Yukawa International Seminar 2007 (YKIS2007) *"Interaction and Nanostructural Effects in Low-Dimensional Systems", Nov.* 5-30, 2007, Kyoto University, Kyoto

Strongly Correlated Electrons on Frustrated Lattices

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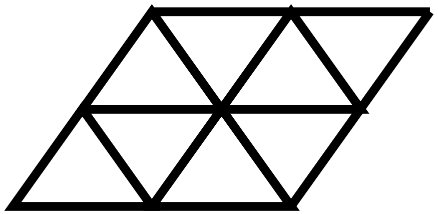
- *the Next Generation Super Computing Project, Nanoscience Program*

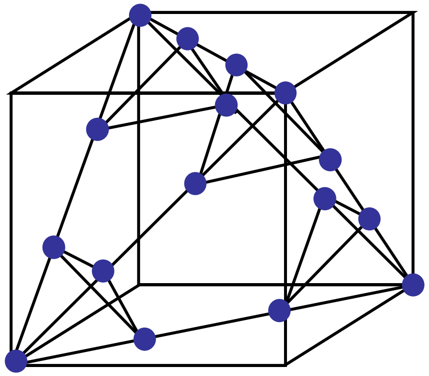
OUTLINE

- Correlated electron systems with geometrical frustration
- [A] Kagomé Lattice Hubbard model
  - exotic spin correlation near metal-insulator transition
- [B] Anisotropic Triangular Lattice Hubbard Model
  - entropy and frustration effects
  - heavy quasiparticle formation and metal-insulator transition
- [C] Trimer phase of bilinear-biquadratic zigzag chain
  - antiferro spin nematic correlation

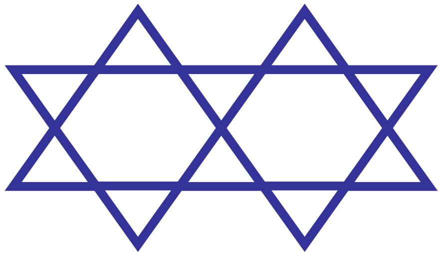
Correlated electron systems with geometrical frustration



Triangular



Pyrochlore



Kagomé

Classical: Many states are degenerate in low-energy sector.
Quantum effects hybridize these states -> new phase/correlations?

Many interesting systems:

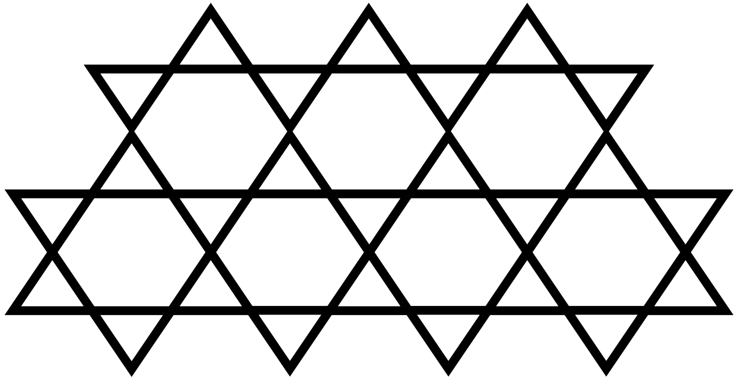
- Superconductivity **$Na_xCoO_2 \cdot yH_2O$, AOs_2O_6**
- Heavy Fermion **LiV_2O_4, etc**
- Quantum spin liquid **κ-$(ET)_2Cu_2(CN)_3$**

PART A

Mott Transition in
Kagomé Lattice Hubbard Model

[Ohashi, Kawakami, and Tsunetsugu, Phys. Rev. Lett. **97** ('06) 066401]

Kagomé Lattice Hubbard Model



$$H = -t \sum_{\langle i,j \rangle,\sigma} c_{i\sigma}^{+} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

- Typical frustrated lattice in 2D

  (thermodynamically degenerate ground states of AF Ising spins)
- 2D analog of pyrochlore lattice
- Effective model of $Na_xCoO_2 \cdot yH_2O$

  Koshibae & Maekawa PRL 91, 257003 (2003)

  Bulut, Koshibae, & Maekawa PRL 95, 37001 (2005)
- Spin systems on Kagomé lattice

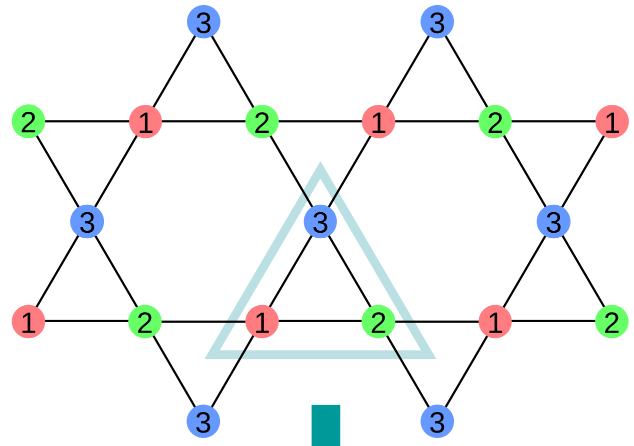
  ⇒ unusual properties (gapped triplet, gapless singlet excitations)

Relation btw charge fluctuations and spin correlations
Effects on quasiparticle coherence
[fix density at half filling n=1]

Method

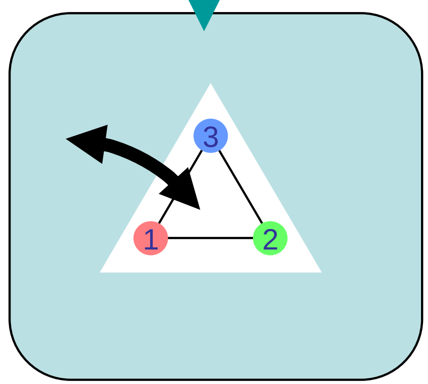
Metal-insulator transition in Kagomé lattice at half filling

- strong correlation
- geometrical frustration
- short-range quantum fluctuations ⇒ DMFT



Cellular dynamical mean field theory (CDMFT)

Kotliar, et al. PRL 87, (2001)
Lichtenstein & Katsnelson, PRB 62, (2000)
...



Effective cluster model

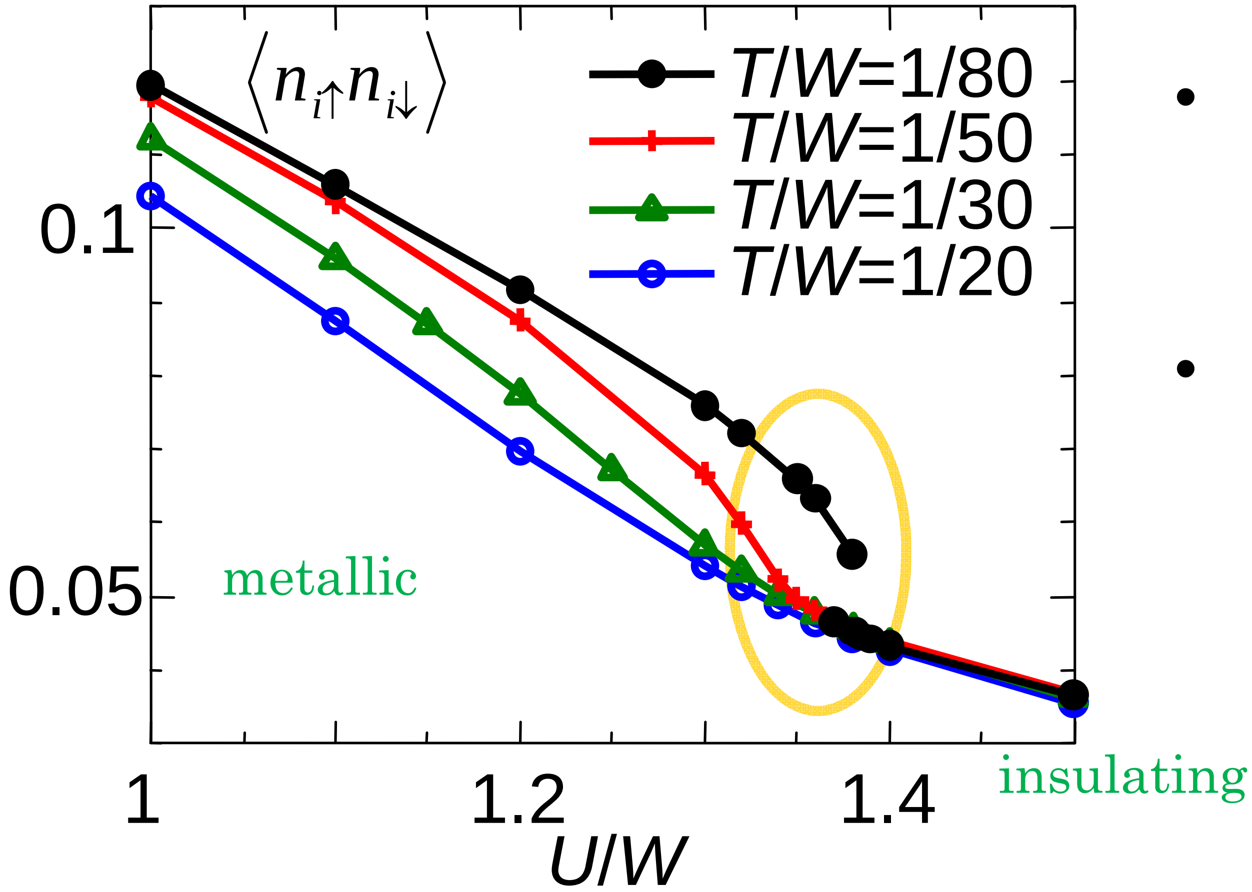
DMFT
Vollhardt
Muller-Hartmann
Kotliar
Georges
Jarrell
...

Self-energy: 3x3 matrix $\Sigma_{ij}(\omega)$

- Spatially extended correlation
- Geometrical frustration

Mott transition

: Double occupancy
measure of charge fluctuations



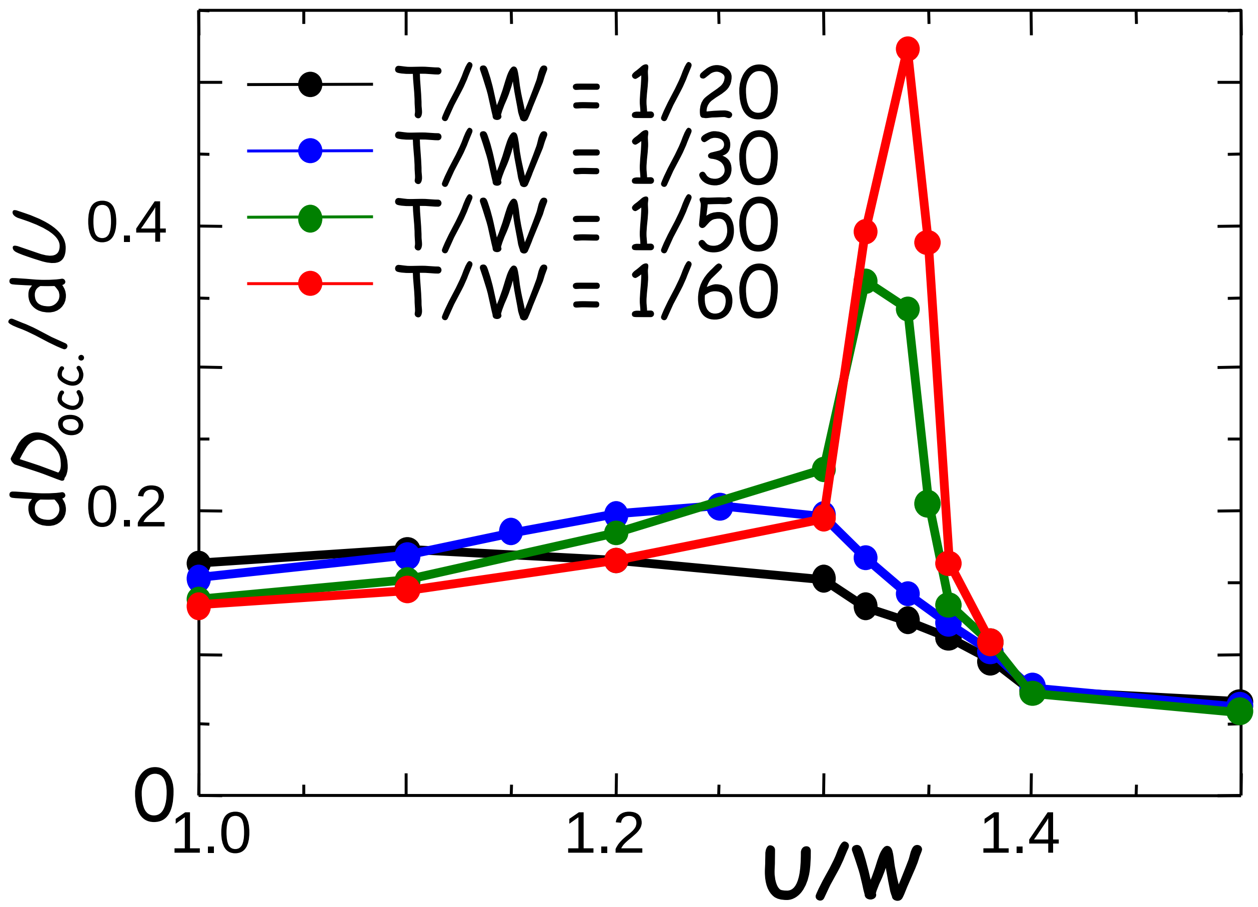
square lattice:
$U_c \sim 0.5\text{-}1.0$

Band width: $W=6t$

- High temperature
  $T/W>1/80$
  crossover $U^* \sim 1.35$
- Low temperature
  $T/W=1/80$
  1st order transition with hysteresis: $U_c \sim 1.37$

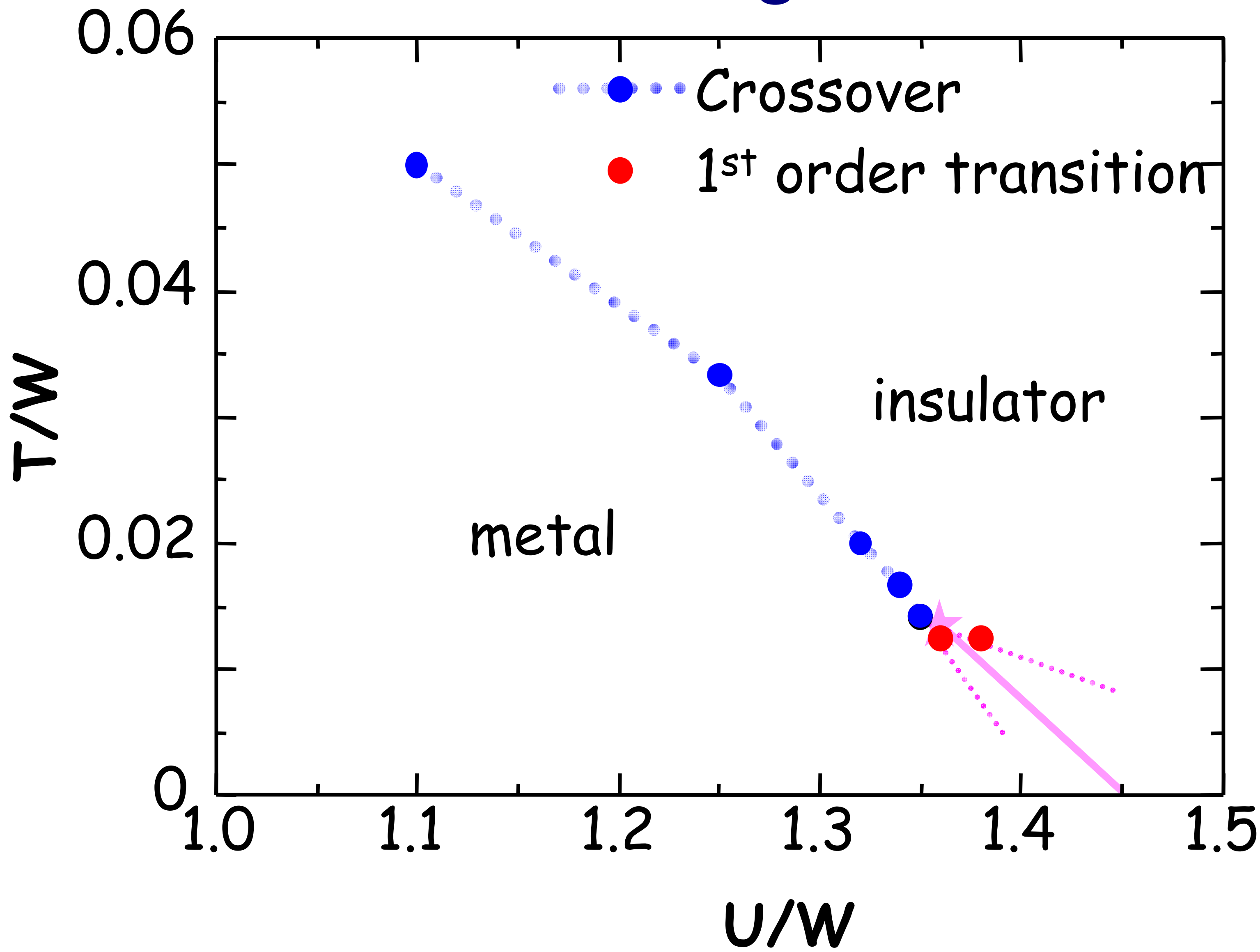
Larger critical value U_c

Crossover



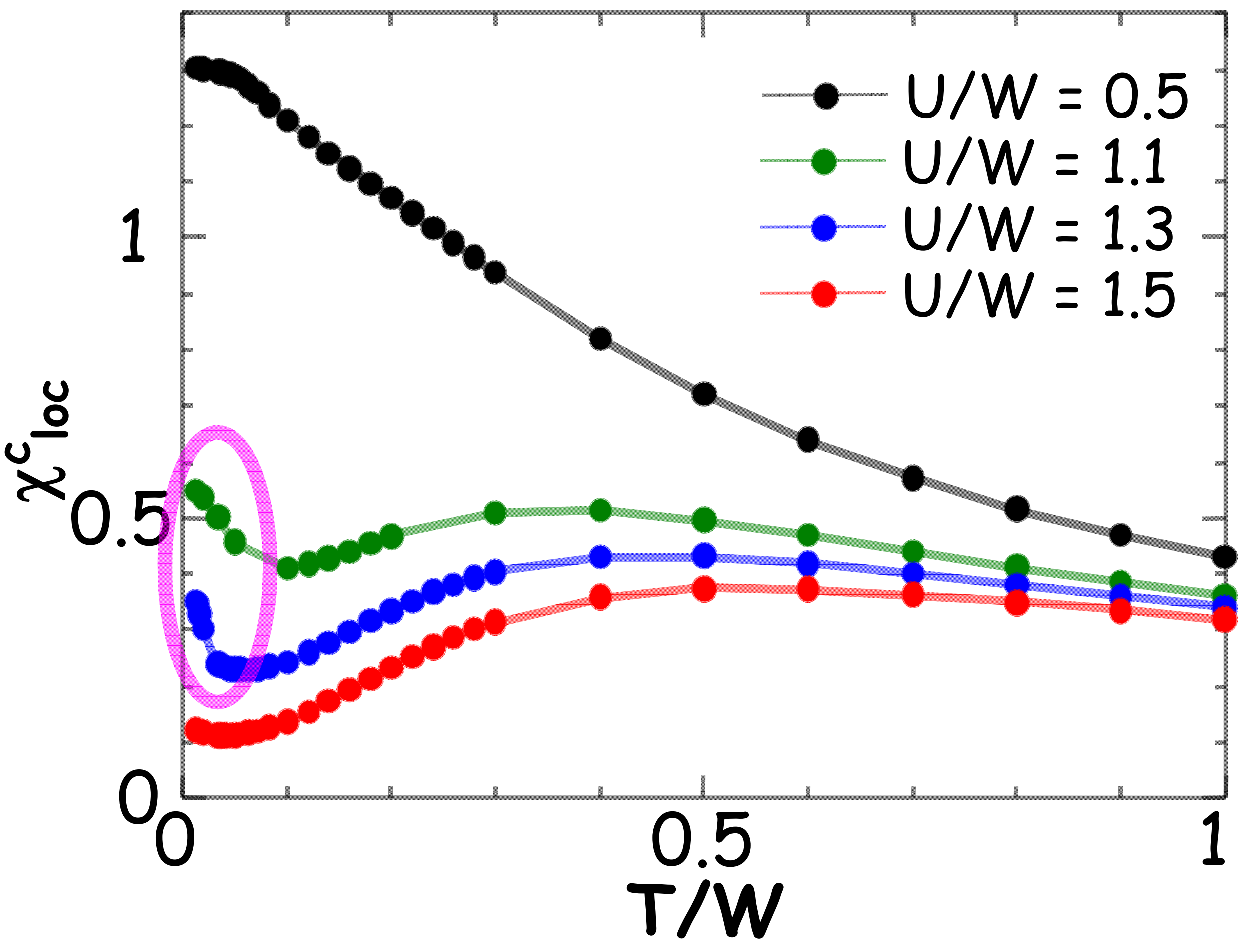
Define metal-insulator crossover points $U^*(T)$ by largest change in double occupancy

Phase diagram



Mott transition in Kagome Hubbard model

Charge susceptibility



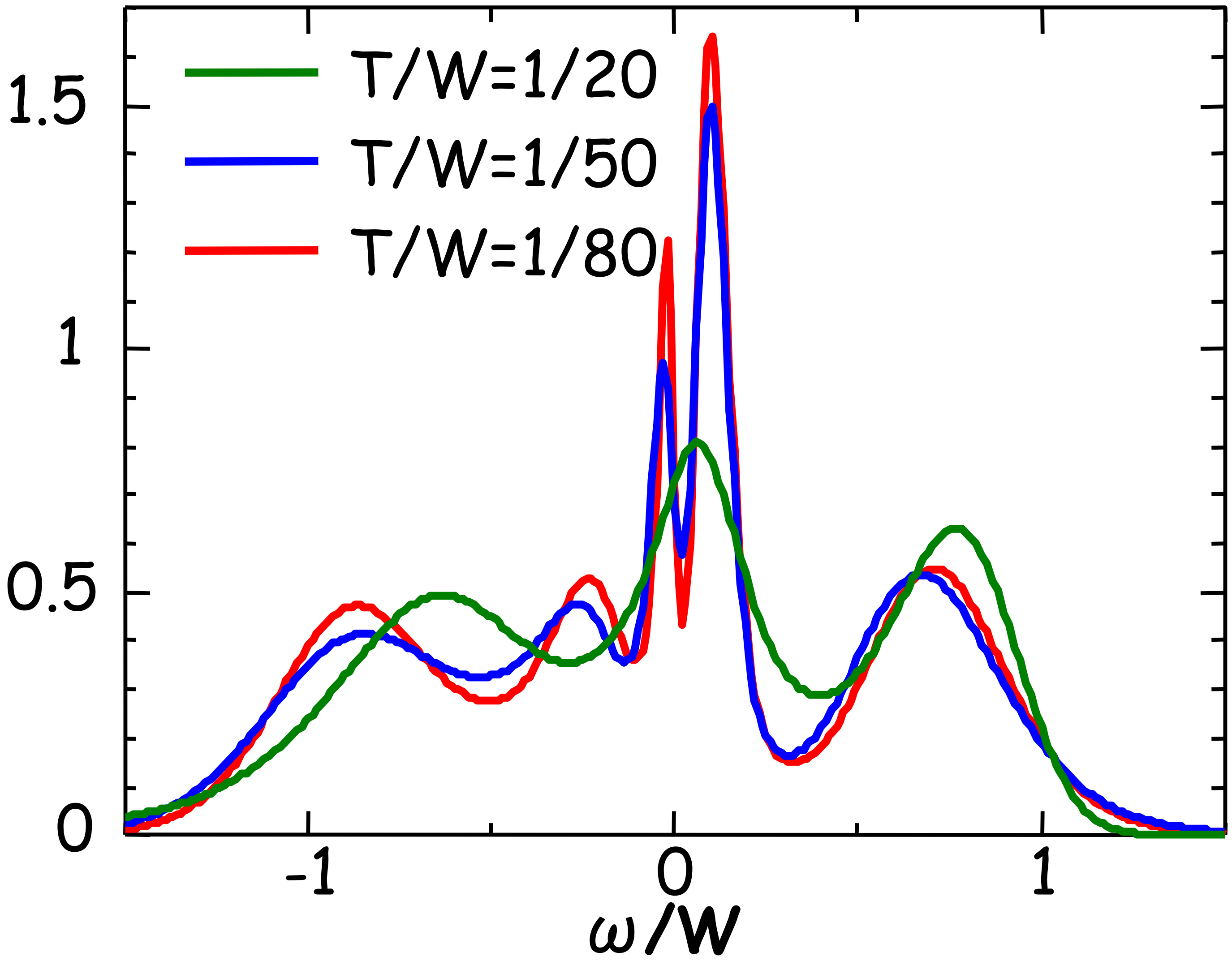
Charge response grows once again
in low-T metallic region

