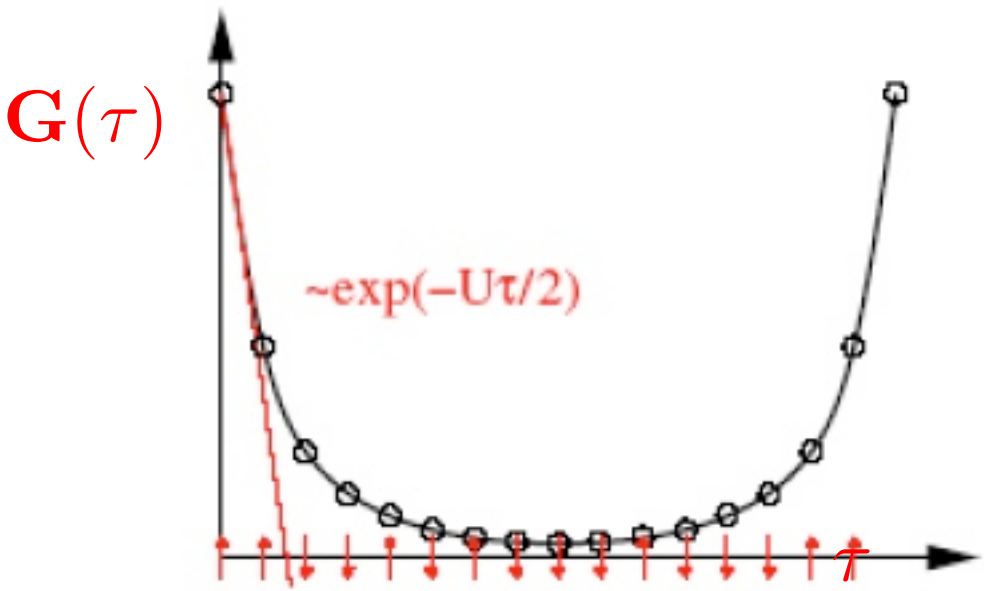
Key advantage: continuous time (many-body adaptive grid)

Previously used methods ('Hirsch-Fye') needed time discretization =>many points needed to represent behavior of Green function at small times



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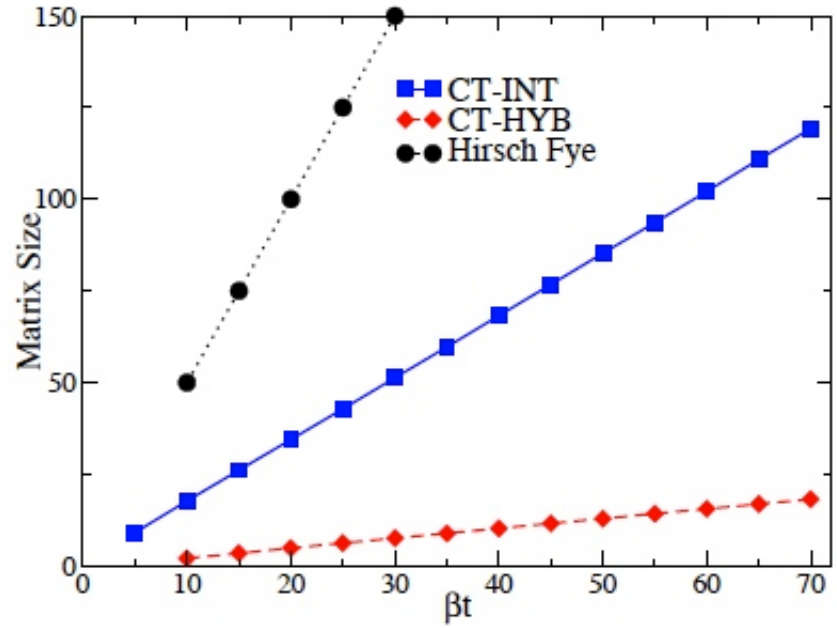
Consequence

All methods involve manipulating matrices:

Cost ~ cube of typical matrix size.

In new approaches: much smaller matrix is needed (same scaling, much lower prefactor)



efficiency improved by orders of magnitude => much more is possible

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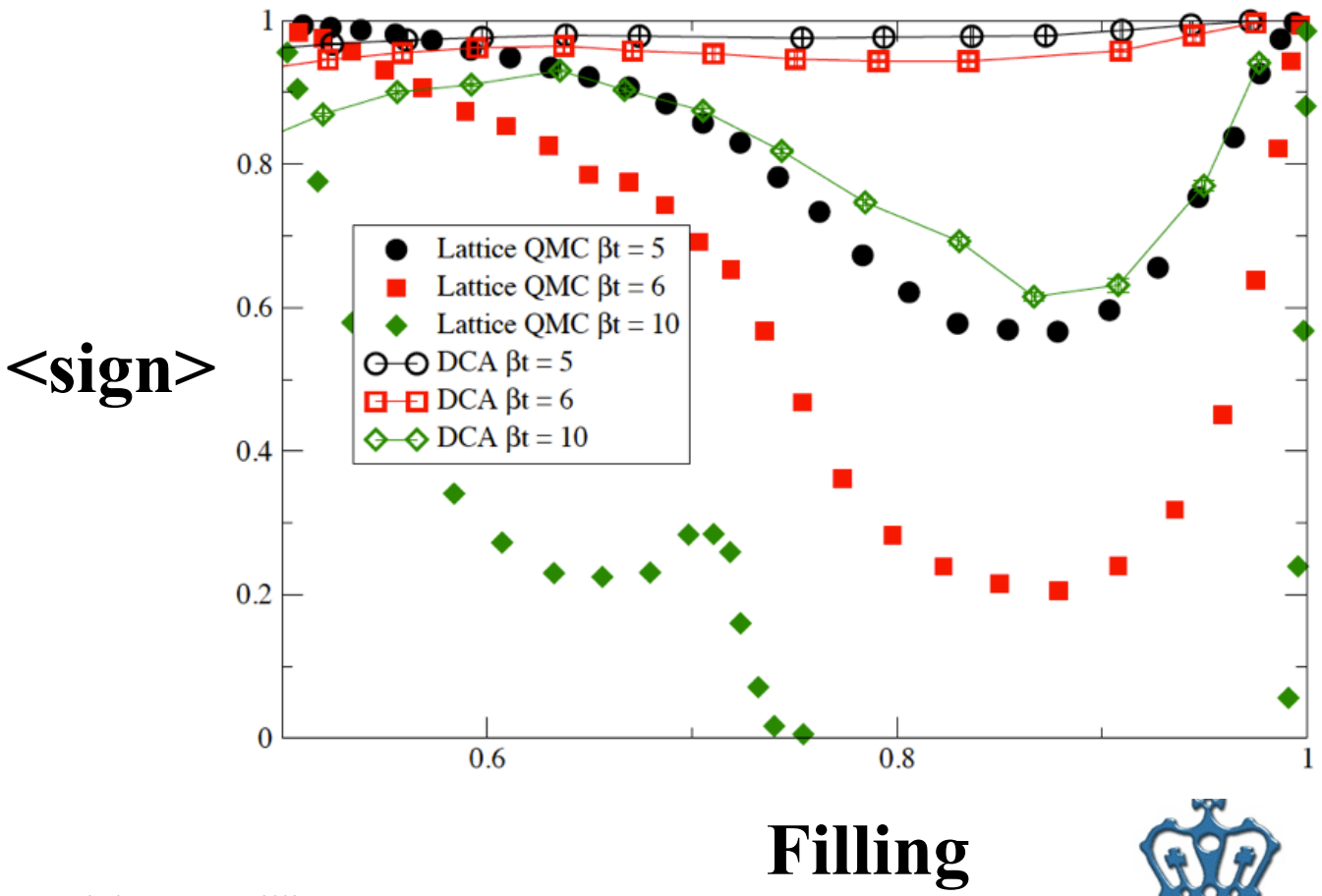
Empirical fact:

Coupling to bath=>

SIGN PROBLEM MUCH BETTER

2d Hubbard model, 36 sites



F. Assaad (QMC)
E. Gull (DMFT)

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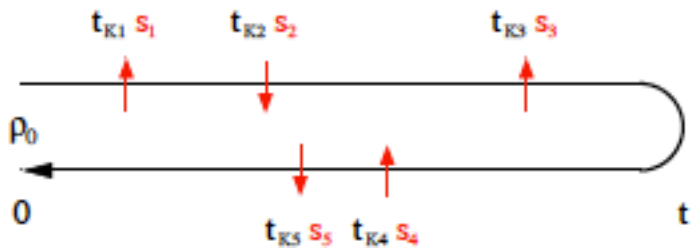
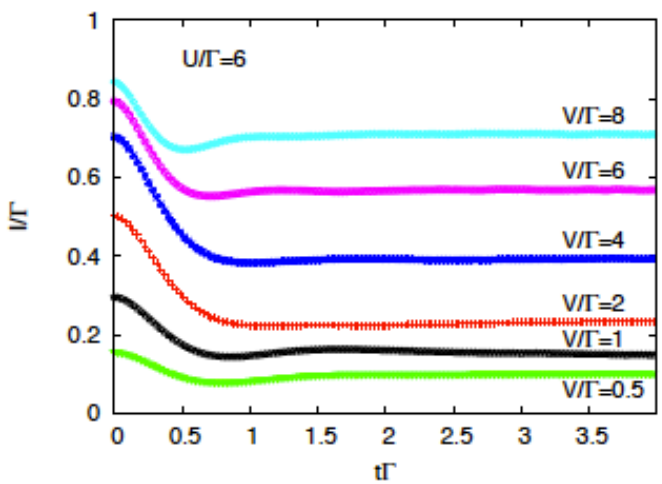
Generalizations

Nonequilibrium (Keldysh contour)

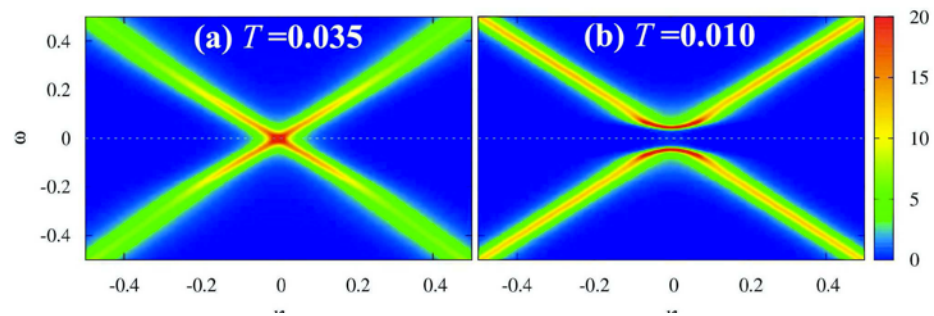




Werner, Oka Millis

Phys. Rev. B79, 035320 (2009).

Kondo-like models (Otsuki et al)



'Bold' diagrammatics: numerical corrections to analytical resummation (Gull and AJM)

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What can we do (at present)

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Technical remarks

Computations are trivially parallelizable:

Basic computation performed on a single core.

Initialization/thermalization time is very low

=>pays to distribute computation over as many as 10^4 cores

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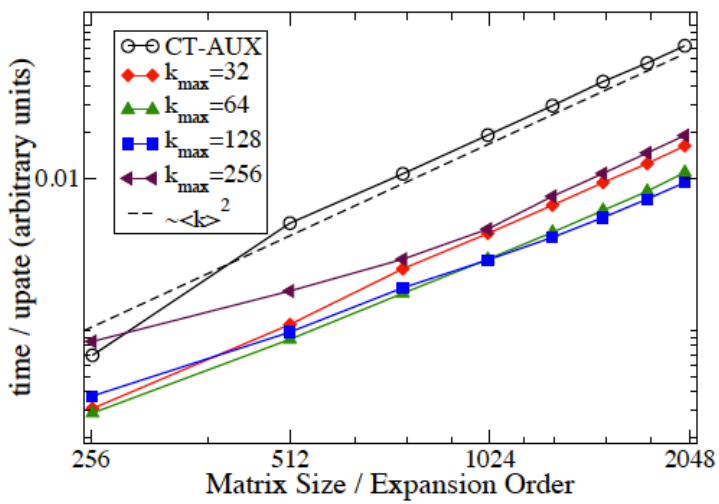
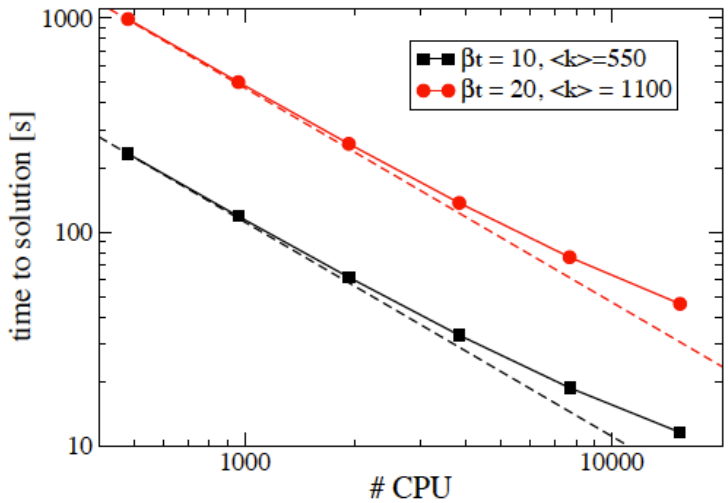
(sub)matrix updates
E. Gull et al PRB 83 075122

***Method deals with large matrices =>computational bottleneck is moving information from memory into cache**

=>Efficiency gain from arranging calculation to maximize # compute operations/memory call.





16 sites, U/t=8

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Typical computational costs (on 2-5 year old machines)

- Single-site 1 orbital: minutes on a laptop
- Single-site, 2 orbital: 16 cores, 4 hours
- 8 site Hubbard normal state (T=t/20): 64 cores, 8 hours
- 8 site Hubbard superconducting state (T=t/60) 128 cores 8 hours
- 8 site Hubbard, measurement for optics vertex (T=t/20), 4096 cores 8 hours (BNL Blue Gene)

These times are 'reasonable'

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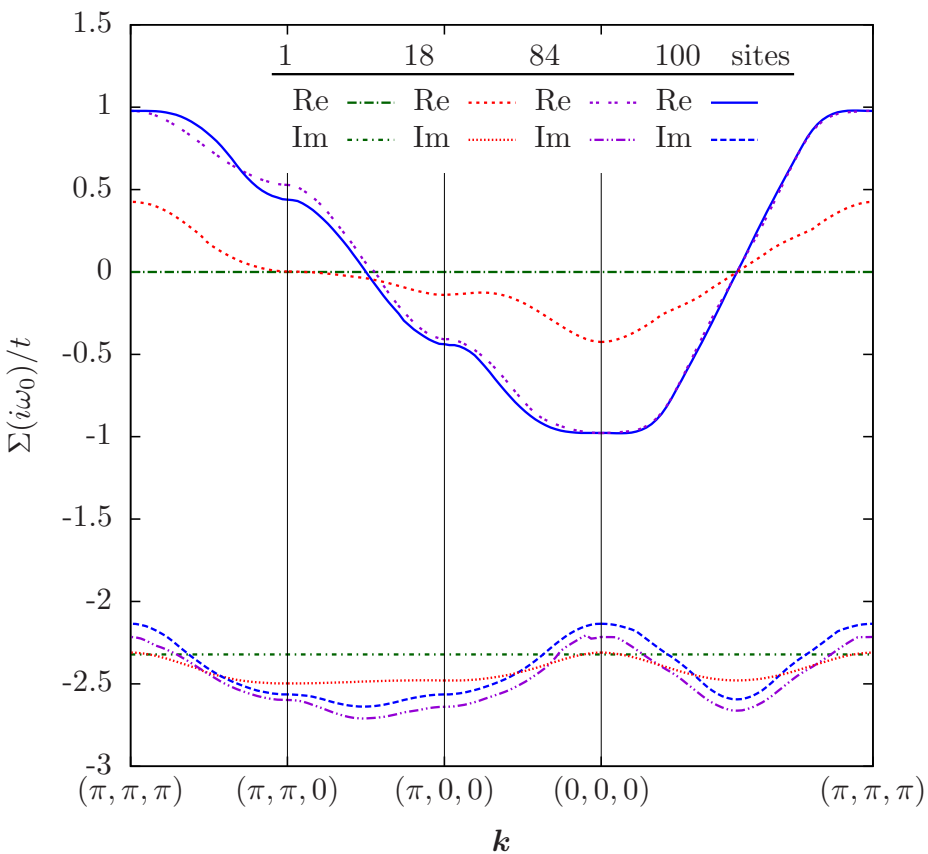
3 dimensional Hubbard model

S. Fuchs, E. Gull, L. Pollett et al PRL 106 030401 (2011)

U=8t cluster sizes up to 100 sites high-ish T=t/2



convergence of self energy with cluster size now demonstrated.

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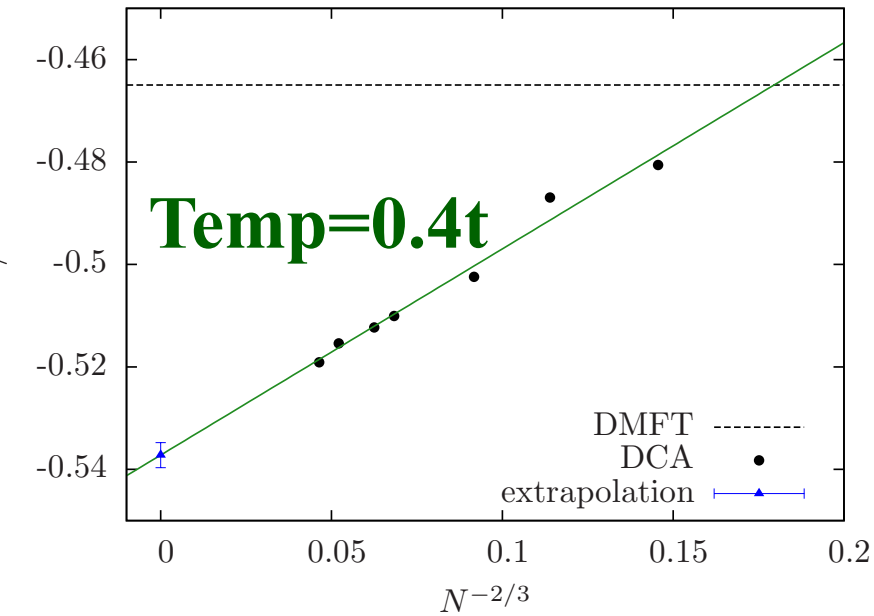
3 dimensional Hubbard model

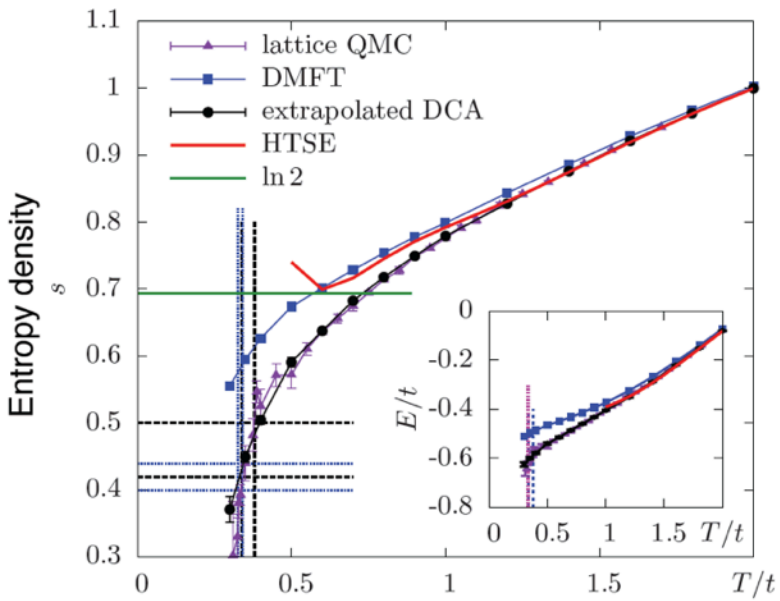
S. Fuchs, E. Gull, L. Pollett et al PRL 106 030401 (2011)

U=8t cluster sizes up to 100 sites high-ish T=t/2

extrapolation of energy



entropy density compared



Controlled extrapolation to thermodynamic limit now possible (at high T)

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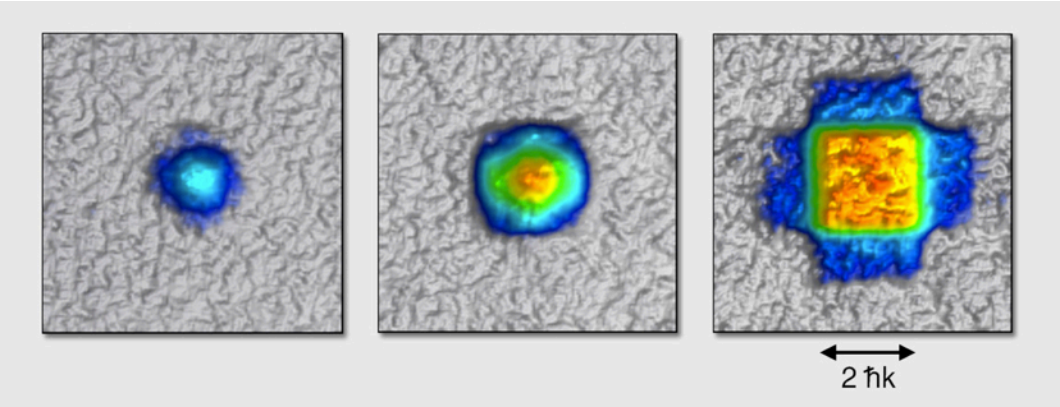
‘Optical emulator’: cold atomic gasses as analogue computers for model systems of condensed matter physics



fermion density distribution

T. Esslinger, Ann. Rev. CMP 129 (2010)

Very promising, but present experiments cannot reach low enough T; also validation needed

DMFT: Optical emulator emulator

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Limitations:

- `Heroic' (large cluster) calculations at present possible for Hubbard model only (on-site density-density interaction =>crucial simplification)
- Realistic (exchange and pair-hopping) interactions can be treated accurately only for models with 5 orbitals (single-site DMFT for transition metal oxides, actinides with truncation)
- Sign problem: severe for low symmetry situations (technically, hybridization function does not commute with local Hamiltonian; physical example Co on surface of Cu).

