On the dual fermion approach to charge order, spin frustration, and transport

NCTS workshop on SCES – Taiwan – ROC – March 2-3, 2016

Stefan Kirchner ⋆ Center for Correlated Matter ⋆ Zhejiang University ⋆ Hangzhou

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Introduction

The observed behavior of strongly correlated systems typically results from a delicate interplay of competing dynamics over regions of phase space:

Kinetic and potential energy in the Hubbard model on the square lattice

Interplay of frustration and interaction in the Hubbard model on a triangular lattice

Studying this interplay when the density matrix is unknown (non-thermal steady states) is particularly challenging

Introduction

Correlation effects in materials are not only interesting from an academic point of view

Correlations in oxides can yield large power factors:

$P = S^2\sigma$ **thermoelectric power factor**

maybe technologically interesting for recovering waste heat

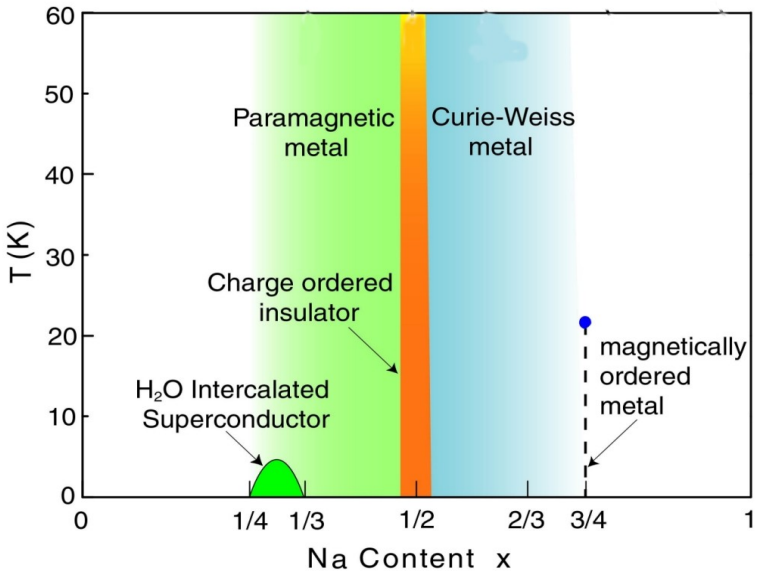
Cobaltates, vanadates, ruthenates, cuprates show large, doping dependent Seebeck coefficient

- an unusually large Seebeck coefficient is found in the cobaltate $\mathrm{Na}_x\mathrm{CoO}_2$ around x=0.5
- the system appears metallic in charge response but insulating in the spin response: „Curie-Weiss metal“
- sodium cobaltate turns superconducting at x≈1/4
- sodium cobaltate is a triangular system → frustration effects are pertinent

Foo et al., PRL (2004), Lee et al., Nature Materials (2006)

Introduction

Sodium cobaltate: $\mathrm{Na}_x\mathrm{CoO}_2$



Triangular system → frustration effects pertinent

System appears metallic in charge response but insulating in its spin response

Unusually large Seebeck coefficient is found near x=0.5

Lee et al., Nature Materials (2006)

Frustrated interactions commonly occur in real materials

in κ-$(\mathrm{BEDT\text{-}TTF})_2\mathrm{Cu}_2(\mathrm{CN})_3$ e.g. it may create a spin-liquid state

Can we calculate the thermopower of a generic strongly correlated electron system possessing frustrated interactions?

Outline

- **Introduction**
- **Dual Fermions**
- **Triangular Hubbard model**
- **Falicov-Kimball model and the two-dimensional Ising lattice**
- **Discussion, Outlook, & Summary**

References:

„*Universal out-of-equilibrium transport in Kondo-correlated quantum dots: renormalized dual fermions on the Keldysh contour*“,
Enrique Muňoz, C. Bolech, & Stefan Kirchner,
PRL (2013) **110**, 016601

„*Critical Exponent of Strongly Correlated Fermion Systems from Diagrammatic Multi-Scale Methods*“
Andrey E. Antipov, Emanuel Gull, & Stefan Kirchner,
PRL (2014) **112**, 226401

„*Competing phases of the Hubbard model on a triangular lattice – insights from entropy*“
Gang Li, Andrey E. Antipov, Alexey N. Rubtsov, Stefan Kirchner, & Werner Hanke,
PRB (2014) **89**, 161118(R)

Acknowledgment

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Gang Li & Werner Hanke	**Würzburg University**
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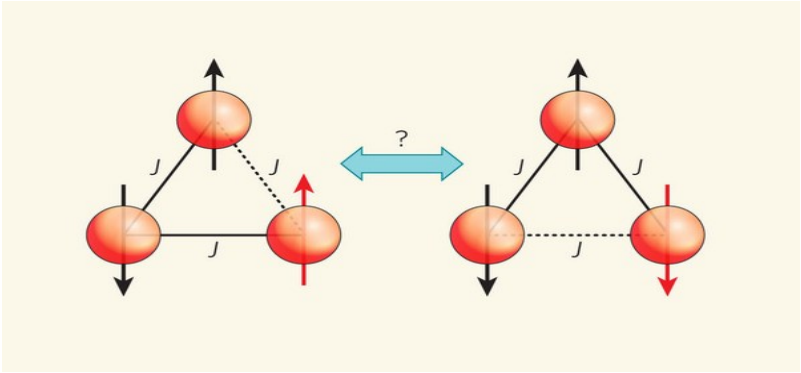
Thermopower of Sodium Cobaltate

Geometric frustration:



large spin-entropy

$$S_{\mathrm{Kelvin}} = \frac{k_B}{e}\frac{\partial S}{\partial n}\Big|_{T,U}$$

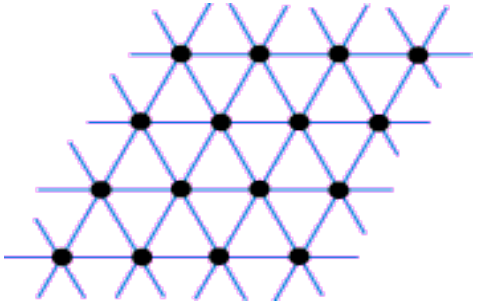
(thermopower via Kelvin formula)

(static limit is taken before the system is uniform)

Effective microscopic model for sodium cobaltate:

Hubbard model on the triangular lattice

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



(1) Solve the strong correlation problem: DMFT

(2) Include the non-local physics in a 2nd step via a dual fermion extension

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Dynamical Mean Field Theory

- **standard approach to strongly correlated lattice fermions**
- reminiscent of Weiss mean field for the Ising model:

$$h_{\mathrm{MF}} = I_{ex} \sum_{<i,j>} \langle S^z \rangle$$

$$(S_i^z - \langle S^z \rangle)(S_j^z - \langle S^z \rangle) \stackrel{!}{=} 0$$

$$H_{\mathrm{Ising}} = -\frac{I_{ex}}{2} \sum_{<i,j>} S_i^z S_j^z \quad \Longrightarrow \quad H_{\mathrm{Ising}}^{\mathrm{MF}} = -h_{\mathrm{MF}} \sum_i S_i^z$$

The interacting spin problem is replaced by a single spin in a self-consistent mean field