Density of States: *U*/*W*=1.1

electron spectral function (k-summed)

measurable by PE/IPE experiments

Evolution of heavy quasiparticles at low temperatures

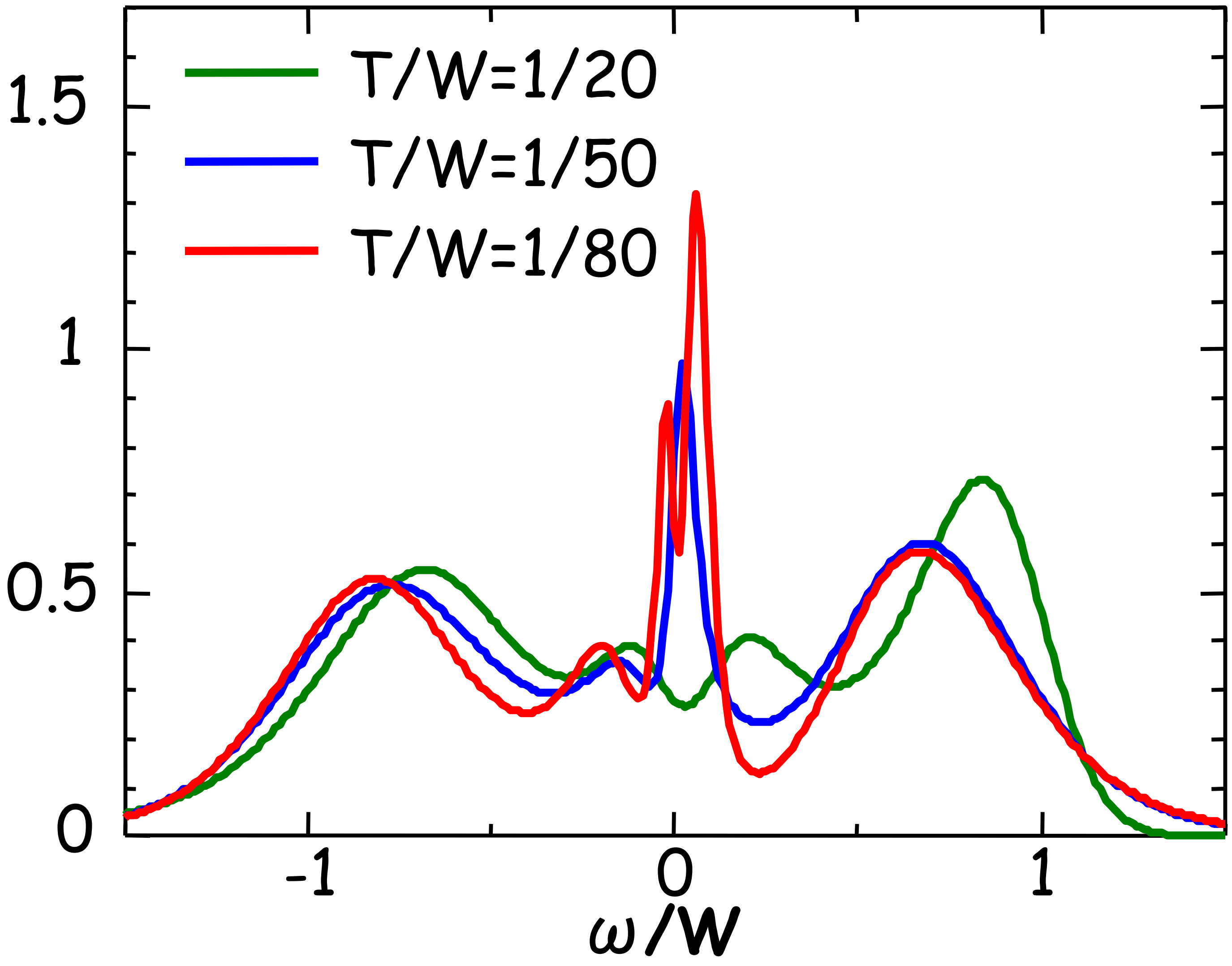


Density of States: *U*/*W*=1.3

Precursor of insulating behavior

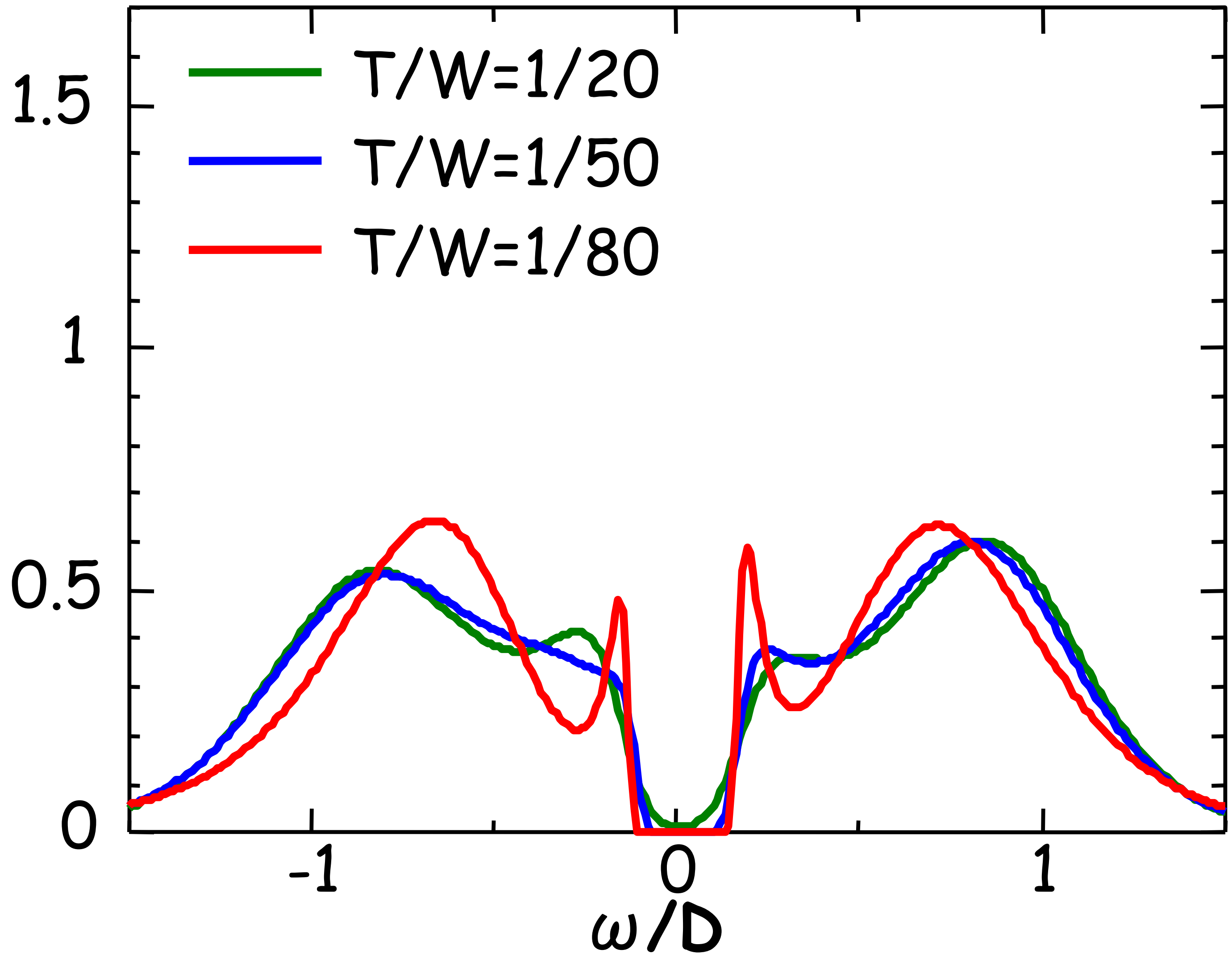
+

Evolution of heavy quasiparticles at low temperatures



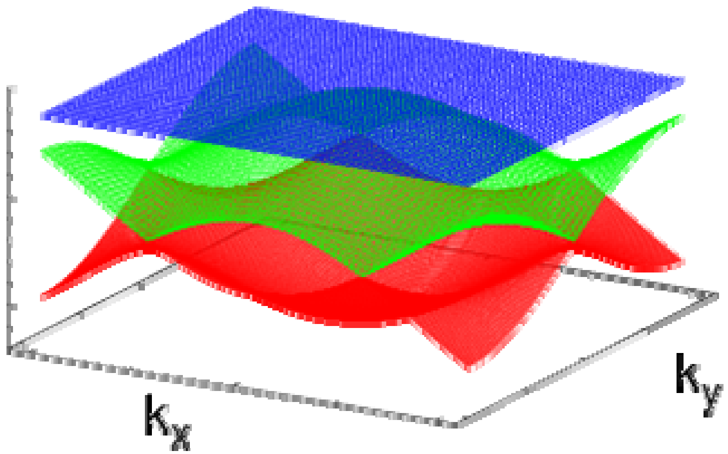
Density of States: *U*/*W*=1.5

Clear formation of Mott gap

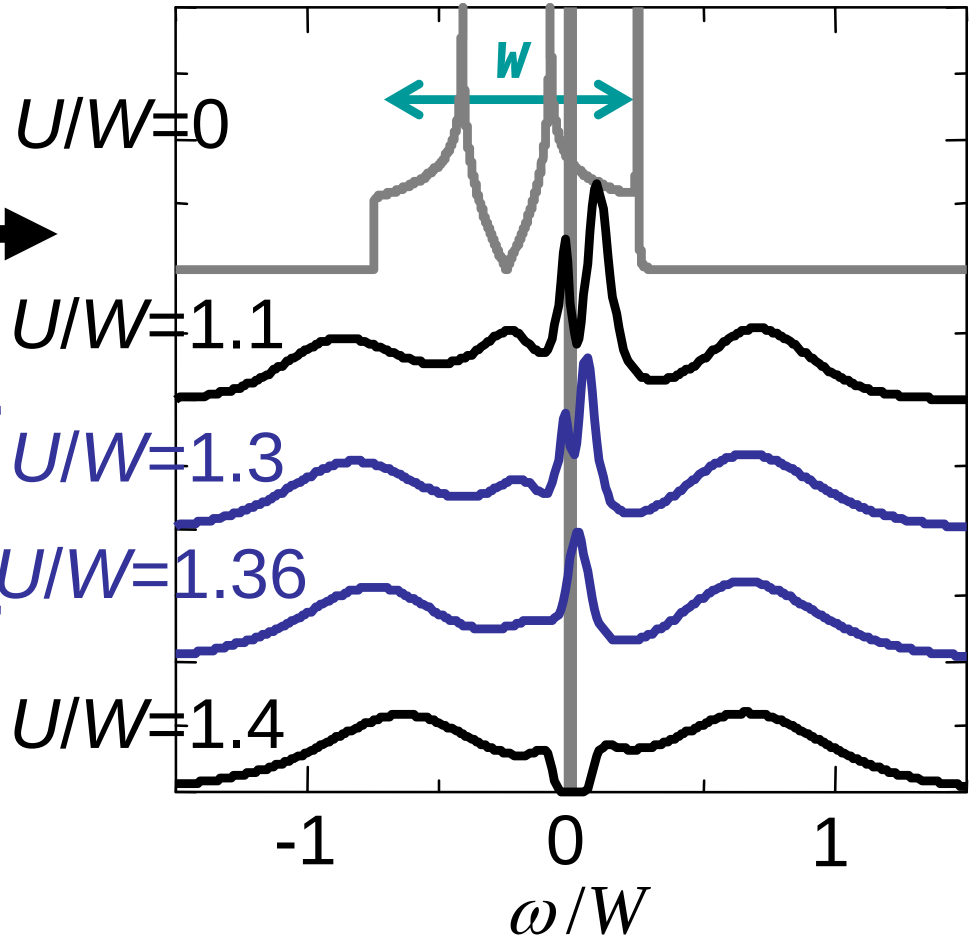


Density of states

Dispersion (U=0)



DOS (T/W=1/80)

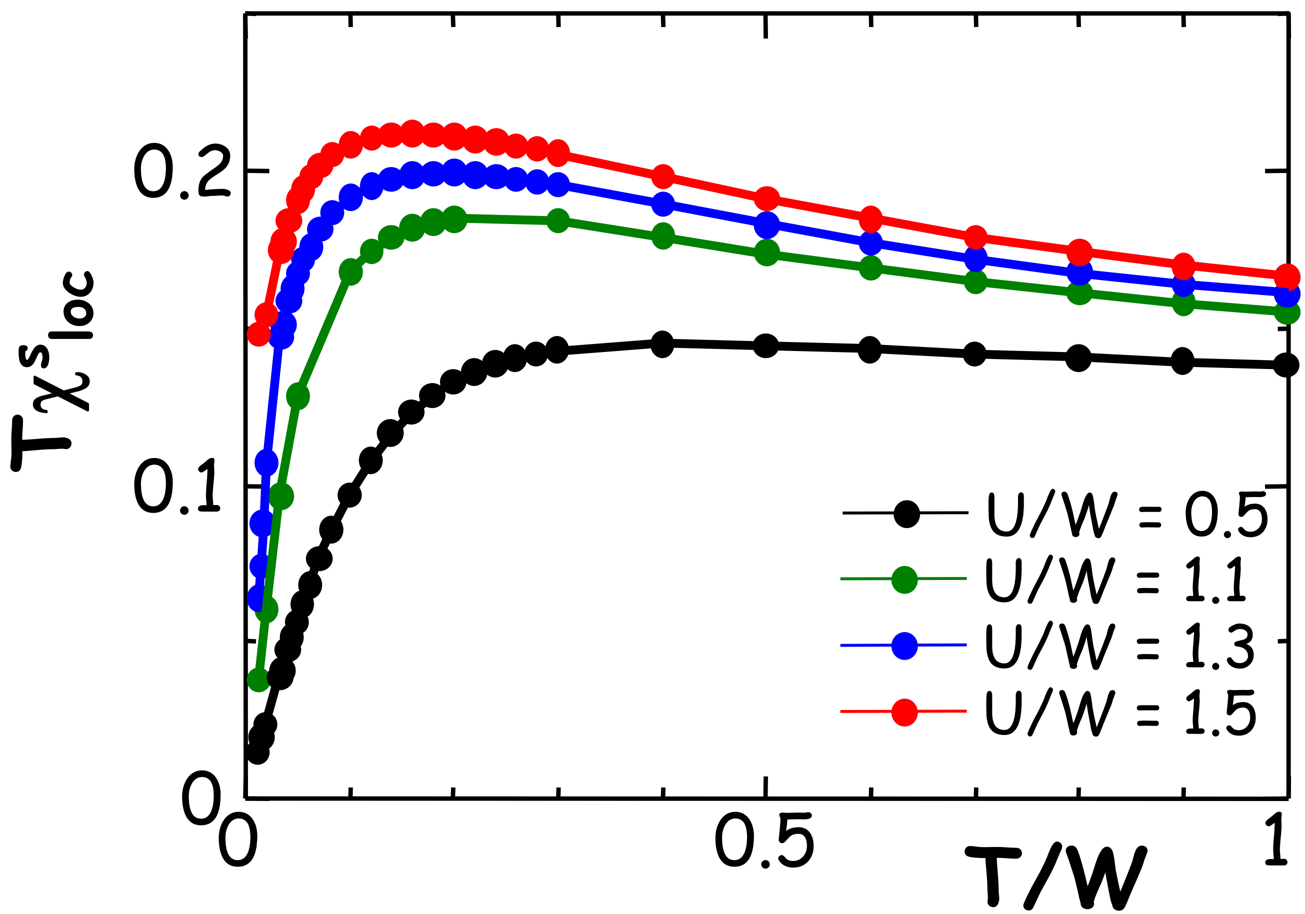


Strongly correlated metal

- Whole bands are renormalized
- Heavy quasiparticles

Insulator: $U_c/W \sim 1.37$

Local spin susceptibility



1-site DMFT:
Free spins in
insulating phase

cluster DMFT:
Spins are
screened/correlated

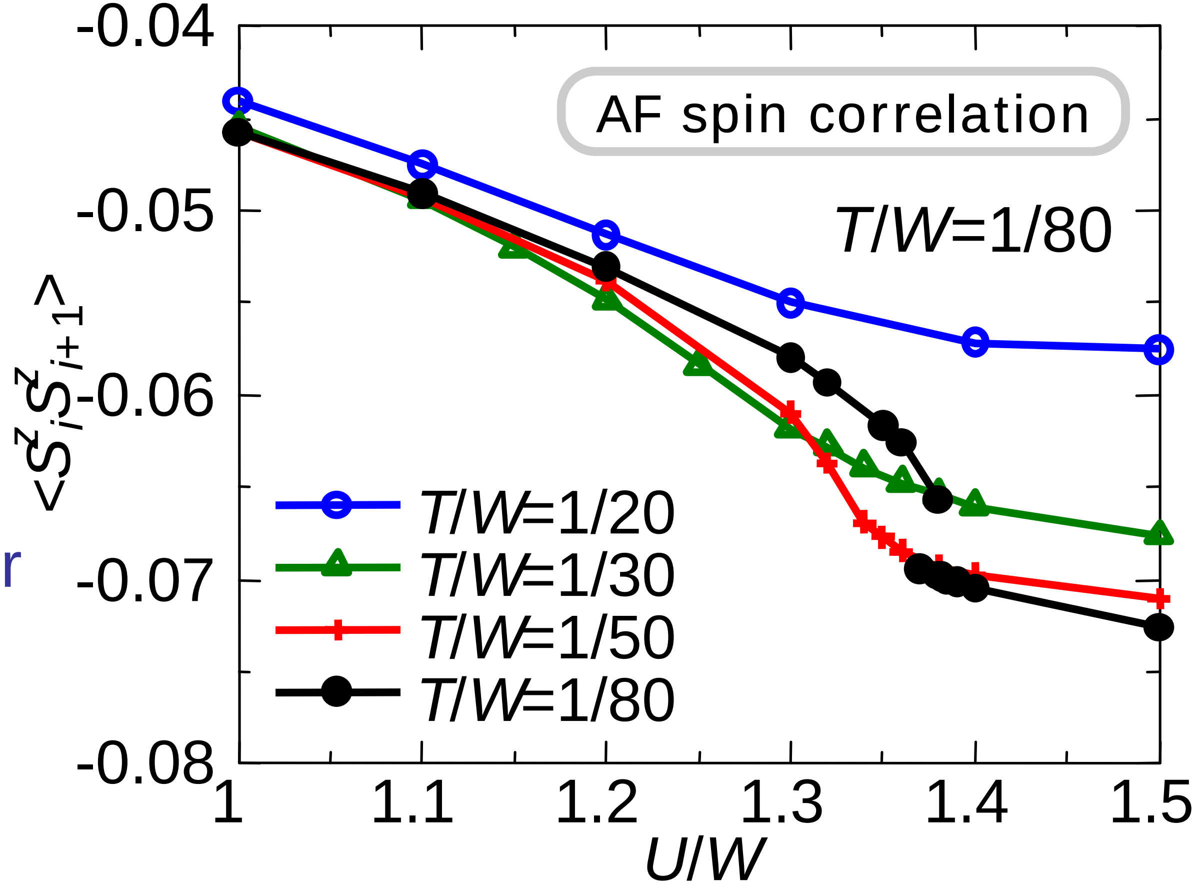
Spin correlation function

Nearest-neighbor spin correlation $\langle S_i^z S_{i+1}^z \rangle$

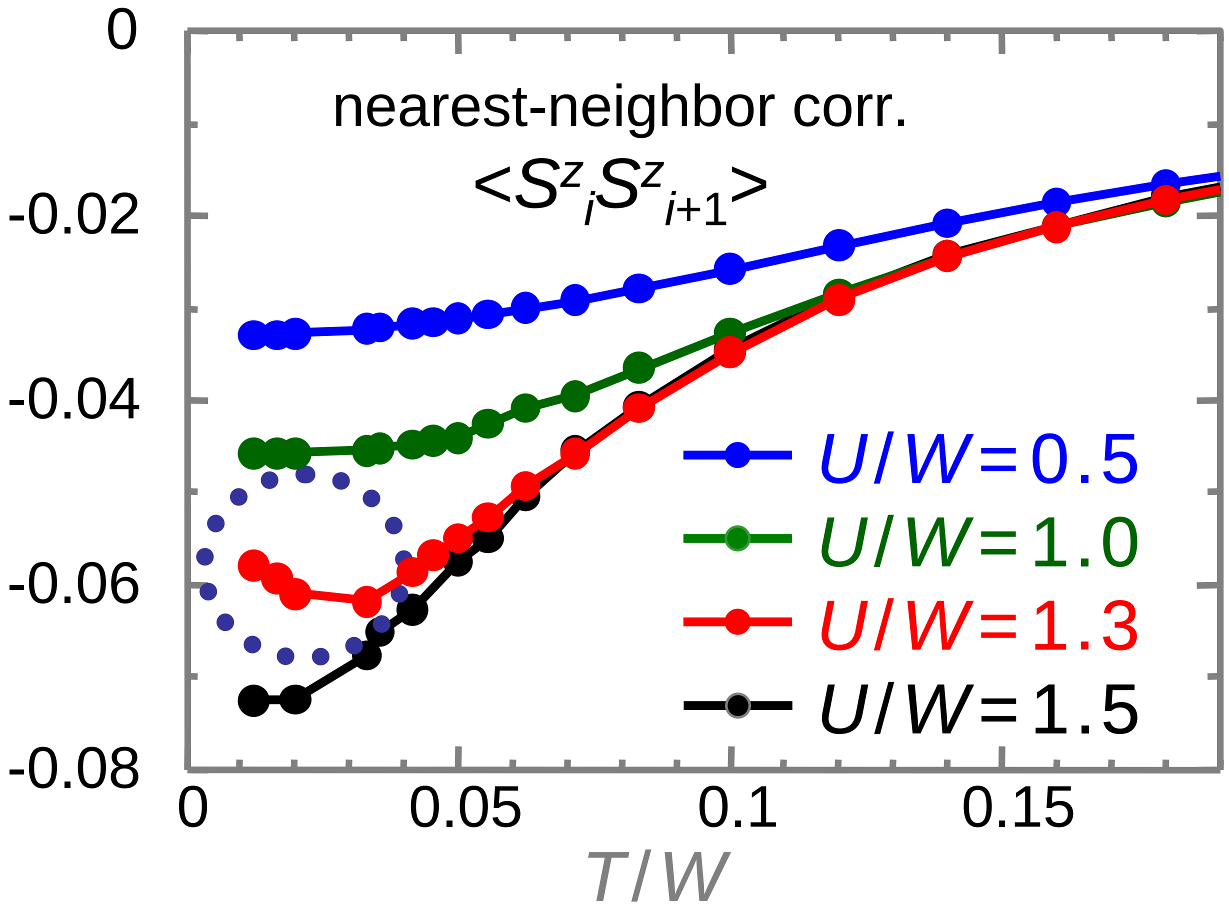
Insulating phase:
AF correlation ⇒ monotonic enhancement

