Spin and orbital freezing in unconventional superconductors

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Kyoto, November 2017

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In collaboration with:

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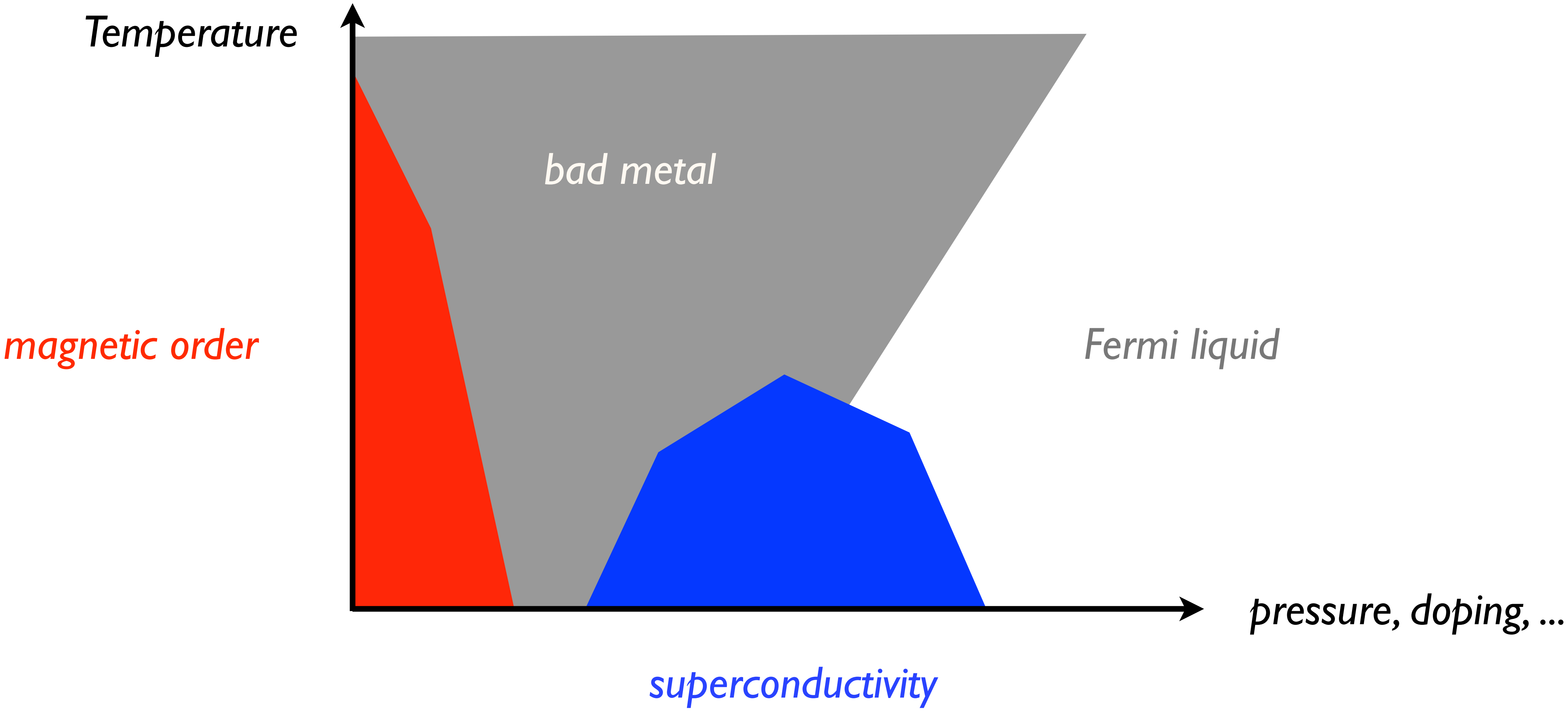
Introduction