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Qualitative Insights from DMFT: Charge-transfer energy and the metal-insulator transition in late transition metal oxides

M. J. Han, X. Wang, C. Marianetti

Computations at CNMS

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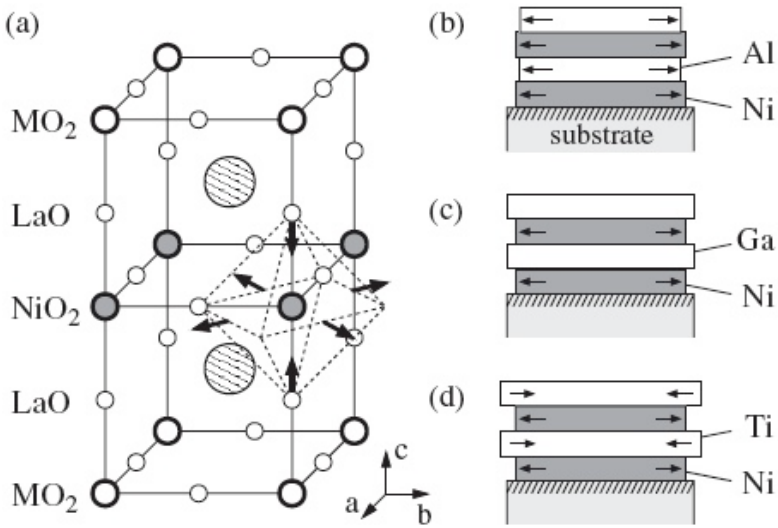
Chaloupka/Khalliulin:
?New high-T_c superconductor?



Phys. Rev. Lett. **100** 016404 (2008)

Idea:

Bulk $LaNiO_3$ Ni $[d]^7$ (1 electron in two degenerate e_g bands).

In correctly chosen structure, split e_g bands, get 1 electron in 1 band--"like" high-Tc

Even if no superconductivity--by how much can we split the orbitals??

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Thus: orbital polarization (differential occupancy of atomic orbitals)

- **Fundamental electronic property**
- **Important for magnetism and conductivity**
- **Largely controlled by 'local' (on-site) physics**

System:
--$ReNiO_3$-based superlattices

Method:
--LDA+DMFT (single site, ok for local physics)

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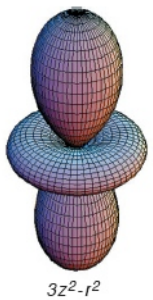
Pseudocubic $ReNiO_3$

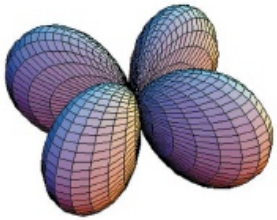
Relevant orbitals: e_g symmetry
Ni-O antibonding combinations

$3z^2$-r^2



Hybridizes strongly along z
Hybridizes weakly in x-y

x^2-y^2



Hybridizes strongly along x-y
Hybridizes very weakly in z

2 orbitals transform as doublet in cubic symmetry
superlattice: breaks symmetry, splits degeneracy

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One electron (band theory) physics

Pseudocubic $LaNiO_3$

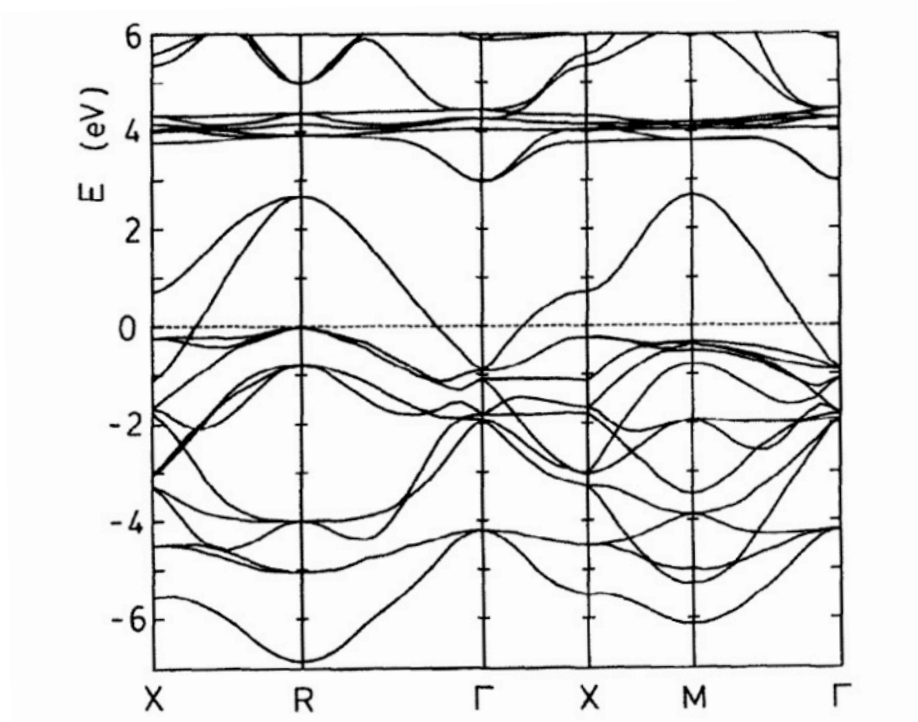

Figure 1: Band structure of a cubic $LaNiO_3$ with a lattice parameter of 3.88Å. The origin of energy is taken at the Fermi energy.

Hamada's result:

Two-fold degenerate e_g band complex

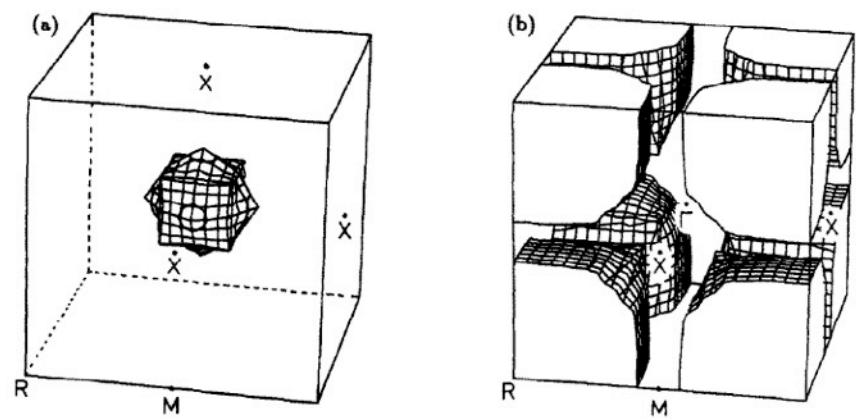

Figure 2: Fermi surfaces of a cubic $LaNiO_3$: (a) a electron pocket at the Γ point containing 0.04 electrons, and (b) a large hole Fermi surface around the R point containing 1.04 holes inside.

Hamada J Phys. Chem Sol **54** 1157

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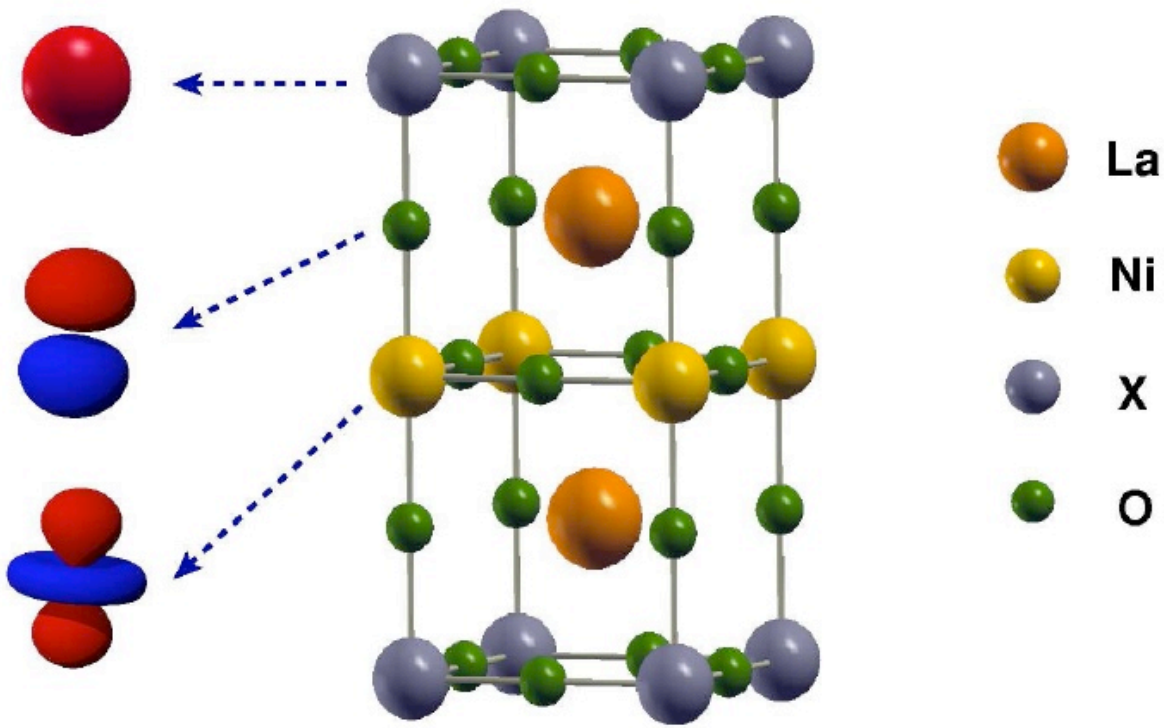
Superlattice

La_2NiXO_6 X=B, Al, Ga, In



X layer breaks cubic symmetry, acts as 'orbital magnetic field' giving orbital polarization

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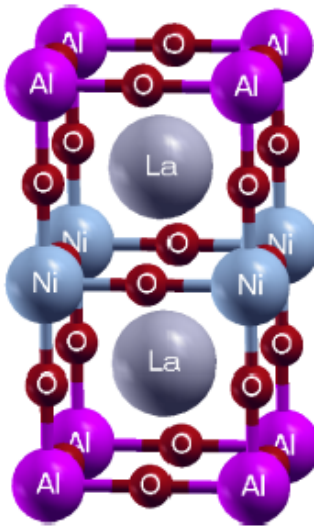
Superlattice



Superlattice: La_2AlNiO_6

$LaAlO_3$ layer: insulating barrier

Question (Khalliulin): how much orbital polarization can we get?

$$P = \frac{n_{x^2-y^2} - n_{3z^2-r^2}}{n_{x^2-y^2} + n_{3z^2-r^2}}$$

MJ Han, X Wang, C. Marianetti and A. J. Millis, arXiv:1105.0016

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p-d hybridization: orbitals are mixed d character in several bands

3 definitions of orbital polarization:

--total orbital occupancy integrated over all energies
--occupancy of near Fermi surface ``bands”
--fermi surface topology

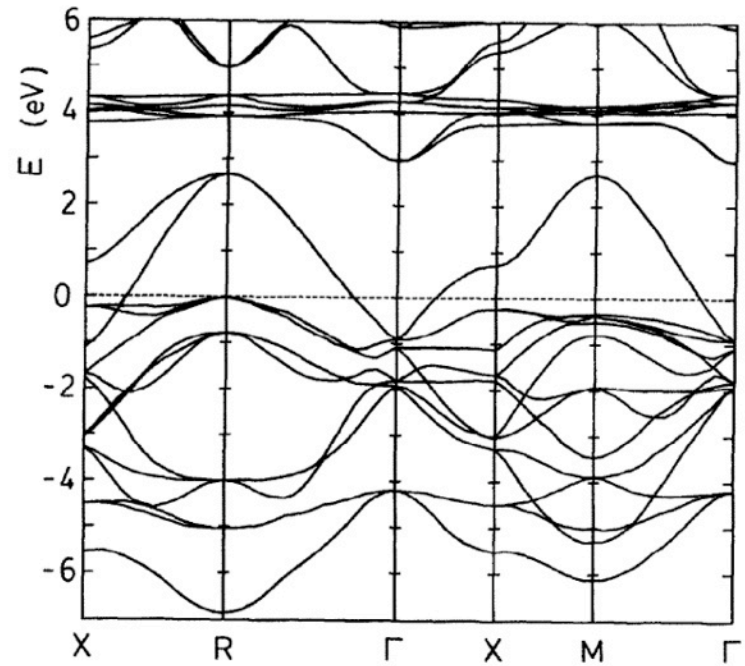

Figure 1: Band structure of a cubic $LaNiO_3$ with a lattice parameter of 3.88Å. The origin of energy is taken at the Fermi energy.

All definitions give consistent picture

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Theoretical Approach (General)

1. Basic electronic structure (DFT or Hartree-Fock)

2. Identify 'active subspace' (important orbitals, interactions).

3. Solve correlation problem in active subspace

4. Embed active subspace solution (self consistently) in electronic structure

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``LDA+DMFT”

- **Correlated subspace: Ni e_g (defined from wide energy range Wannier functions) (=>0.5Angstrom size)**
- **Local interaction U, J**
- **DMFT: solves many-body problem via mapping to impurity model + self consistency condition**
- **Embedding via DMFT self-consistency condition**

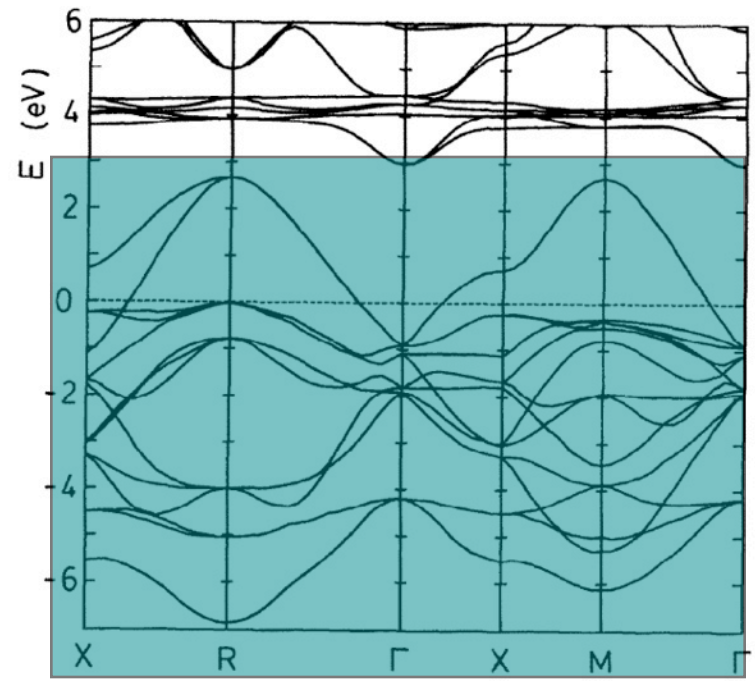

Figure 1: Band structure of a cubic $LaNiO_3$ with a lattice parameter of 3.88Å. The origin of energy is taken at the Fermi energy.

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Technical notes

1. Actual procedure: map band theory to p-d tight binding model. Other orbitals irrelevant

$$\mathbf{H} = \varepsilon_{\mathbf{d}} \mathbf{d}^{\dagger}\mathbf{d} + \varepsilon_{\mathbf{p}} \mathbf{p}^{\dagger}\mathbf{p} + \mathbf{t}_{\mathbf{pd}} \left(\mathbf{d}^{\dagger}\mathbf{p} + \mathbf{H.c}\right) + \mathbf{t}_{\mathbf{pp}} \mathbf{p}^{\dagger}\mathbf{p} + \mathbf{U}_{\mathbf{dd}} \mathbf{d}^{\dagger}\mathbf{d}^{\dagger}\mathbf{d}\mathbf{d}$$

2. Key embedding issue: ``double counting correction”

$$\textbf{value of } \boldsymbol{\Delta} = \varepsilon_p - \varepsilon_d$$

3. Minor embedding issue: definition of orbital polarization

4. Rotationally invariant Hunds coupling treated correctly

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‘Double Counting’ correction

Hartree shift: $\varepsilon_d \rightarrow \varepsilon_d + U < \hat{N}_d >$

part of this is included in the band calculation

=>

must adjust ε_d–but by how much?