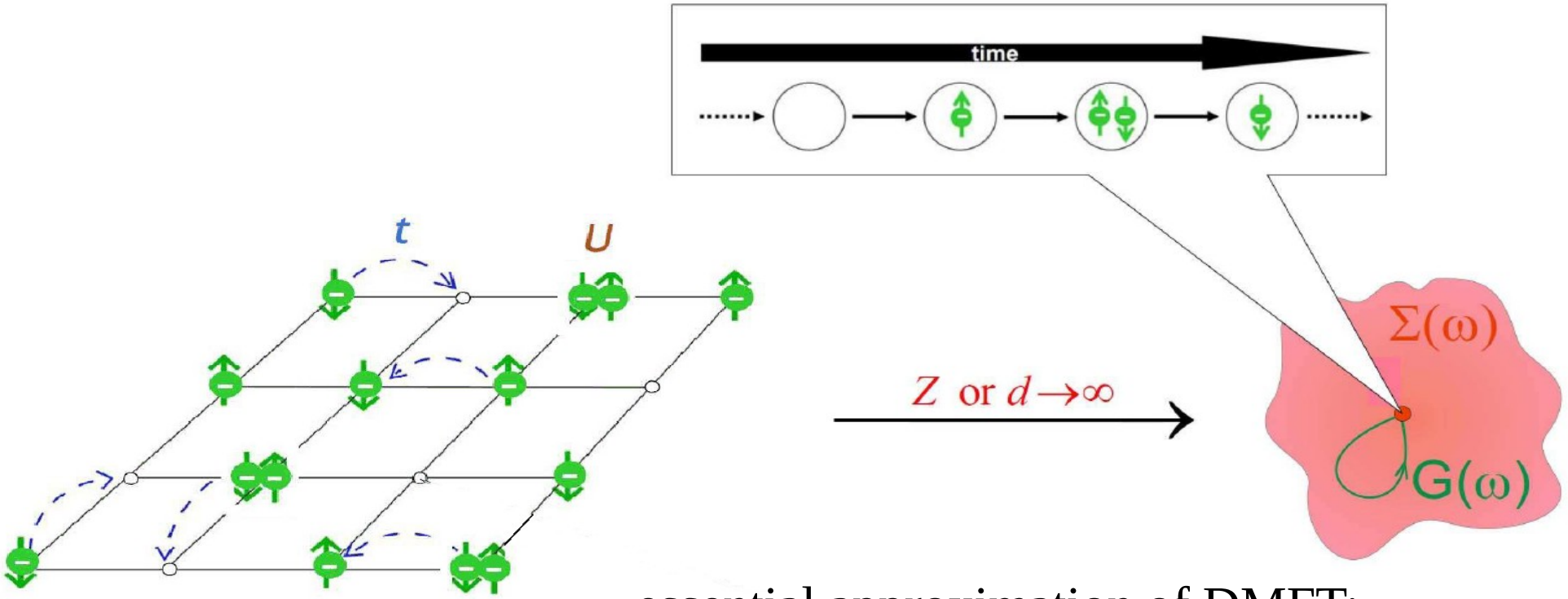
spatial fluctuations are neglected:

- solution in D=2 is very different from the Onsager solution
- becomes exact in the limit of infinitely many nearest neighbors (D=∞)

Dynamical Mean Field Theory

[Metzner,Vollhardt (1989), A. Georges et al. (1996)]

lattice problem is replaced by a self-consistent quantum impurity problem



essential approximation of DMFT:

self-energy becomes a local, k-independent quantity:

$$\Sigma(k, i\omega_n) \stackrel{!}{=} \Sigma(i\omega_n)$$

⟹ exact in the limit of infinite coordination

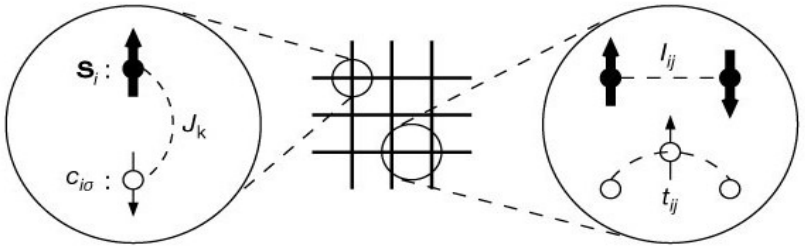
Dynamical Mean Field Theory

neglect of k-dependence in Σ leads to effective quantum impurity problem:

- captures Mott-Hubbard transition but insulating state has entropy issue
- description of e.g. anti-ferromagnetism and superconductivity are problematic
- interplay of correlation and frustration is not captured

DMFT extensions:

- extended DMFT (EDMFT) [Qimiao Si et al.]



dynamic competition of Kondo and RKKY interaction

- Cluster approaches (CDMFT, DCA)

  [Lichtenstein & Katnelson, Kotliar et al.]
  [Hettler et al.]

  self-energy at discrete k-point
  may break translational invariance

- Dynamical vertex approximation (DΓA)

  [Held et al.]

Dual Fermion Extension

perturbative expansion around the DMFT solution

[Rubtsov, Katnelson, Lichtenstein, 2008]

Hubbard model:

$$H = \sum_{R_i,R_j} \sum_{\sigma} t_{i,j} c^{\dagger}_{i,\sigma} c_{j,\sigma} + U \sum_{R_i} n_{i,\uparrow} n_{i,\downarrow} \quad \rightarrow \quad Z = \int \mathcal{D}[\hat{\psi}^{\dagger}, \hat{\psi}] e^{iS[\hat{\psi}^{\dagger}, \hat{\psi}]}$$

single-particle part $h_{\vec{k}}$

exact lattice action:

$$S[\hat{\Psi}^{\dagger}, \hat{\Psi}] = \sum_{i} S_{\mathrm{imp}}[\hat{\Psi}^{\dagger}_{i}, \hat{\Psi}_{i}] - \sum_{\omega,\vec{k}} \hat{\Psi}^{\dagger}_{\omega,\vec{k}} (\Delta(\omega) - h_{\vec{k}}) \hat{\Psi}_{\omega,\vec{k}}$$

hybridization function Δ(ω)

impurity action at each lattice side with arbitrary hybridization function

$$\Delta(\omega) = \sum_{k} \frac{V_k V_k^*}{\omega - \epsilon_k}$$

couples „impurities“ – spatial correlations enter here;
setting up a PT around S_{imp} non-trivial since S_{imp} is non-Gaussian

Dual Fermion Extension

[Rubtsov, Katnelson, Lichtenstein, 2008]

$$S[\hat{\Psi}^\dagger, \hat{\Psi}] = \sum_i S_{\mathrm{imp}}[\hat{\Psi}_i^\dagger, \hat{\Psi}_i] - \underbrace{\sum_{\omega,\vec{k}} \hat{\Psi}^\dagger_{\omega,\vec{k}} (\Delta(\omega) - h_{\vec{k}}) \hat{\Psi}_{\omega,\vec{k}}}_{\textbf{perturbation}}$$

Rewrite perturbation via fermionic Hubbard-Stratonovich identity
This brings in new fermionic degrees of freedom: „dual fermions“

$$S_{\mathrm{dual}}[\hat{f}^\dagger, \hat{f}] = -\sum_{\omega,\vec{k}} \hat{f}^\dagger_{\omega,k} G_f^{-1} \hat{f}_{\omega,k} + \sum_i V[\hat{f}_i^\dagger, \hat{f}_i]$$

V depends on all n-point reducible impurity vertices

$$\Longrightarrow \mathbf{G}_f(\omega,\vec{k}) = \mathbf{G}_f(\omega,\vec{k})^{(0)} + \mathbf{G}_f(\omega,\vec{k})^{(0)} \mathbf{\Sigma}_f(\omega,\vec{k}) \mathbf{G}_f(\omega,\vec{k})$$

bare dual fermion Green's function:

$$G_f(\omega,\vec{k})^{(0)} = -g_\omega \left[g_\omega + (\Delta(\omega) + h_{\vec{k}})^{-1} \right]^{-1} g_\omega$$

Dual Fermion Extension

$$S_{\mathrm{dual}}[\hat{f}^\dagger, \hat{f}] = -\sum_{\omega,\vec{k}} \hat{f}^\dagger_{\omega,k} G_f^{-1} \hat{f}_{\omega,k} + \sum_i V[\hat{f}_i^\dagger, \hat{f}_i]$$

Dual fermions give a formal expansion around the DMFT solution (= the reference system) in terms of the n-point reducible vertices of the reference system!

- Reminiscent of a Taylor expansion
- Self-energy is a continuous function of **k**
- in practice, it is impossible to include terms beyond the 4-point vertex:

$$V[\hat{f}^\dagger, \hat{f}] = \frac{1}{4}\gamma^{(4)}_{1234}\hat{f}^\dagger_1 \hat{f}^\dagger_2 \hat{f}_4 \hat{f}_3$$

- in fact, it is even impossible to include all self-energy diagrams generated by the 4-point vertex!