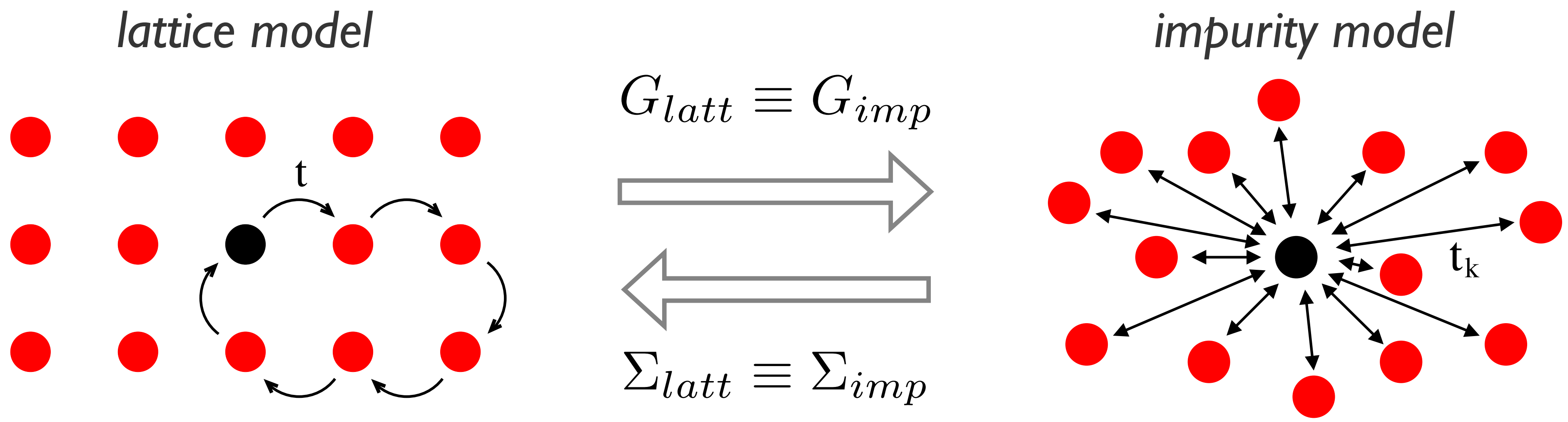
- Generic phase diagram of unconventional superconductors
  - Superconducting dome next to a magnetically ordered phase
  - Non-Fermi liquid metal above the superconducting dome



Method

Georges and Kotliar, PRB (1992)

- Dynamical mean field theory DMFT: mapping to an impurity problem



- Impurity solver: computes the Green's function of the correlated site
- Bath parameters = "mean field": optimized in such a way that the bath mimics the lattice environment

Method

Werner et al., PRL (2006)