This talk: investigate a range of double counting corrections

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Two dimensional parameter space: U and d-level energy

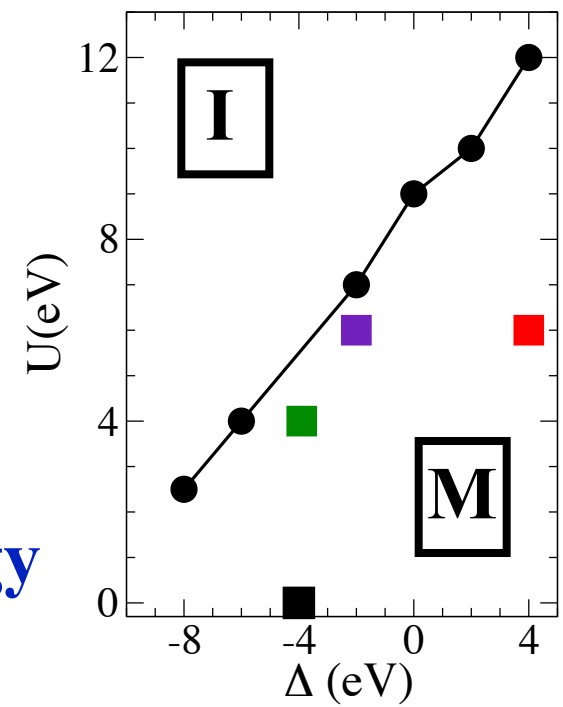
Interaction $\frac{U}{2}N_d(N_d-1)$
acts to change d occupancy

=> 'double counting' question: as one adds 'U' to a model do you

(a) reduce N_d

(b) shift d-level energy (keep N_d fixed)

Phase Diagram



Here--consider two trajectories in parameter space:
N_d constant or E_d constant

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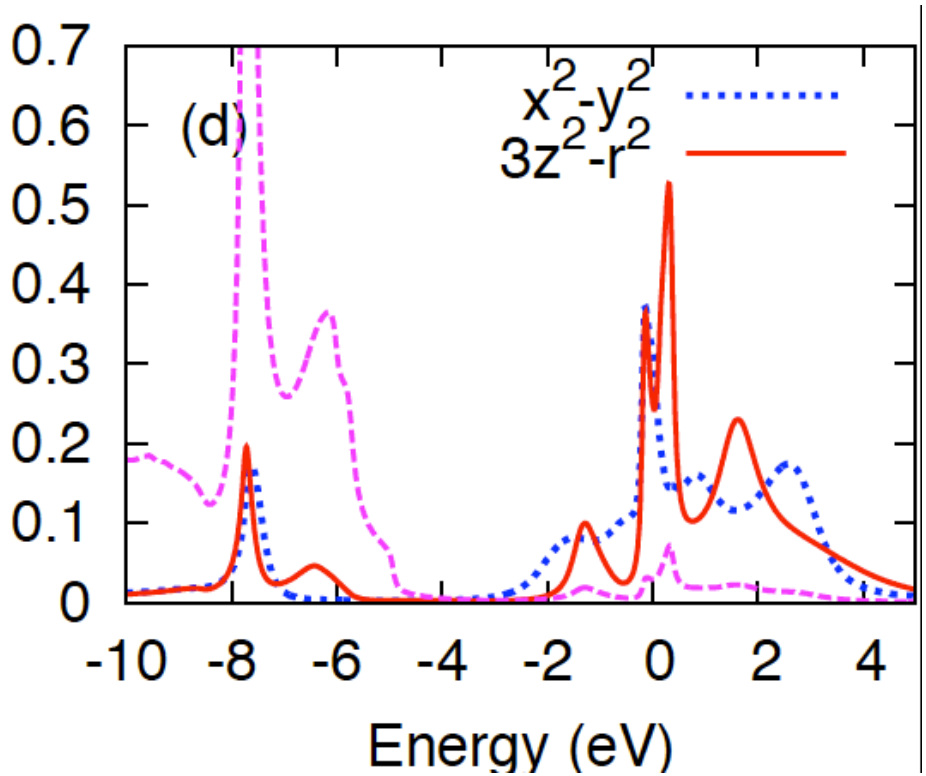
Orbital polarization: multiband model

3 definitions:

--total orbital occupancy integrated over all energies
--occupancy of near Fermi surface ``bands”
--fermi surface topology



All definitions give consistent picture

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Key point: Ni-O covalency

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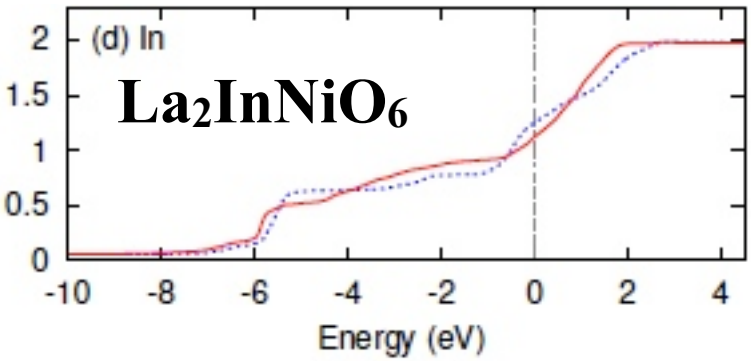
Key point: Ni-O covalency

Occupancy per orbital



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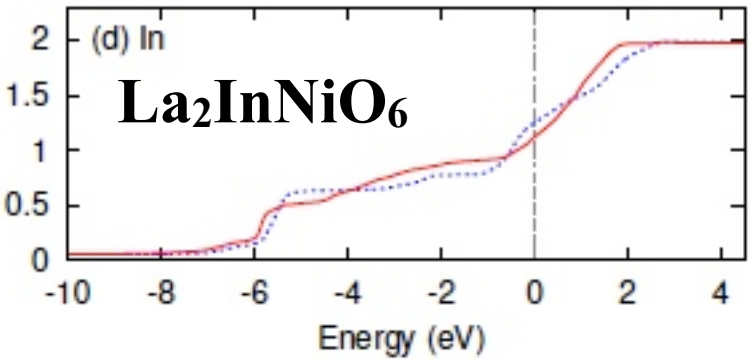
Key point: Ni-O covalency

Occupancy per orbital

Band theory: electron moves from O to Ni,

configuration is $d^8\bar{L}$



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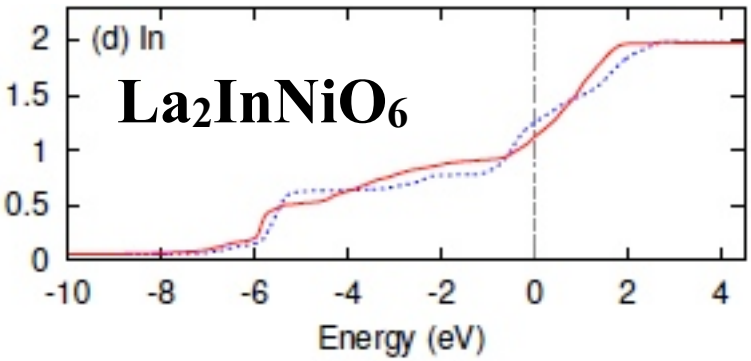

High-spin d^8: no orbital polarization possible

'Hubbard' model does not capture this physics because $d^8\underline{L}$ state does not exist

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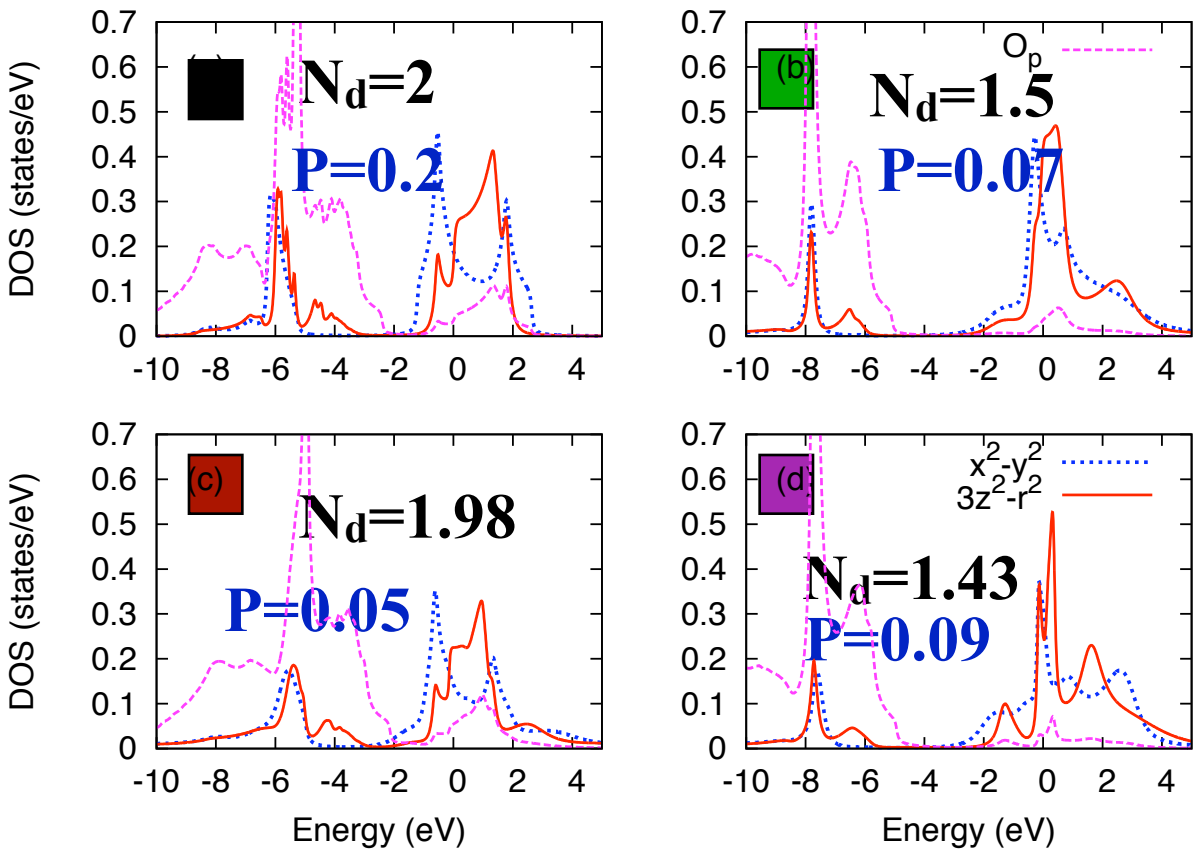
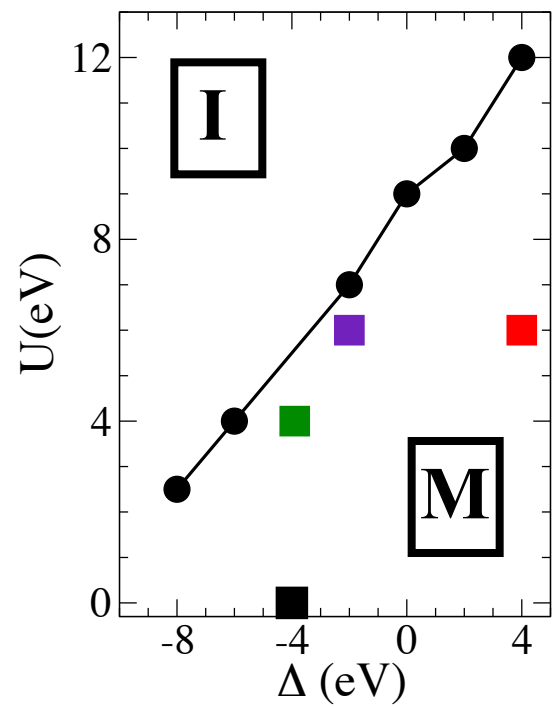
Spectral and orbital polarization along two trajectories

Spectral functions



Phase Diagram



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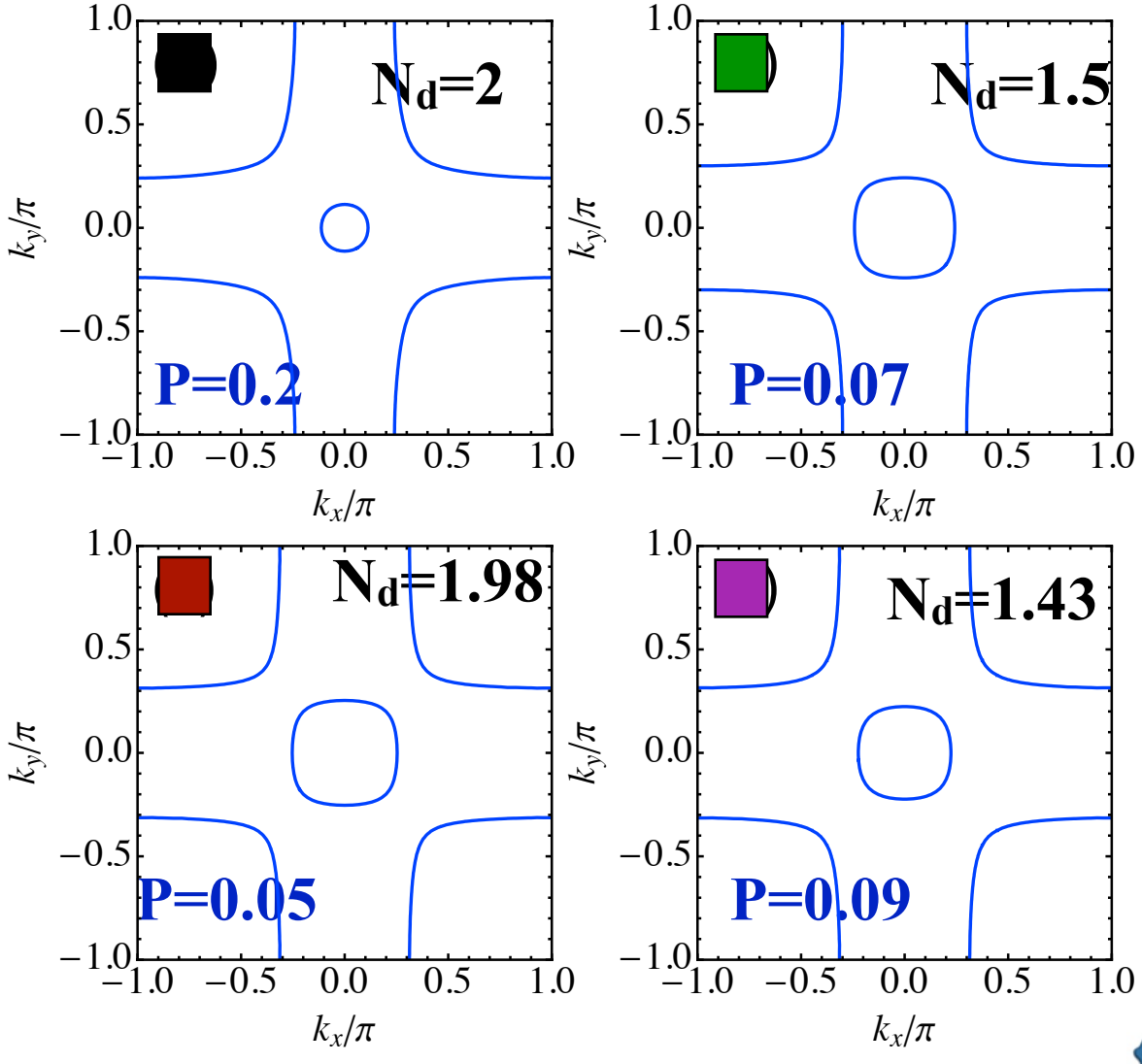
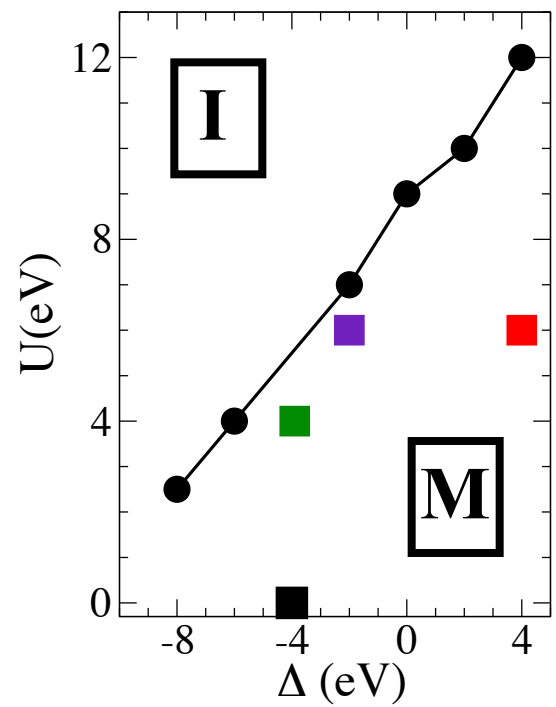
Fermi Surface



Phase Diagram



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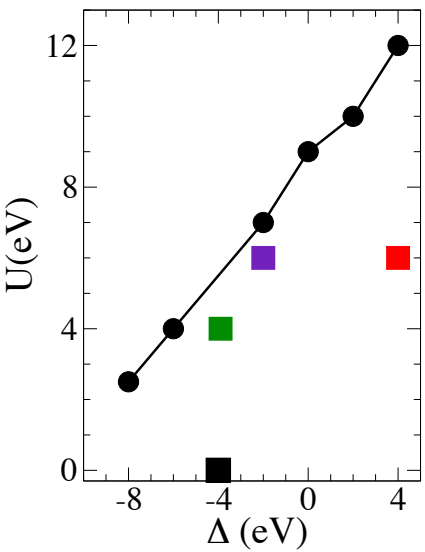
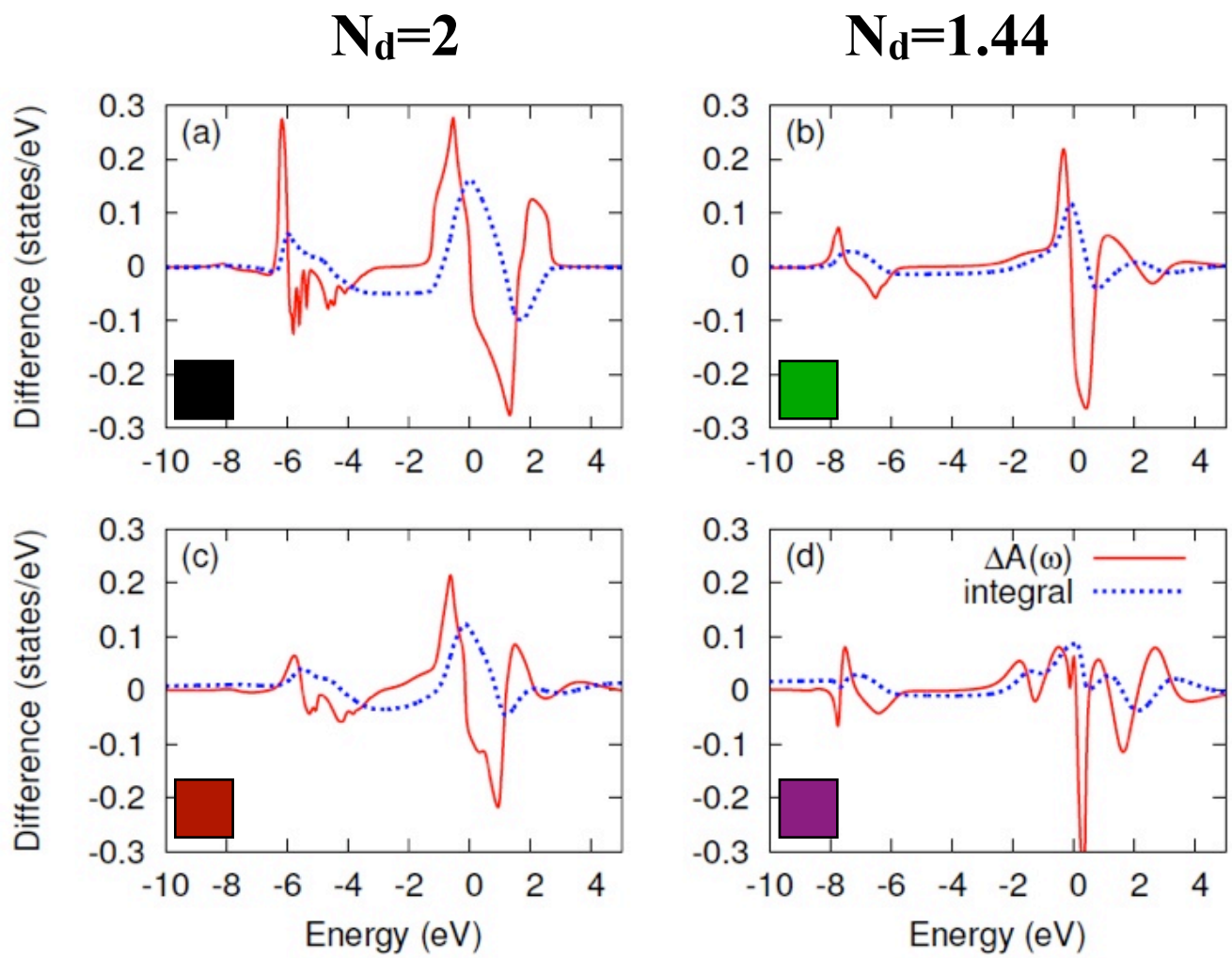
Differential polarization





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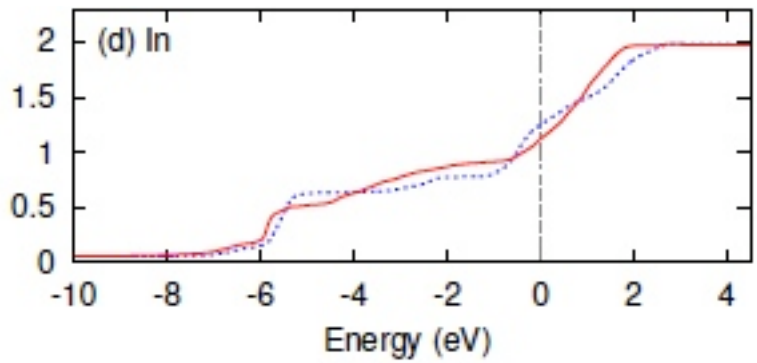
Why no polarization?



e_g occupancy near 2.
high-spin $d^8\underline{L}$ configuration is important. This configuration cannot be polarized.
`Hubbard' description misses this physics.