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Triangular Hubbard Model

existing studies are either limited to half-filling

e.g. L. Tocchio et al. (2013)

or zero-temperature,

e.g. J. Merino et al. (2006)

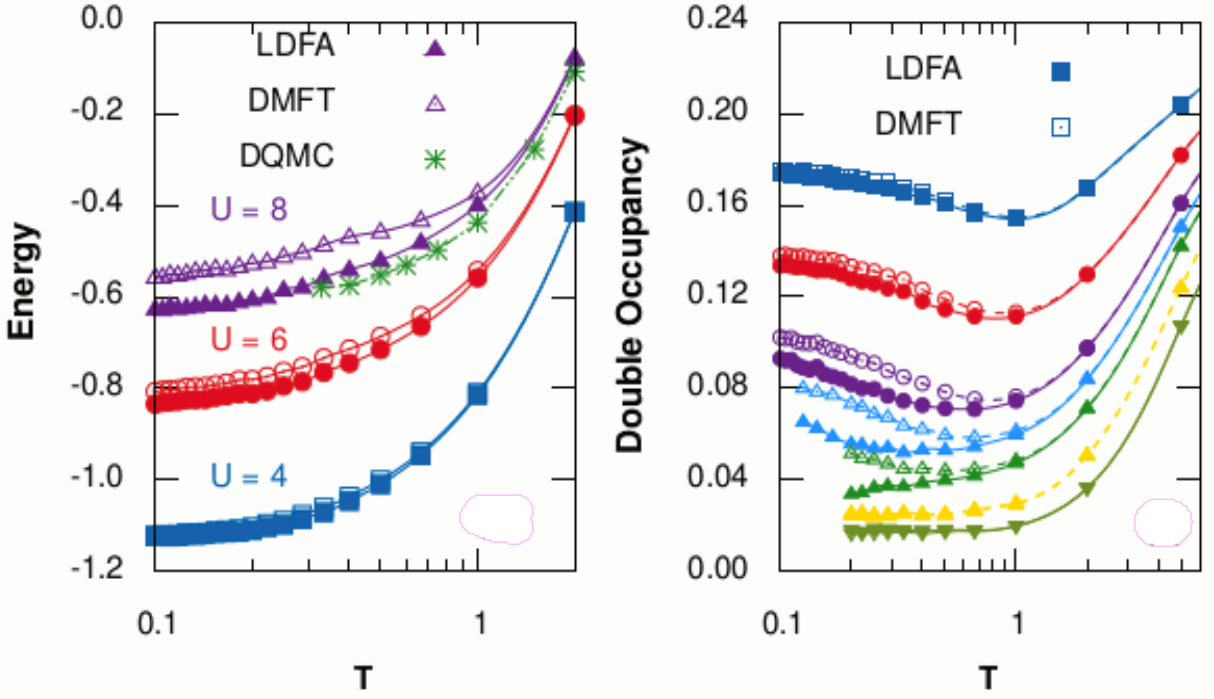
or are finite-cluster studies

e.g. K. Aryanpour et al. (2006)

(favor SL phase, see honeycomb lattice; variational approaches favor ordered phases)

here:

- no finite-size clusters
- no minus-sign problem away from half-filling
- spin- and charge excitations accessible



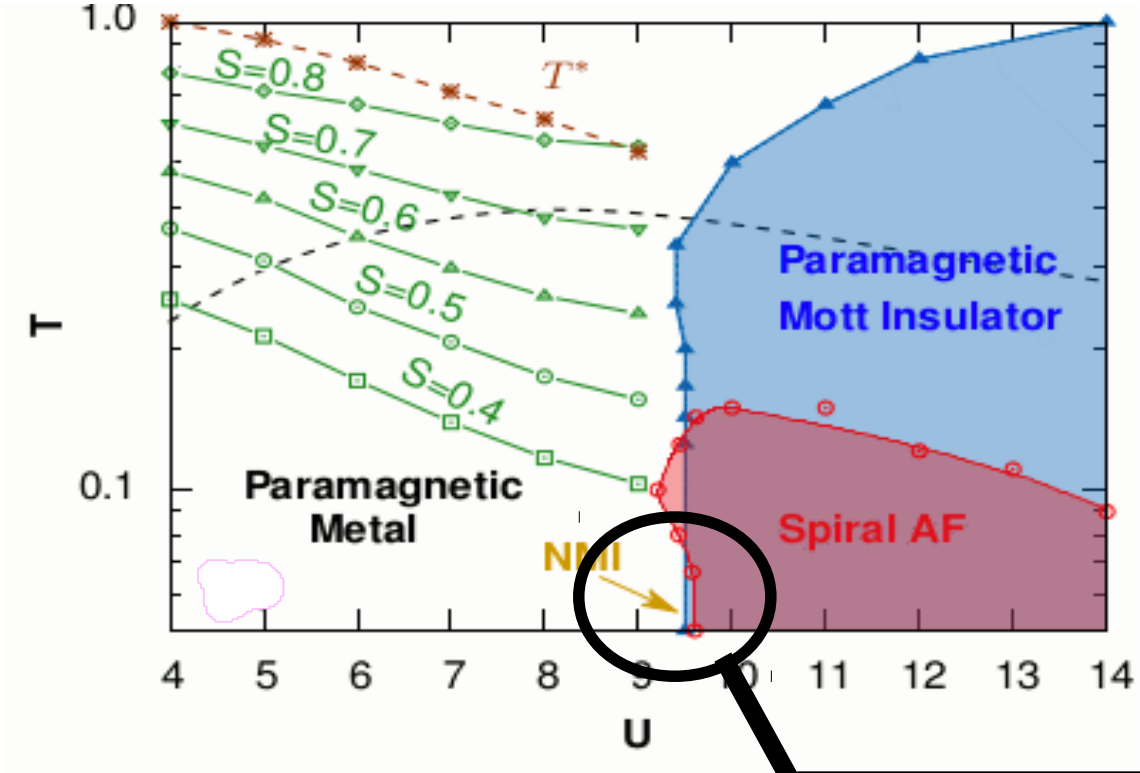
LDFA results are always lower in energy than DMFT results
and agree well finite-cluster QMC results of K. Aryanpour (2006)

G. Li et al.,PRB (2014) **89**, 161118(R)

Triangular Hubbard Model

T-U Phase diagram at half-filling

effect of frustration: spiral AF only above U/t=9.55
square lattice: AF order at any non-vanishing U/t (dashed line: phase boundary)



T*-line marks minimum of $\frac{\partial D}{\partial T}$

entropy is obtained from total energy by integration and

$$S(T=0,n) = -n\ln(n/2) - (2-n)\ln(1-n/2)$$

Candidate for the spin-liquid state: non-magnetic Mott insulator for 9.4<U/T<9.55

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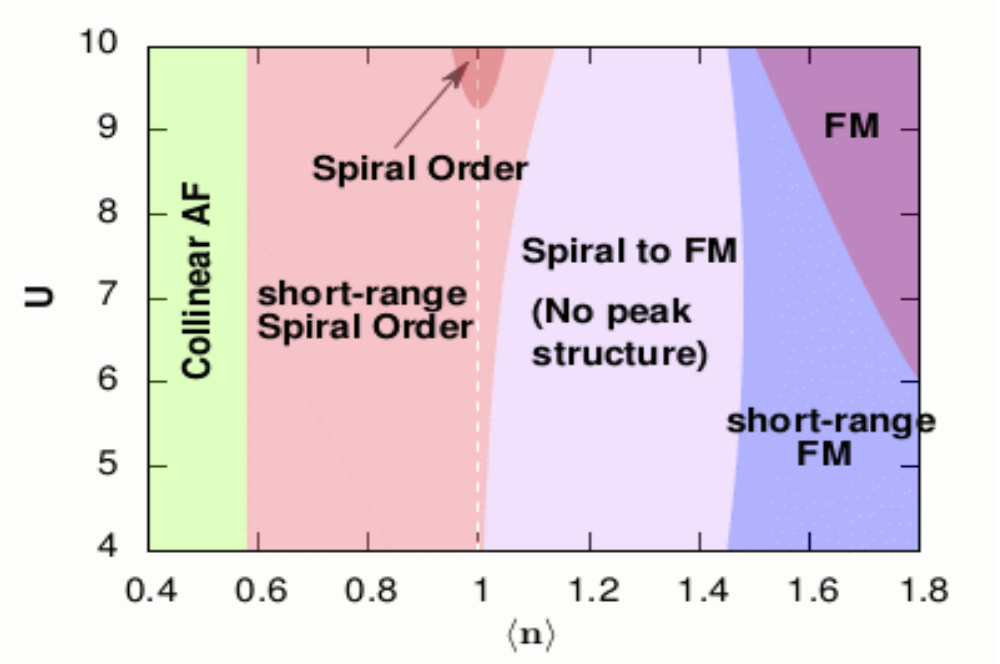
Triangular Hubbard Model