- CT-QMC solvers allow efficient simulation of multiorbital models

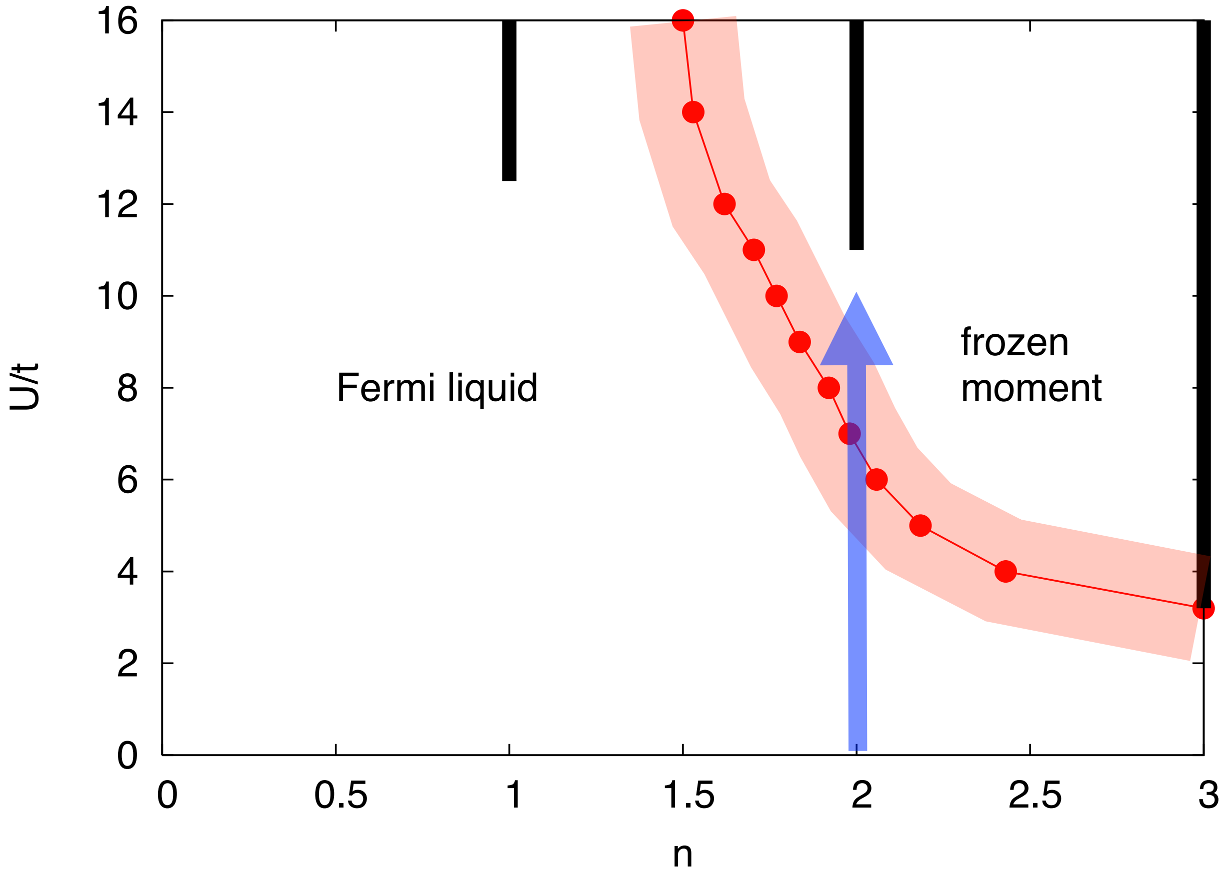
$$
\begin{aligned}
H_{\mathrm{loc}} = & -\sum_{\alpha,\sigma} \mu n_{\alpha,\sigma} + \sum_{\alpha} U n_{\alpha,\uparrow} n_{\alpha,\downarrow} \\
& + \sum_{\alpha>\beta,\sigma} U' n_{\alpha,\sigma} n_{\beta,-\sigma} + (U'-J) n_{\alpha,\sigma} n_{\beta,\sigma} \\
& - \sum_{\alpha\neq\beta} J(\psi^\dagger_{\alpha,\downarrow}\psi^\dagger_{\beta,\uparrow}\psi_{\beta,\downarrow}\psi_{\alpha,\uparrow} + \psi^\dagger_{\beta,\uparrow}\psi^\dagger_{\beta,\downarrow}\psi_{\alpha,\uparrow}\psi_{\alpha,\downarrow} + h.c.)
\end{aligned}
$$

- Relevant cases:
  - 4 electrons in 3 orbitals: *$SrRu_2O_4$*
  - 3 electrons in 3 orbitals, *J*<0: *A_3C_{60}*
  - 6 electrons in 5 orbitals: *Fe*-pnictides

3-orbital model

Werner, Gull, Troyer & Millis, PRL (2008)