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Conclusions

1. Large orbital polarization in Nickelate superlattices unlikely

2. 'Hubbard' modelling not appropriate

3. Proximity to Metal-Insulator transition does not enhance polarization (much)

4. Questions

- **Where are the materials on the U-Nd phase diagram**
- **Experimental values of P**

5. Other Systems (titanates, vanadates)--in progress!

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New insights into the doping-driven metal insulator transition in high T_c (copper oxide) superconductors

With E. Gull, A. Georges, O. Parcollet, P. Werner, M. Ferrero

Computations at CNMS

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Characteristic features of high-Tc superconductivity

1. Superconductivity created by adding carriers to nontrivial insulator
2. Characteristic scaling with doping: optical conductivity strongly doping dependent; carrier mass much less so
3. `Pseudogap' for hole doped materials
4. Precursor: scattering rate anisotropy

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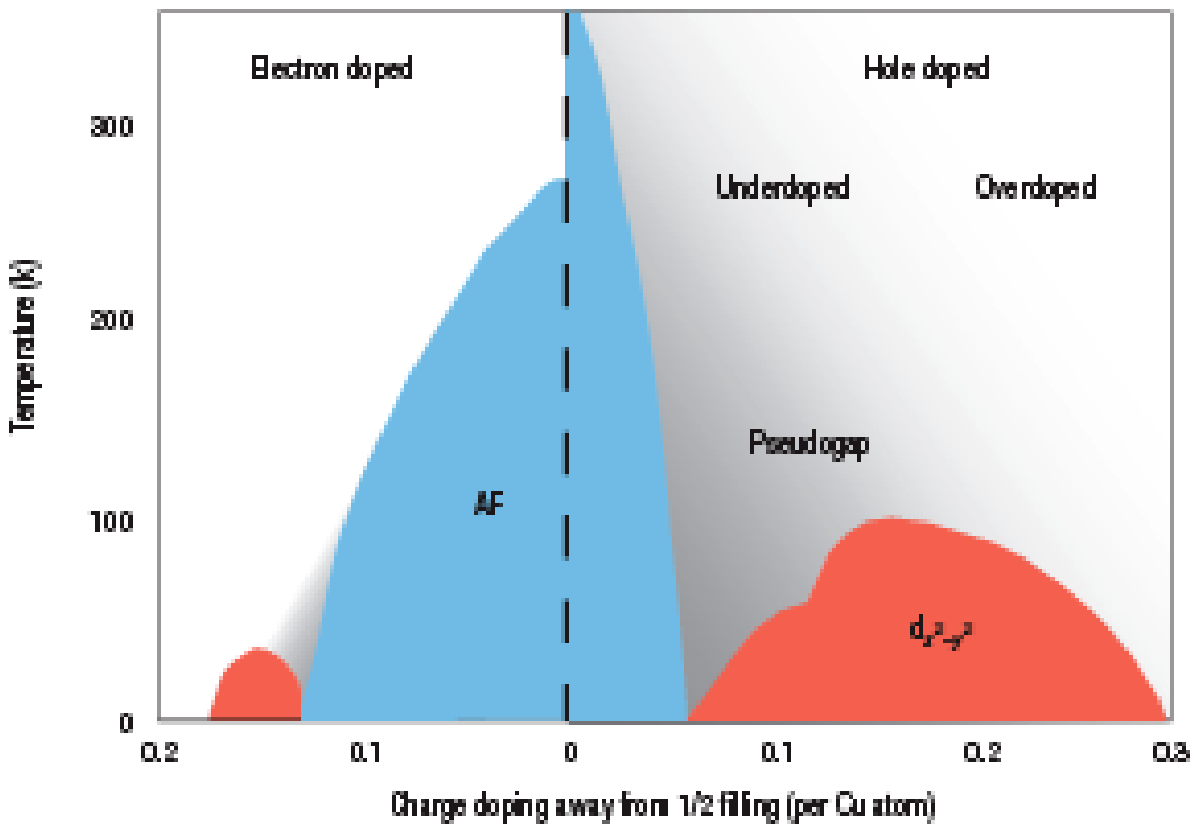
Phase diagram



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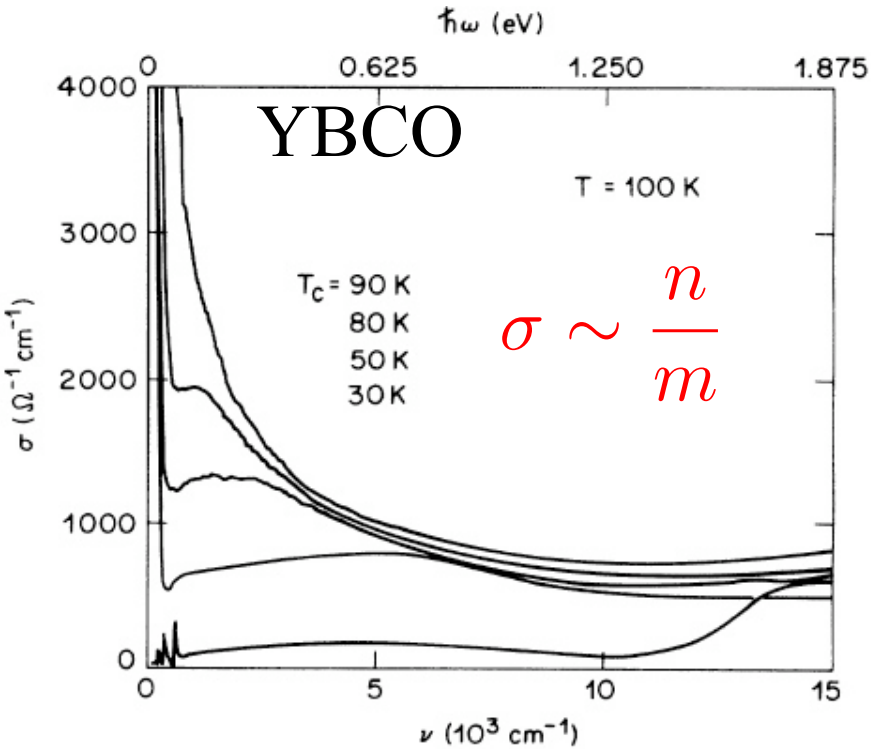
Probes of carrier motion

Optical conductivity:

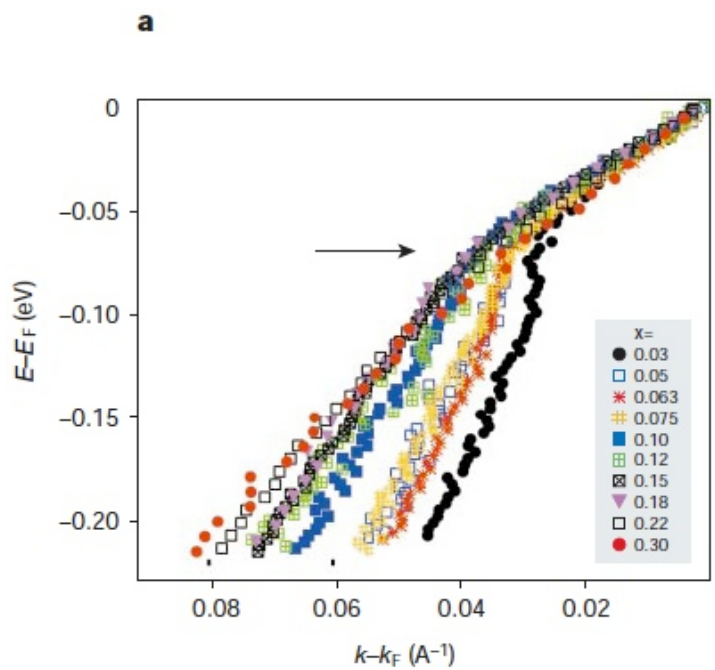


$$\sigma \sim \frac{n}{m}$$

Orenstein, Thomas, Millis et. al,
Physical Review B42, 6342-62 (1990).

n/m: strongly doping dep.

Photoemission:



carrier velocity: weakly doping dependent

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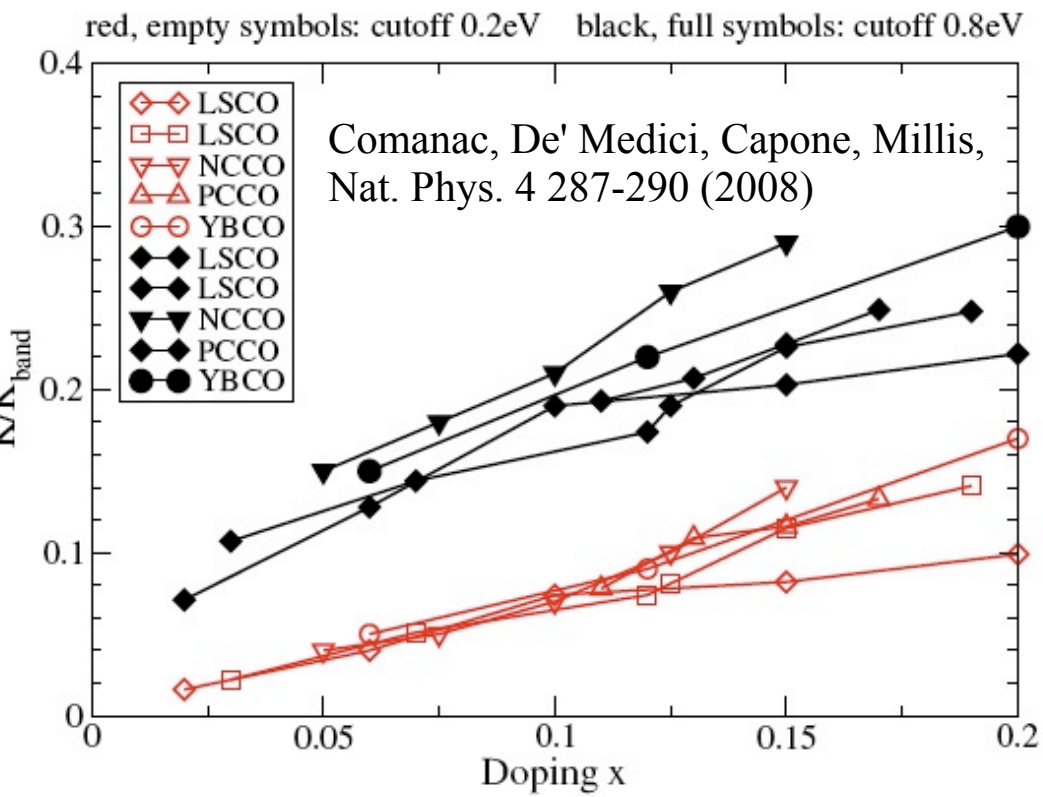
Characterize by 'spectral weight'

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



$\mathbf{\Omega} = 0.2eV$:
$K \sim x$

$\mathbf{\Omega} = 0.8eV$:
$K \sim x + 0.1$

Low freq conductivity ~x but quasiparticle velocity is not

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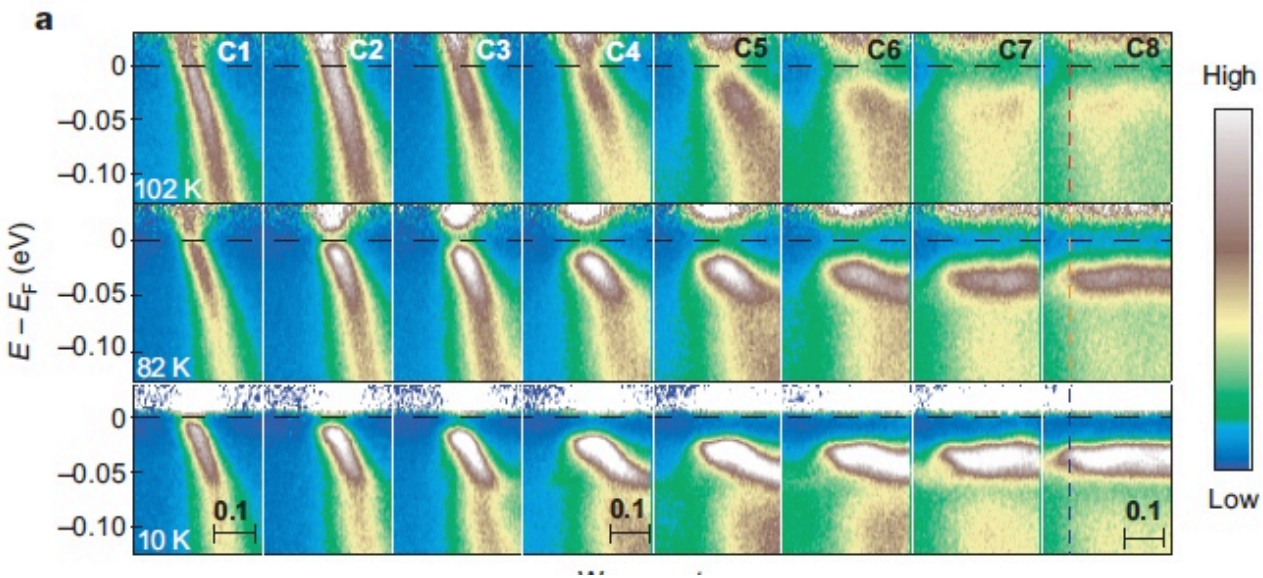
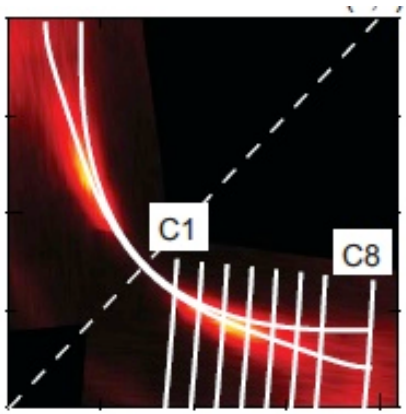
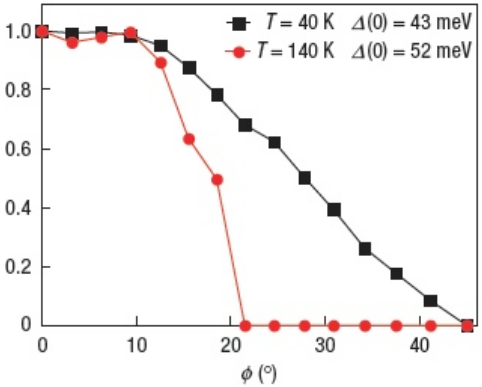
`Pseudogap'

Suppression of density of states in zone corner

Angle-resolved photoemission sample w/ 90K T_c:







S. Lee et al, Nature 450, p. 81 (2007)

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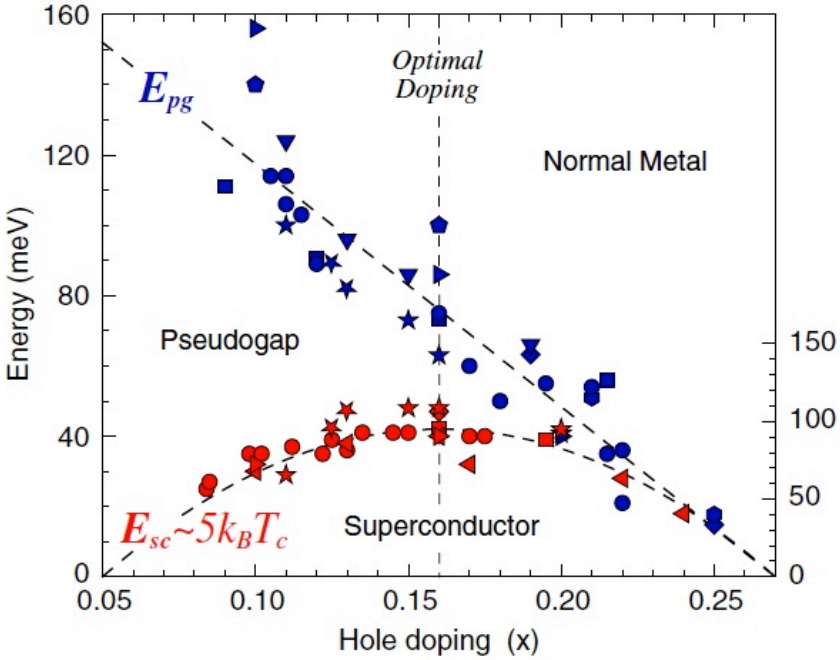
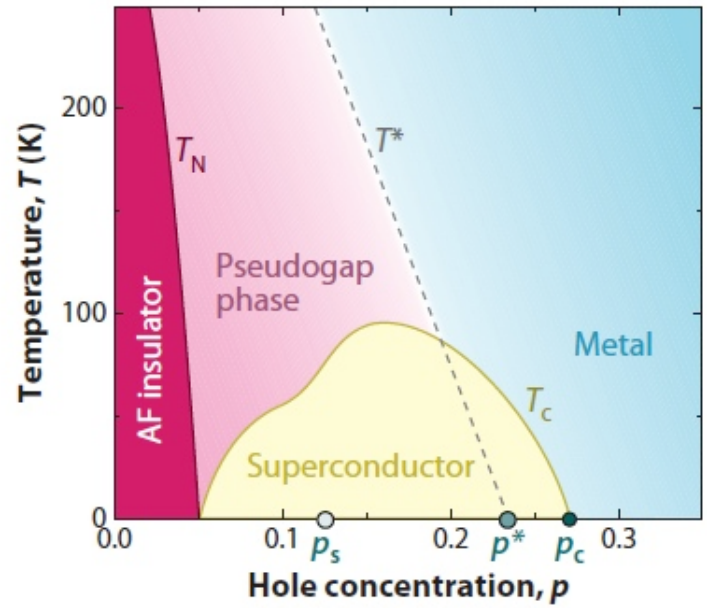
`Pseudogap'

Magnitude increases as doping decreases

Onset temp. increases as doping decreases



Huefner et al Rep. Prog. Phys. 71 062501 (2008)



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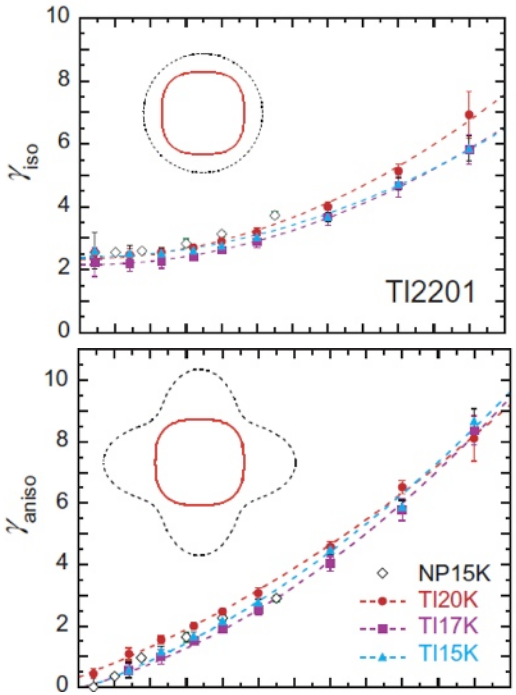
Precursor of pseudogap in momentum-space dependent scattering rate



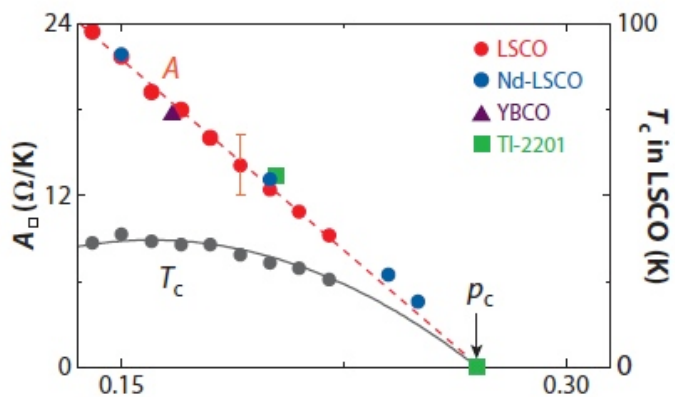
Temperature

M. J. French et al., N. J. Phys.

11 055057 (2009)

Idea: from anisotropy of magnetoresistance, can tease out variation of electronic scattering rate around fermi surface.