Metallic phase:
Nonmonotnic behavior

Recover of itinerancy



Temperature Dependence of Spin Correlations



- Antiferromag.
- **insulator:**

Monotonic growth of AF correlation

- **correlated metal:**

Nonmonotonic T-dep

Suppressed AF correlation at low-T

↓

Characteristic for frustrated systems near MIT

- recovery of coherence
- relax frustration

Dynamical Susceptibility near Mott Transition

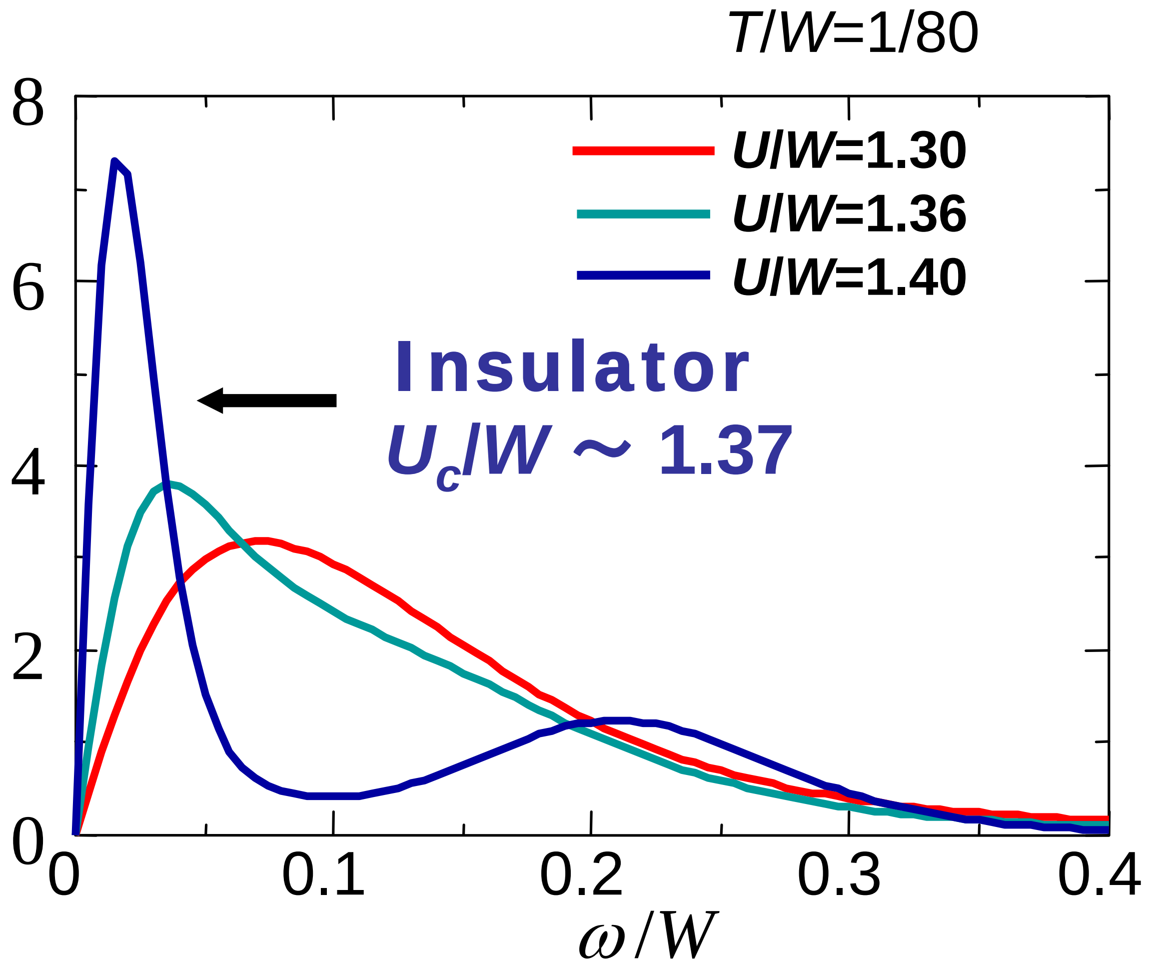
Imaginary part of local susceptibility

$$\chi_{loc}(\omega) = -i\int \left\langle \left[S_i^z(t), S_i^z(0) \right] \right\rangle e^{-it\omega} dt$$

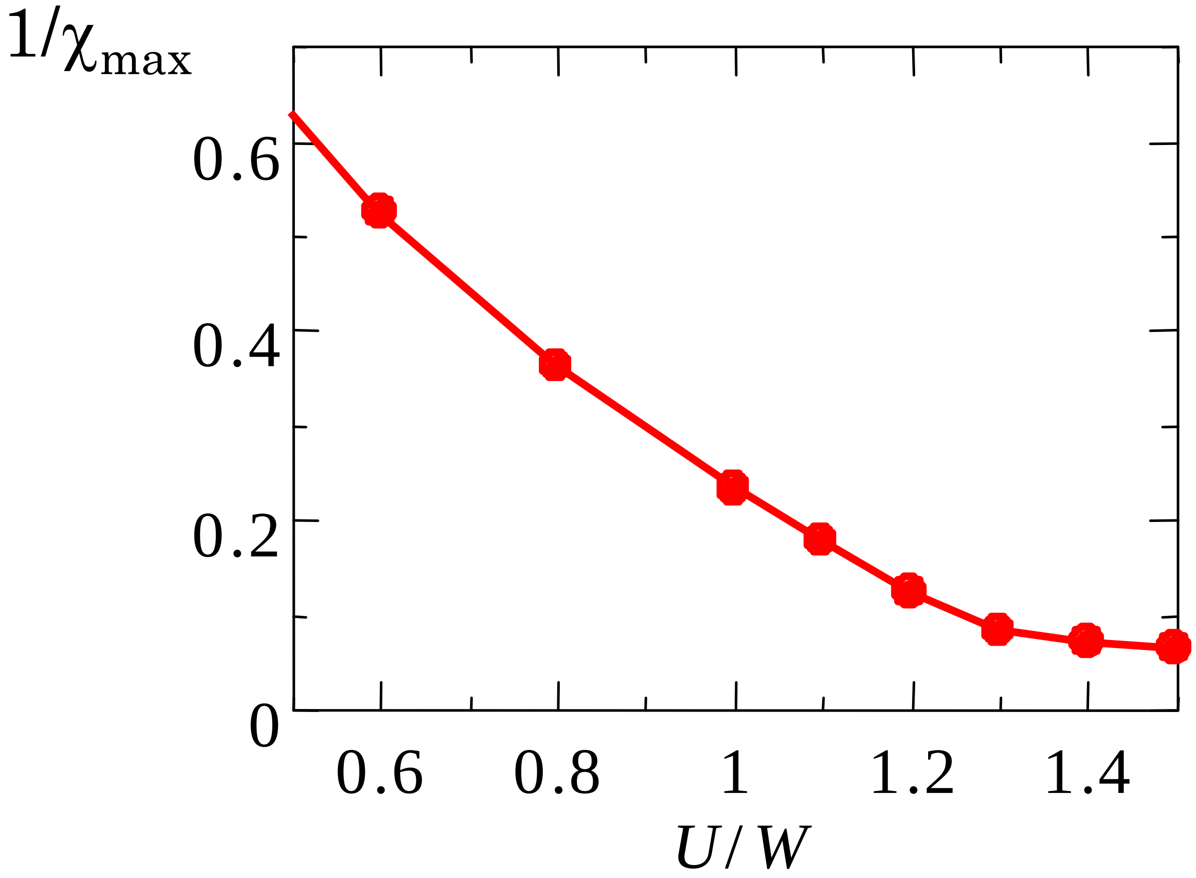
metal ⇒ insulator

Double peak

Metallic phase:
Renormalized single peak



Suppressed Magnetic Instability



T/W=1/30

Mott transition : $U_c/W \sim 1.35$

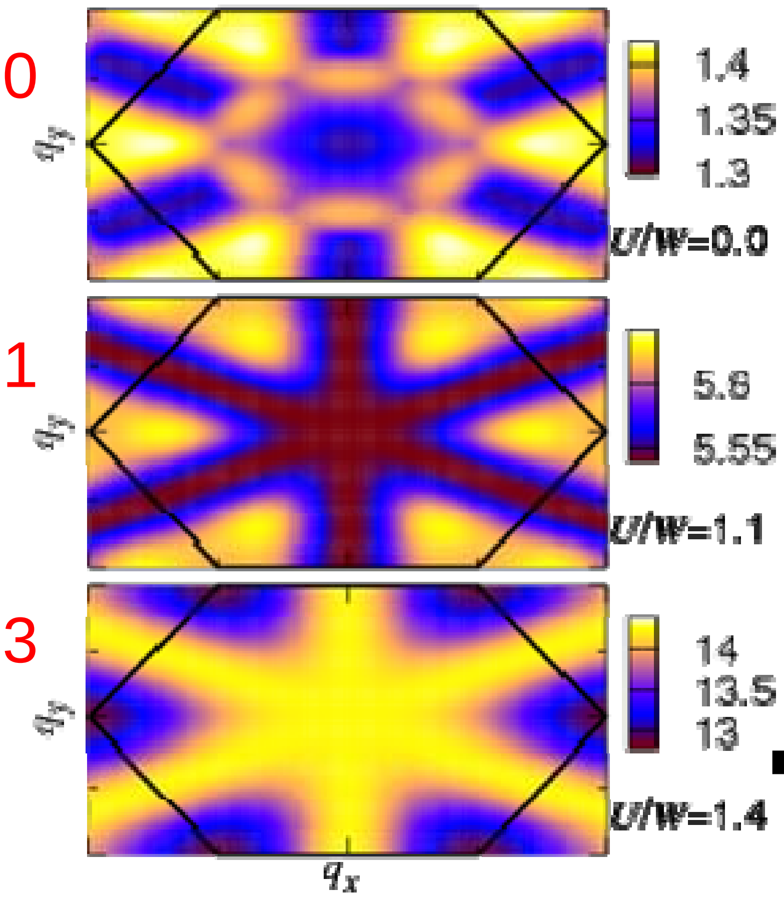
Wavevector dependence of dominant mode

$\chi_{\max}(\boldsymbol{q})$

U/*W*=0.0

U/*W*=1.1

U/*W*=1.3



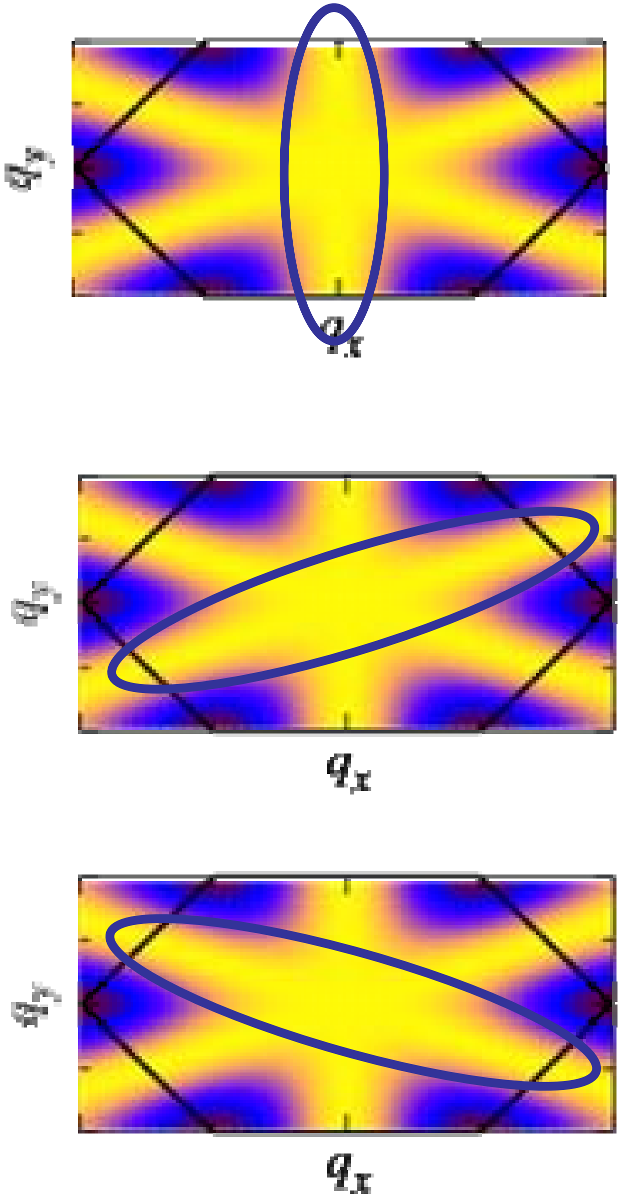
Metallic phase

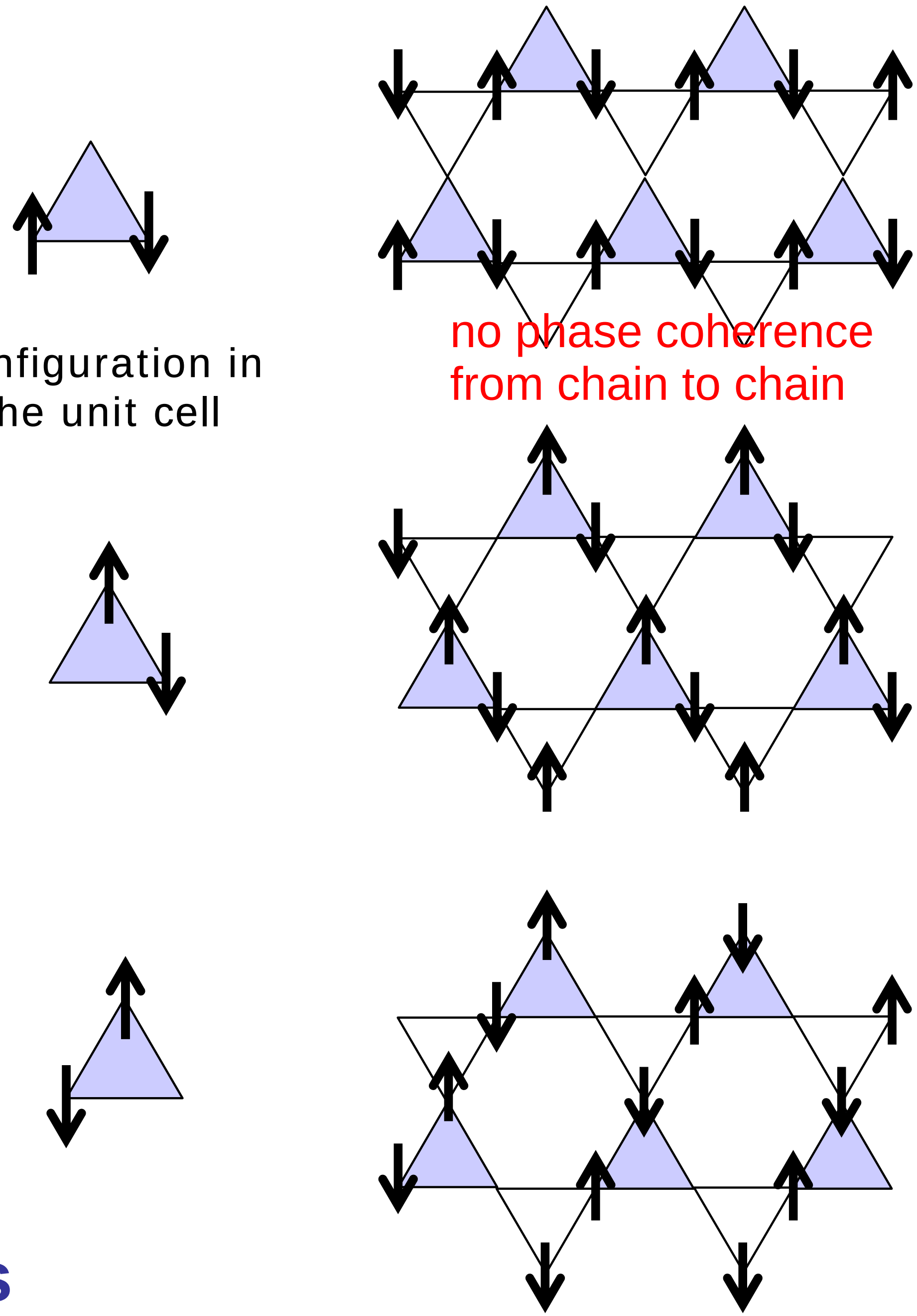
Leading magnetic mode:
6 points ⇒ nesting

Insulating phase
Leading magnetic mode:
Three lines ⇒
1-dimensional order

temperature: *T*/*W*=1/30

Spin correlations in the real space



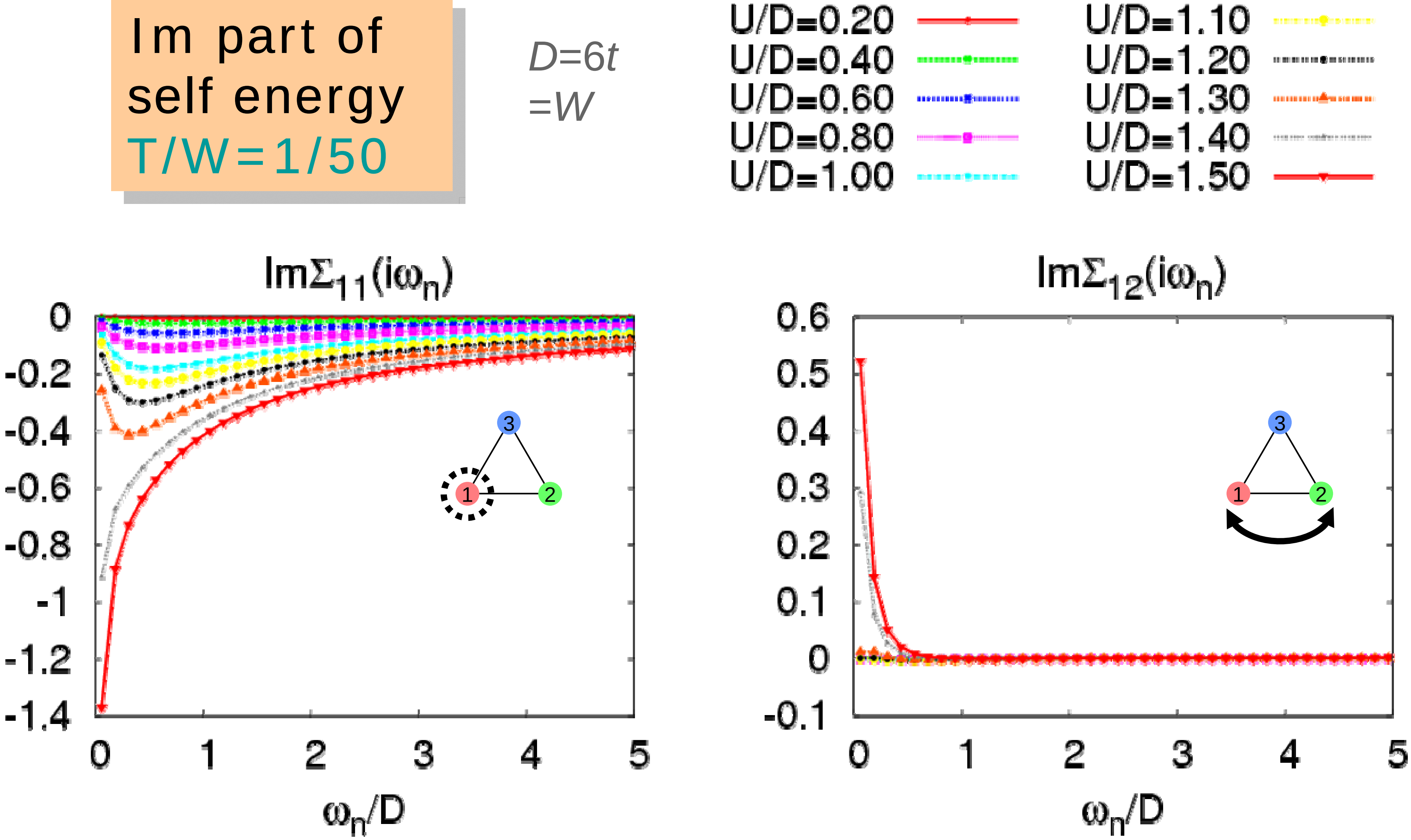


1-dim. spin correlations

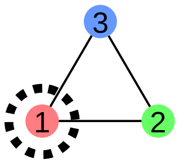
Self Energy of Single-Particle Green's Fn.