- Phase diagram for $U' = U - 2J, J/U = 1/6, \beta = 50$



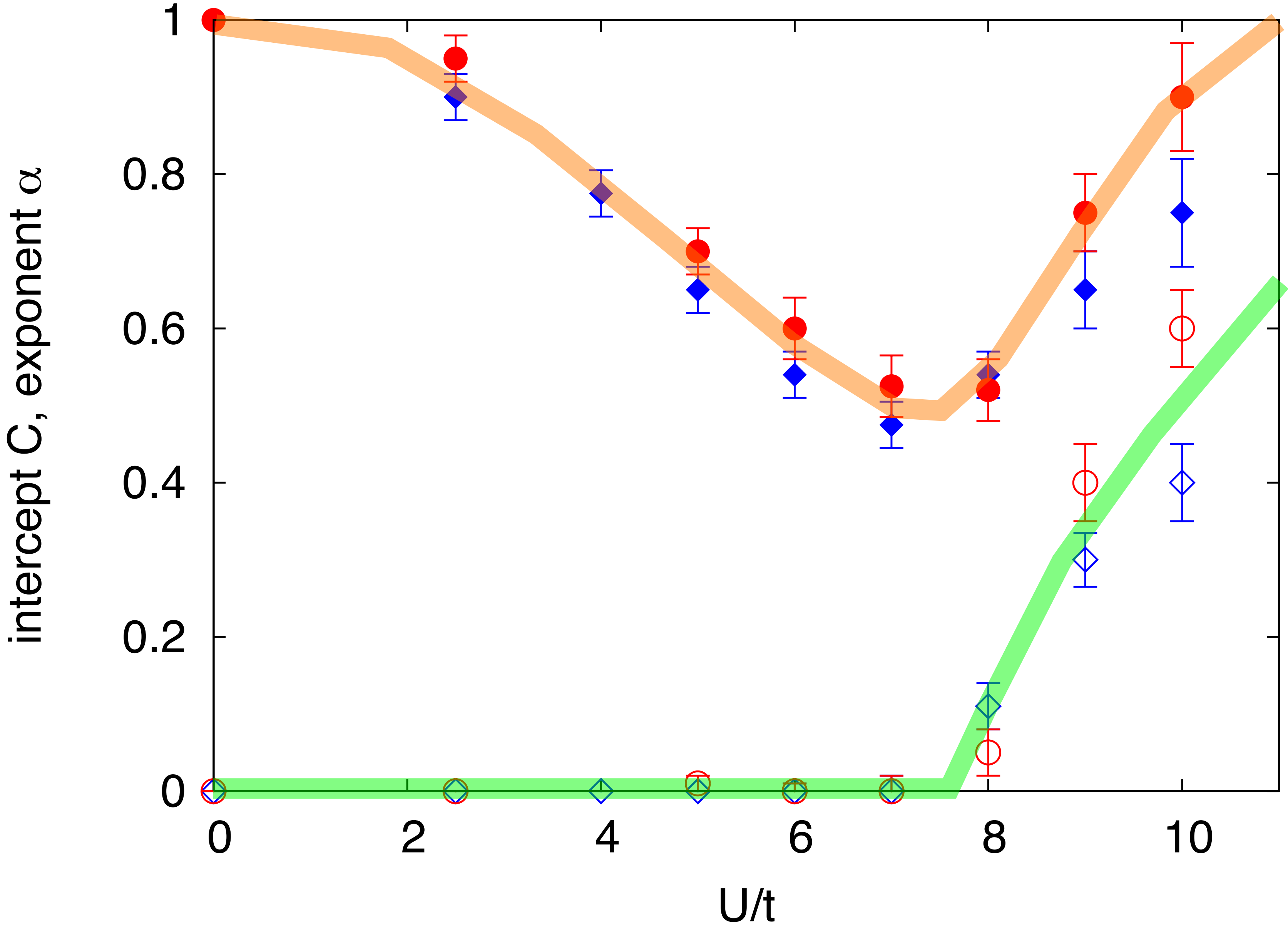
- Metallic phase: "transition" from Fermi liquid to spin-glass
- Narrow crossover regime with self-energy

$$\mathrm{Im}\Sigma/t \sim (i\omega_n/t)^{\alpha},\ \alpha \approx 0.5$$

3-orbital model

Werner, Gull, Troyer & Millis, PRL (2008)