Result: unconventional term (rate ~T not T^2) associated with (0,pi) turns on as doping is decreased.



L. Taillefer Ann. Rev. Condens.Matter Phys. 2010. 1:51–70

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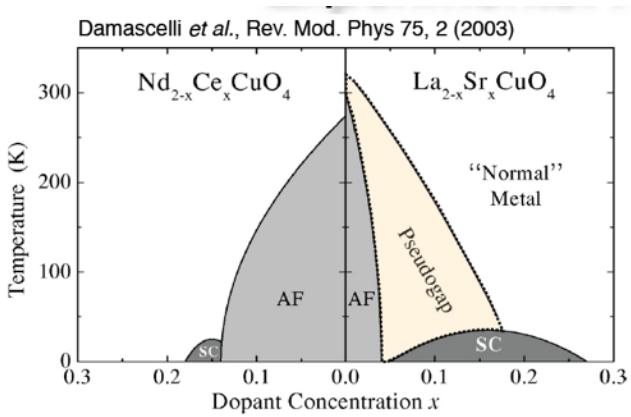
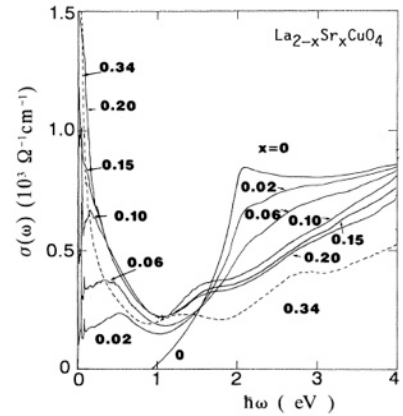
Are these phenomena properties of a theoretical model?

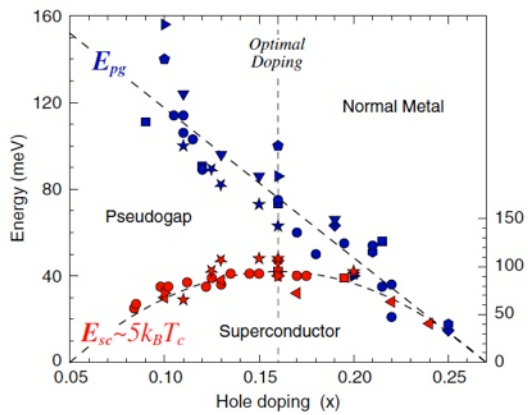
Phase diagram



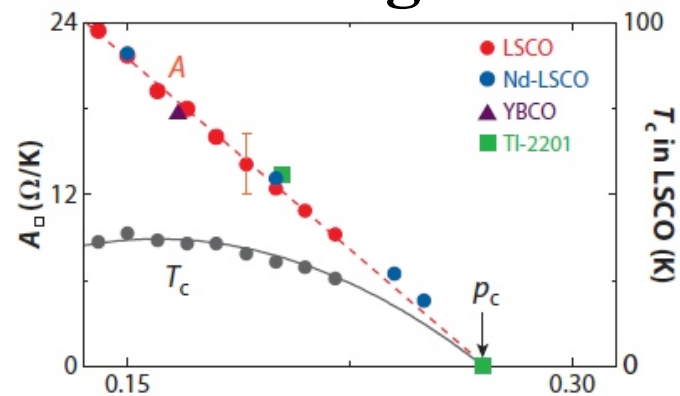
Conductivity



Pseudogap



Scattering rate



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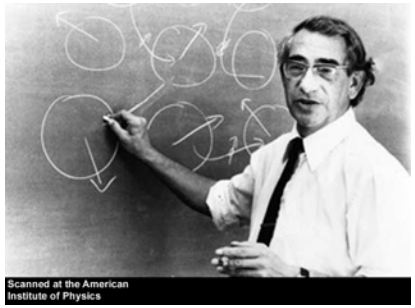
www.picsearch.com

For high T_c materials, Model Hamiltonian: 'Hubbard model'



http://theor.jinr.ru/~kuzemsky/jhbio.html

One (spin degenerate) orbital per lattice site.
hopping t short ranged.

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

Important parameters:
--relative interaction strength U/t
--electron density n

Hopping parameters from band theory

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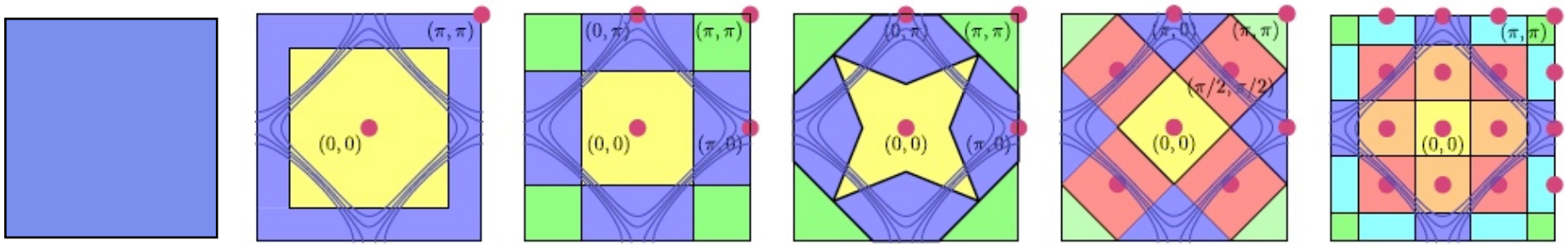
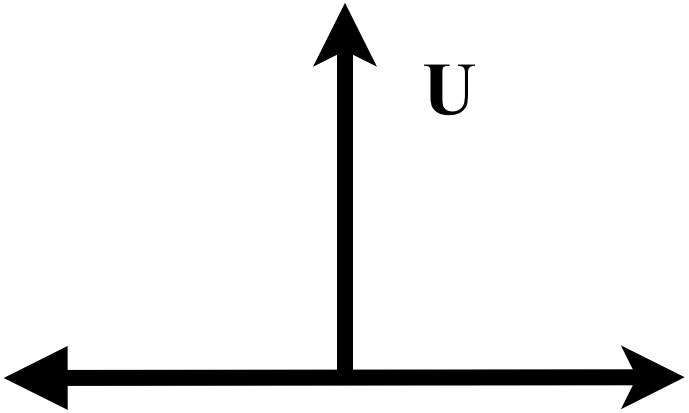
1,2,4,8,16 site cluster DMFT



1 2 4 4* 8 16



Carrier concentration

Focus on quantities for which all clusters give (qualitatively) the same answers

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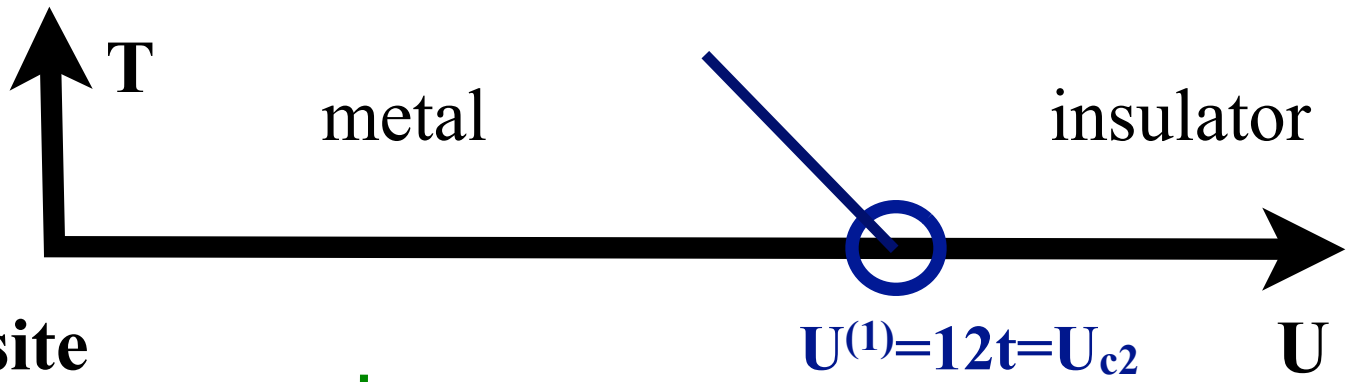
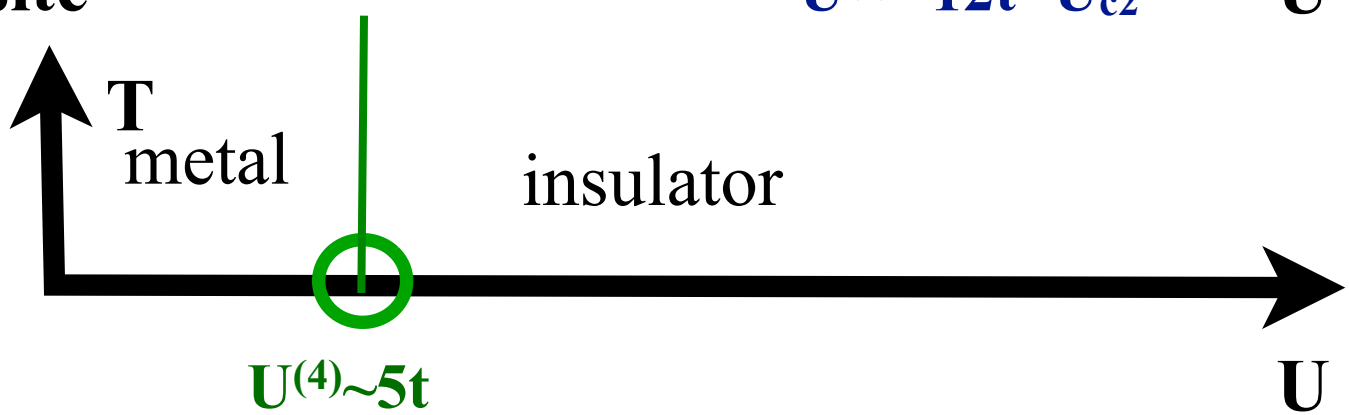
The metal-insulator transition

1 and 4 site approx: different from all others

1-site



4-site



Only 1 transition

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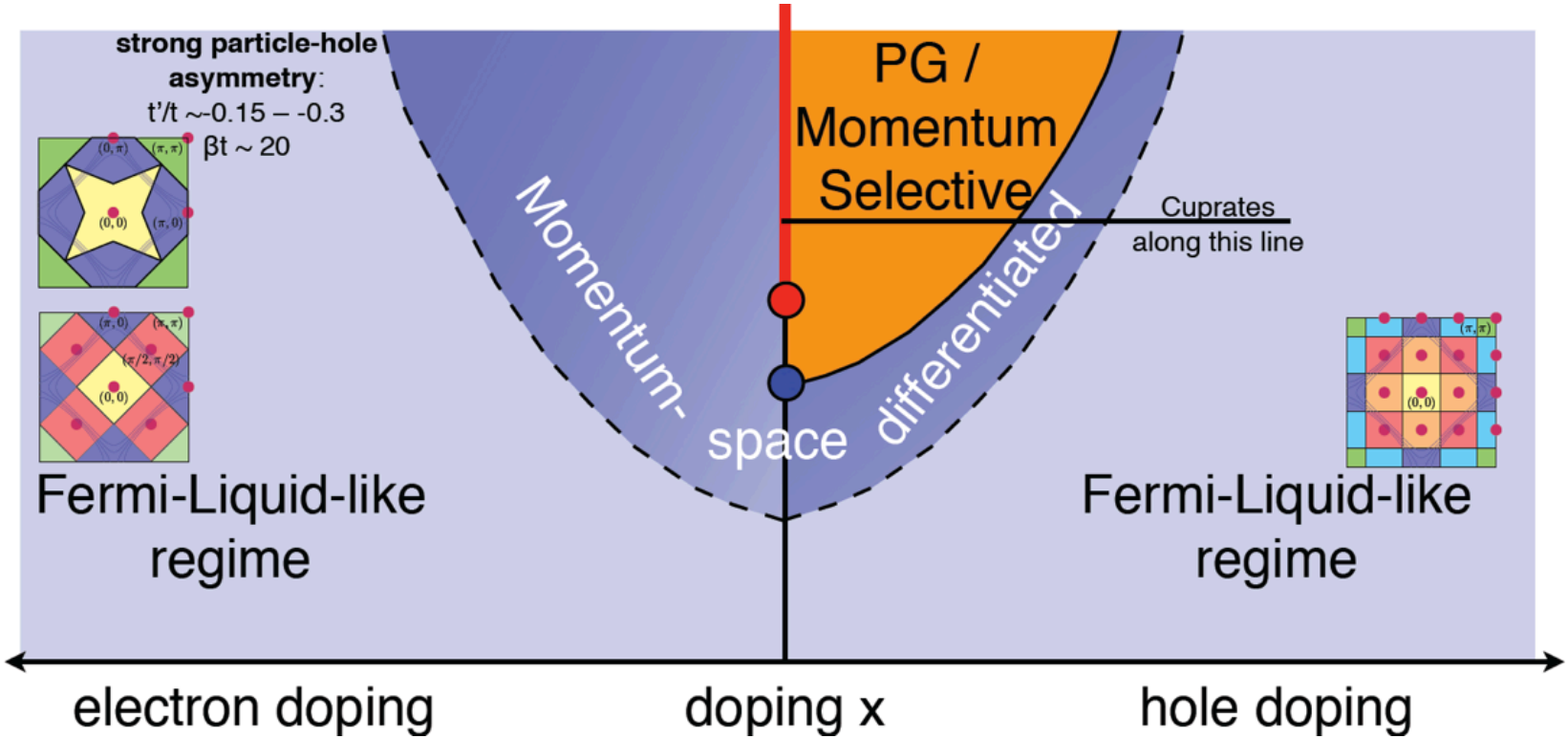
Generic solution: metal-insulator transition is multi-stage



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Sector selective transition

$$\varepsilon_k = -2t\left(\cos k_x + \cos k_y\right) - 4t' \cos k_x \cos k_y$$

look first at

$$\beta G(\tau = \frac{\beta}{2}) = \beta \int \frac{dx}{4\pi} \frac{A_{sector}(x)}{cosh\frac{x}{2T}} = \int \frac{dy}{4\pi} \frac{A_{sector}(2Ty)}{coshy}$$

Integral is peaked at y~1 =>T->0
picks out fermi level density of states
Directly measured.
No analytic continuation. No interpolation

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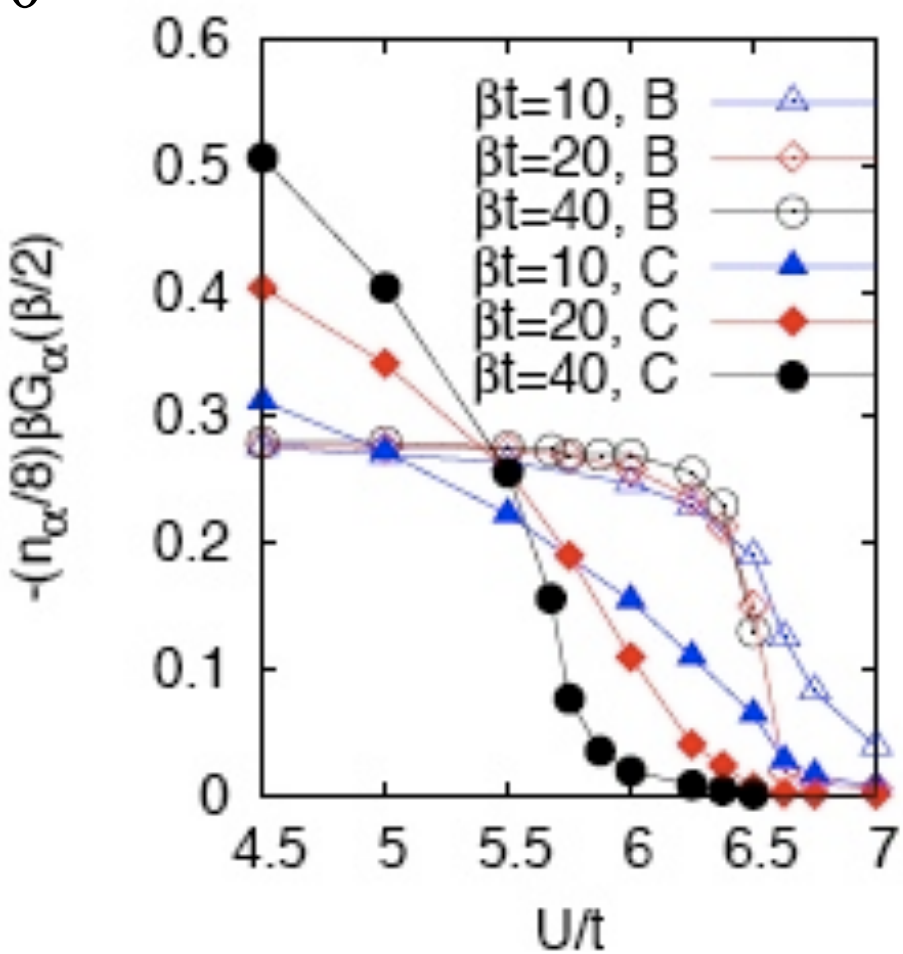
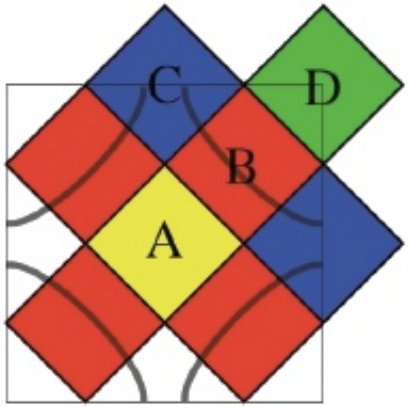
n=1, vary U
sector-selective transition