Im part of self energy
T/W=1/50

D=6*t*
=*W*



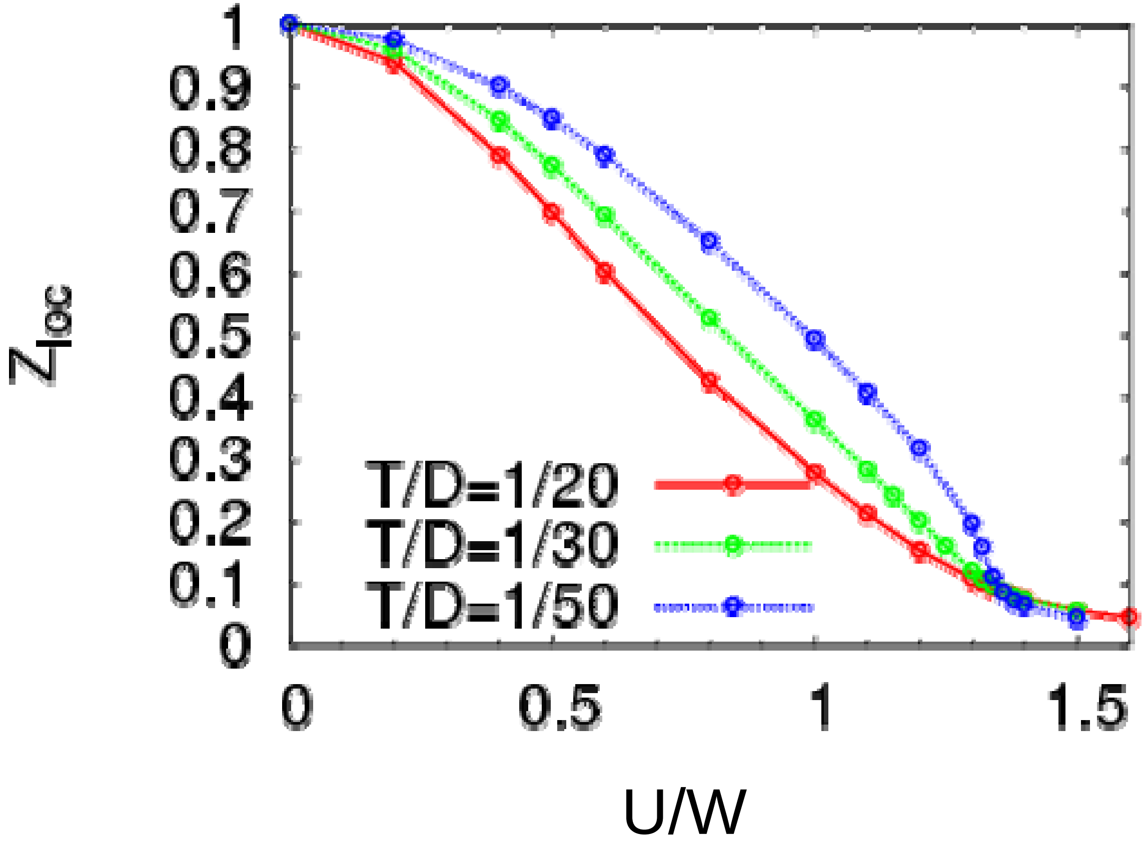
Quasiparticle Renormalization Factor



self energy

$\Sigma_{11}(i\omega_n)$ → Renormalization factor Z_{loc}

Mass enhancement
∽ 10-20
near Mott transition



Summary (1)

Kagome lattice Hubbard model

Cellular dynamical mean field theory

- Metal-insulator transition
  - 1st order transition :$U_c/W \sim 1.37$
- Strongly correlated metal
  - Whole bands are renormalized
  - large mass enhancement
  - nonmonotonic temperature dependence of spin correlation functions
- Magnetic instability
  - one-dimensional spin correlations

PART B

Mott Transition in Anisotropic Triangular-Lattice Hubbard Model

- Phase Boundary Topology
- Heavy Quasiparticles

[Ohashi, Momoi, Tsunetsugu, and Kawakami, cond-mat.st-el/0709.1700]

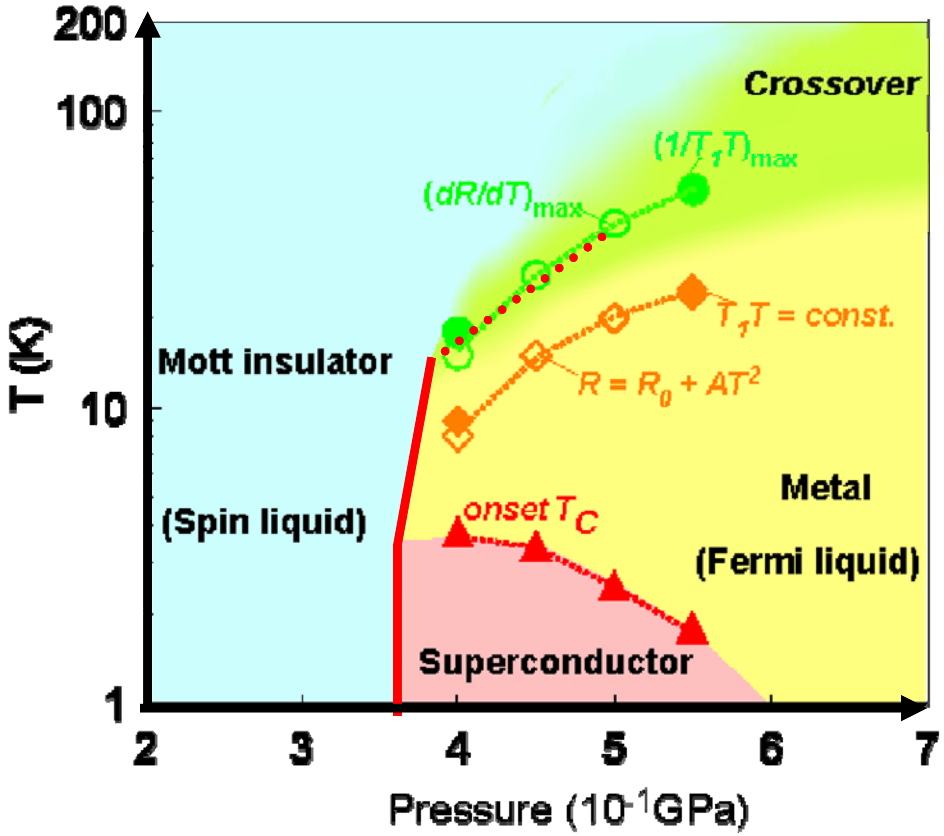
Mott transition in κ-type organic materials

STRONG frustration

nearly perfect regular triangle

κ–$(ET)_2$-$Cu_2(CN)_3$

Y. Kurosaki et al., PRL 95, 177001 (2005)

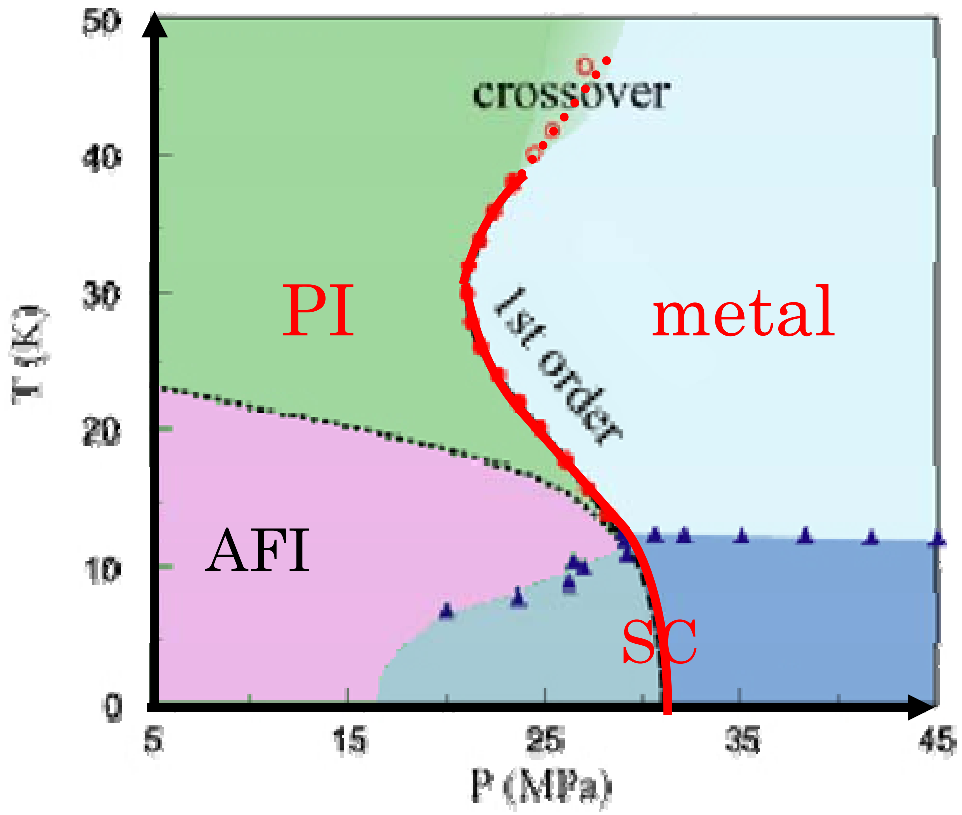


INTERMED. frustration

distorted towards square

κ-$(ET)_2$-Cu$[N(CN)_2]$Cl

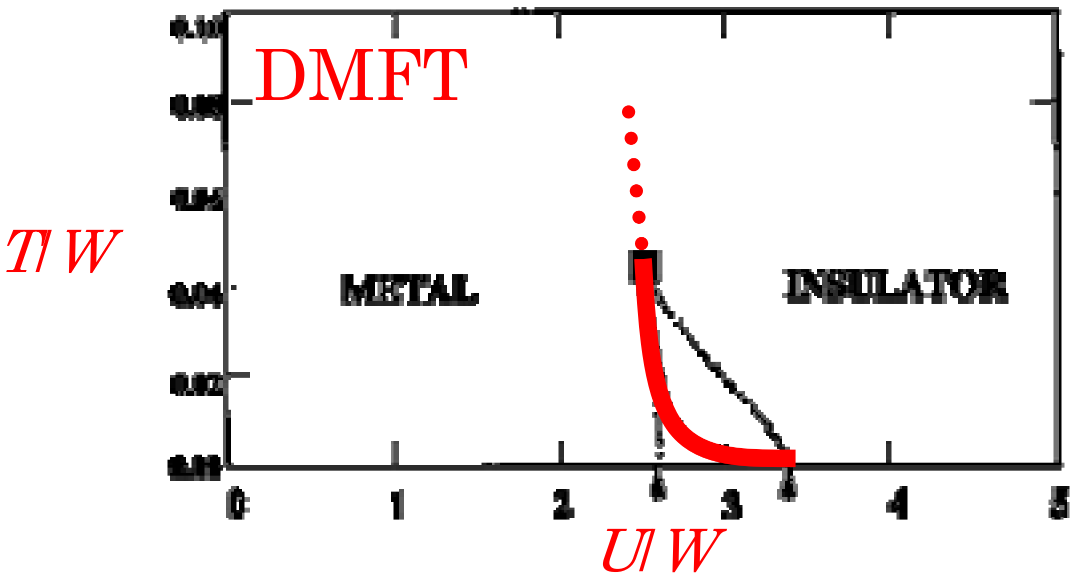
F. Kagawa et al., PRB 69, 064511 (2004)



Reentrant !!

Mott transition line in Phase Diagram

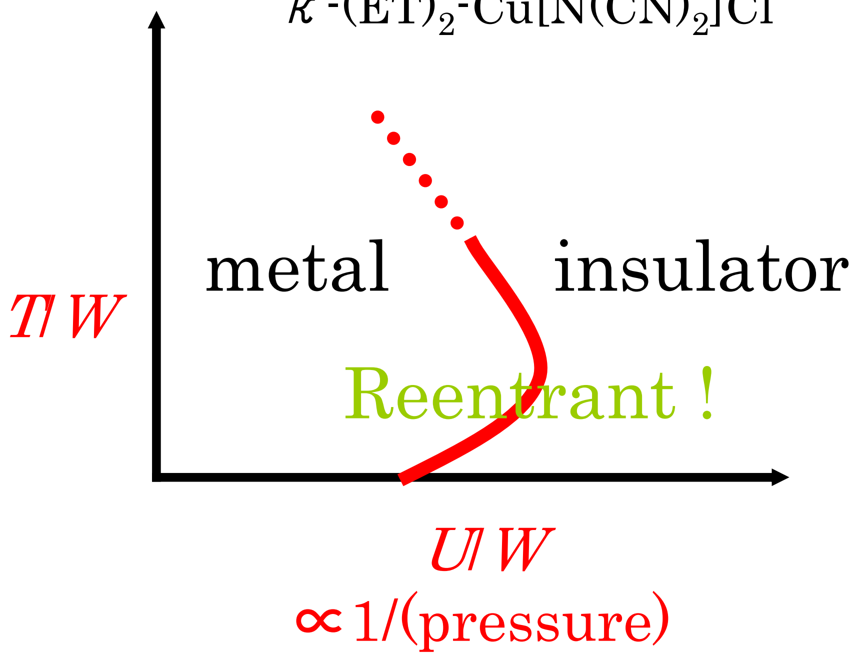
d=∞ Hubbard model



Georges et al., RMP, 68, 13 (1996)

organic conductor

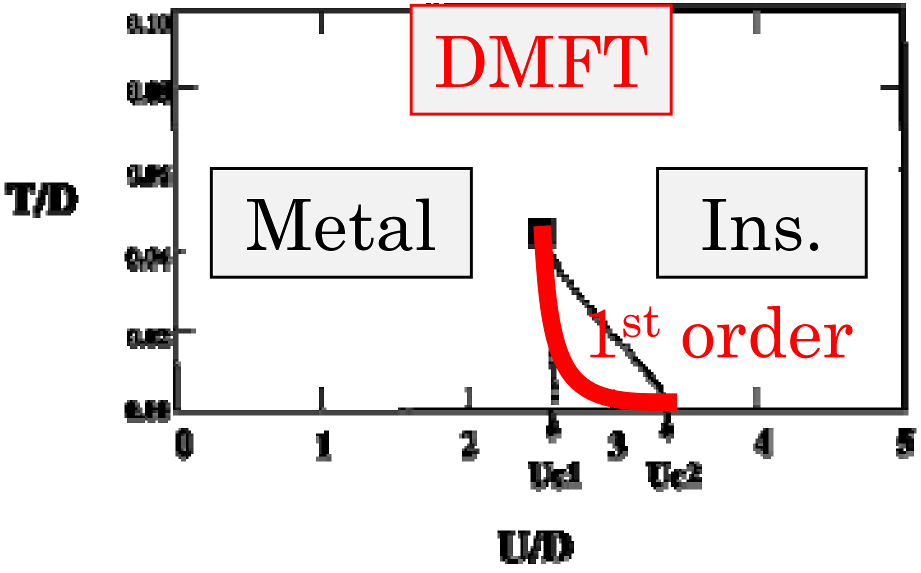
κ-$(ET)_2$-$Cu[N(CN)_2]Cl$



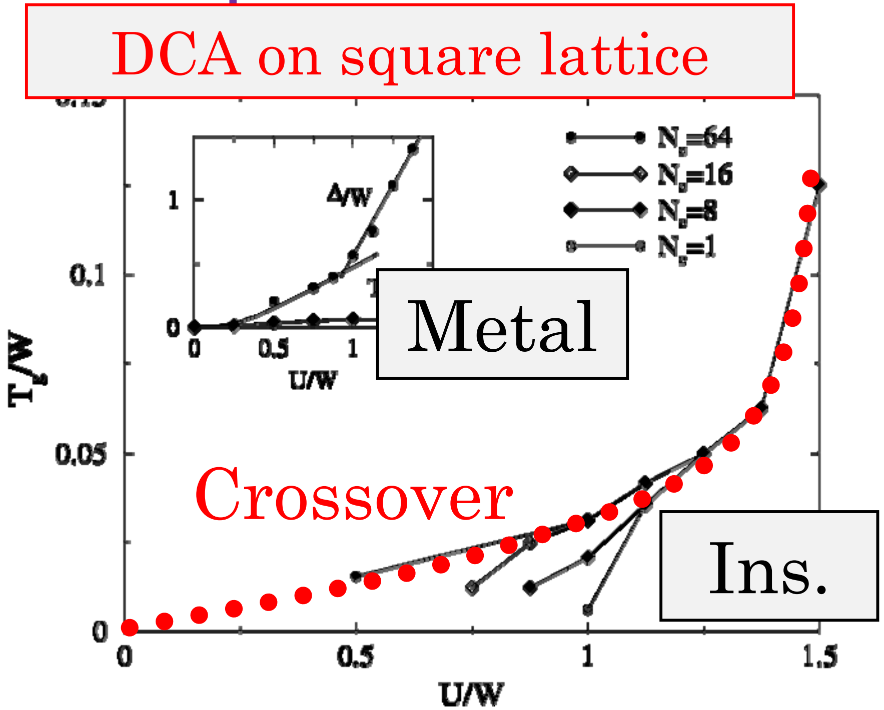
Anisotropic Hubbard model → Cellular-DMFT

effects of 1-site approx. or frustration?

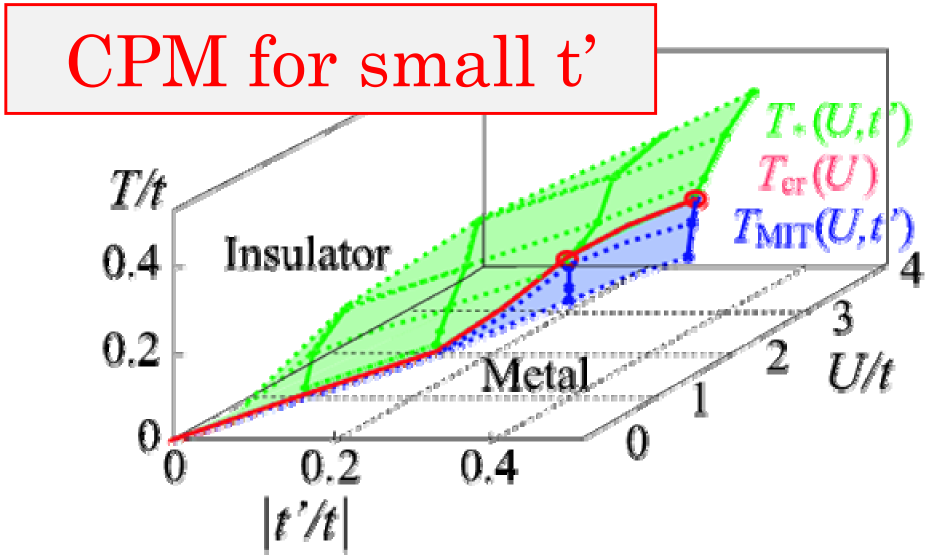
Mott transition at finite temperature



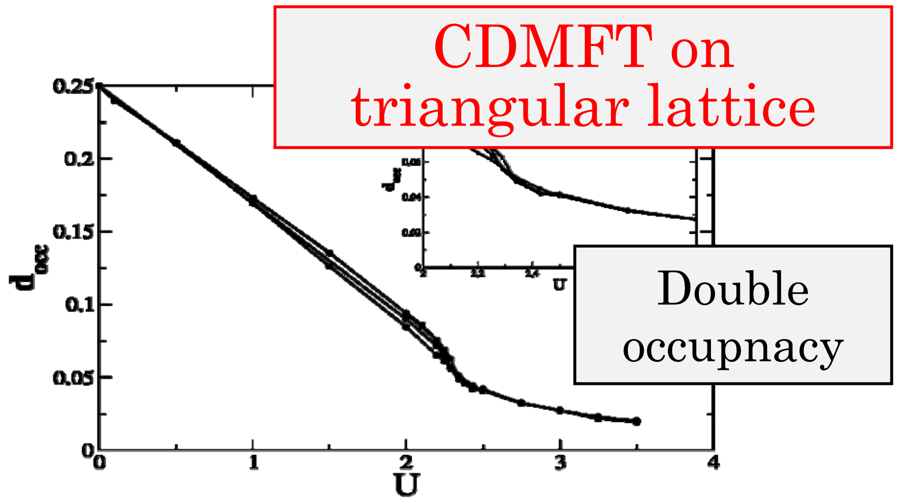
Georges et al., RMP, 68, 13 (1996)



Moukouri & Jarrell PRL 87, 167010 (2001)



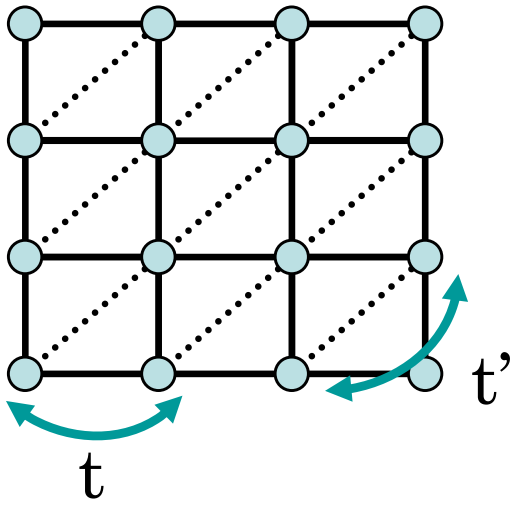
Onoda & Imada, PRB 67, 161102 (2003)



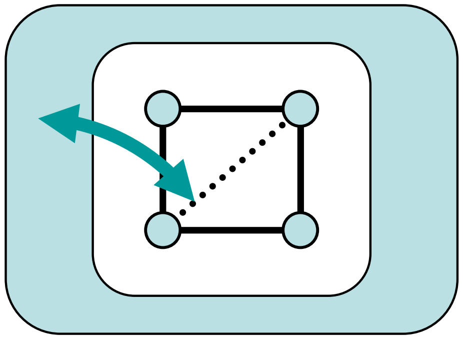
Parcollet et al., PRL 92, 226402 (2004)

Anisotropic Triangular Lattice Model

t-t'-U Hubbard model



4-site
cluster



effective cluster
model

anisotropic triangular lattice

- t'/t=0: regular square
- t'/t=1: regular triangular

t'/t controls frustration

κ–(BEDT-TTF)$_2$-Cu$_2$(CN)$_3$

t'/t ~ 1

κ-(BEDT-TTF)$_2$-Cu[N(CN)$_2$]Cl

t'/t ~ 0.8

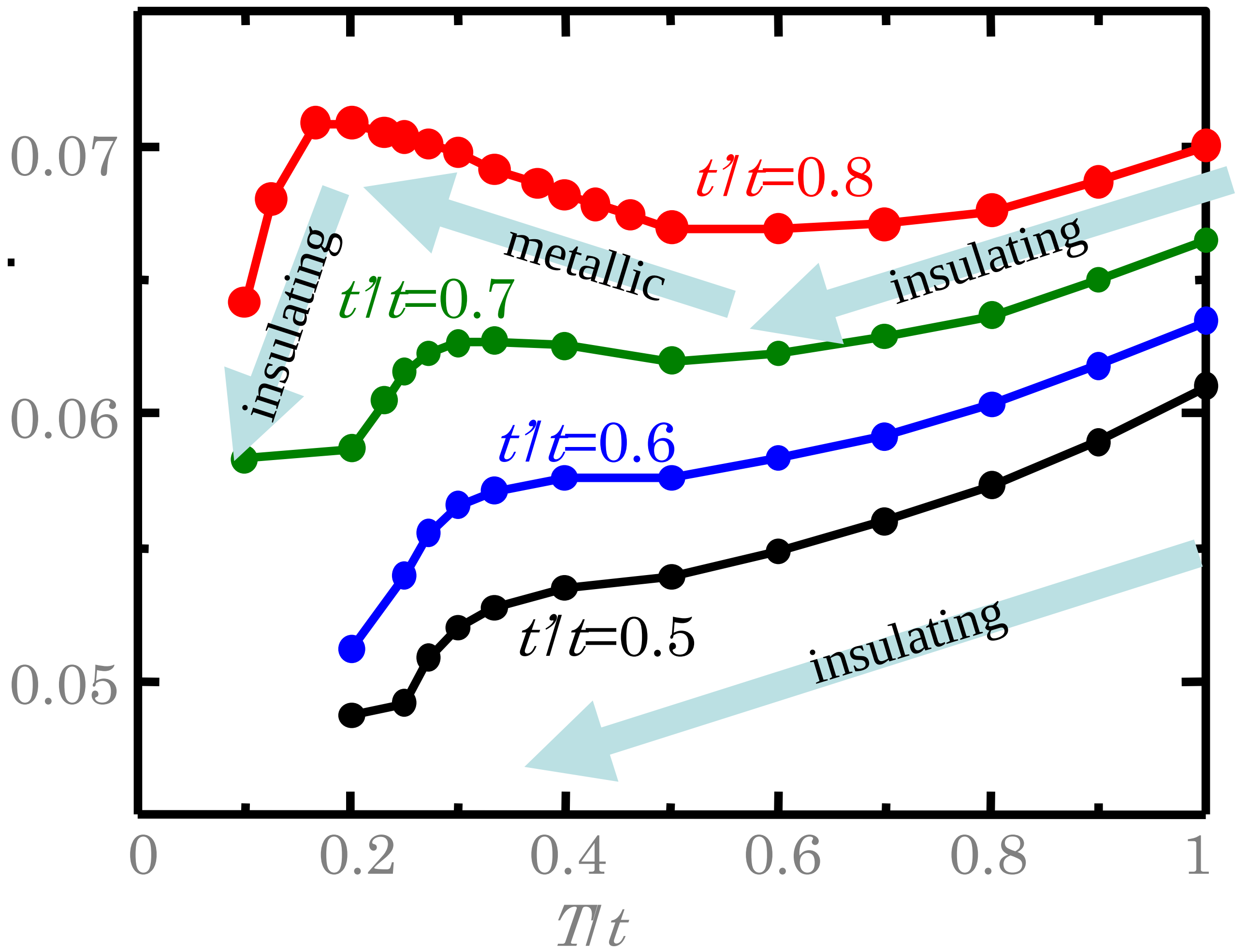
Temperature-Dependence of Double Occupancy

Insulator-Metal-Insulator Transition?