Magnetic phase diagram of the doped model at T/t=0.1

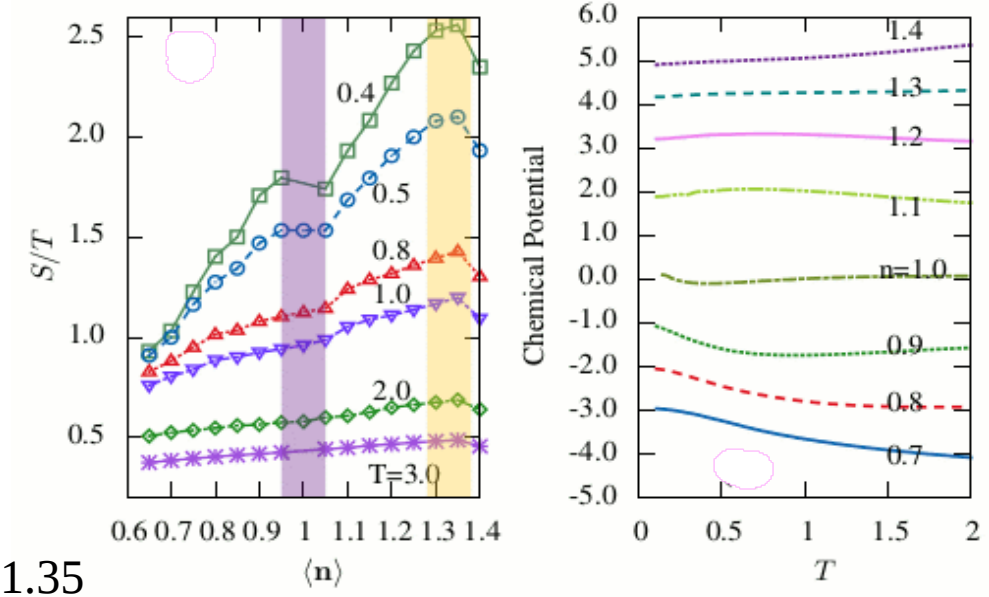


Thermopower via Kelvin formula:

$$\mathbf{S}_{\mathrm{Kelvin}} = \frac{\mathbf{k_B}}{\mathbf{e}} \frac{\partial \mathbf{S}}{\partial \mathbf{n}}\bigg|_{\mathbf{T},\mathbf{U}}$$

Maxwell relation:

$$\frac{\partial \mathbf{S}}{\partial \mathbf{n}}\bigg|_{\mathbf{T},\mathbf{U}} = -\frac{\partial \mu}{\partial \mathbf{T}}\bigg|_{\mathbf{n},\mathbf{U}}$$



- large, negative S at n≈1.5 and a sign change at n≈1.35
- reminiscent of what has been measured for Na_xCoO_2 (Lee et al. Nature Materials (2006))

G. Li et al.,PRB (2014) **89**, 161118(R)

Dual Fermion Extension

How well does the dual fermion method describe phase changes?

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Hubbard Model

$$\mathbf{H} = \sum_{<\mathbf{i},\mathbf{j}>,\sigma} \mathbf{t_{i,j}} \mathbf{c}^{\dagger}_{\mathbf{i},\sigma} \mathbf{c}_{\mathbf{j},\sigma} + \mathbf{U} \sum_{\mathbf{i}} \mathbf{n}_{\mathbf{i},\uparrow} \mathbf{n}_{\mathbf{i},\downarrow}$$

At half-filling and for $U \to \infty$:

model undergoes a phase transition for **d>2** with an O(3) order parameter

$$\chi(T) \sim (T - T_c)^{-\gamma}$$

$$\xi \sim (T - T_c)^{-\nu}$$

$$\chi(r, T = T_c) \sim |r|^{-(d-2+\eta)}$$

η characterizes the deviation from „trivial“ Gaussian behavior.

For the Hubbard model in d=3: $\eta_{\mathrm{Hubbard}} \approx 0$

Falicov-Kimball Model

Hubbard model:

$$\mathbf{H} = \sum_{<\mathbf{i},\mathbf{j}>,\sigma} \mathbf{t_{i,j}} \mathbf{c}^{\dagger}_{\mathbf{i},\sigma} \mathbf{c}_{\mathbf{j},\sigma} + \mathbf{U} \sum_{\mathbf{i}} \mathbf{n}_{\mathbf{i},\uparrow} \mathbf{n}_{\mathbf{i},\downarrow}$$

simplification: $c = c_{\uparrow}; \quad f = c_{\downarrow}$ **and localize all w1 f-states**

Falicov-Kimball model

$$\mathbf{H} = \sum_{<\mathbf{i},\mathbf{j}>} \mathbf{t_{i,j}} \mathbf{c}^{\dagger}_{\mathbf{i}} \mathbf{c}_{\mathbf{j}} + \sum_{\mathbf{i}} (\mathbf{E_i} - \mu_{\mathbf{f}}) \mathbf{f}^{\dagger}_{\mathbf{i}} \mathbf{f}_{\mathbf{i}} + \mathbf{U} \sum_{\mathbf{i}} \mathbf{c}^{\dagger}_{\mathbf{i}} \mathbf{c}_{\mathbf{i}} \mathbf{f}^{\dagger}_{\mathbf{i}} \mathbf{f}_{\mathbf{i}}$$

itinerant up-spin electrons scatter on fixed down-spin electrons

FK model frequently employed for
liquid-solid transitions and binary alloys

Falicov-Kimball Model

at each lattice site, $\langle n_f \rangle = w = 0$ or 1

as a result, the model is exactly solvable at the DMFT level

$$Z = Z^{w=1} + Z^{w=0}; \langle w \rangle = Z^{w=1}/Z$$

$$\langle \mathcal{O} \rangle = \langle w \rangle \mathcal{O}^{w=1} + (1 - \langle w \rangle)\mathcal{O}^{w=0}$$

and the reducible (DMFT) 4-point vertex can be constructed directly

$$\gamma_{\mathbf{\Omega}}^{\mathrm{DMFT}}(\omega) = -\frac{\mathbf{1}}{\mathbf{T}} \frac{\mathbf{\Sigma}_\omega - \mathbf{\Sigma}_{\omega+\mathbf{\Omega}}}{\mathbf{g}_\omega - \mathbf{g}_{\omega+\mathbf{\Omega}}}$$

[Brandt & Mielsch, 1989]
[Freericks & Zlatic 2003]

In any dimension (and at half-filling):

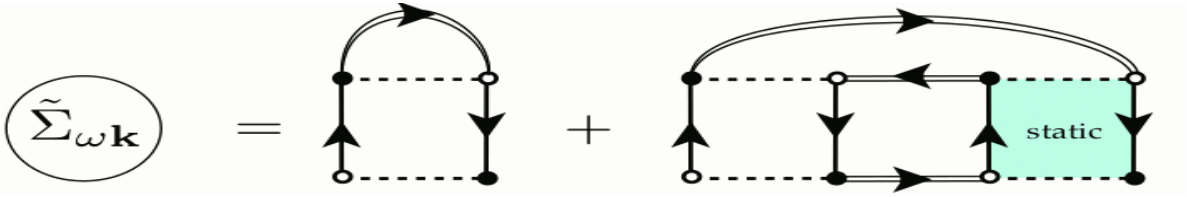
For U/t → ∞ the model is equivalent to the Ising model with $J=t^2/(4U)$

Phase transition with known non-mean field exponents that depend on dimensionality in D=2, 3 and approach mean-field behavior in D=4!