t'=0





van Hove singularity in sector C=>T-dependence

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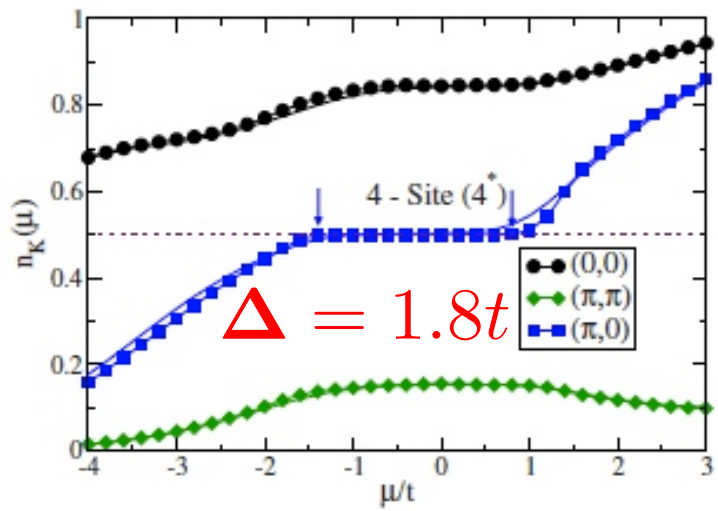
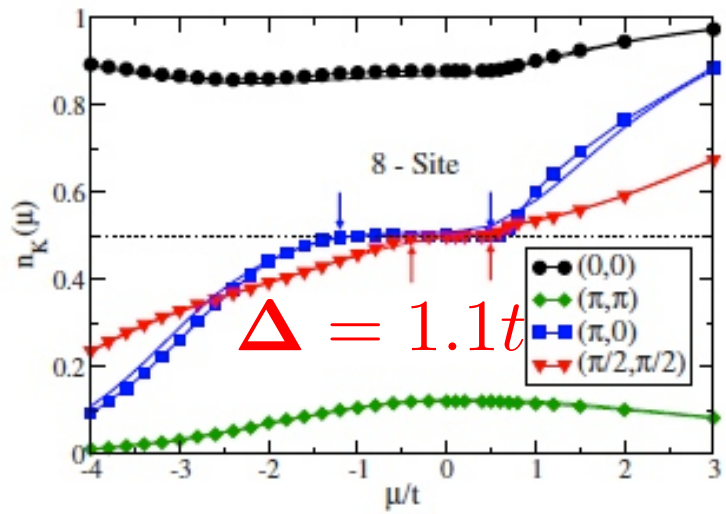
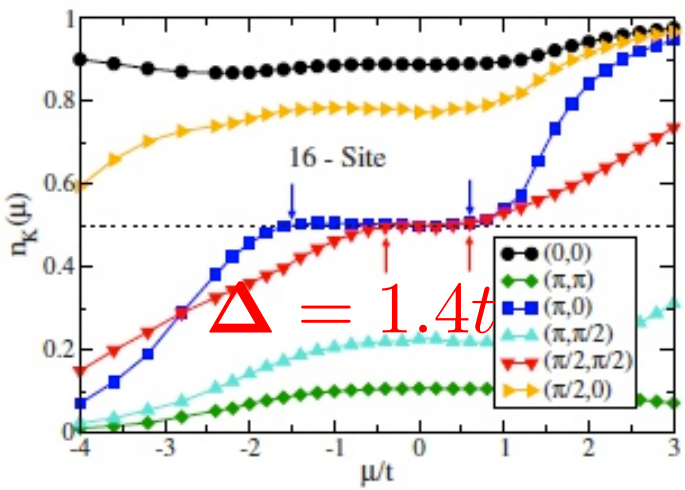
Momentum sector occupancy vs chemical potential







All sizes: n=1=>gap (different in different momenta) =>paramagnetic (Mott) insulator, reasonable estimate of gap~1.4t

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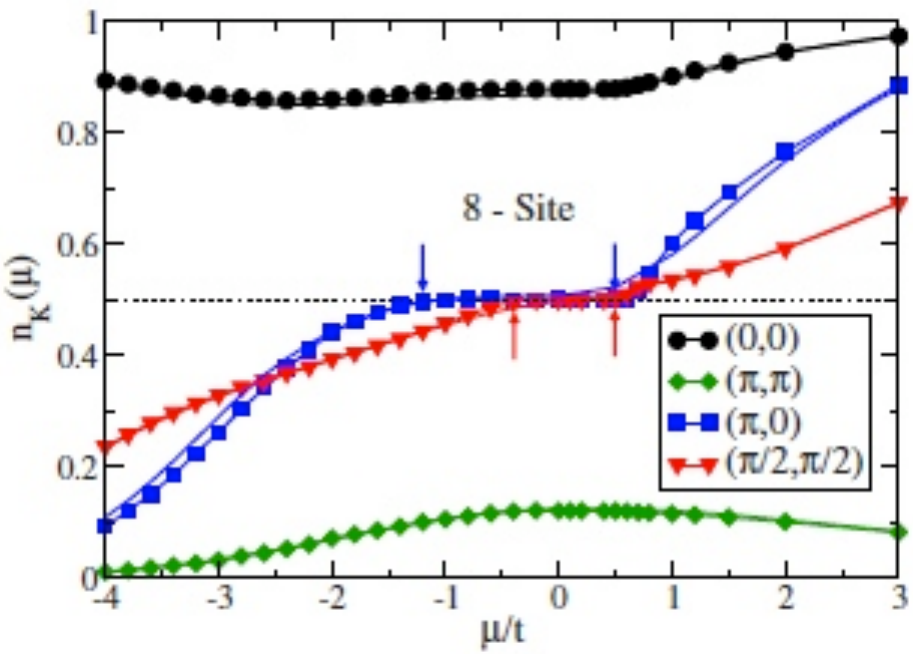
‘Sector selective’ transition: region near (0,Pi) remains gapped on doping



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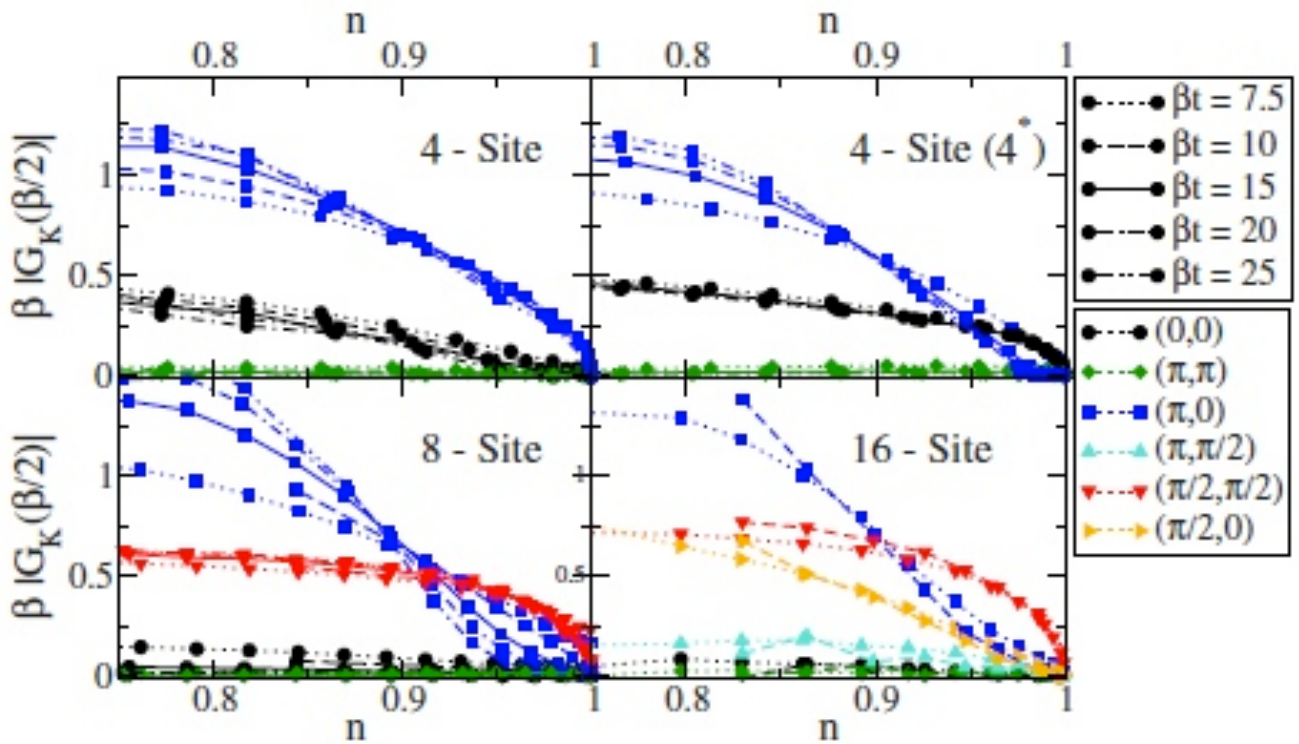
Additional evidence for 'sector selectivity'



$$\beta G(\tau = \frac{\beta}{2}) = \beta \int \frac{dx}{4\pi} \frac{A_{sector}(x)}{cosh\frac{x}{2T}} = \int \frac{dy}{4\pi} \frac{A_{sector}(2Ty)}{coshy}$$

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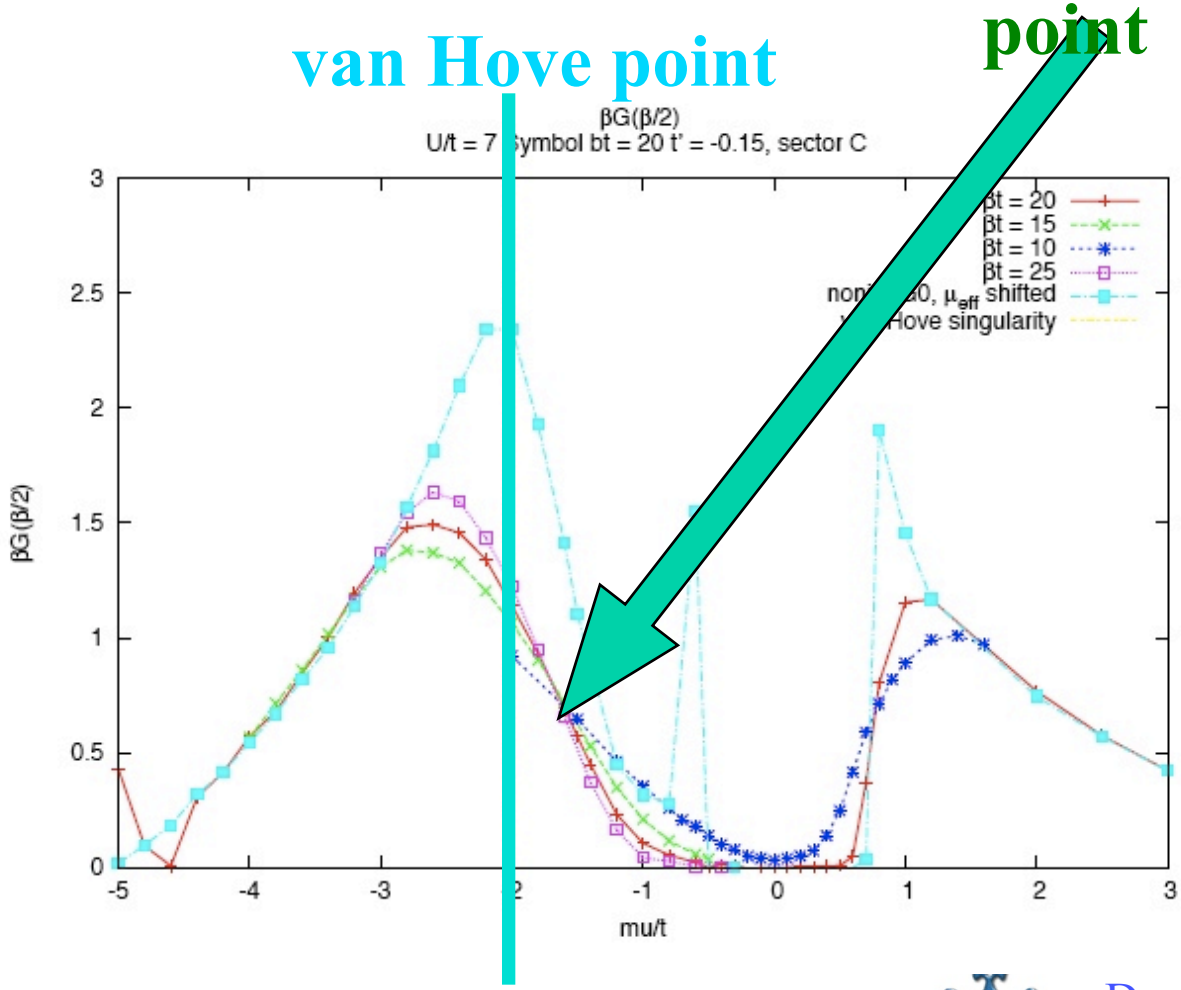
Transition not controlled by van Hove physics



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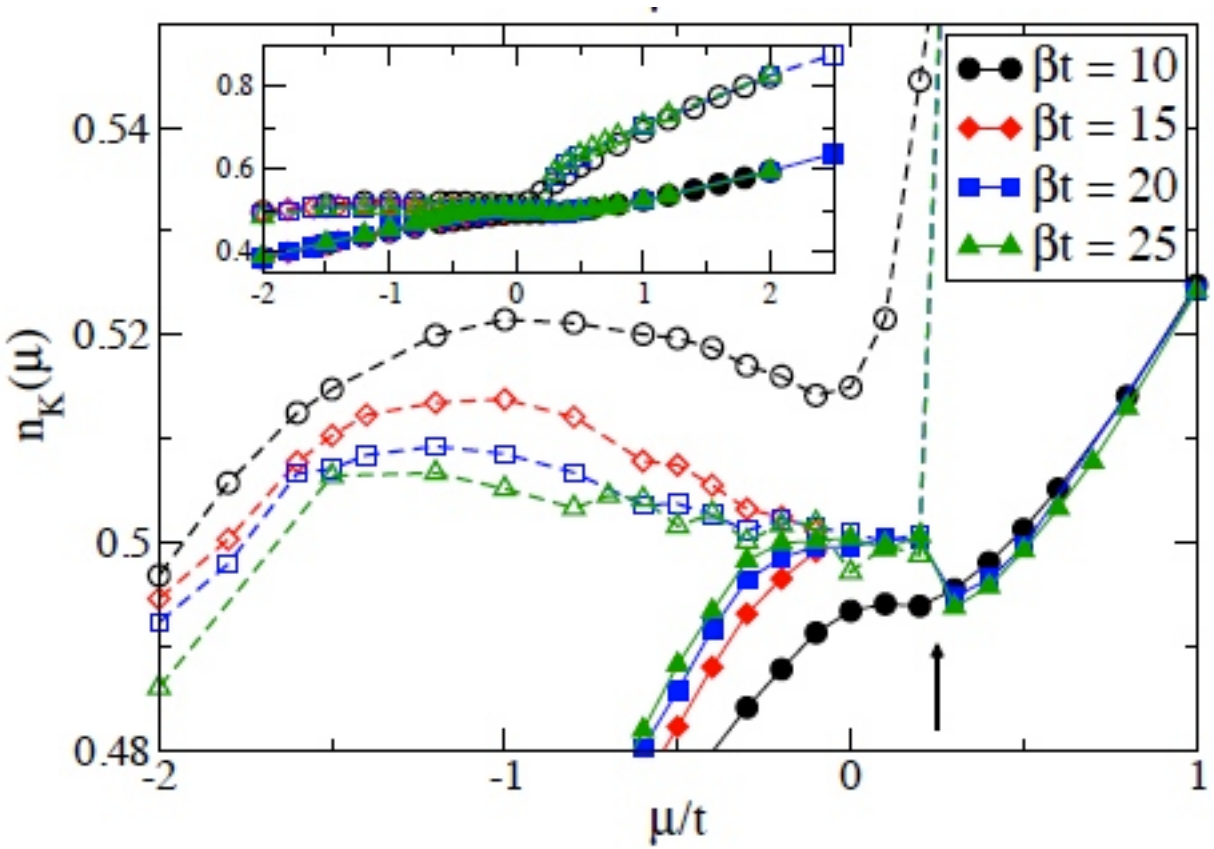
t'=-0.3t



e-doping: transition strongly first order

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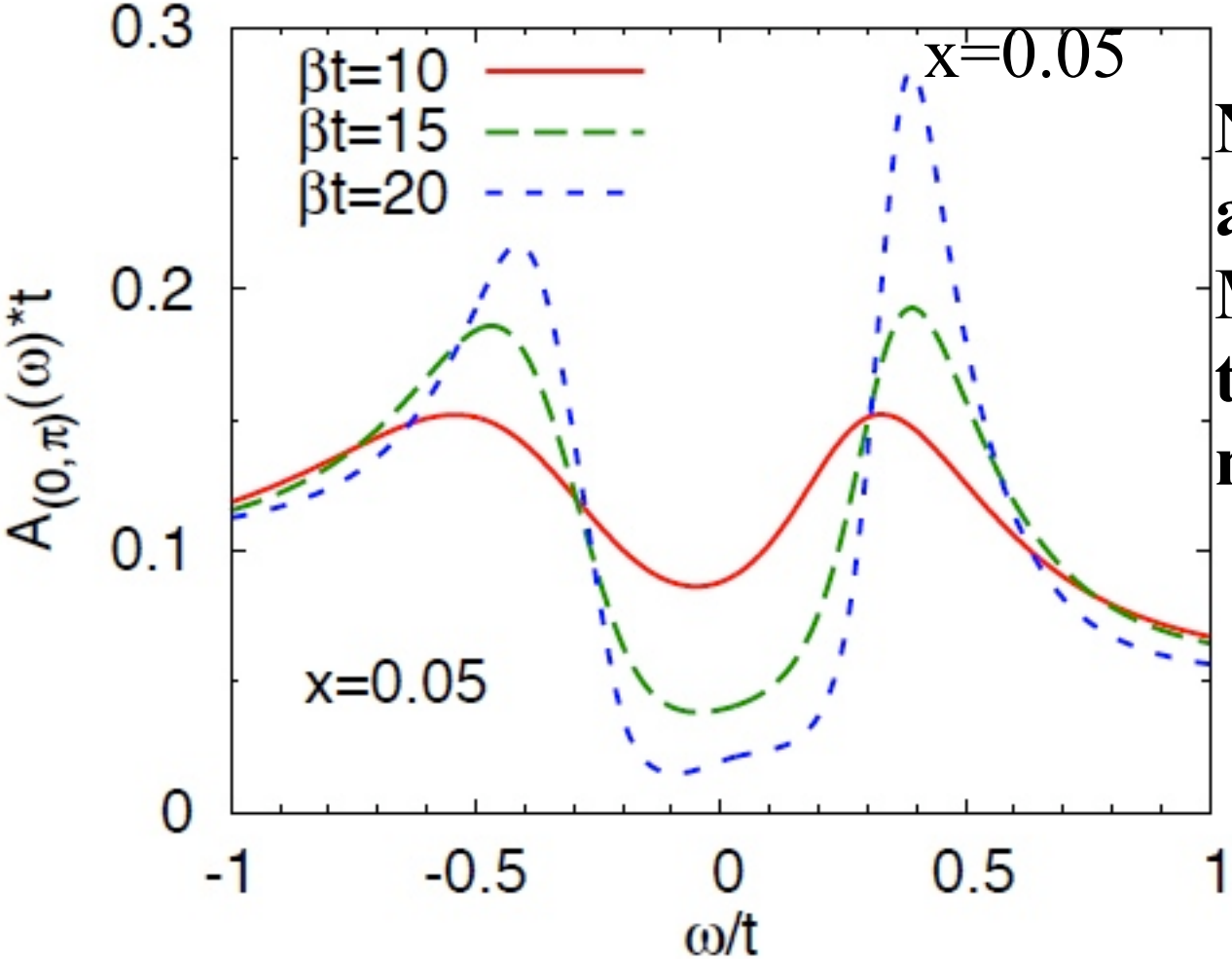
Closer look at the pseudo--or is it real--gap: maximum entropy analytical continuation



Note: gap 'fills in' as T increases. Magnitude (peak to peak distance) not changed much