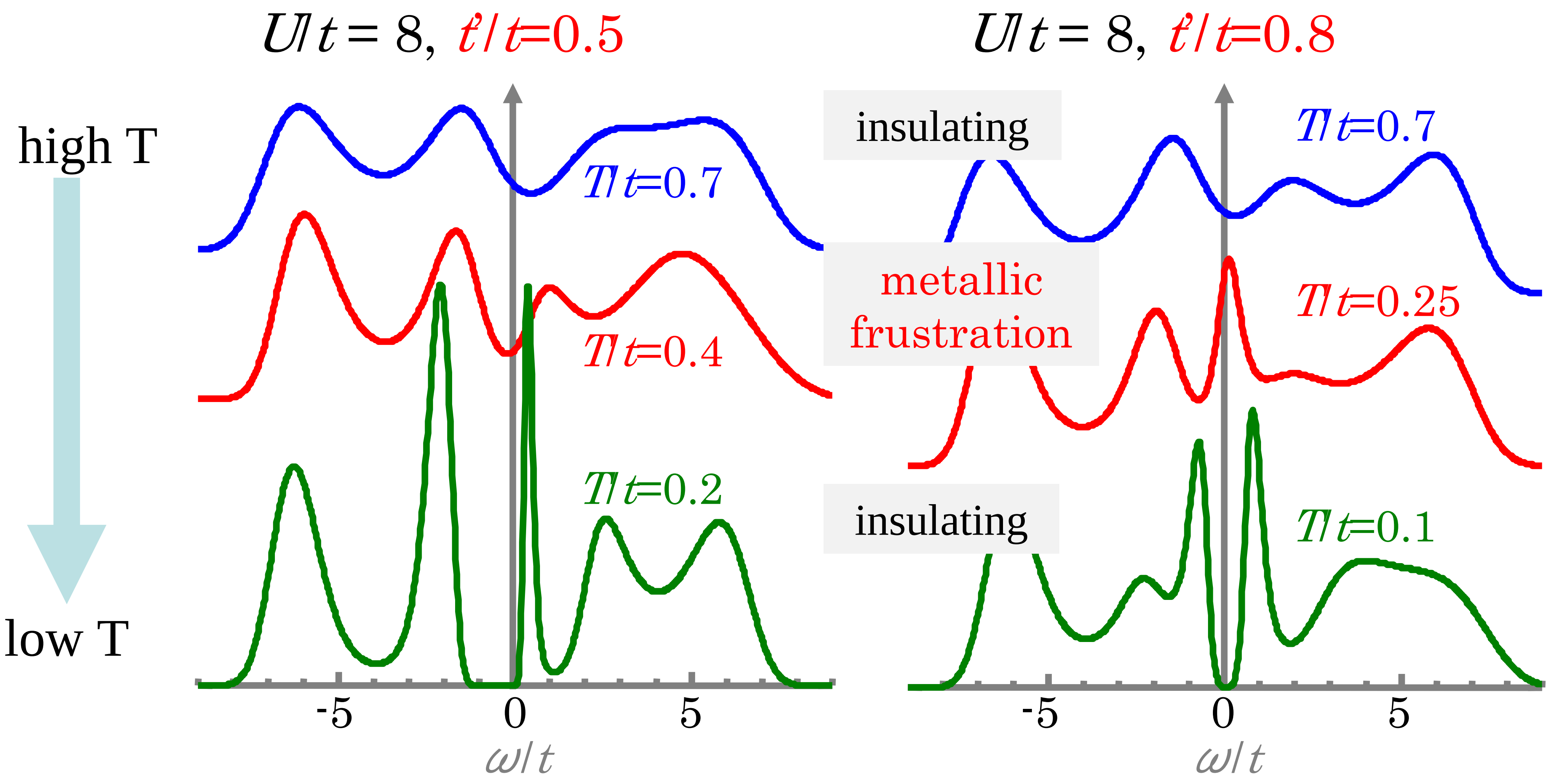
- large *t*'-strong GF: nonmonotonic T-dep.

- small *t*'-weak GF: almost monotonic insulating

Repulsion $U/t = 8$



Electron Spectral Function

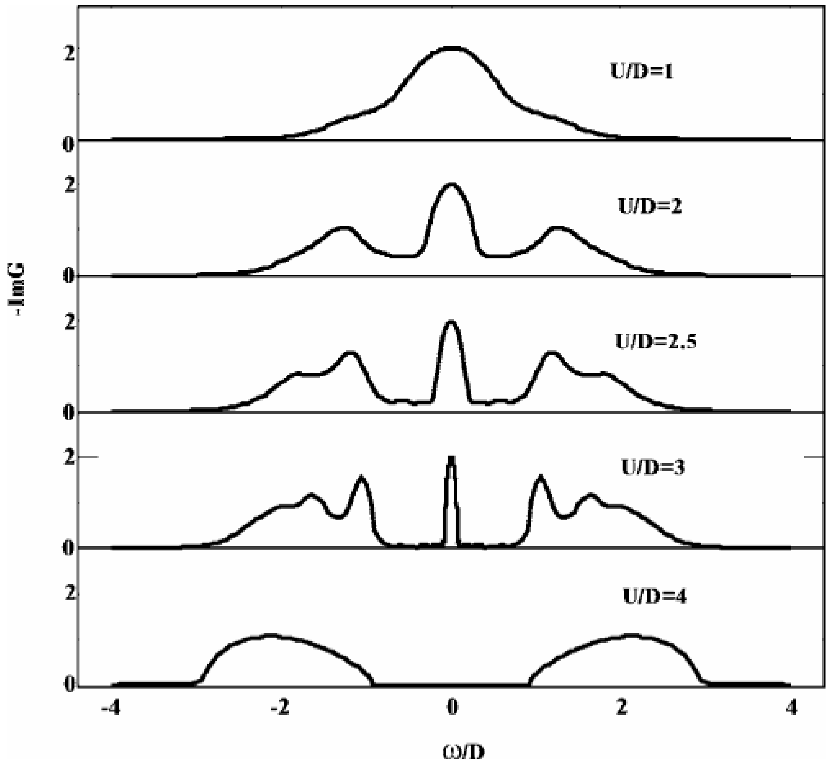


gap emerges :insulating

Reentrant: I→M → I

Local Spectral Function – single site DMFT

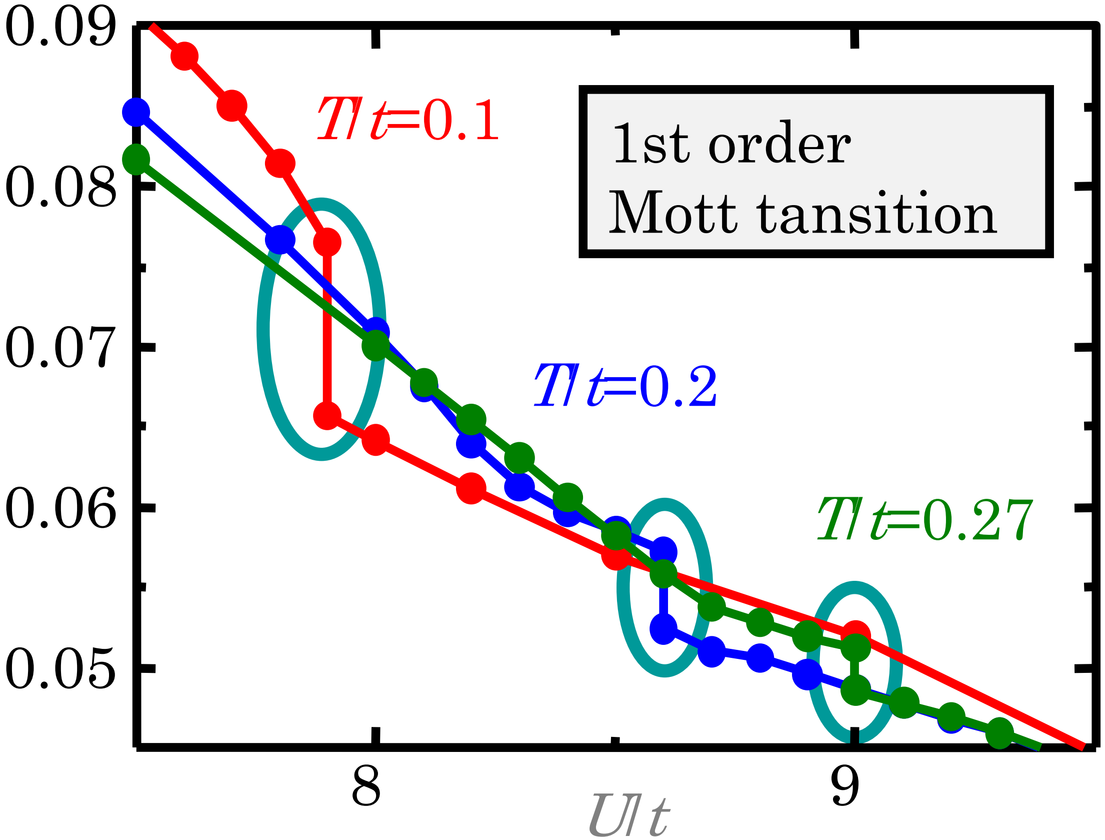
Mott transition is driven by transfer of spectral weight between high-energy Mott band and low-energy quasiparticle band



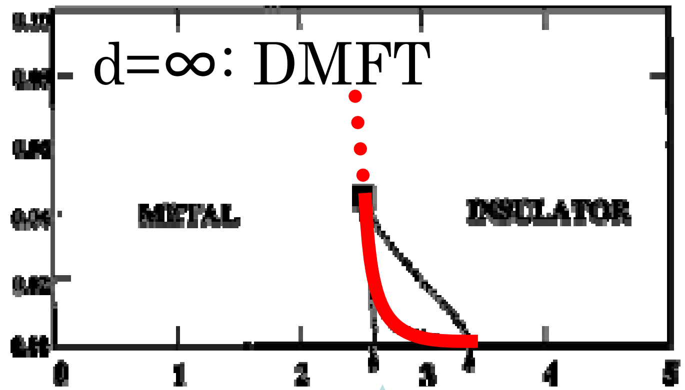
[Zhang, Rosenberg, and Kotliar, PRL, 1993]

Mott Transition

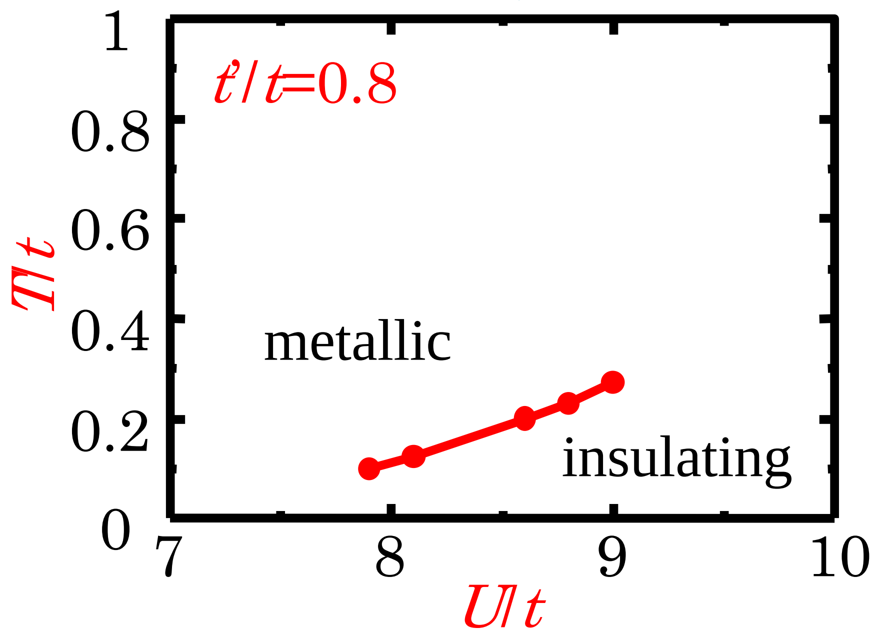
U-dep of double occupancy



higher T ⇒ larger U_c

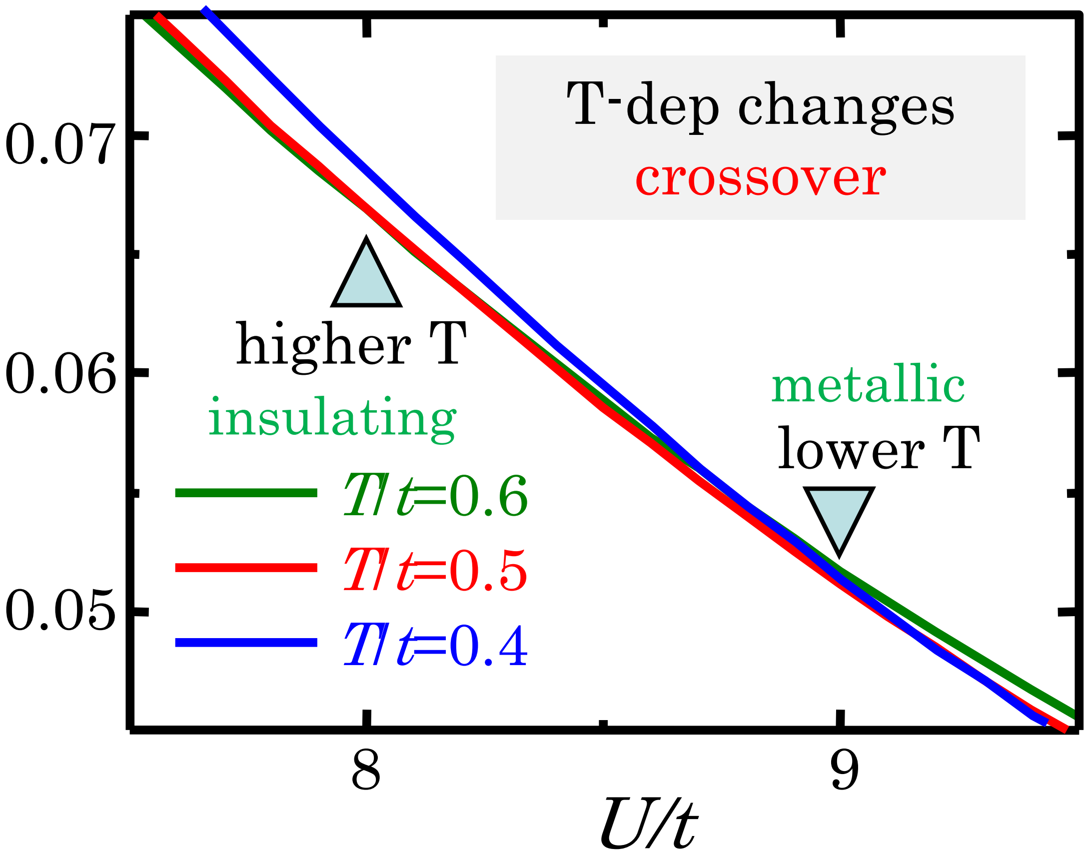


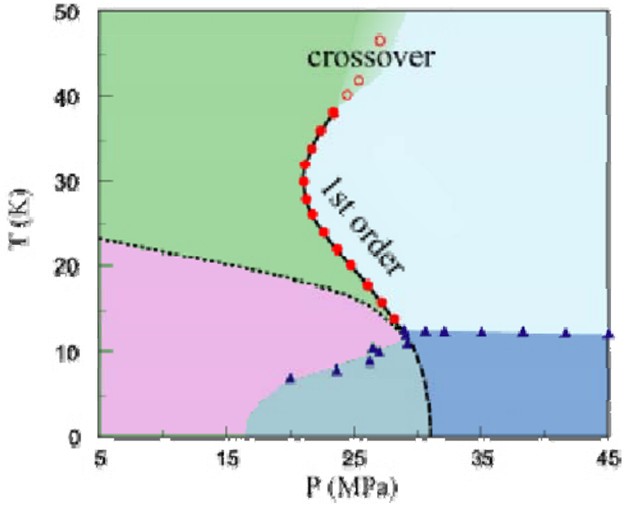
T-dep reversed

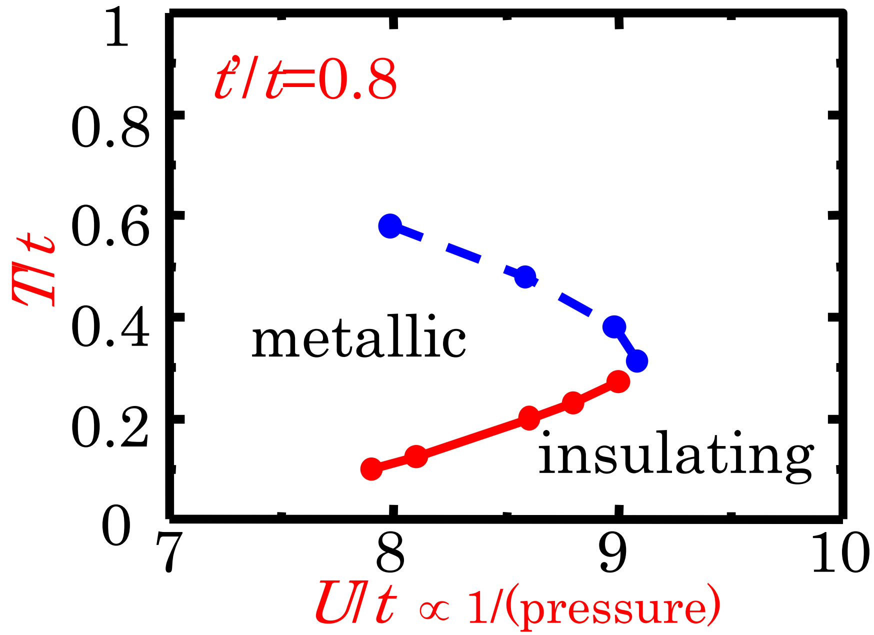


Crossover at a higher temperature

U-dep of double occupancy





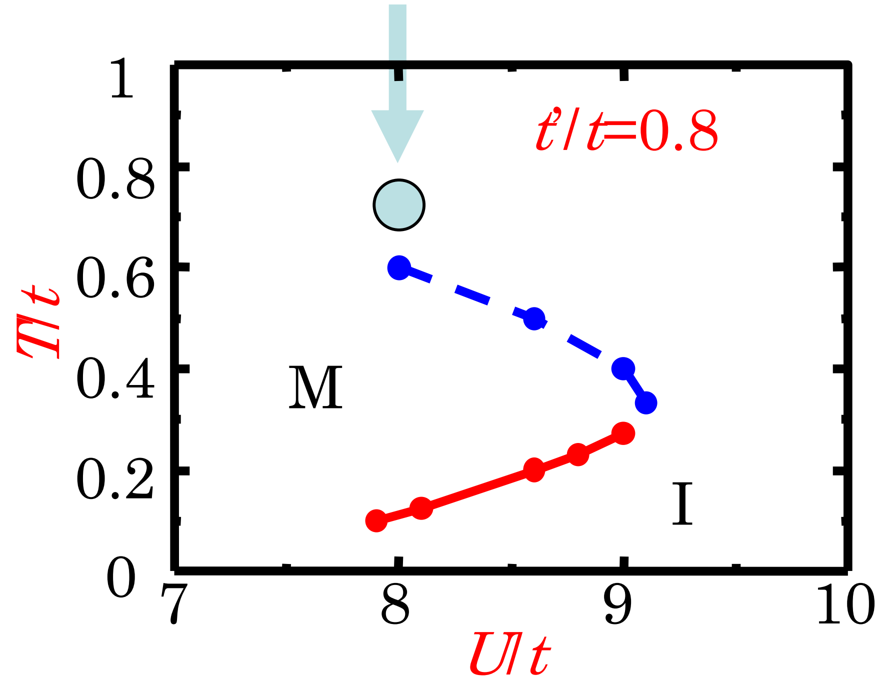


Reentrant !!