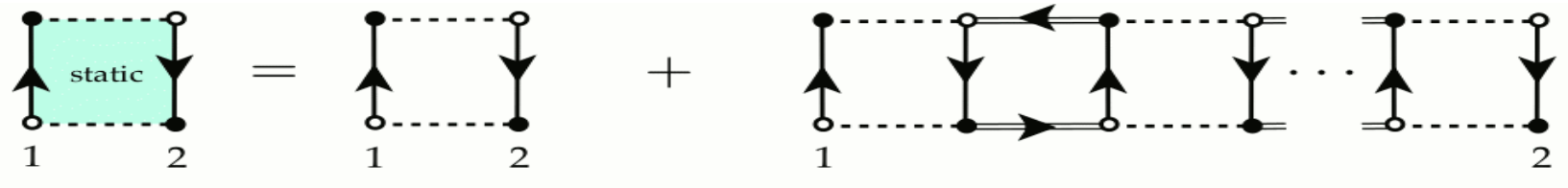
$$\eta_{\mathbf{FK}}^{\mathbf{d=2}} = 1/4$$

Falicov-Kimball Model

The LDFA approximation (static ladder diverges at the phase transition):



Bethe-Salpeter equation:

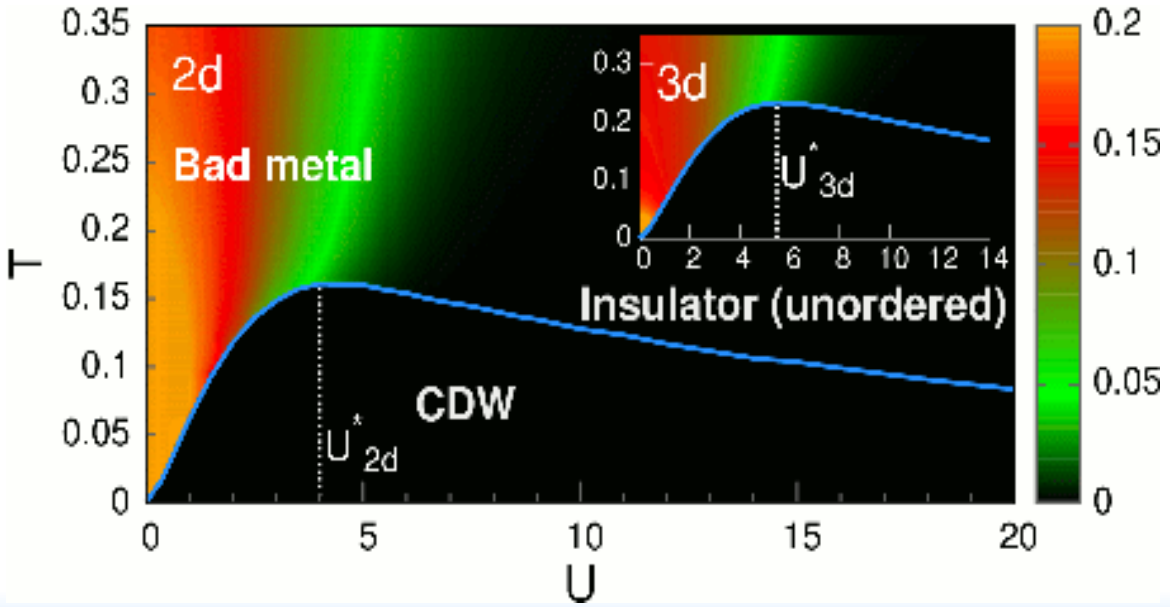


$$G_{\omega,\mathbf{k}} = (\Delta_w - \epsilon_{\mathbf{k}})^{-1} + g_\omega^{-2}(\Delta_w - \epsilon_{\mathbf{k}})^{-2}\tilde{G}_{\omega\mathbf{k}}$$

Estimate of the DOS at the Fermi energy:

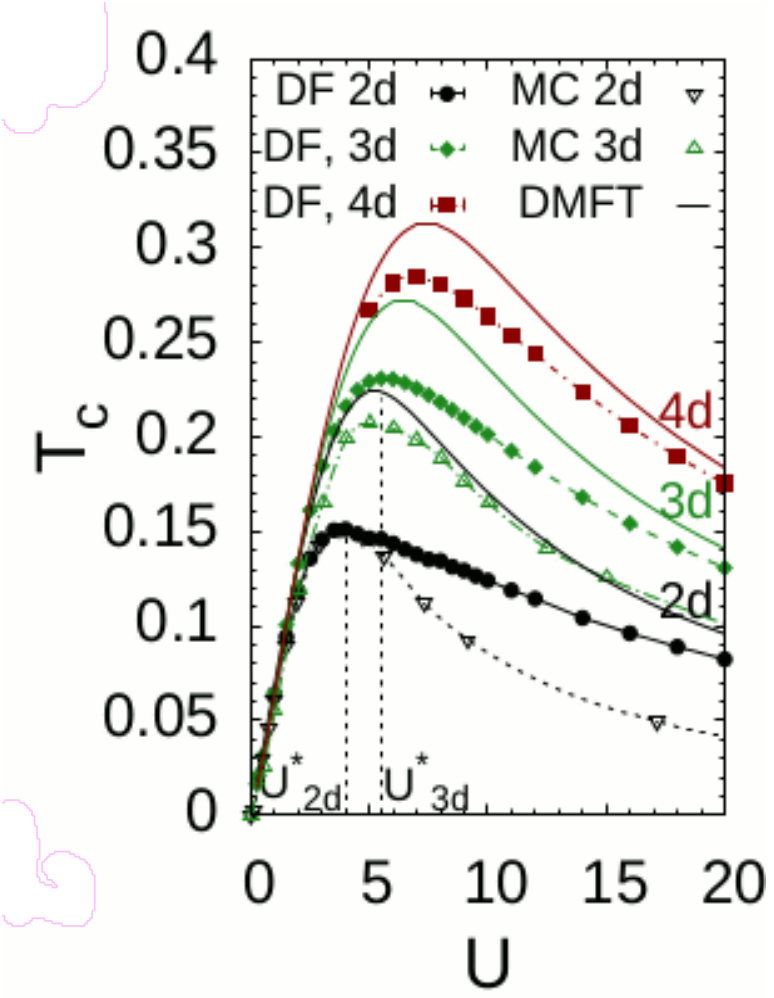
$$A(0) \approx -\beta G_{\mathrm{loc}}(\tau = \beta/2)/\pi$$

Solid blue line:
$T_c(U)$ of transition into checkerboard order



Falicov-Kimball Model

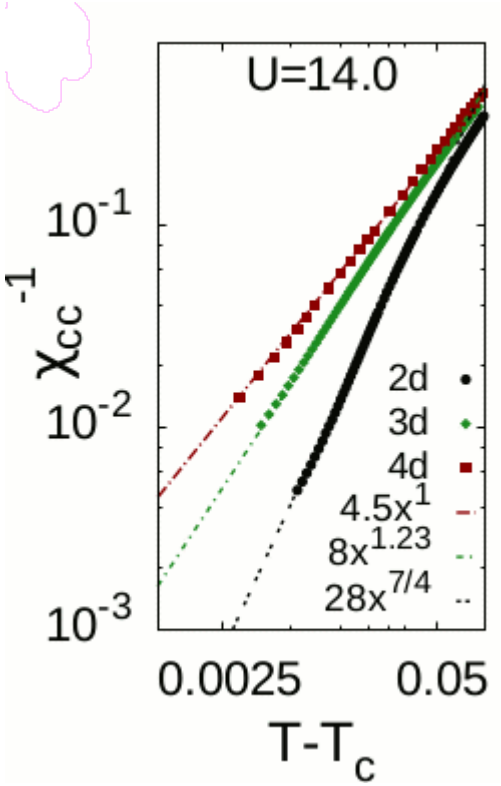
critical temperature $T_c(U)$ obtained from DMFT, LDFA, & Monte Carlo in D=two, threee, and four dimensions:



MC in 2D from Maśka et al. (2006)

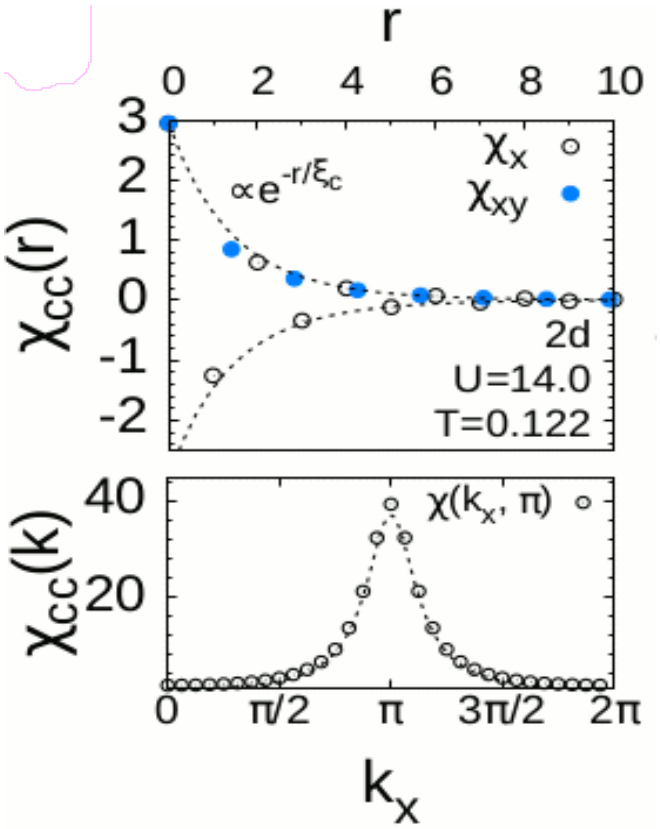
MC in 3D from Žonda et al. (2009)