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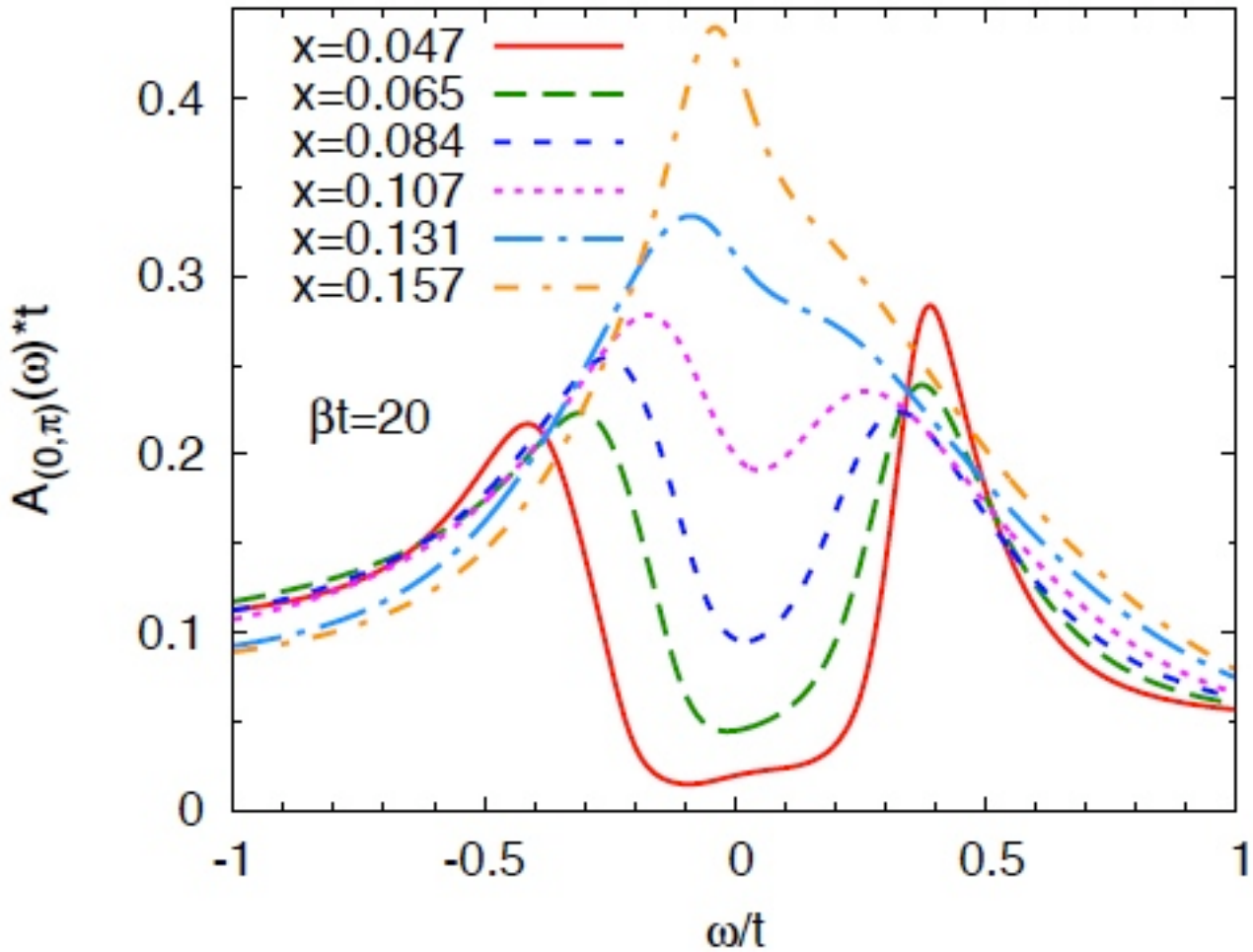
Doping dependence



Gap decreases with increasing doping--but has filled in, not closed at x~0.11 boundary of sector selective phase

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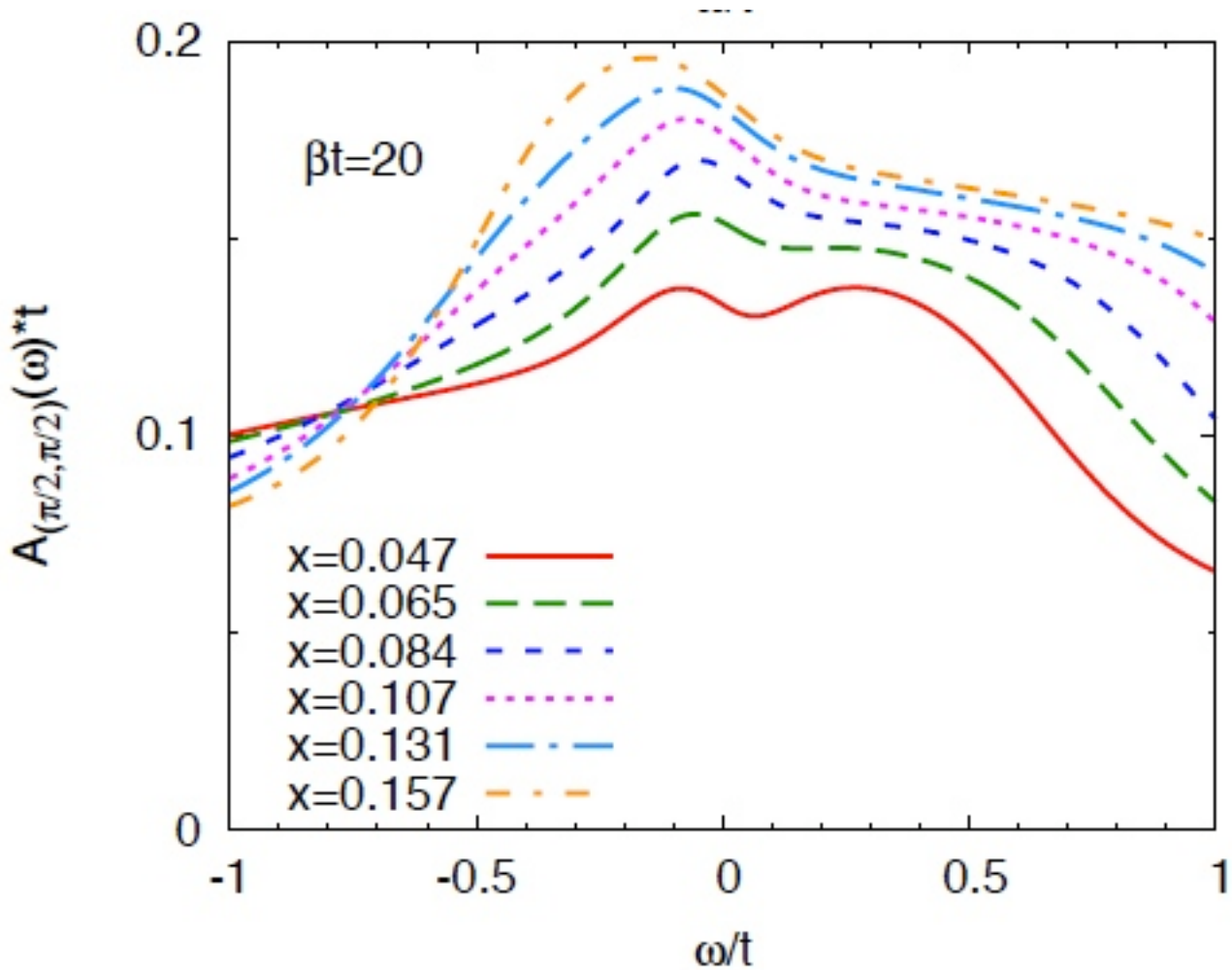
Zone diagonal sector



Possibly hint of gap at lowest dopings, but otherwise no gap

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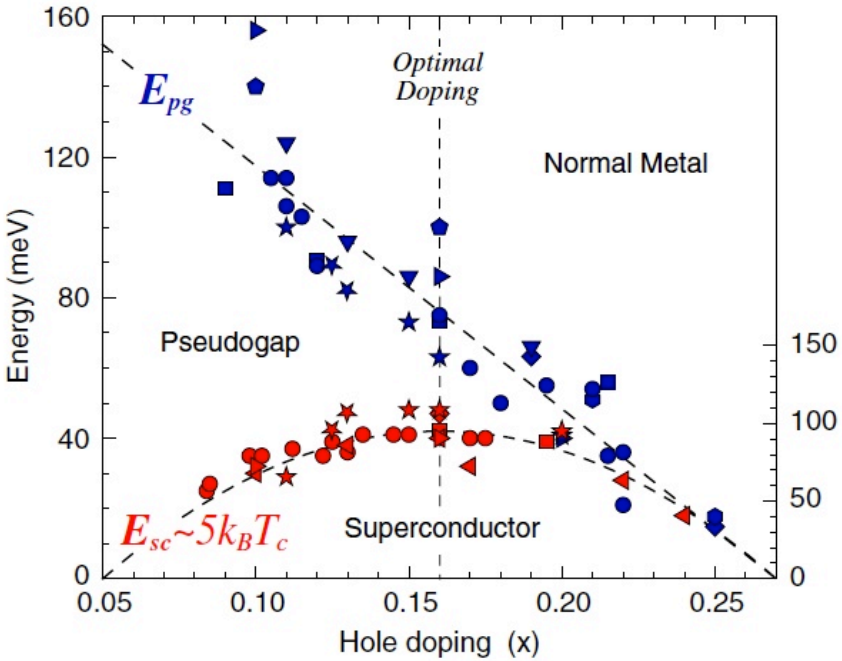
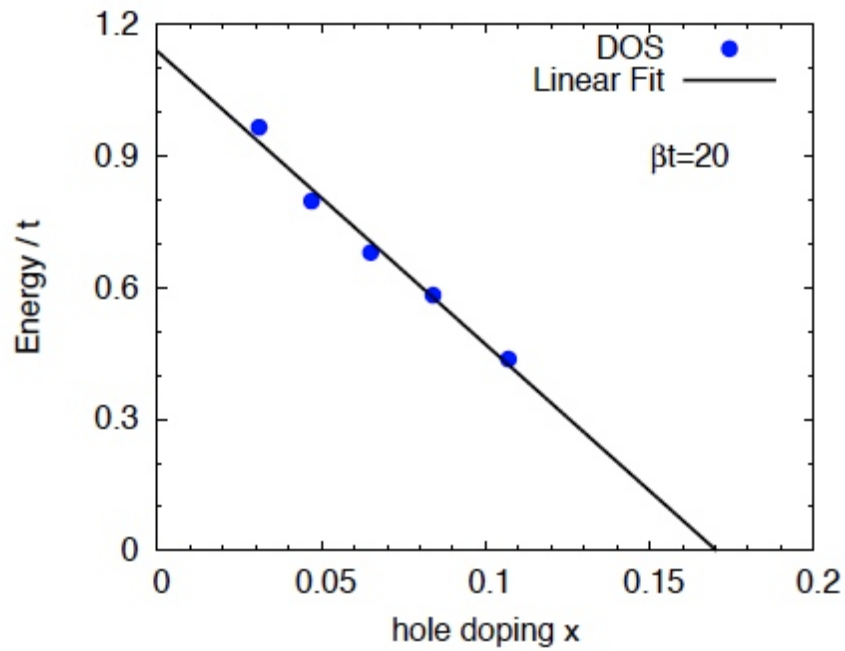
Summary of gap size

Compilation of data



Huefner et al Rep. Prog. Phys. 71 062501 (2008)

Calculation (t=300meV)



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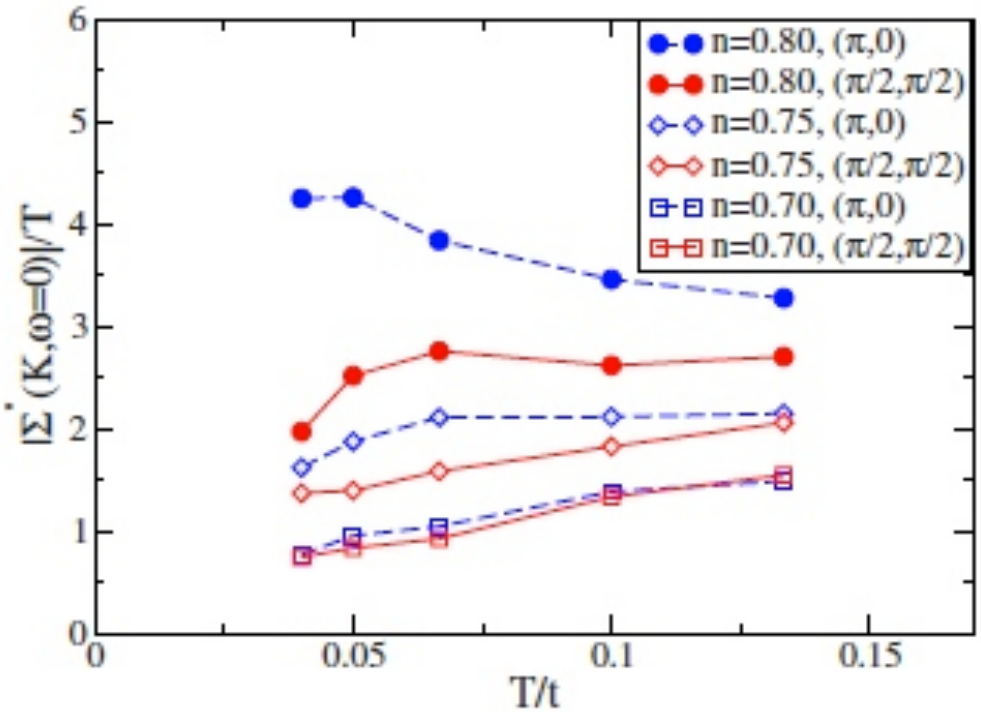
Higher doping: electron scattering rate divided by T



High doping regime of isotropic scattering
Intermediate doping: anisotropy in magnitude, T-dep

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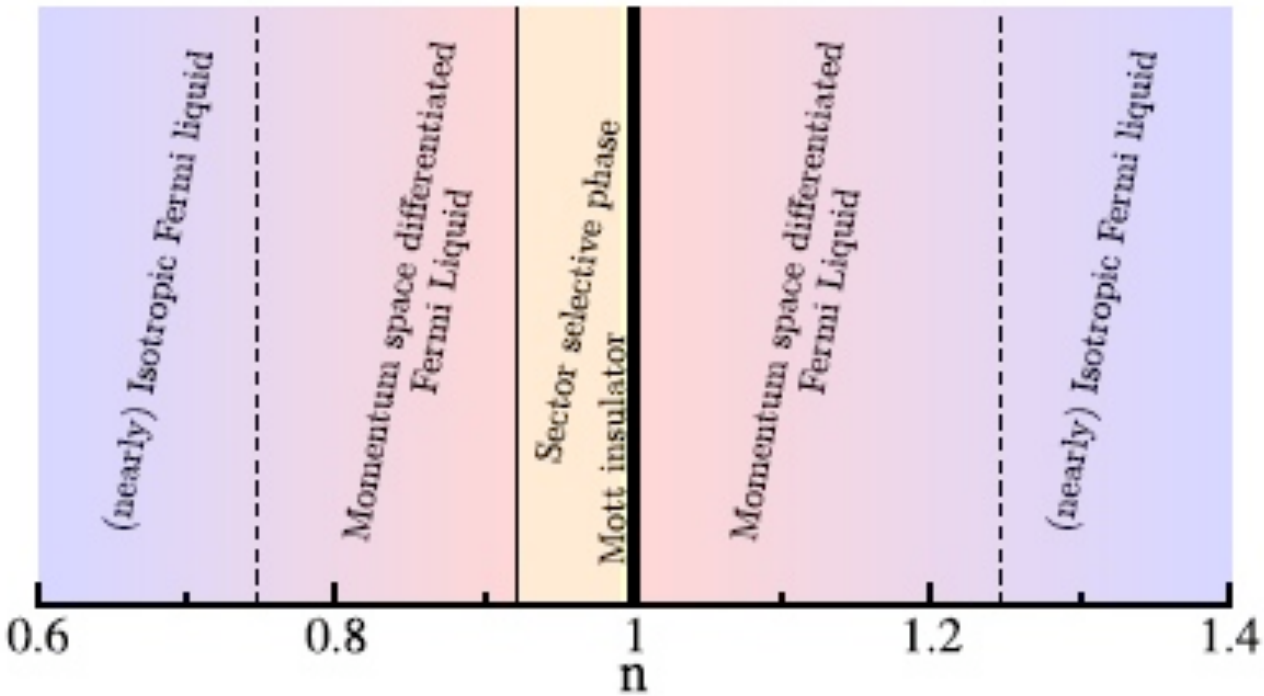
Doping phase diagram
U=7t



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Two-particle response: optical conductivity and raman

Computing: BNL Blue gene; NSERC Cray XMP-6 (via CNMS)

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Theory

Expectation value of current: $\vec{j}(t) = Tr\left[\vec{\mathbf{J}}(A)\mathbf{G}(t;\{\vec{A}\})\right]$

Current operator J and Green function G computed in presence of vector potential A

Hamiltonian: $\hat{H}[\{\mathbf{A}\}] = \hat{T}[\{\mathbf{A}\}] + \hat{U}$

Hopping: depends on A

Interaction: particle positions, not velocities

$$\vec{J} = \frac{\delta \hat{T}}{\delta \vec{A}} \qquad G = \left[i\partial_t - \hat{T}(\vec{A}) - \Sigma(\vec{A})\right]^{-1}$$

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Expanding

Vertex function: $$\mathbf{\Gamma} = \frac{\delta \Sigma}{\delta \vec{A}}$$

=>usual 3 contributions to conductivity

$$\chi_{\text{dia}}(t-t') = \text{Tr}\left[\mathbf{K}\mathbf{G}(t=0)\right]\delta(t-t')$$

$$\chi_{\text{bubble}}(t-t') = \text{Tr}\left[\vec{\mathbf{J}}\mathbf{G}(t-t')\vec{\mathbf{J}}\mathbf{G}(t'-t)\right]$$

$$\chi_{\text{vertex}}(t-t') = \text{Tr}\left[\vec{\mathbf{J}}\mathbf{G}(t-t_1)\vec{\mathbf{\Gamma}}(t_1-t',t'-t_2)\mathbf{G}(t_2-t),\right]$$

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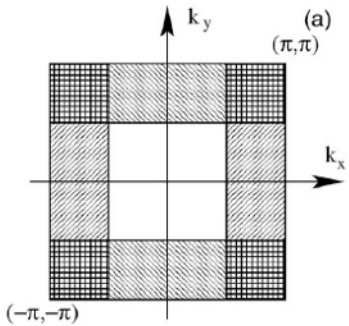
Vertex function: $$\Gamma = \frac{\delta \Sigma}{\delta \vec{A}}$$

In DCA: $$\Sigma(k,\omega) = \sum_{a}^{N} \Sigma_a(\omega)\phi_a(k)$$

=>2 contributions to vertex:

From k dependence (delta functions where patches meet)



From dependence of cluster self energy on A

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Vertex from k-dependence: line where patches "a" and "b" meet

$$\vec{\Gamma}^k(\omega+\Omega,\omega) = \vec{n}^{ab}\left(\Sigma_b(\omega+\Omega) - \Sigma_a(\omega)\right)\delta\left((\vec{k}-\vec{k}^{ab})\cdot\vec{n}^{ab}\right)$$

(Previous literature neglected this)

Vertex from cluster self-energy: compute by linearizing self consistency equation in A

$$\delta\mathcal{G}_\alpha^{-1} - I_\Sigma[\{\delta\Sigma_\alpha\}] = -\vec{I}_v^\alpha\cdot\vec{A}$$

$$\vec{I}_v = -G_a^{-1}\left(\int_a (dk)G(k)\frac{\partial\varepsilon}{\partial\vec{k}}G(k)\right)G_a^{-1}$$

$$I_\Sigma = \delta\Sigma^a + G_a^{-1}\left(\int_\alpha (dk)G(k)\delta\Sigma^\alpha G(k)\right)G_a^{-1}$$