- Fit self-energy by $-\mathrm{Im}\Sigma(i\omega_n) = C + A(\omega_n)^{\alpha}$



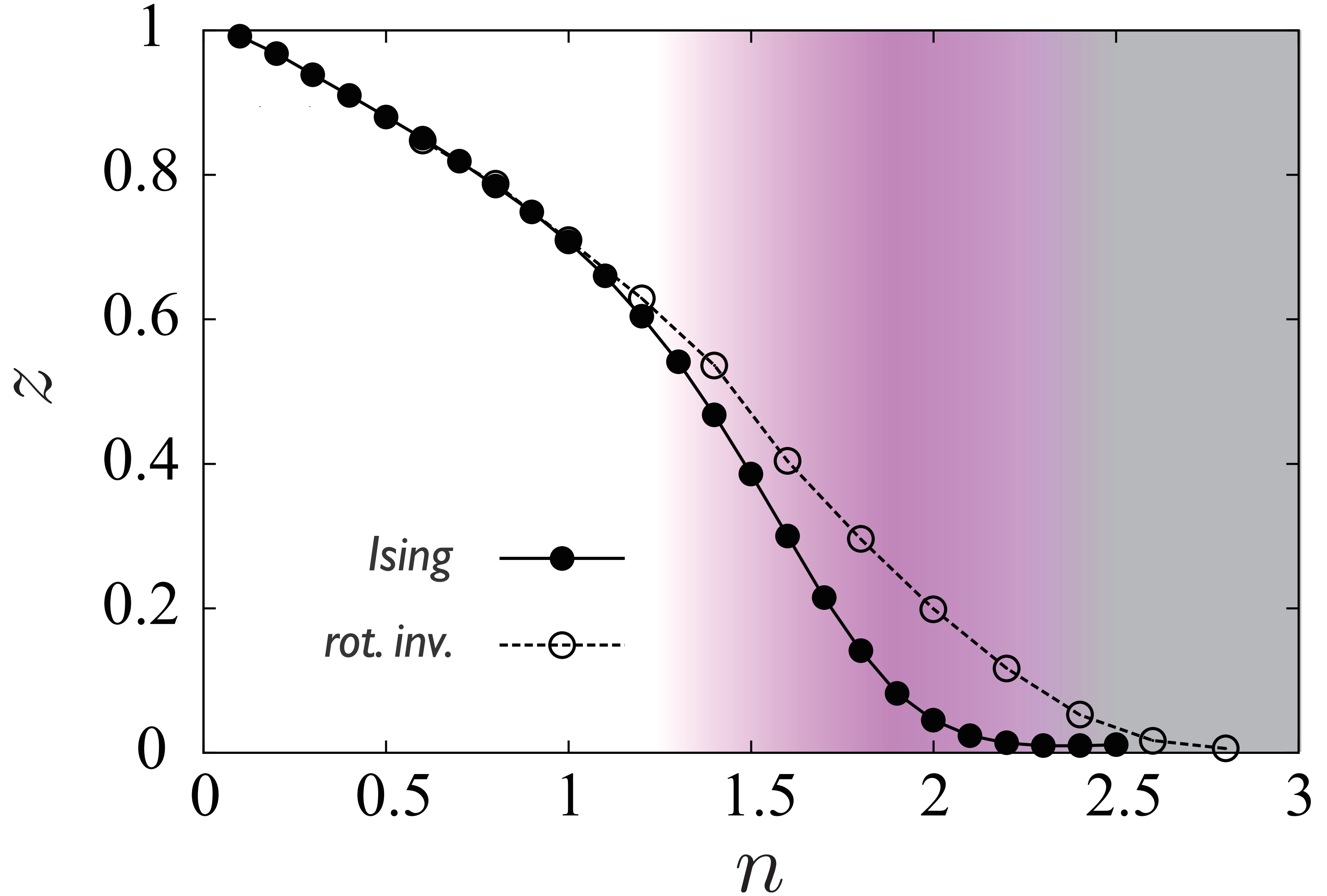
- Square-root self-energy coincides with on-set of frozen moments

3-orbital model

Hoshino & Werner, PRL (2015)