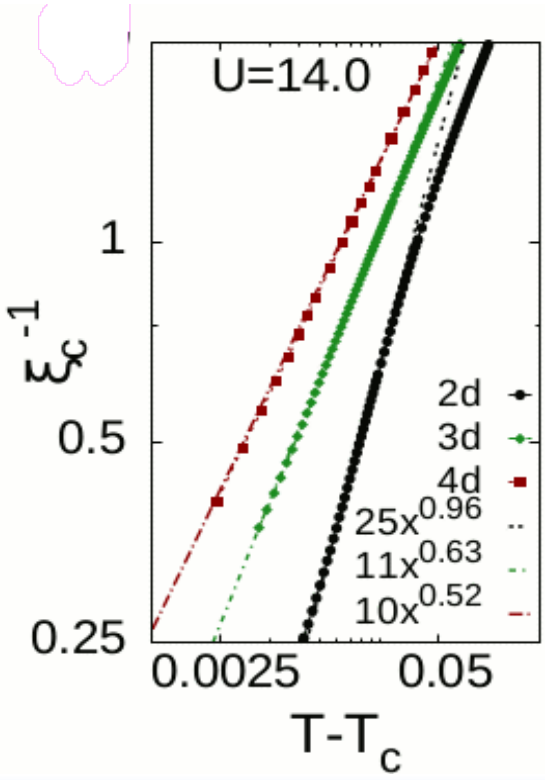
Falicov-Kimball Model



static c-electron charge susceptibility diverges as $T \rightarrow T_c$

static charge susceptibility decays exponentially as one goes away from the ordering wave vector (π,π)

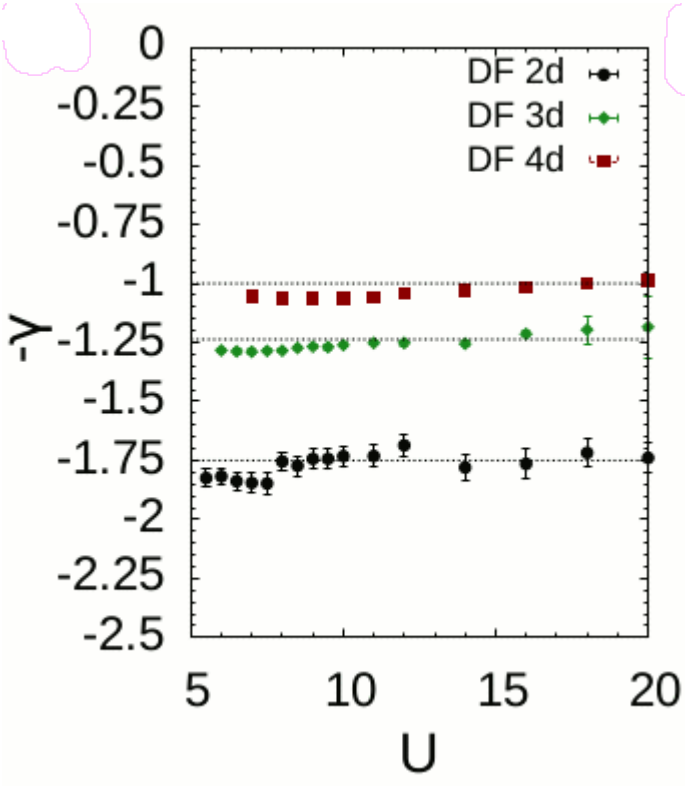


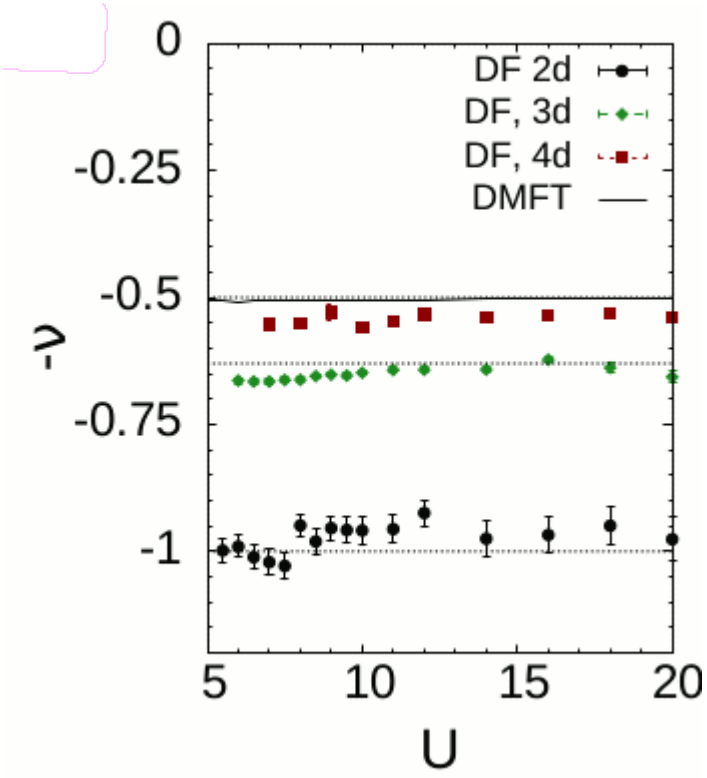


correlation length diverges in a powerlaw fashion as $T \rightarrow T_c$

extract critical exponents

Falicov-Kimball Model



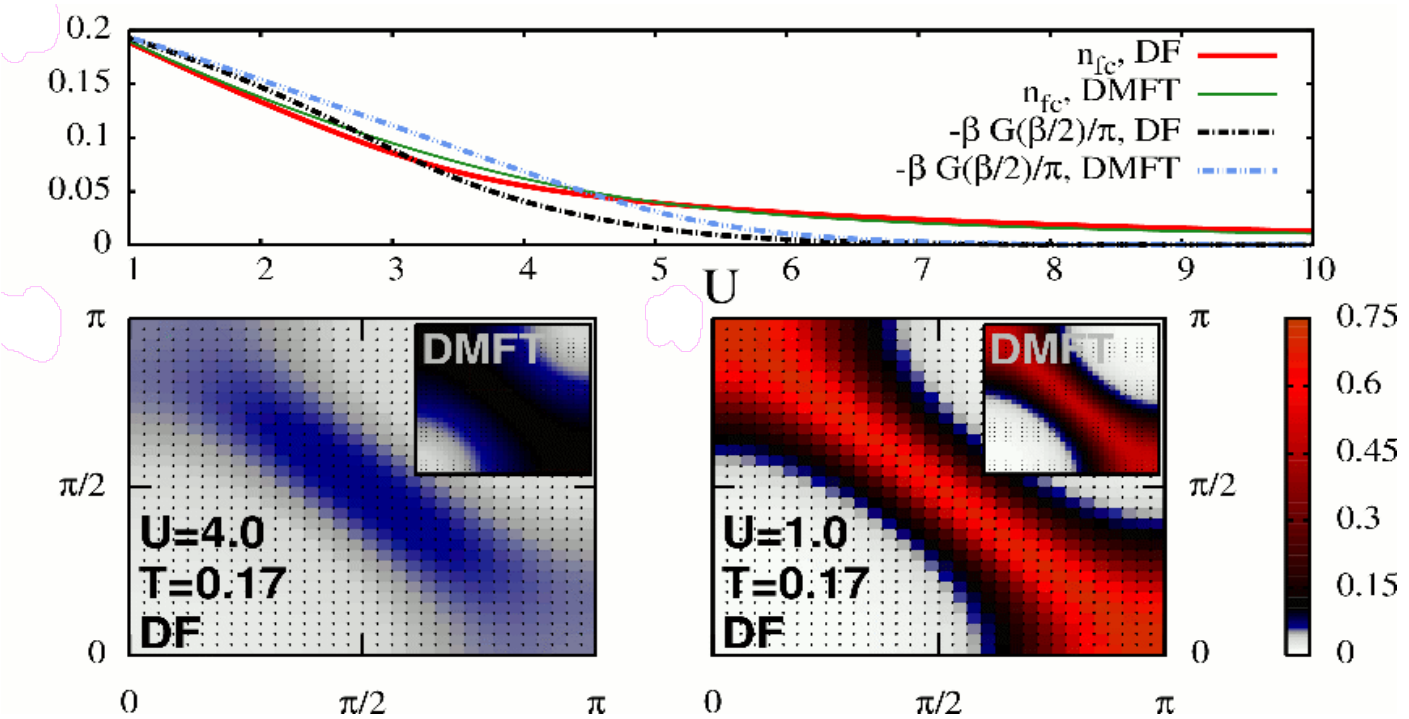


Critical exponents ν and γ agree well with the known values of the Ising model in two and three dimensions! In four dimensions (and above) the correct mean-field behavior is recovered!