Electron Spectral Function $A_k(\omega)$: high-T insulating phase

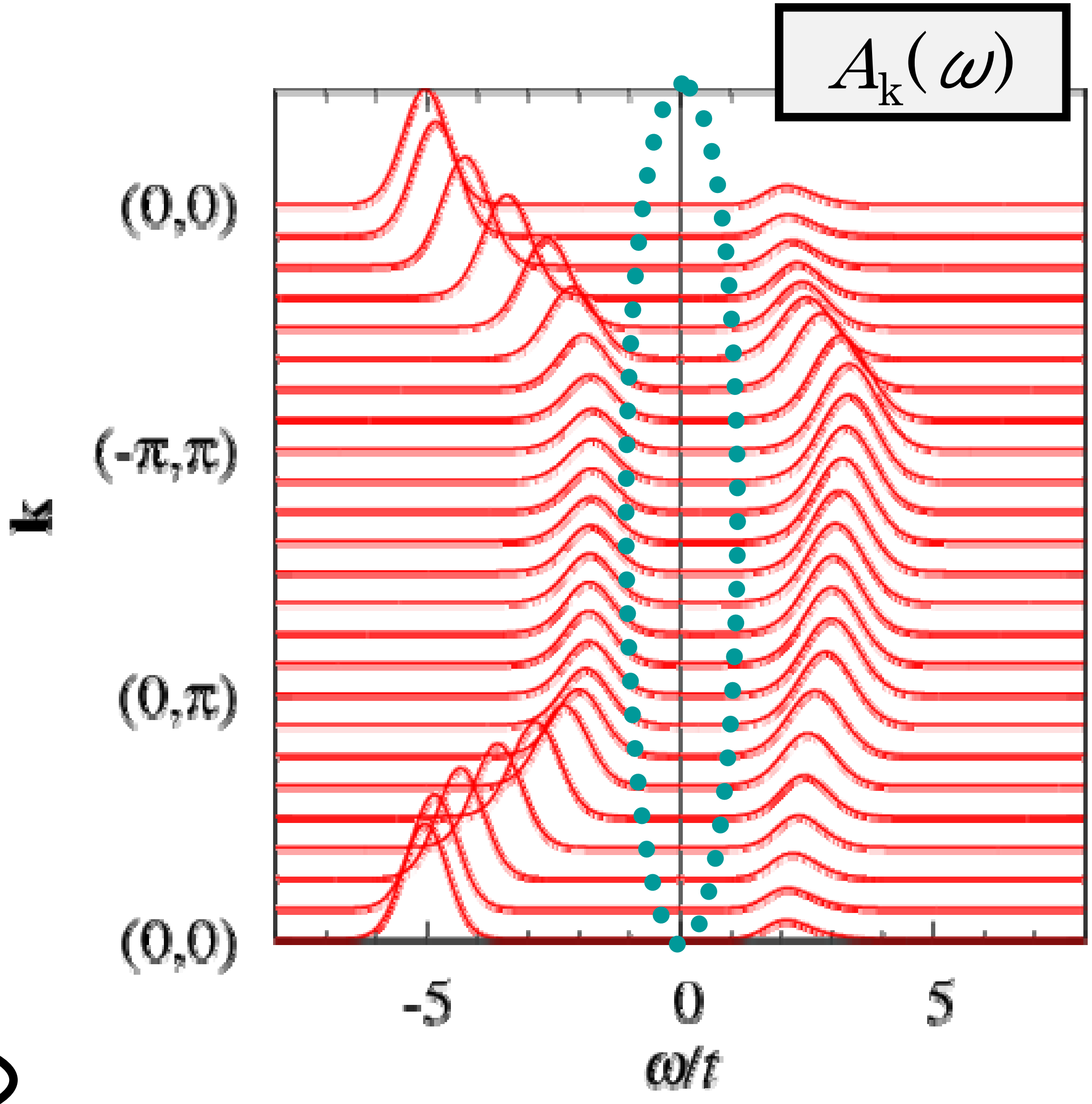
high-T insulating phase

U/t=8.0, T/t=0.7

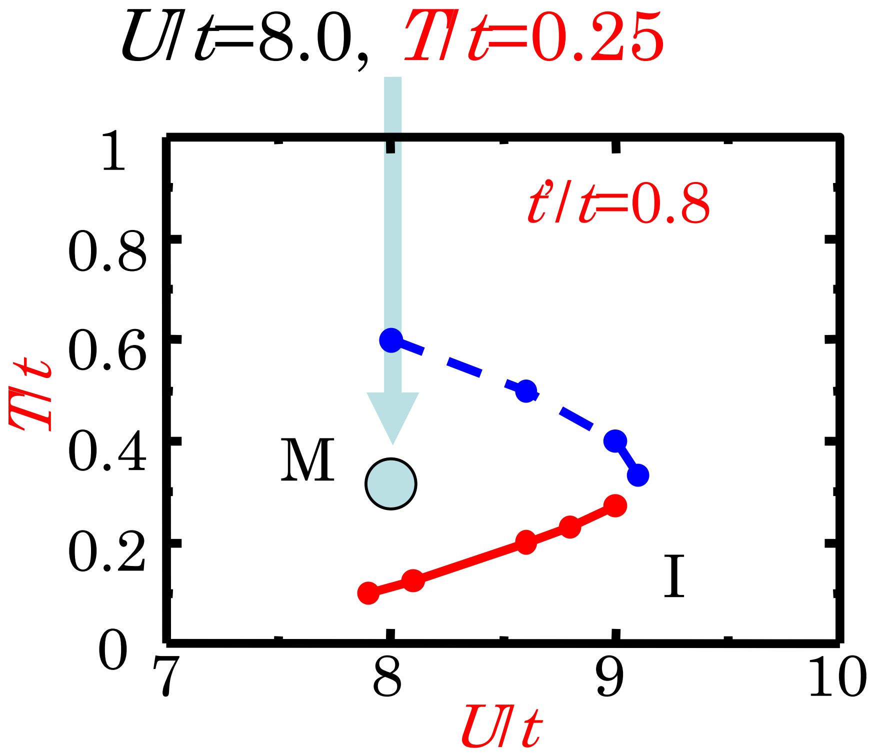


- no quasiparticles
- Hubbard gap

Local moment

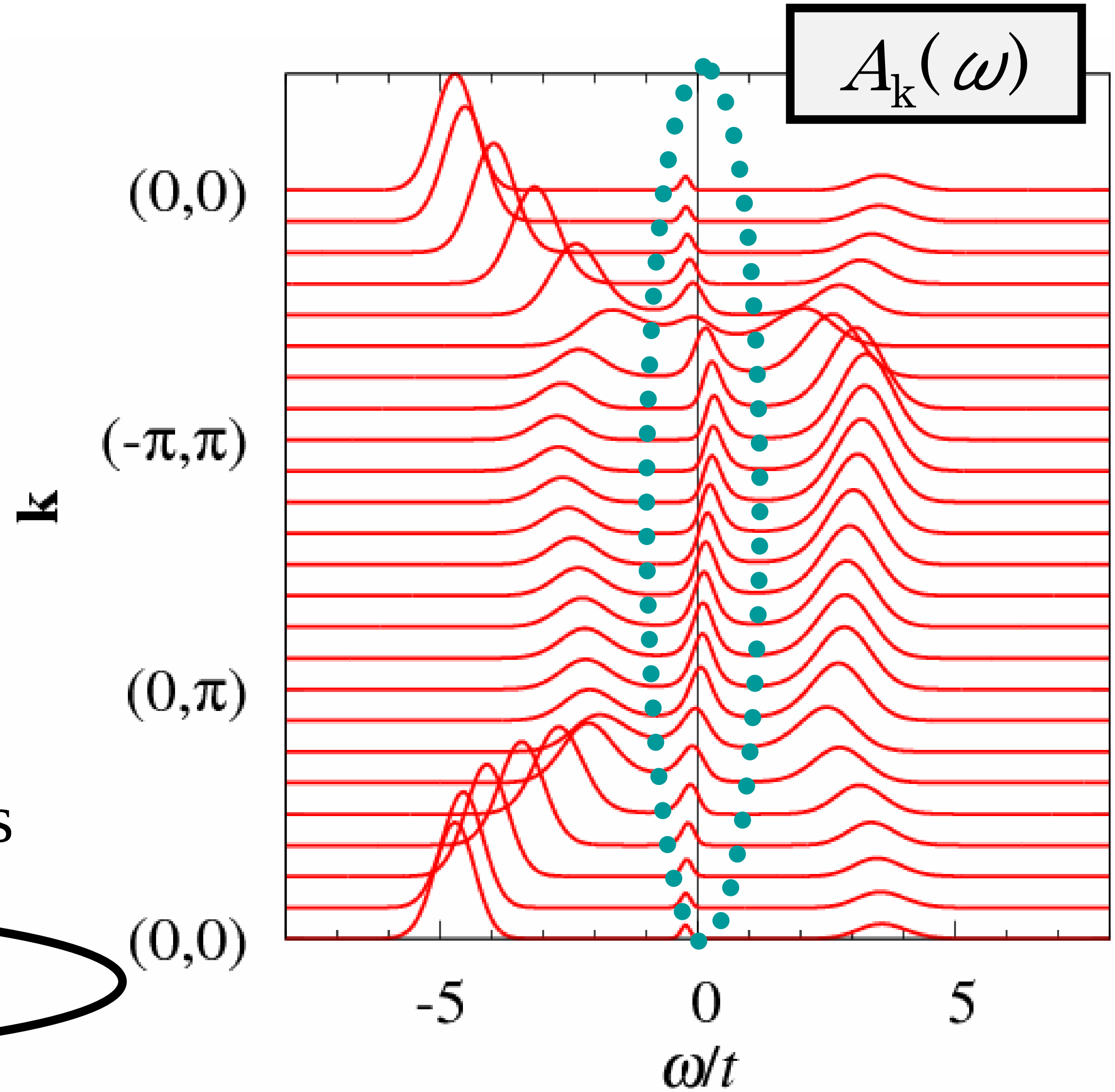


Electron Spectral Function $A_k(\omega)$: intermediate-T metallic phase

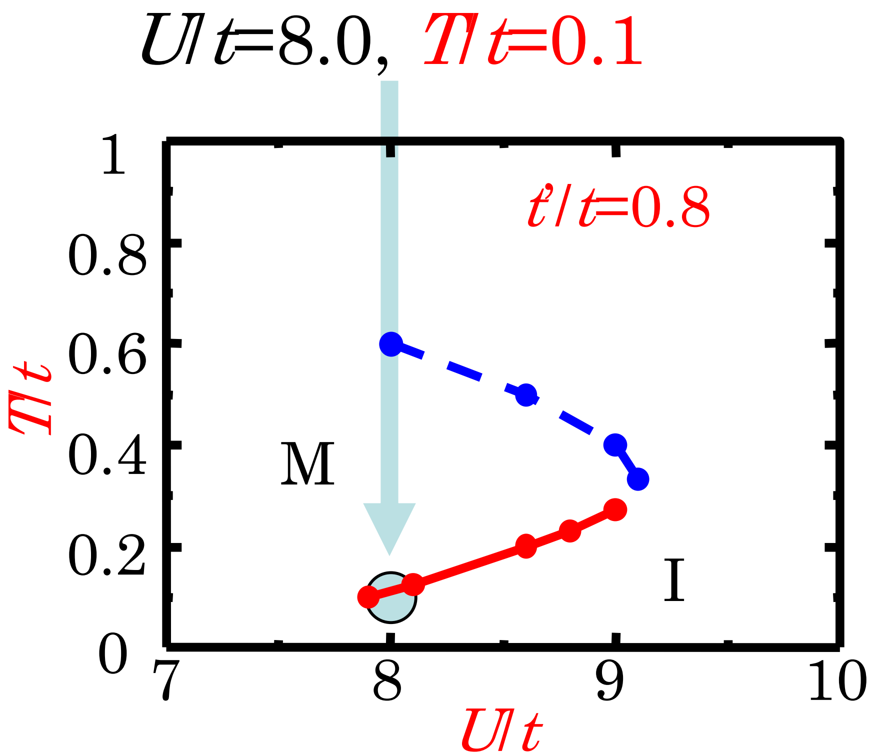


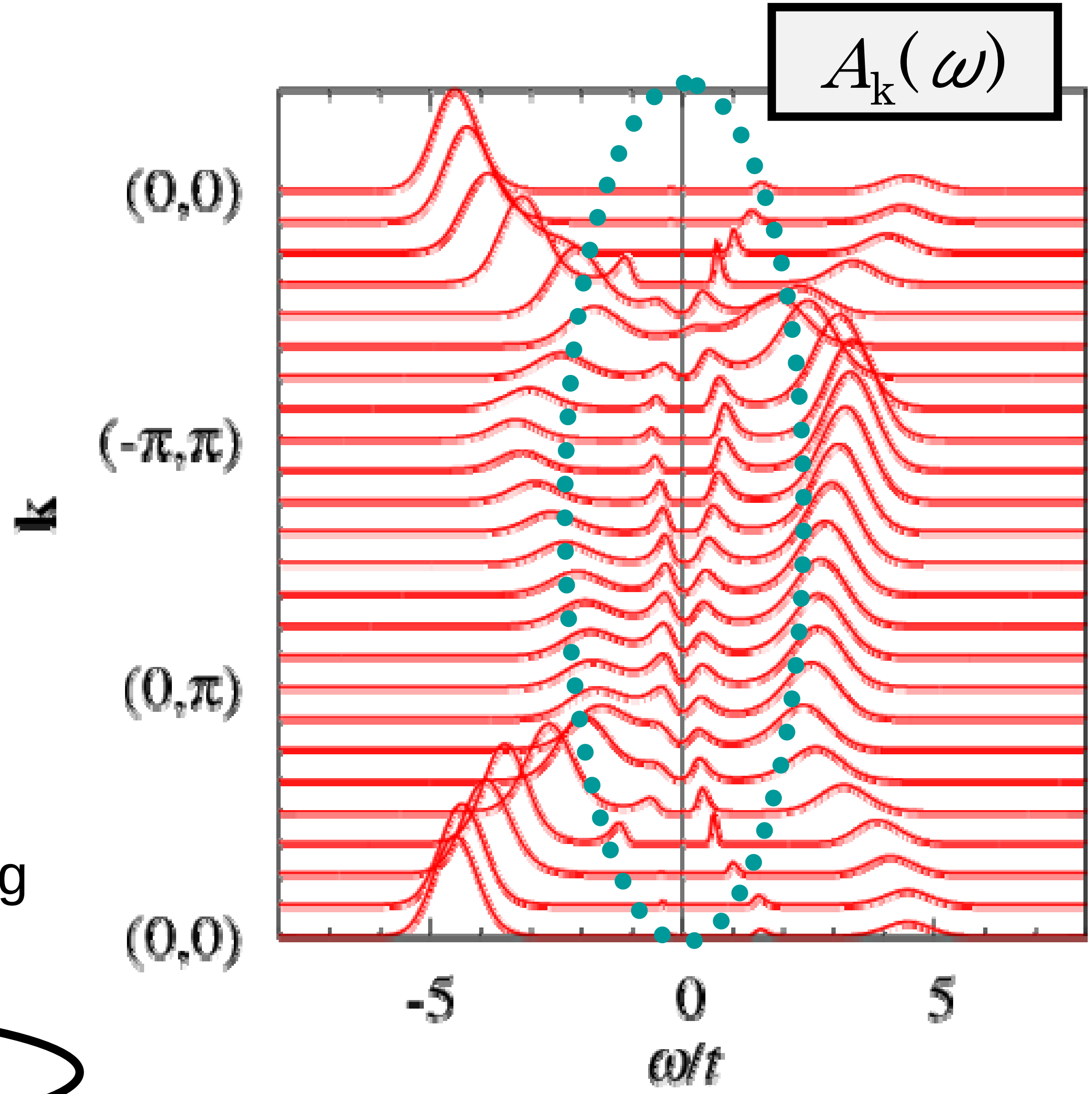
● sharp quasiparticle peaks

GF-induced metal



Electron Spectral Function $A_k(\omega)$: low-T insulating phase

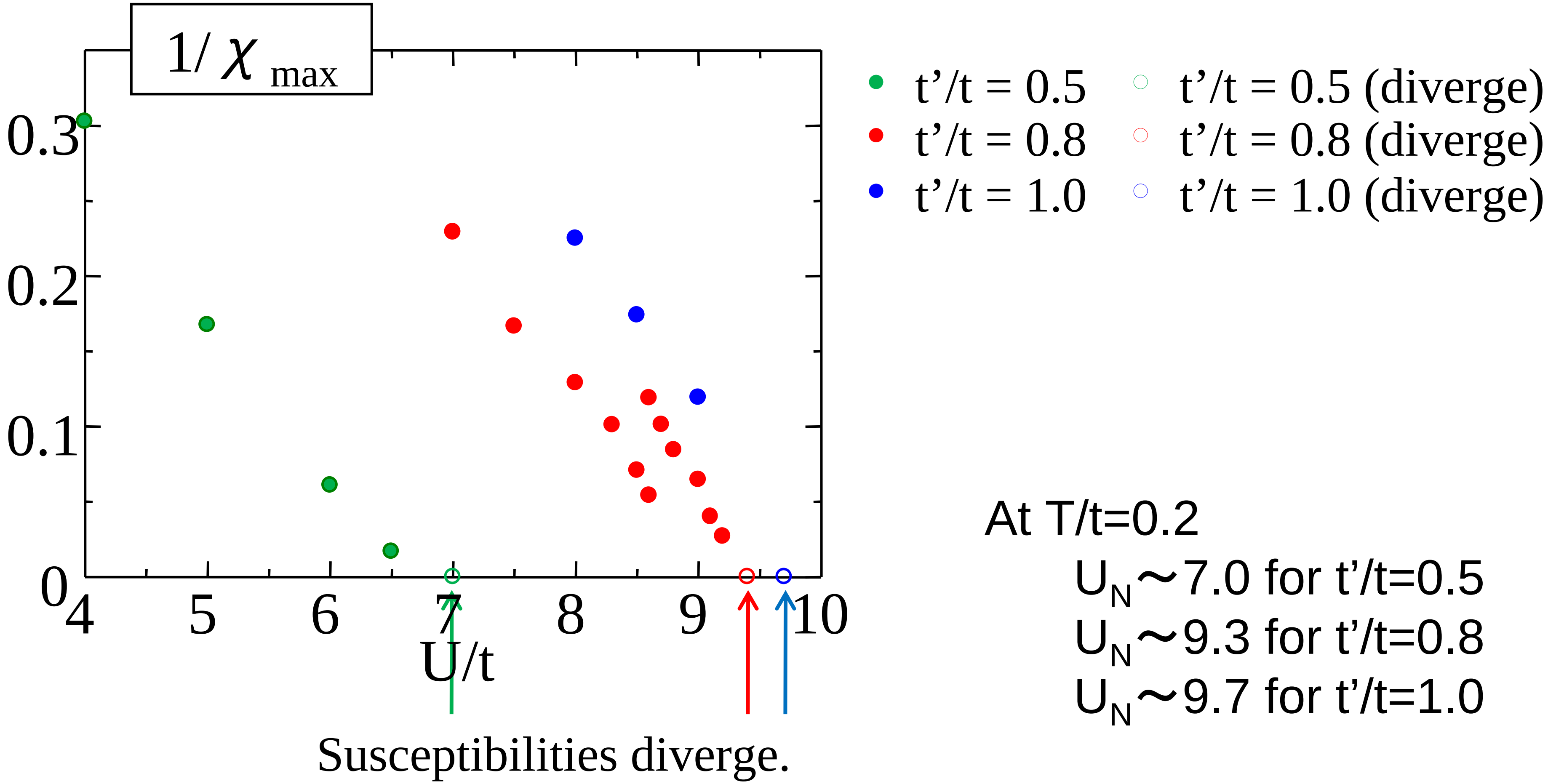




- quasiparticle peak splits
- different from high-T insulating phase

Magnetic exchange

Magnetic susceptibility for different t’ at T/t=0.2



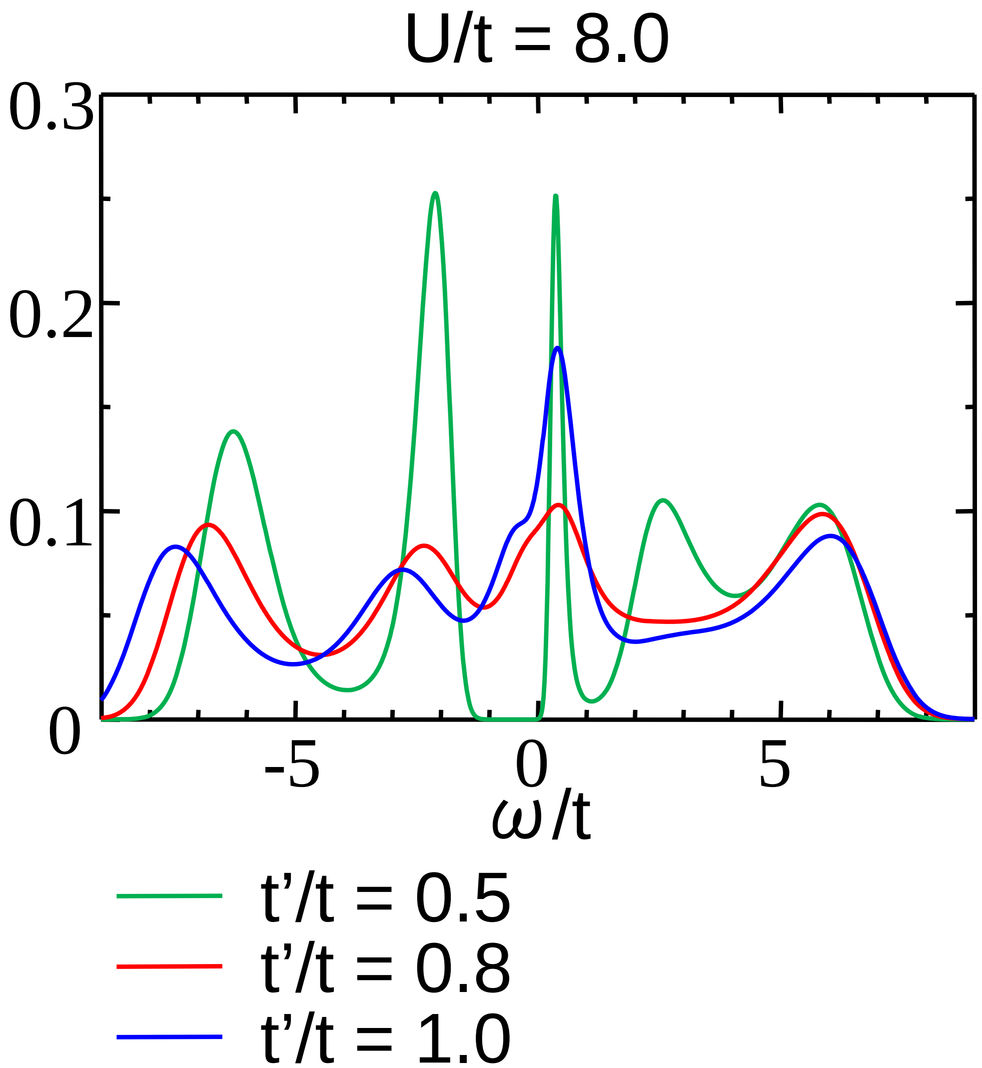
At T/t=0.2

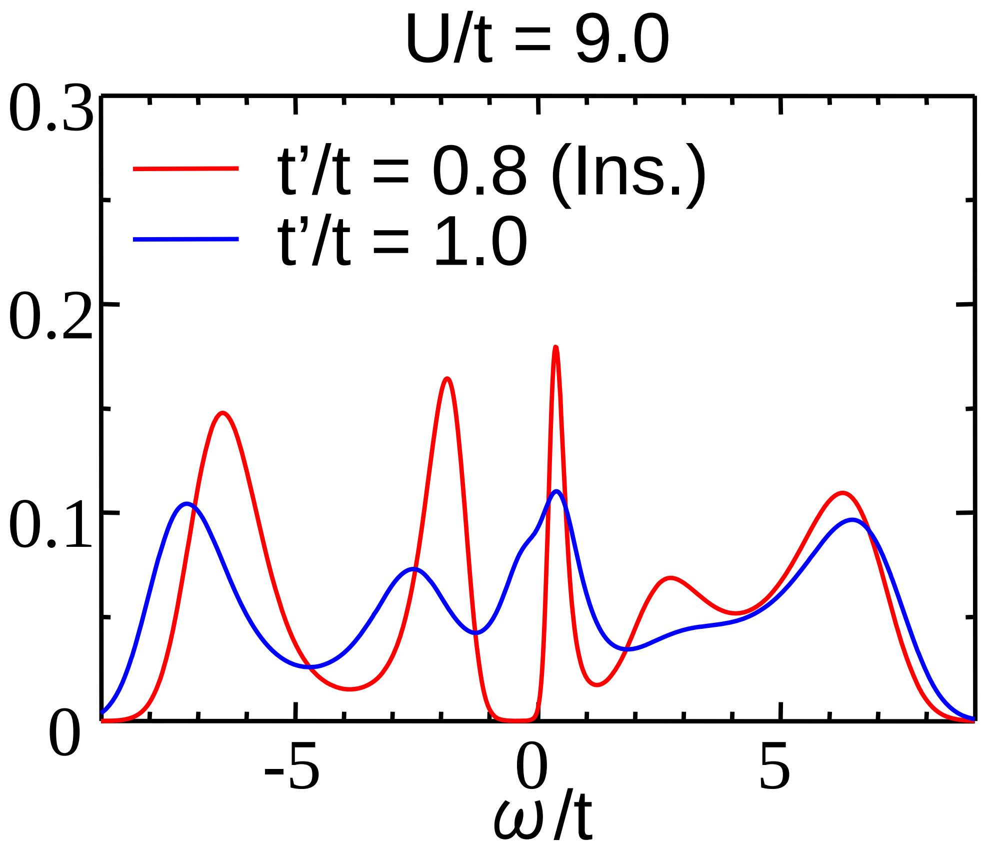
$U_N \sim 7.0$ for t’/t=0.5

$U_N \sim 9.3$ for t’/t=0.8

$U_N \sim 9.7$ for t’/t=1.0

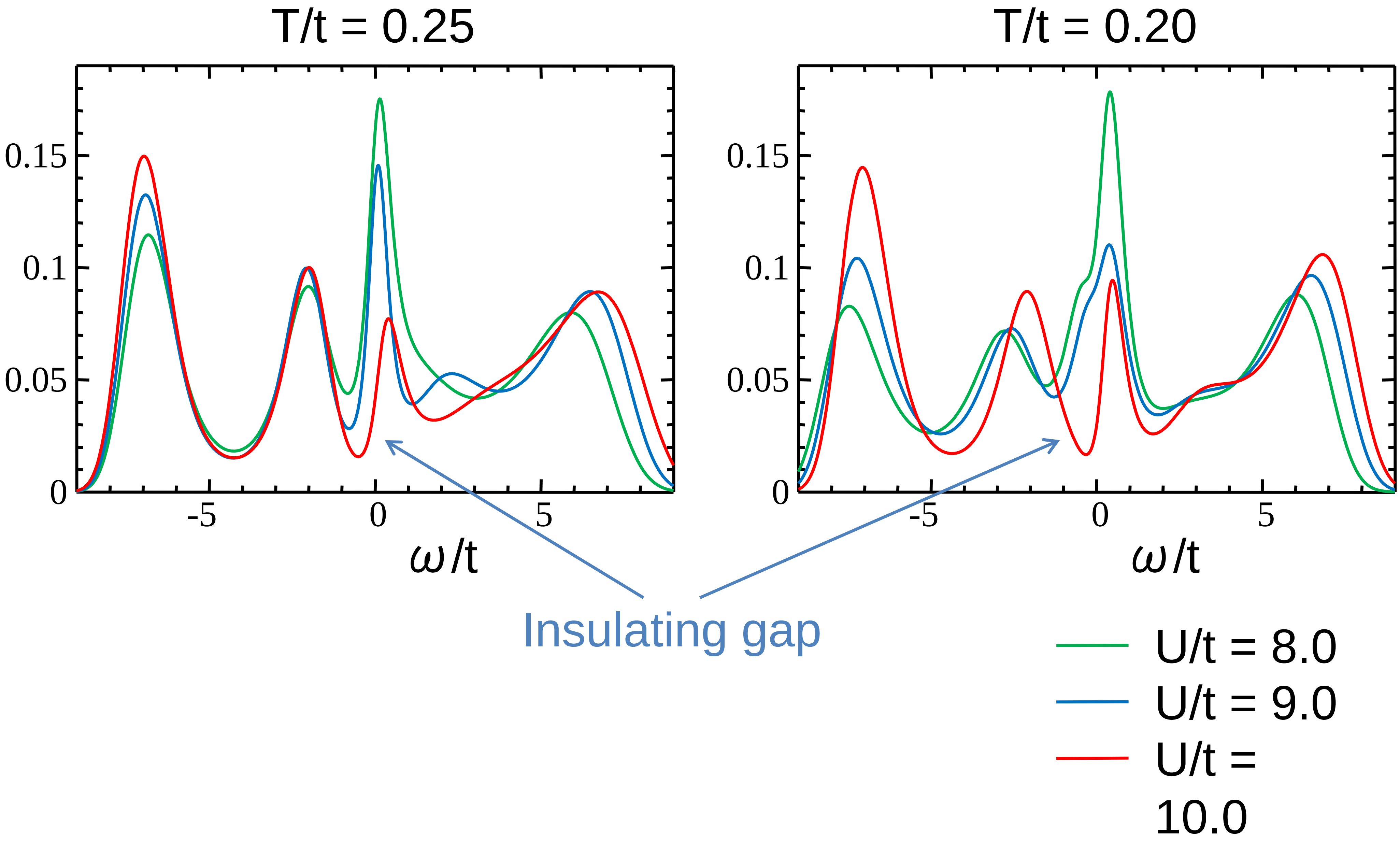
Density of States for different t’ at T/t=0.2