- Spin-freezing leads to a small “quasi-particle weight” z

$$z \approx 1/(1 - \mathrm{Im}\Sigma(i\omega_0)/\omega_0)$$

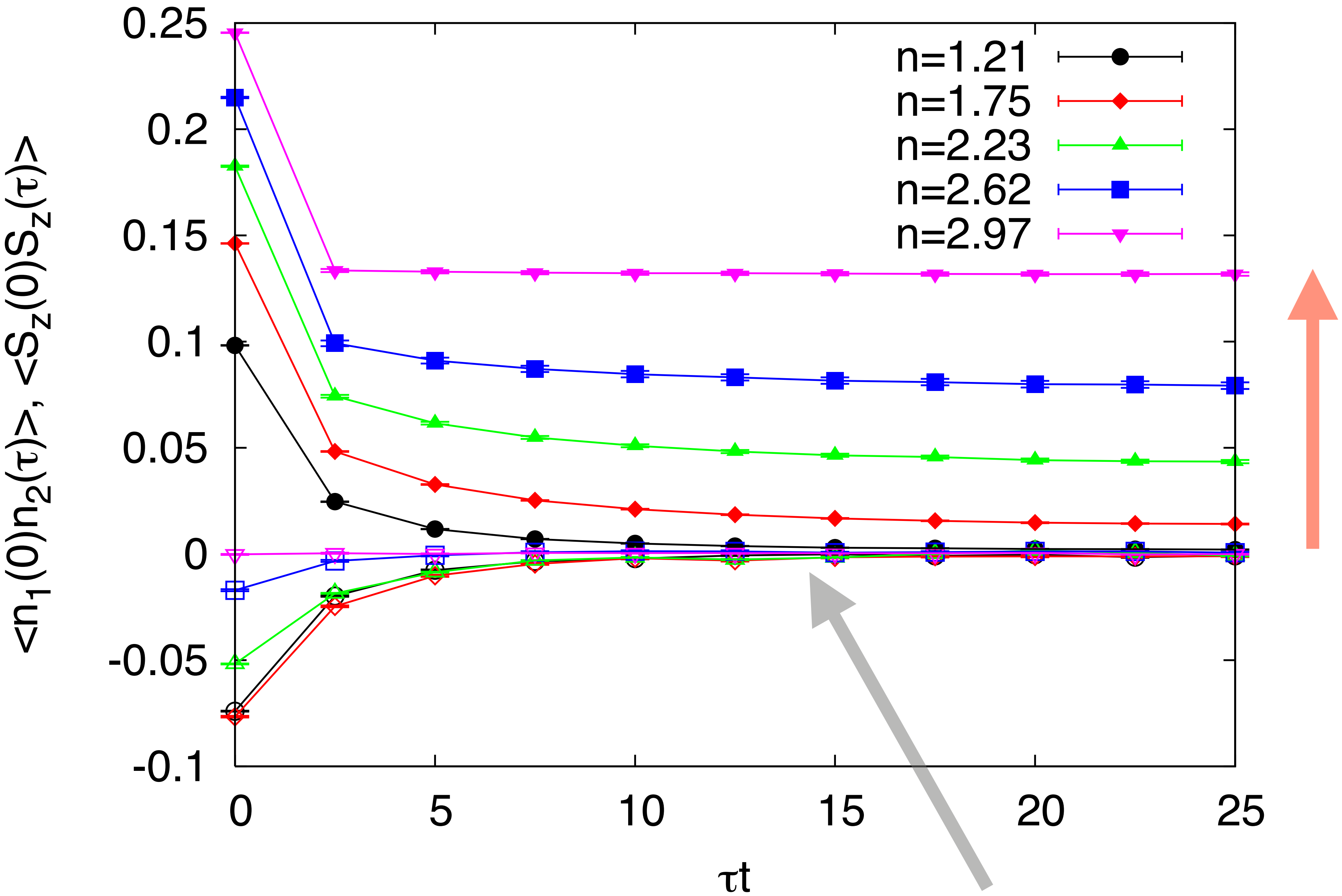


no quasi-particles in spin-frozen regime

3-orbital model

Werner, Gull, Troyer & Millis, PRL (2008)

- Spin-spin and orbital-orbital correlation functions



freezing of spin moments

no freezing of orbital moments

3-orbital model

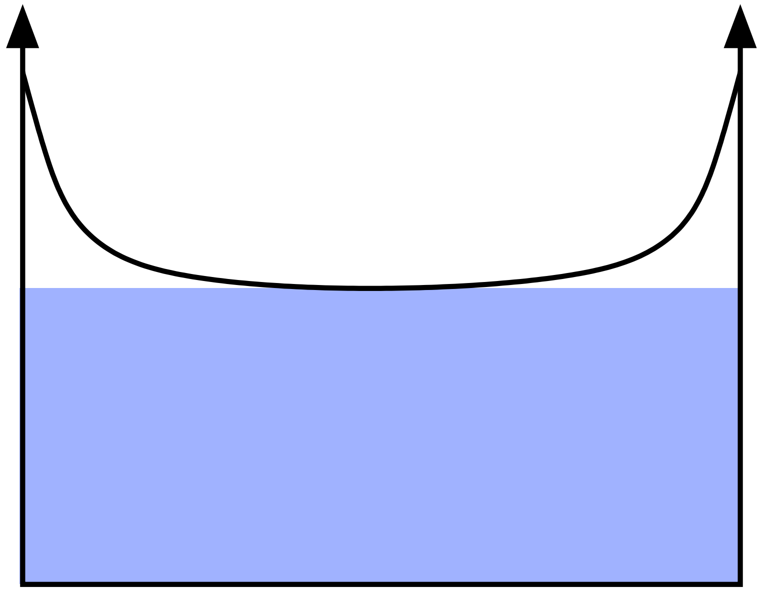
Hoshino & Werner, PRL (2015)