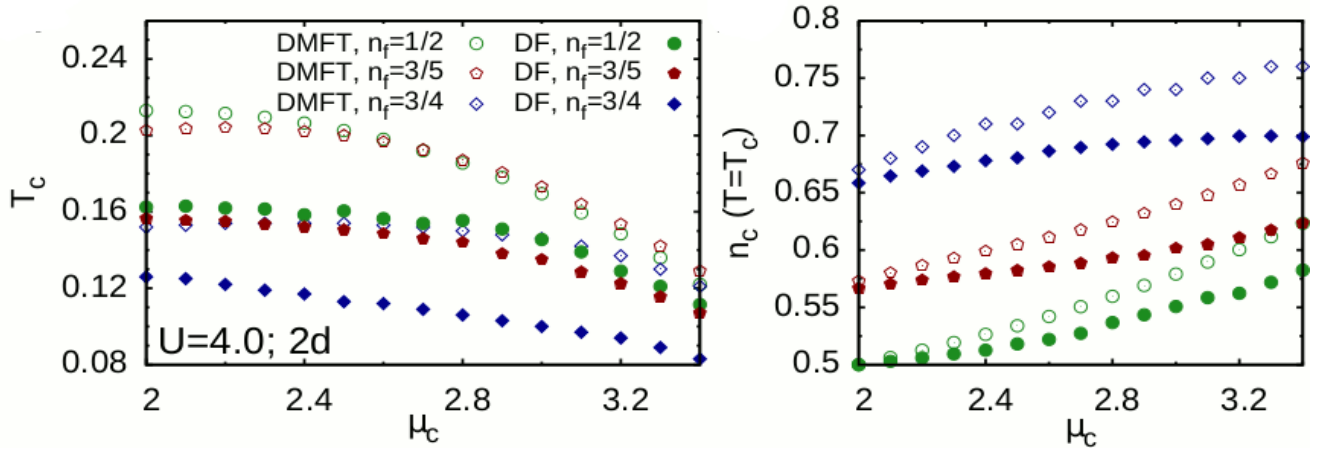
$$\eta_{DF}^{d=2} = 2 - \gamma/\nu \approx 0.25 \neq 0$$

DMFT-Dual Fermion comparison

momentum dependence:



filling dependence:



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Discussion

We can correctly describe phase changes in the Falicov-Kimball model

What about the Hubbard model?

$$\gamma_{\mathbf{\Omega}}^{\mathrm{DMFT}}(\omega) = -\frac{\mathbf{1}}{\mathbf{T}} \frac{\mathbf{\Sigma}_{\omega} - \mathbf{\Sigma}_{\omega+\mathbf{\Omega}}}{\mathbf{g}_{\omega} - \mathbf{g}_{\omega+\mathbf{\Omega}}} \longrightarrow \gamma_{\mathrm{Hubbard}} = \phi(\omega_1, \omega_2, \Omega)$$

The local 4-point vertex in the FK model is simpler than in the Hubbard model but finite temperature transitions are independent of dynamics due to critical slowing down

In fact, for a classical phase transition, only number of space dimension and (local) order parameter symmetry should matter

The spin-rotational invariance of the local DMFT γ_4-vertex in the Hubbard model is vital for capturing the correct Heisenberg exponents

Dual fermion method should give correct(ish) O(3) exponents in three dimensions

Discussion

What about the Hubbard model (at half-filling and large interaction strength) in two dimensions?

DMFT predicts a phase transition but in two dimensions the order is melted away by spin waves (Mermin-Wagner theorem)

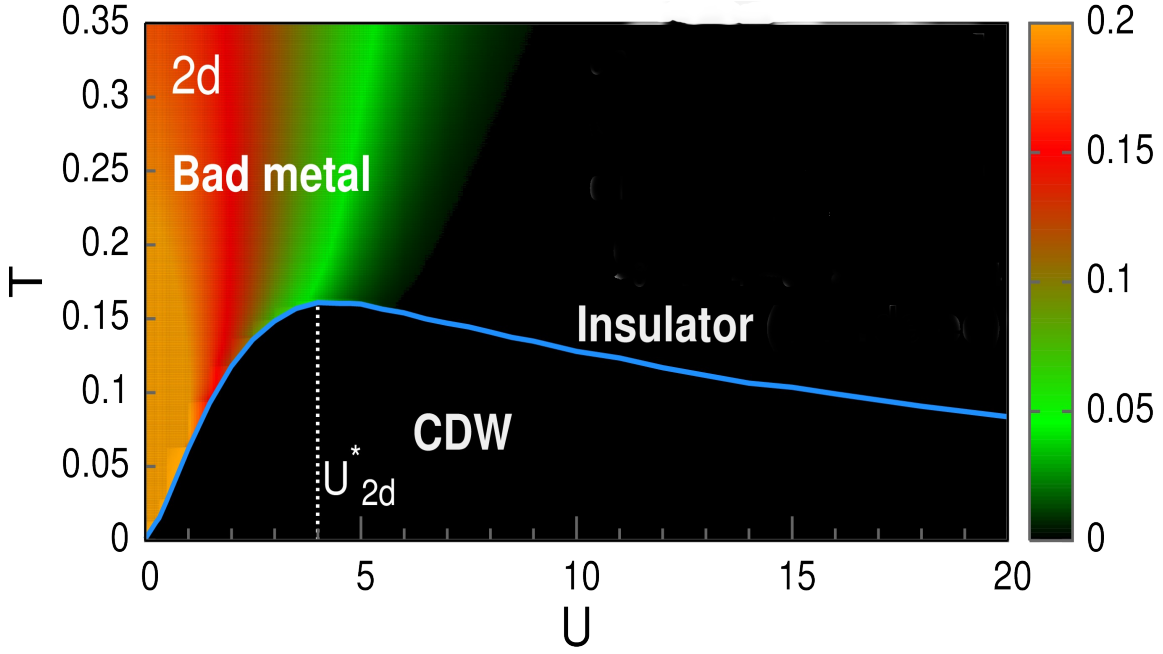
Does the dual fermion scheme descibre the absence of the finite-temperature transition in d=2?

- Most likely not
- This is now a dynamic problem, need to describe spin wave spectrum
- Recall, we started to build around the DMFT solution:
  capturing the hydrodynamic regime (low frequency **and** long wavelength) seems extremely challenging

Similar arguments would apply to a zero-temperature phase transition where dynamics is already part of the equilibrium fluctuation spectrum

Outlook

Dual fermion phase diagram in two dimensions



Above T_c :
interaction driven transition from metal to insulator

CDW is checker-board charge order

at large U: Ising-like transition
at small U: CDW due to nested Fermi surface?

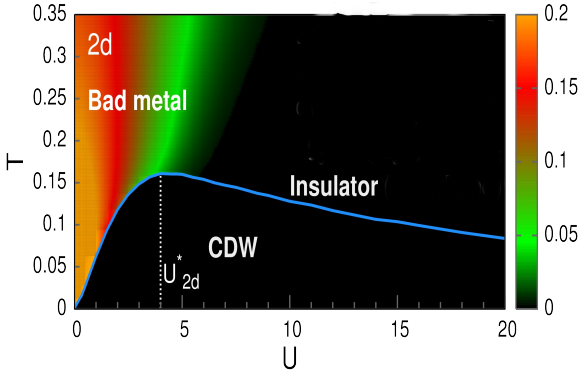
MC in 2D: transition at small U is first order [Maśka et al. (2006)]

Outlook

How does the system evolve from a metal to an insulator as a function of U?

$$H = \sum_{<i,j>} t_{i,j} c_i^\dagger c_j + \sum_i (E_i - \mu_f) n_f^i + U \sum_i c_i^\dagger c_i n_f^i$$

$$[H, n_f^i] = 0$$



For given configuration of $\{n_f^i\}$: conduction electrons see set of static impurities

→ single electron in random potential: grand canonical distribution ≡ annealed disorder

Conduction electrons can be integrated out exactly for each $\{n_f\}$ configuration.