Geometrical frustration tends to stabilize the metallic phase

DOS on the triangular lattice (t'/t=1.0)

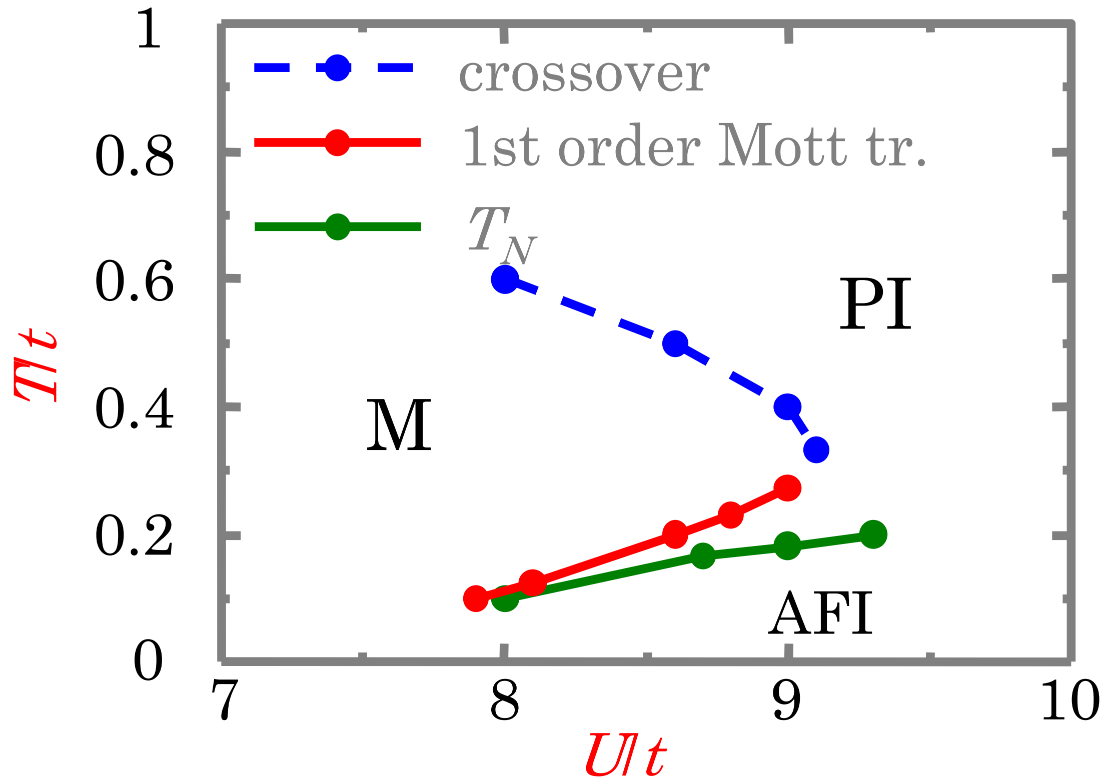


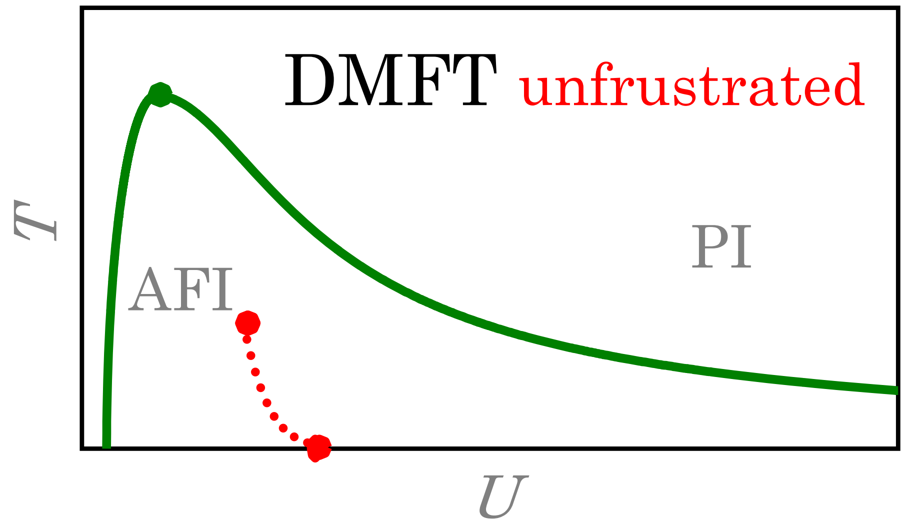
Magnetic Order

Cellular-DMFT → • magnetic LRO at T>0

weak 3-dim couplings stabilize LRO

anisotropy: $t'/t=0.8$





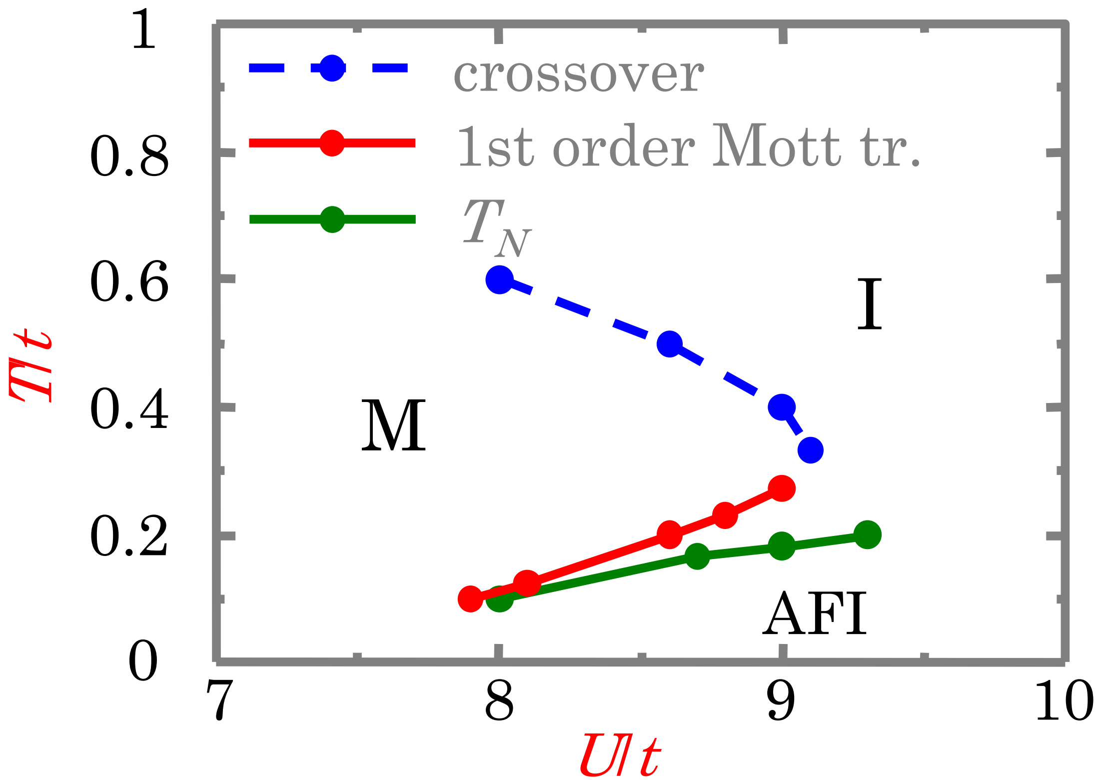
Georges et al. RMP 68, 13 (1996)
Zitzler et al. PRL 93 016406 (2004)

Mott transition is masked.

Frustrated system: Mott transition is NOT masked
paramagnetic insulator phase

Comparison with ∞–dim frustrated stsytems

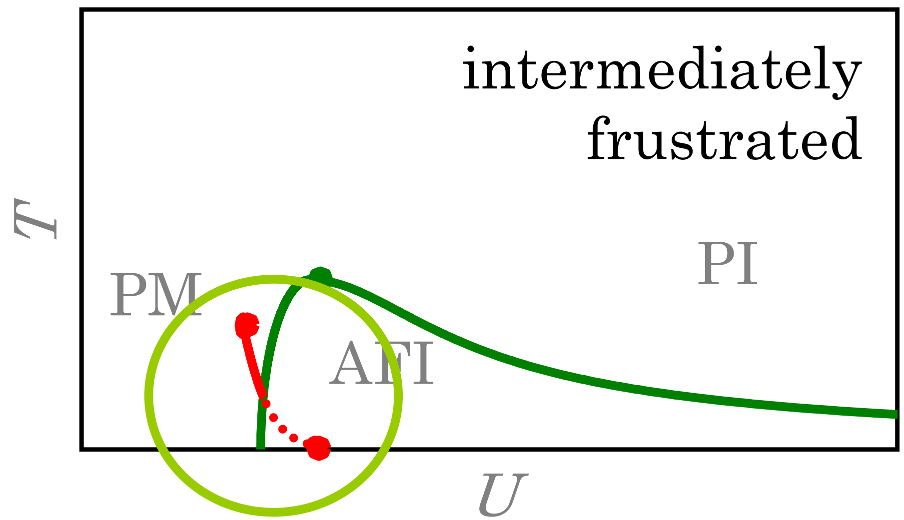
anisotropy: t'/t=0.8



Cellular-DMFT

DMFT: frustrated Bethe lattice

Zitzler et al. PRL 93 016406 (2004)



effects of short-range fluctuations

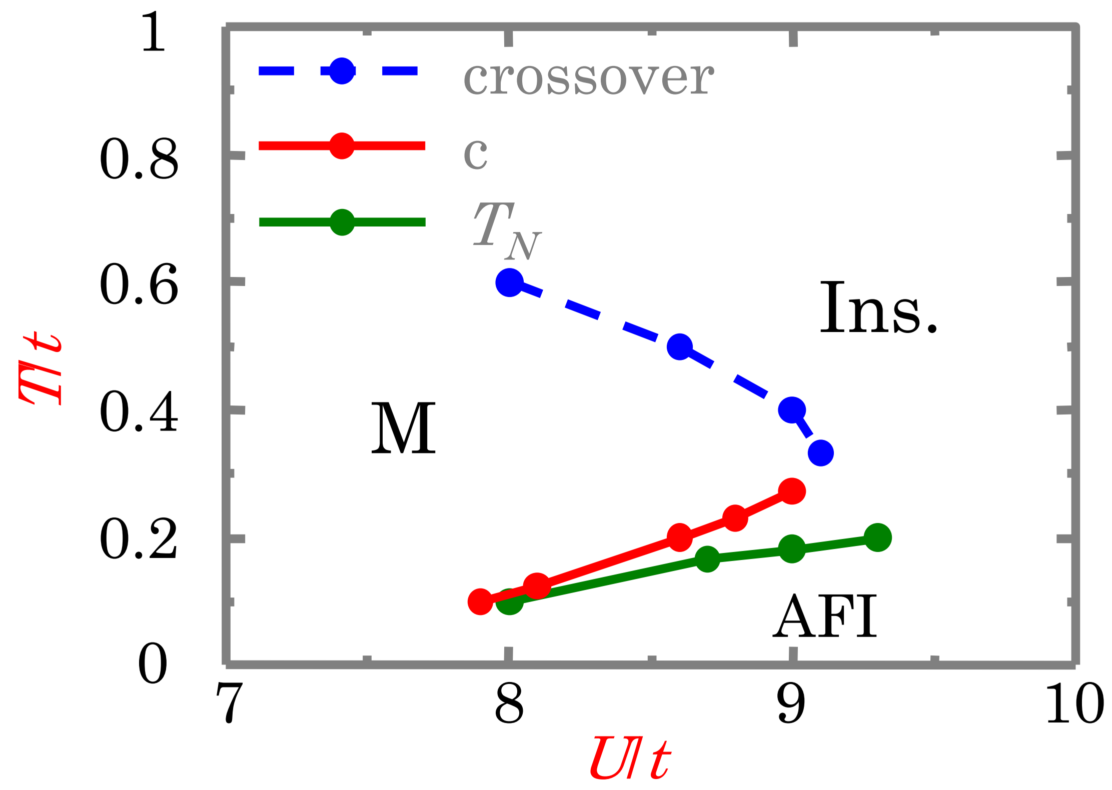
- U_c-curve changes its direction
- nonmagnetic insulating phase

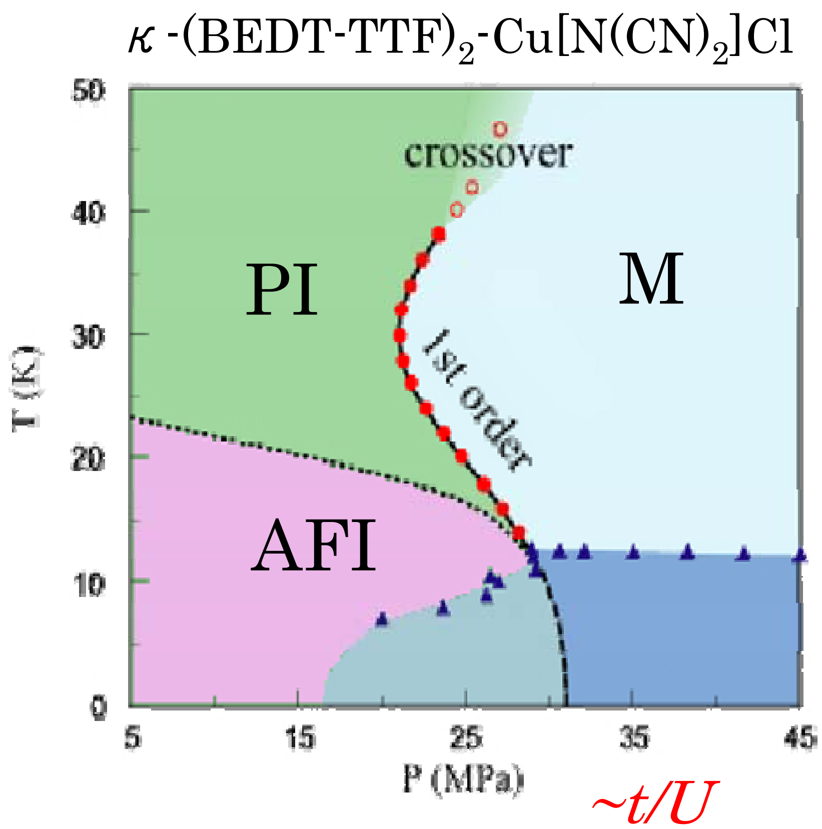
Comparison with Organic Materials

Cellular-DMFT →
- magnetic order at T>0
- stabilized by weak 3-dimensionality

anisotropy: $t'/t=0.8$

Maier et al., PRL 85, 1524 (2000)





consistent with experiments

Summary (2)

Anisotropic Triangular Lattice Hubbard model (mainly t'/t=0.8)
Cellular dynamical mean field theory

- Metal-insulator transition
  - different slope of transition line from unfrustrated systems – entropy effects
- Intermediate Correlation Regime:
  - "reentrant" insulator → metal → insulator transition
  - heavy quasiparticle formation in the intermediate metallic phase
  - gap formation inside heavy qp band
- Magnetic instability
  - transition to paramagnetic insulator phase
  - magnetic phase appears at lower temperature

PART C

Trimer Phase of bilinear-biquadratic zigzag chain

collab. with Philippe Corboz (ETH Zurich)
Andreas Läuchli (EPF Lausanne)
Keisuke Totsuka (YITP, Kyoto U.)

[Corboz, Lauchli, Totsuka and Tsunetsugu, cond-mat.st-el/0707.1195
in press in Phys. Rev. B]

3-sublattice Antiferro Nematic Order

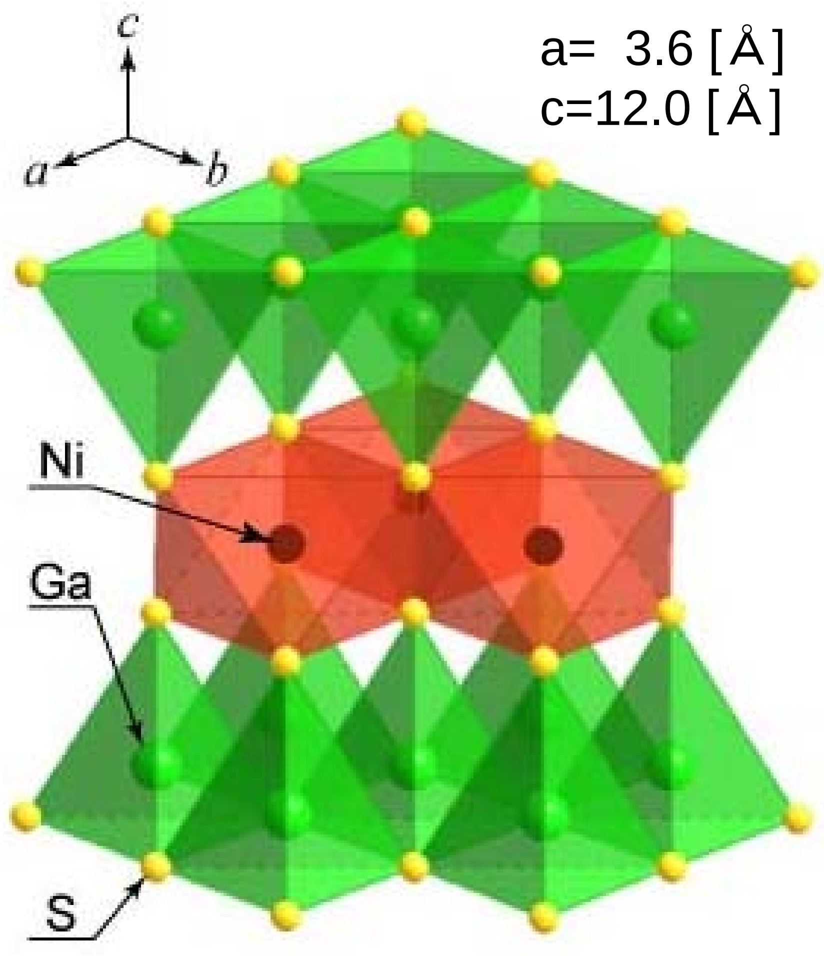
[Tsunetsugu and Arikawa, JPSJ 75, 083701 (2006)]

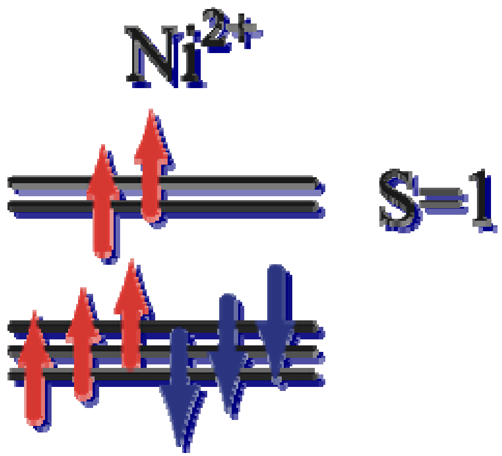
$NiGa_2S_4$ - structure

- S=1 spin system (Ni^{2+})
- Quasi-2D triangular structure

Ref: Nakatsuji et al., Science **309**, 1697 ('05)

a= 3.6 [Å]
c=12.0 [Å]



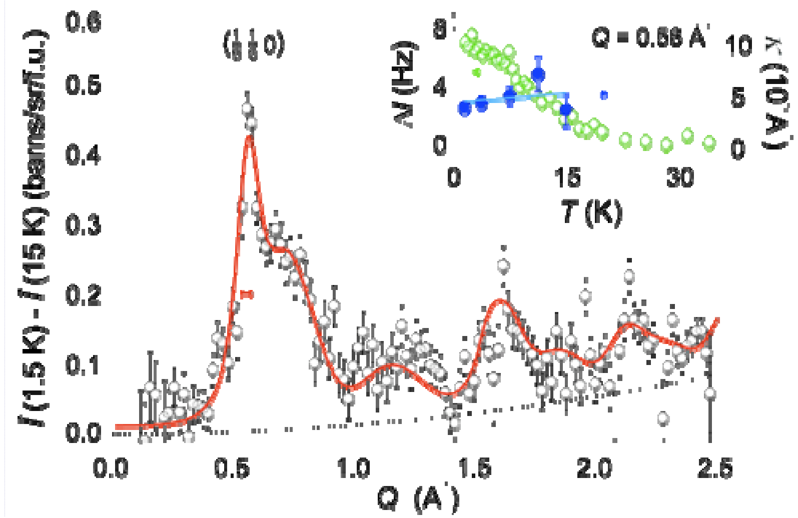


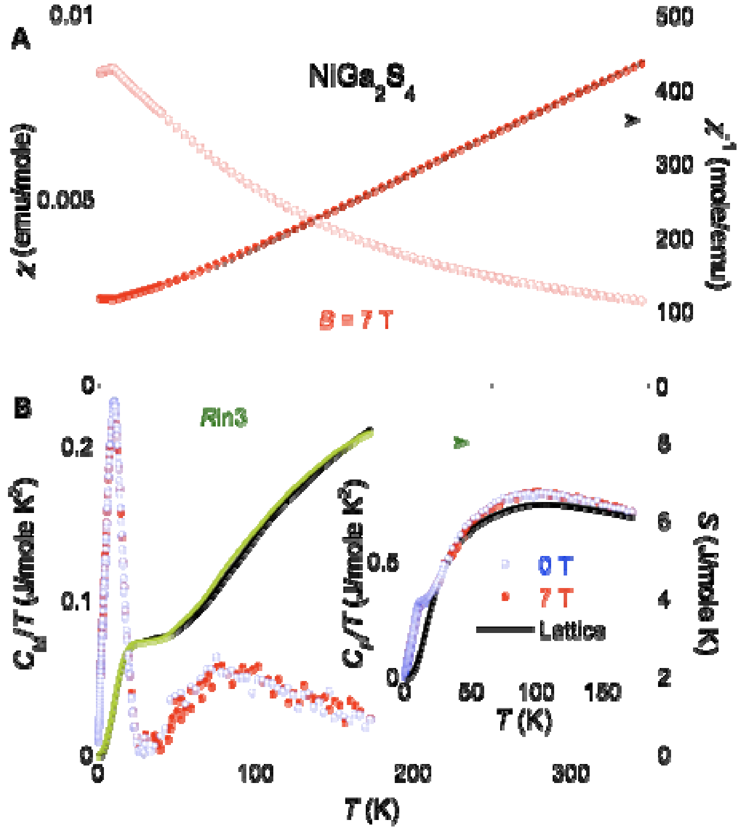
NO orbital degrees of freedom

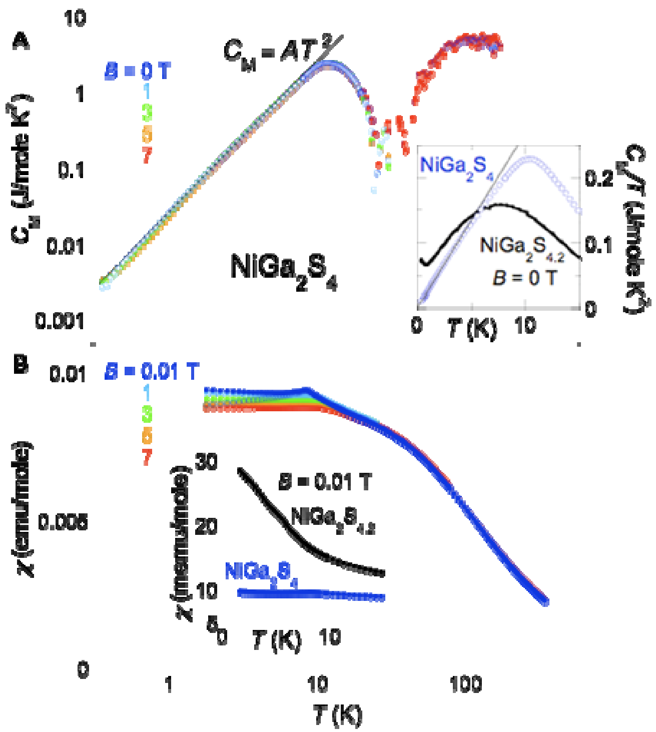
$NiGa_2S_4$: spin liquid behavior

Nakatsuji et al., Science **309**, 1697 ('05)

- No phase transition down to 0.35[K]
- $C(T) \propto T^2$
  -> presence of gapless excitations
- Finite $\chi \approx 8\times10^{-3}$[emu/mole] at T≈0
- Finite $\xi \approx 25$[Å] at T≈0
- Spatial modulation in spin correlations Q≈(1/6,1/6,0)







Difficulty of Ordinary Scenarios

[A] magnetic LRO with $T_c < 0.3$K

- $T = 0$: magnetic LRO (eg, 120-degree structure)
- $T > 0$: paramagnetic (Mermin-Wagner)

consistent:
no singularity in $C(T)$ and $\chi(T)$

NOT consistent:
non-divergent ξ (T) neutron scattering

[B] spin gap state

(a)gap(S=1 excitations) > 0
gap(S=0 excitations) > 0
(eg. Haldane chain,
Shastry-Sutherland system $SrCu_2(BO_3)_2$)

(b)gap(S=1 excitations) > 0
gap(S=0 excitations) = 0
(eg. S=1/2 Kagome)

consistent:
non-divergent ξ(T)

NOT consistent:
(a) : $C(T) \propto T^2$
(b) : $\chi(T \to 0)$ = finite

Possibility of Unconventional Order

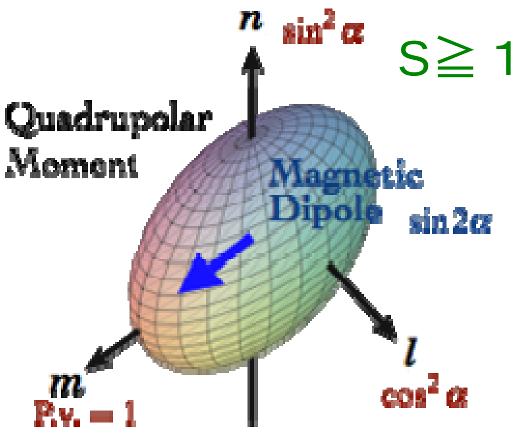
- Hidden **non-"magnetic"** order？
  Antiferro order of spin **quadrupoles**
- spontaneous breaking of spin rotation symmetry
- spin inversion sym. is NOT broken

Blume, Chen&Levy...