- Consider the local susceptibility

$$\chi_{\text{loc}} = \int_0^\beta d\tau \langle S_z(\tau) S_z(0) \rangle$$

and its dynamic contribution

$$\Delta\chi_{\text{loc}} = \int_0^\beta d\tau [\langle S_z(\tau) S_z(0) \rangle - \langle S_z(\beta/2) S_z(0) \rangle]$$

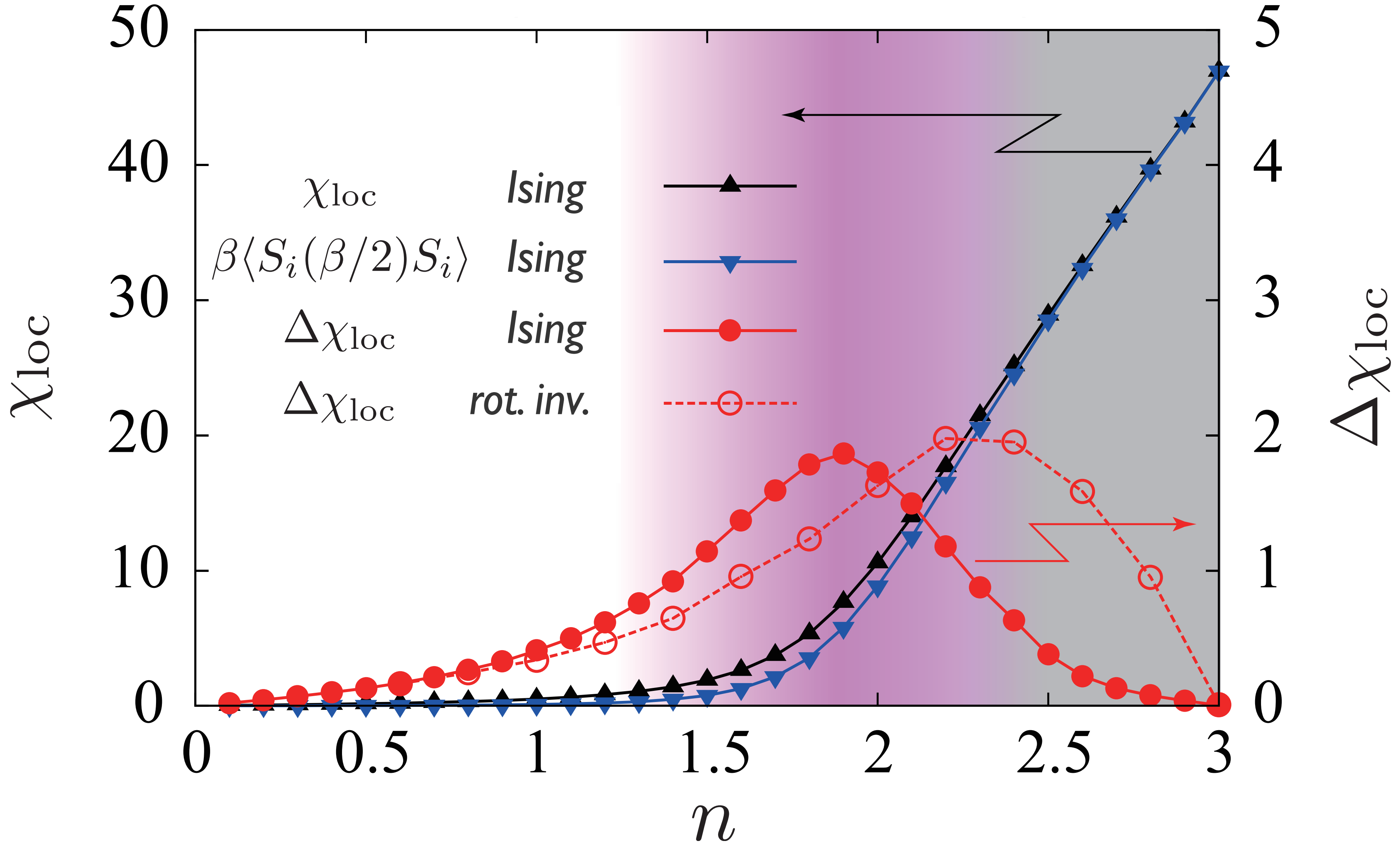


subtract the (frozen) long-time value

3-orbital model

Hoshino & Werner, PRL (2015)