→ **Classical Monte-Carlo over the set of $\{n_f\}$**

Access to thermodynamics, critical exponents, inverse participation ratio, ...

Outlook

'Inverse Participation Ratio' (IPR)

single-particle wave function with quantum number n

$$I_n = \frac{\sum_i |\psi_n(r_i)|^4}{(\sum_i |\psi_n(r_i)|^2)^2}$$

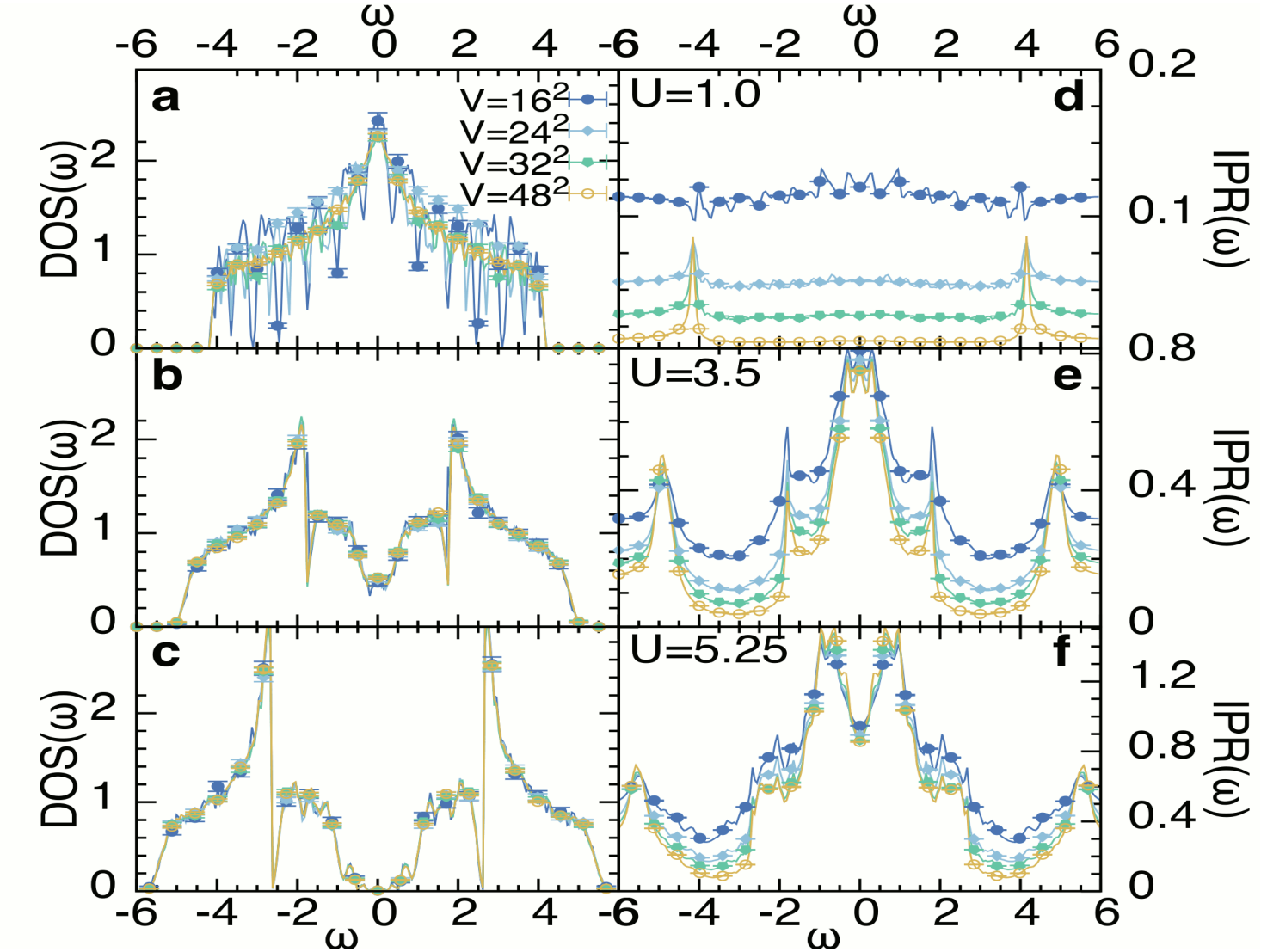
measure of degree of localization of a single-particle wavefunction

$$\lim_{L \longrightarrow \infty} I_n \sim \begin{cases} \text{const. for localized states} \\ 1/L^d \text{ for extended states} \end{cases}$$

We can also study the energy-resolved IPR $I_n(\omega)$

We can average the energy-resolved IPR divided by the DOS(ω) over the probability distribution P($\{n_f^i\}$)

Outlook

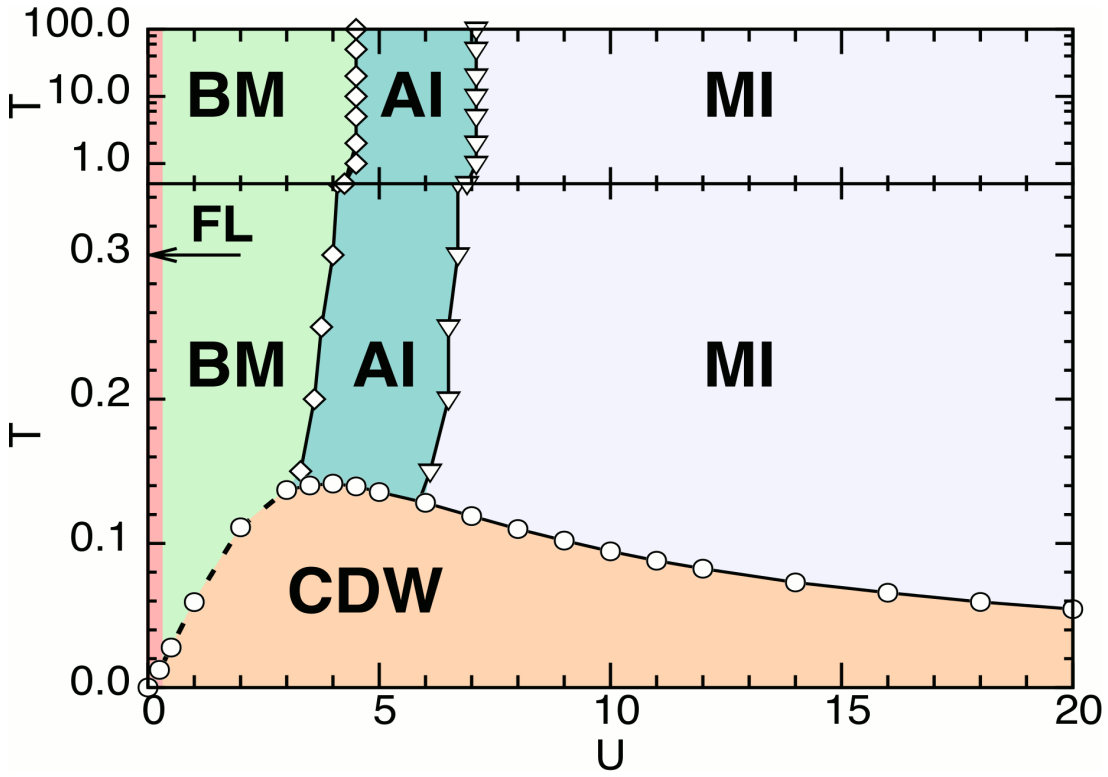


itinerant

localized

Andrey E. Antipov, Younes Javanmard, Pedro Ribeiro, & Stefan Kirchner, in preparation.

Outlook



The BM phase is expected to disappear in the $L \to \infty$ limit

CDW is nowhere a simple charge density wave, no nesting

Andrey E. Antipov, Younes Javanmard, Pedro Ribeiro, & Stefan Kirchner, in preparation.

Summary

- Multi-scale methods are a useful approach to strongly correlated matter
- The LDFA captures the non-mean field character of the transition in the Falicov-Kimball model
- The LDFA can be used to address the non-local interplay of strong correlation and frustration
- The LDFA predicts a spin-liquid phase in the triangular Hubbard model

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Thank You!

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