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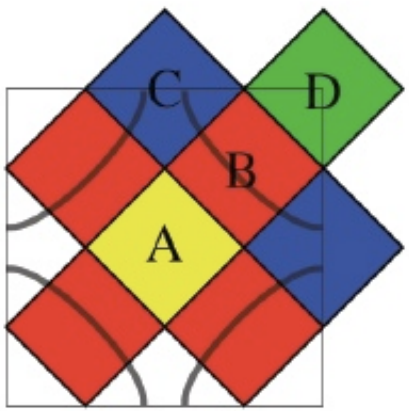
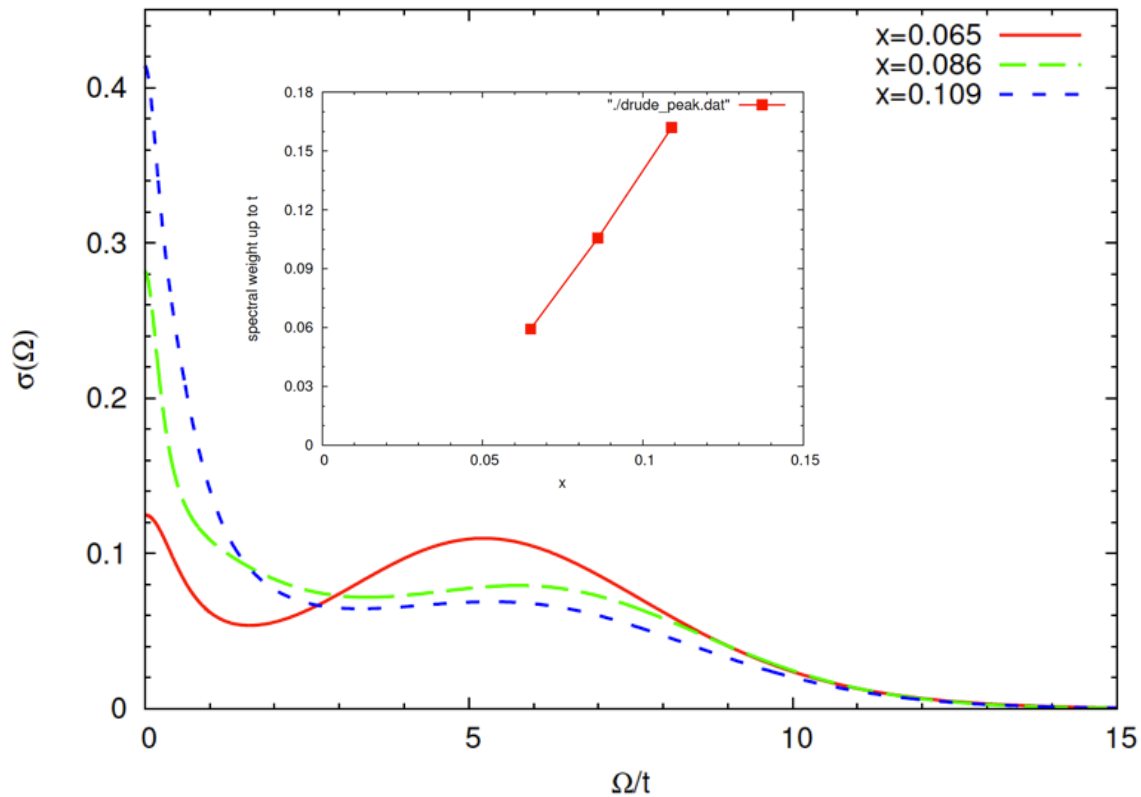
Conductivity: preliminary results 8 site, U=7t, T=t/20 ~200K



N Lin, E. Gull, AJM



Key point: strong suppression of 'Drude' weight

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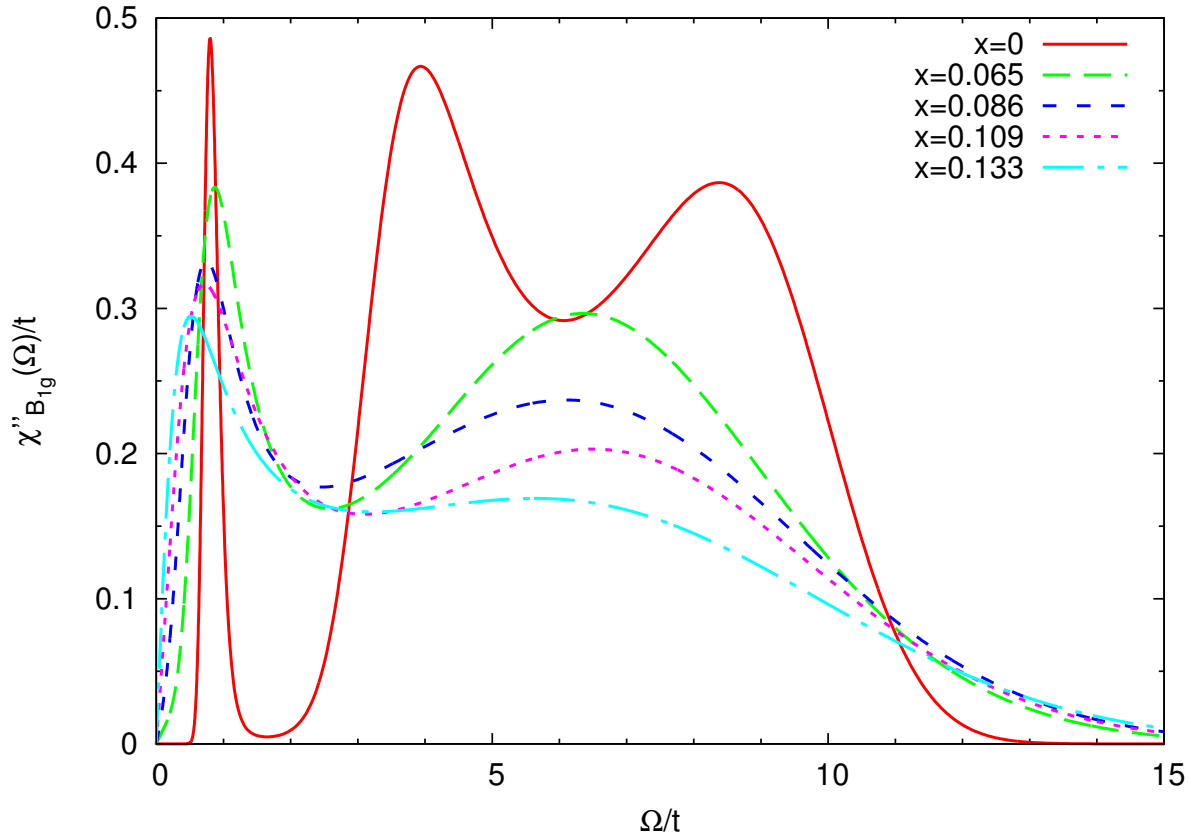
Raman spectra



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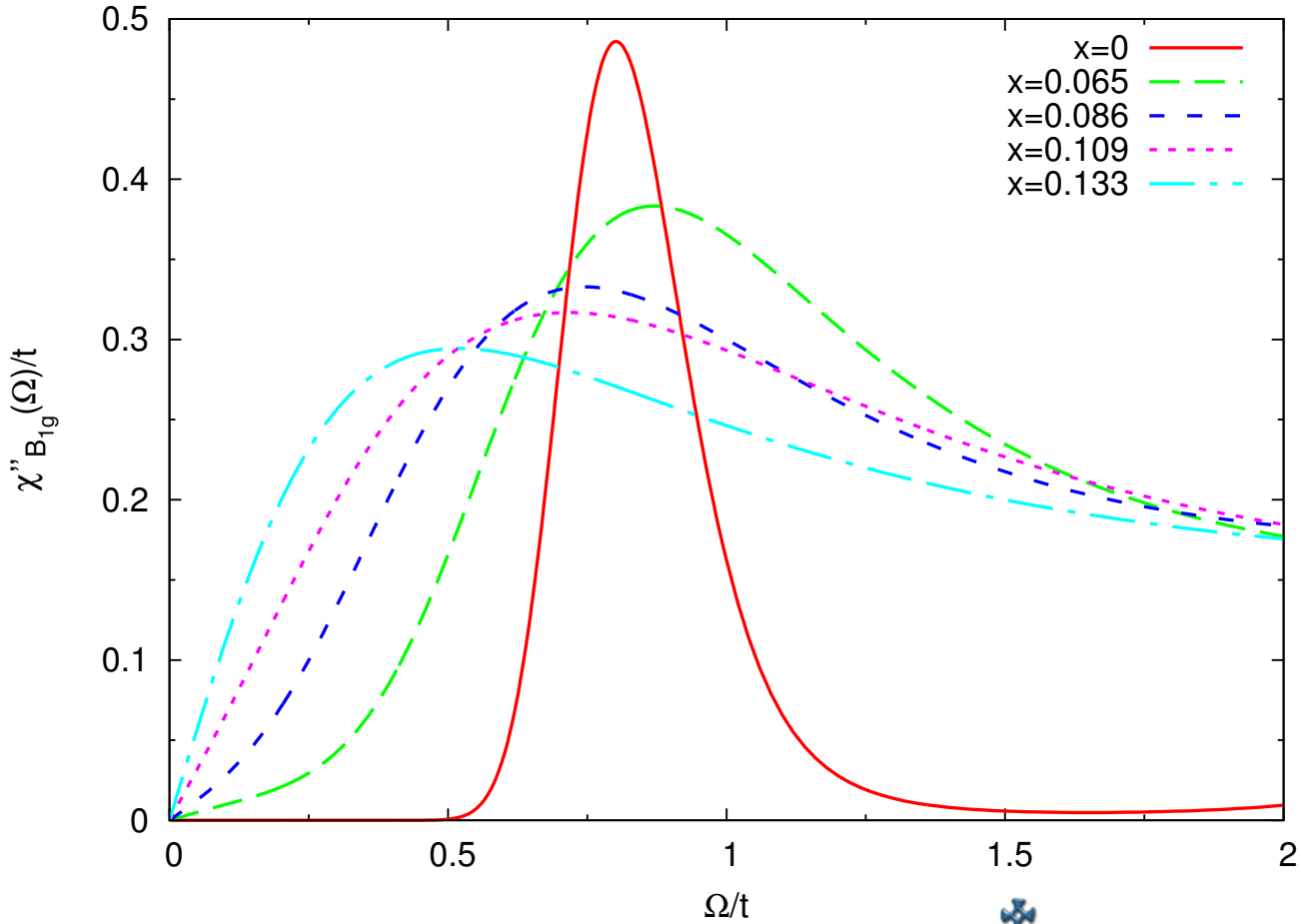
Raman spectra: low frequency regime



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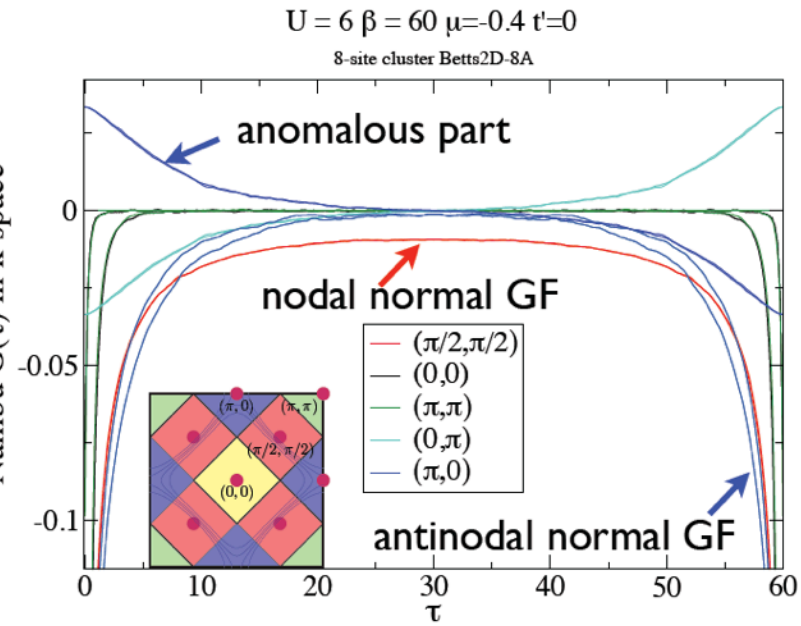
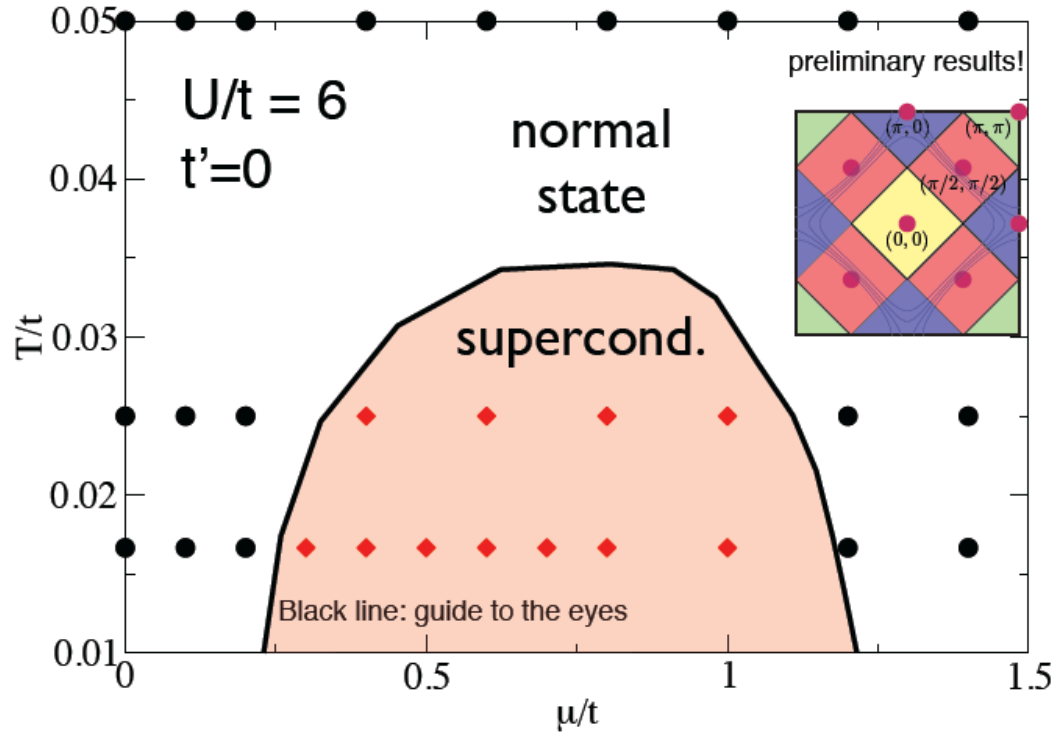
Superconductivity





E. Gull, O. Parcollet, AJM

16 site, larger U now in progress

Next steps (w T. Maier): understand and control pairing interaction, interplay with pseudogap

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Summary: Hubbard model

***Large enough clusters, strong enough interactions=>'real' results**

***Pseudogap <=> sector selective Mott transition. Features very similar to observations**

***Superconductivity (and interplay with pseudogap) now in progress**

***2 particle response functions becoming available**

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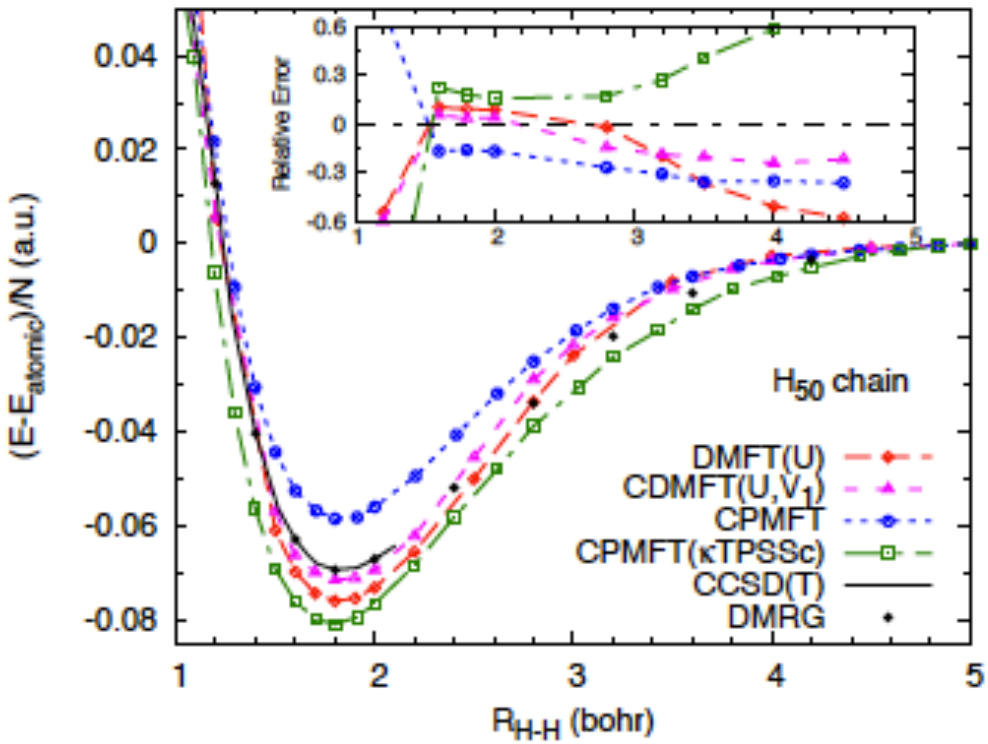
Quantum Chemistry

Quantum chemical test-bed: H_n molecule



Nan Lin, C. Marianetti, A. J. Millis, and D. Reichman, Phys. Rev. Lett. **106**, 096402 (2011).

In process: transition metal dioxide molecules, TM dimers => Understand 'double counting'

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Historical Aside:
Density Functional Theory

Late 1960s, early 1970s (after the Kohn-Sham paper): it became clear that
--there was a formal structure, potentially quite useful
--many questions about reliability of approximations and hence meaning of results
--many difficulties in implementation

Scientific approach: do stuff. improve methods. see what works and what doesnt

Result: broadly useful tool

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Dynamical mean field theory; more broadly,

Is in roughly this place

Formal structure established; convergence understood for model systems.

many applications to model Hamiltonians

'LDA+DMFT' (define model Hamiltonian from underlying band structure calculation) now a working tool (up to 'double counting correction')

(Georges, cond-mat:0403123; K. Held et al, Phys. Status Solidi 243, 2599-2631 (2006); G. Kotliar et al ,Rev. Mod. Phys. 78, 865 (2006))

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Directions:

Longer ranged (density-density) interactions

Cluster calculations for partly-filled d-shells (Slater-Kanamori interactions)

Quantum Chemistry and realistic systems

Big open question: sign problem in realistic situations

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Remark: computational resources

*Simple DMFT: accessible to university group with modest cluster

*DMFT for interesting problems (partially filled d-shells; moderate clusters, some SC): 'capacity computing' such as CNMS provides.

*Technical frontier: vertex functions...leadership-type resources.

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Computing and modern materials theory

Big opportunities--new methods applied to novel materials and nanostructures.

Computational resources needed: not necessarily 'leadership' but beyond what university groups can typically sustain

Need for software and training infrastructure.

Need: 'stewardship role' for materials theory

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Gottfried Wilhelm von

Leibniz' dream...

"If controversies were to arise, there would be no more need of disputation between two philosophers than between two accountants. For it would suffice to take their pencils in their hands, and say to each other, 'Let us calculate.'"

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Now this is not the end (of theory of correlated electron materials). It is not even the beginning of the end. But it is, perhaps, the end of the beginning.

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