Non-magnetic order: $\langle \mathbf{S} \rangle = 0$

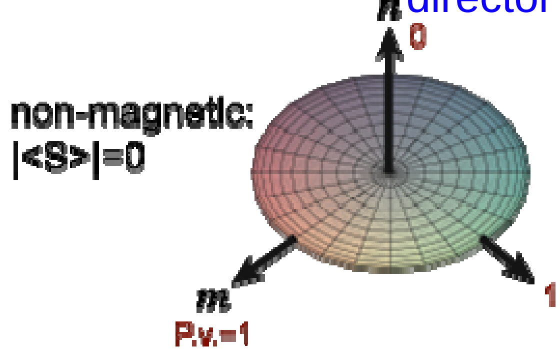
Order parameter: $Q_{\mu\nu} = \frac{1}{2}\langle S_\mu S_\nu + S_\nu S_\mu \rangle - \frac{1}{3}\delta_{\mu\nu} S(S+1)$

anisotropy of spin fluctuations



S≧ 1

$$\sum_\mu Q_{\mu\mu} = S(S+1)$$



Phenomenological Model

low-energy effective model

$$H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + K \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2$$

Biquadratic term

(cf. 4th order of hopping process)

$$(\mathbf{S}_i \cdot \mathbf{S}_j)^2 = \sum_{\mu\nu} (S_i^\mu S_i^\nu)(S_j^\mu S_j^\nu)$$

$$= \tfrac{1}{4} \sum_{\mu\nu} Q_i^{\mu\nu} Q_j^{\mu\nu} - \tfrac{1}{2} \mathbf{S}_i \cdot \mathbf{S}_j$$

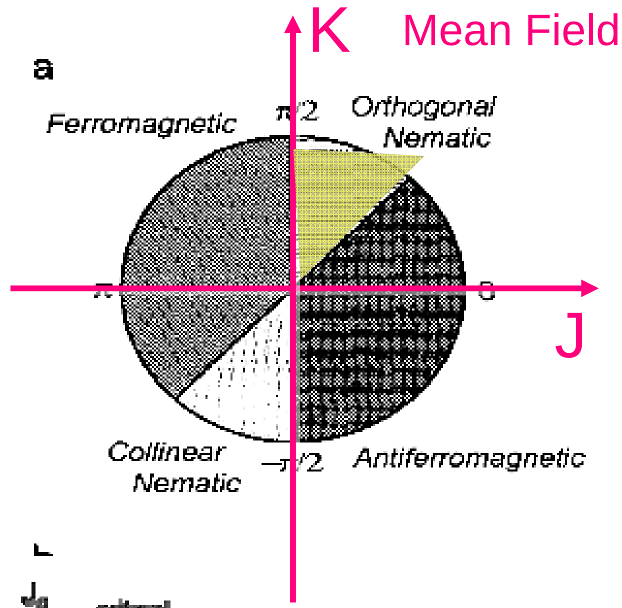
$$= \tfrac{1}{4} (\mathbf{n}_i \cdot \mathbf{n}_j)^2 - \tfrac{1}{2} \mathbf{S}_i \cdot \mathbf{S}_j$$

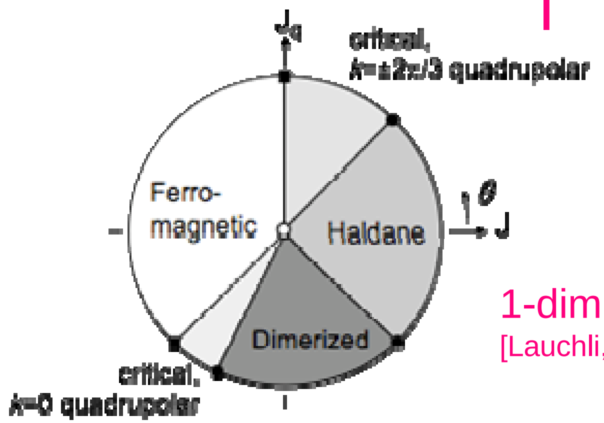
quadrupole couplings

Chen & Levy ('71)
Matveev ('74)
Andreev & Grishchuk ('84)
Fath & Solyom ('95)
Schollwock, Jolicoeur & Garel ('96)
Harada & Kawashima ('02)
Lauchli, Schmidt & Trebst ('03)

Bilinear-Biquadratic model

Mean Field



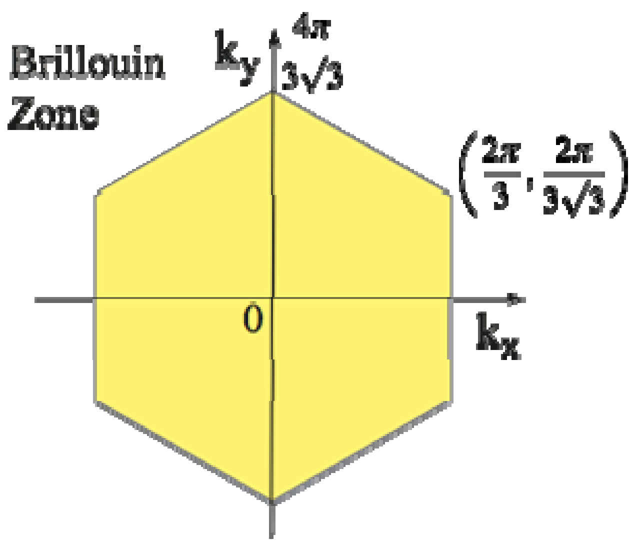


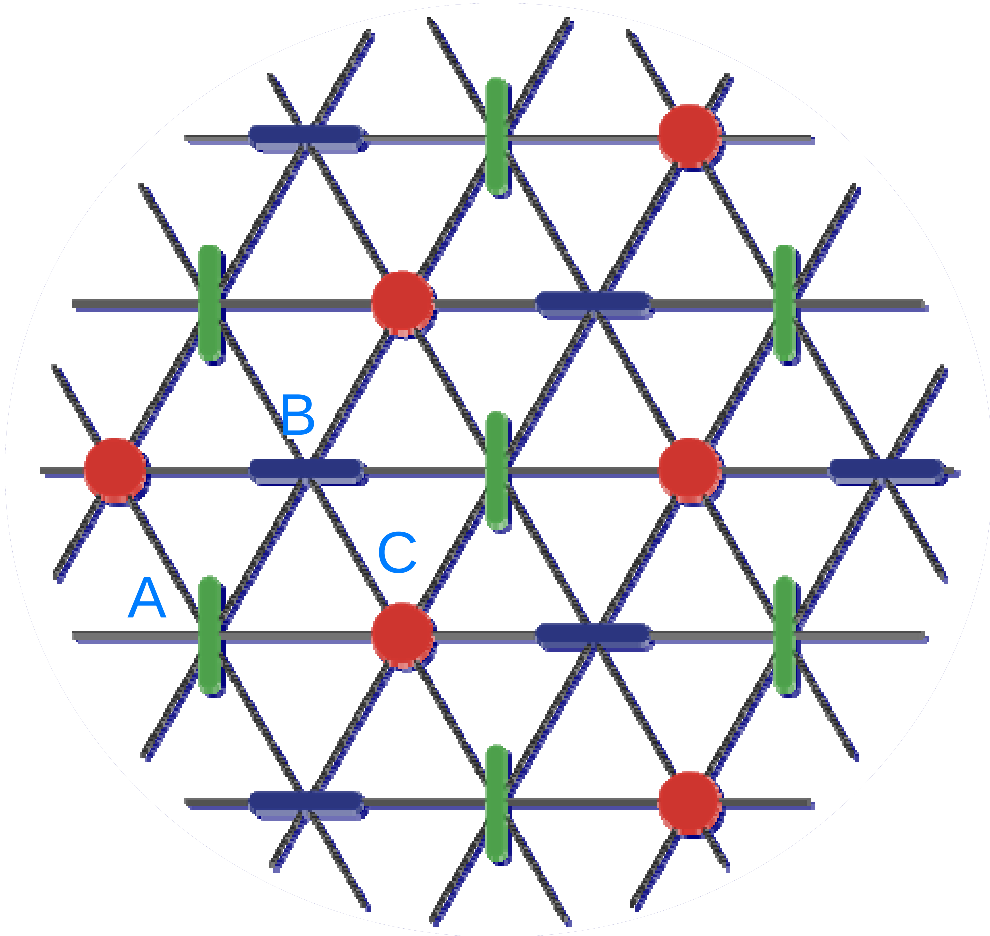
1-dim system
[Lauchli, Schmid, Trebst, '03]

Antiferro Nematic Order

- 3-sublattice order
  magnetic quadrupoles

- K>0
  0<J<K





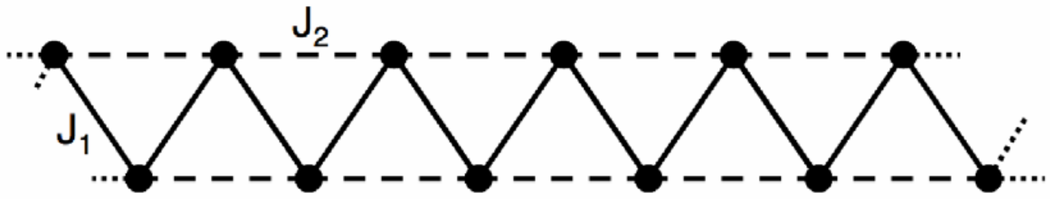
order parameter $\langle 2S_3^2 - S_1^2 - S_2^2 \rangle$

sublattice dependent principal axes

1D Analog of 3-sublattice Nematic Order?

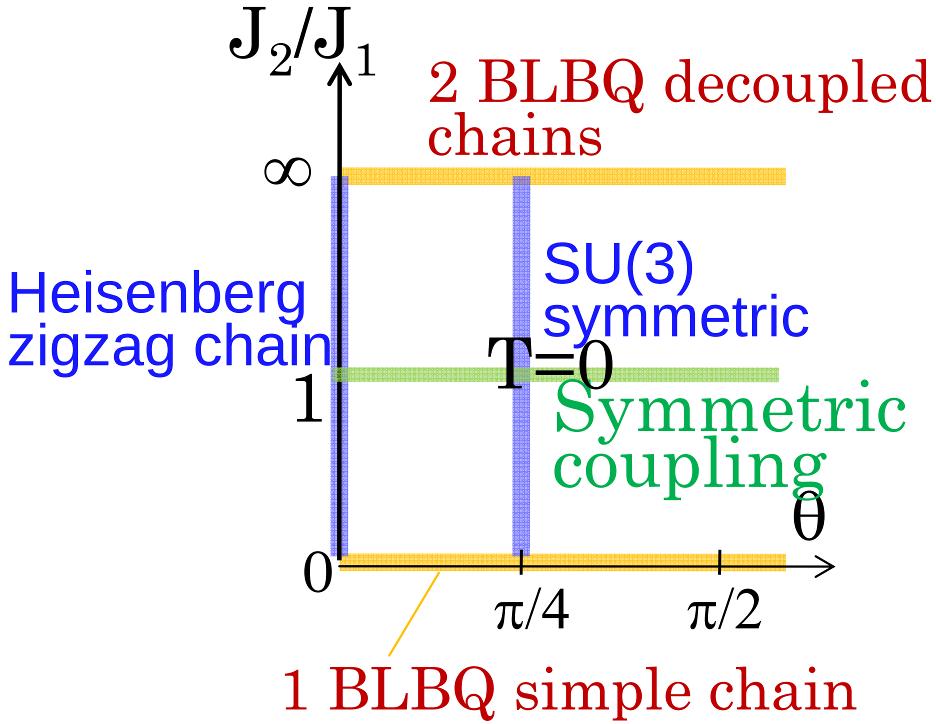
Model

S=1 bilinear-biquadratic (BLBQ) zigzag chain:

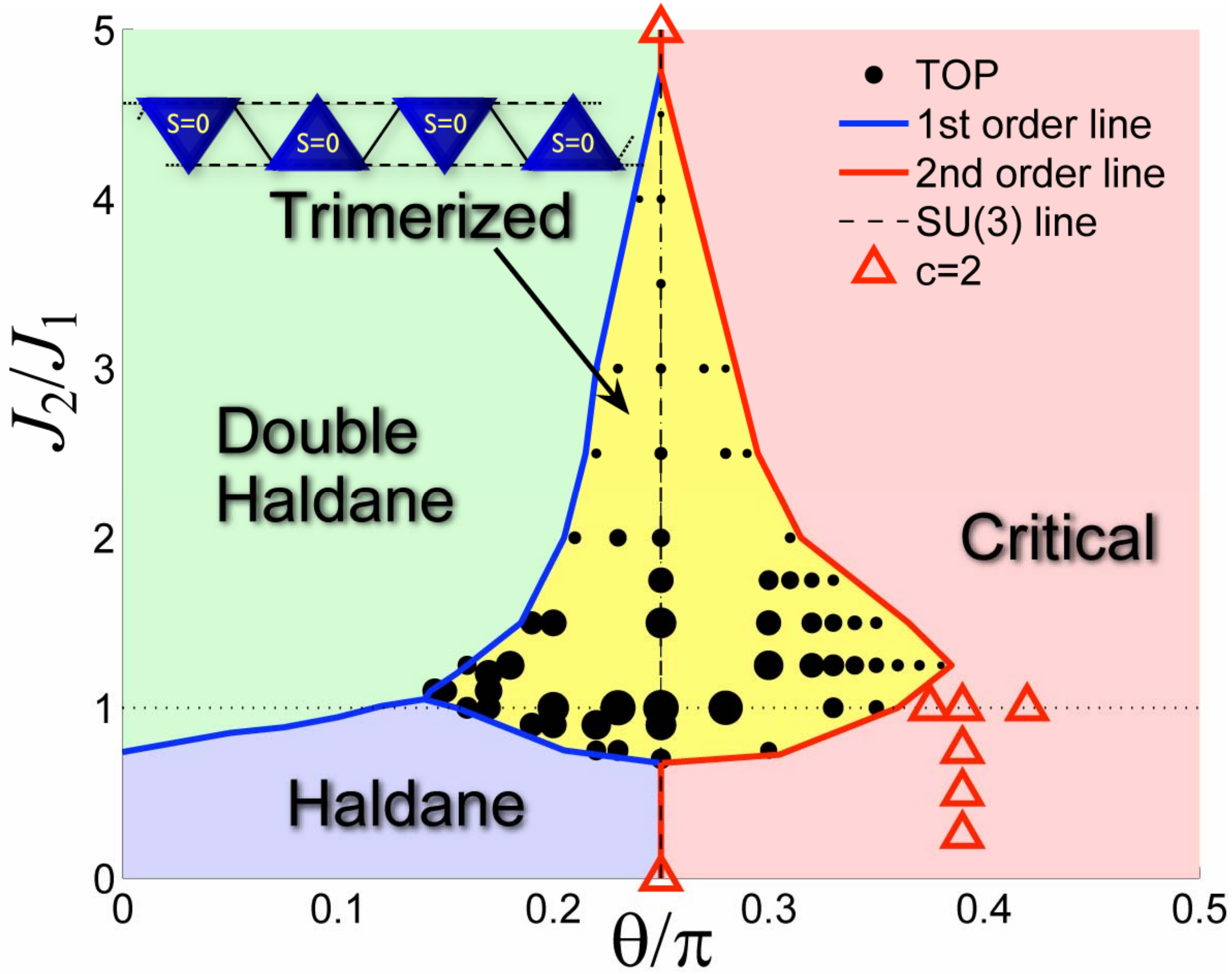


$$H = \sum_{\langle i,j \rangle}^{\text{1st,2nd}} J_{ij} \left[\cos\theta \mathbf{S}_i \cdot \mathbf{S}_j + \sin\theta \left(\mathbf{S}_i \cdot \mathbf{S}_j \right)^2 \right]$$

$$J_{ij} = \begin{cases} J_1 & \text{inter-chain} \\ J_2 & \text{intra-chain} \end{cases}$$



Phase Diagram (S=1 BLBQ zigzag spin chain)



RG Flow

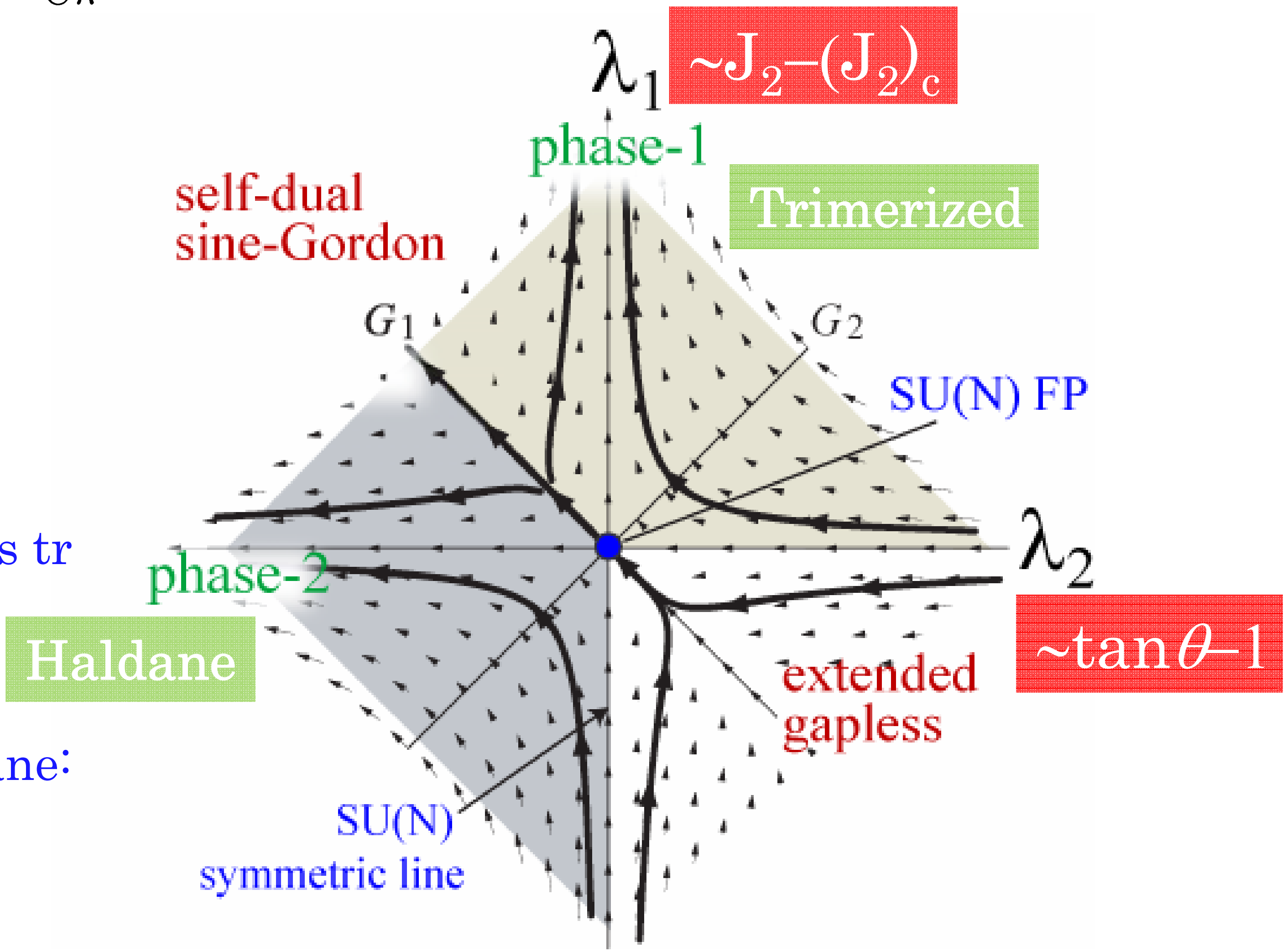
$$\begin{cases} \dot{G}_1 = \dfrac{N-2}{8\pi} G_1^2 + \dfrac{N+2}{8\pi} G_2^2 \\ \dot{G}_2 = \dfrac{N-1}{4\pi} G_1 G_2 \end{cases}$$

[Itoi and Kato, PRB, 1997,
Totsuka and Lecheminant 2007]

N=3

liquid ↔ trimerized:
liquid ↔ Haldane:
Kosterlitz-Thouless tr

trimerized ↔ Haldane:
1st order



Summary

S=1 BLBQ zigzag spin chain

•Existence of trimerized phase

•Order of transitions

•Reentrant transition to liquid phase in the large-J2 regionat SU(3) sym. line