- Consider the local susceptibility χ_{loc} and its dynamic contribution $\Delta\chi_{\text{loc}}$



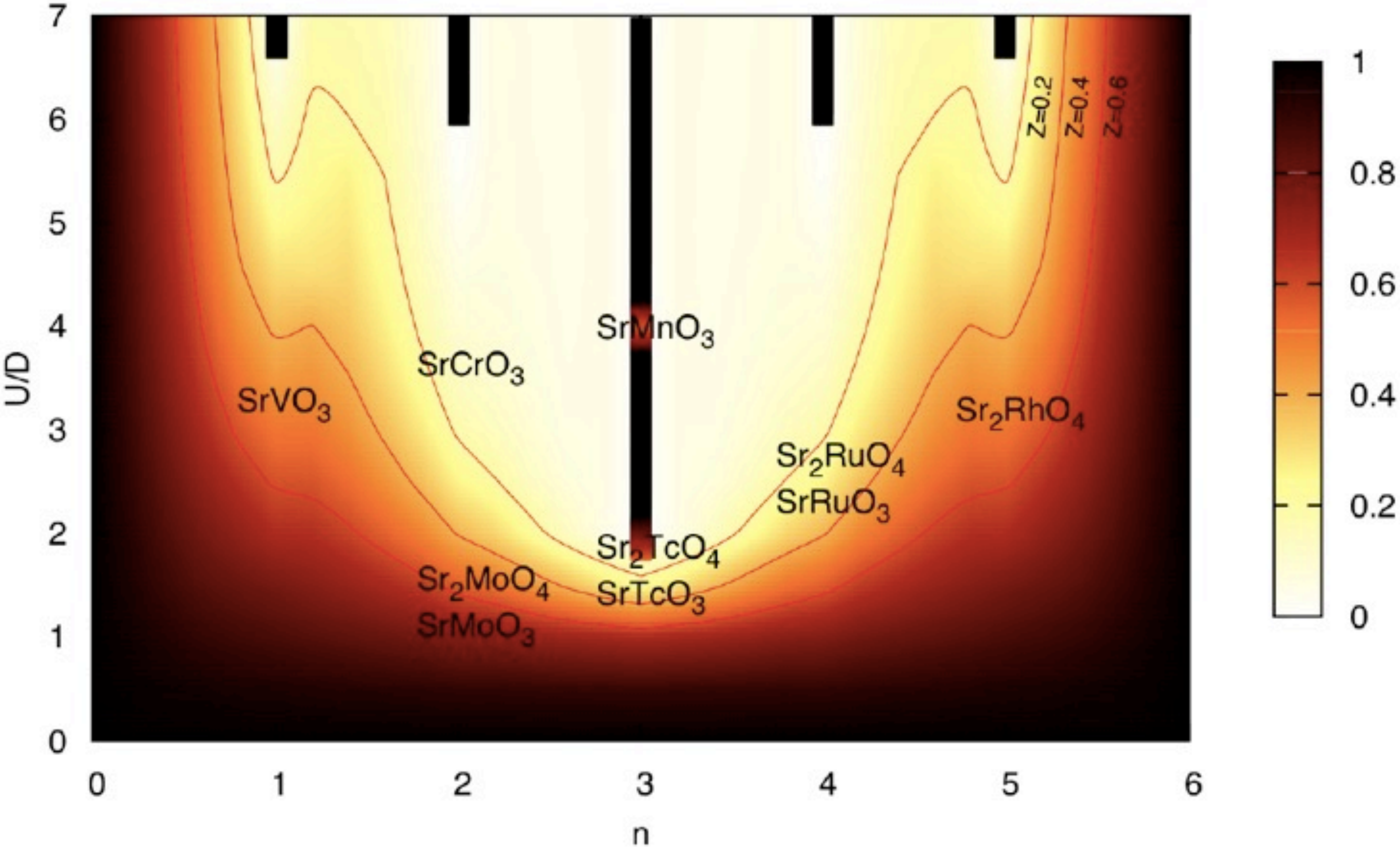
- Crossover regime is characterized by large local moment fluctuations

3-orbital model

Werner, Gull, Troyer & Millis, PRL (2008)

- “quasi-particle weight” *z*

  from De’ Medici, Mravlje & Georges, PRL (2011)



- Hund coupling *J*: Strongly correlated metal far from the Mott transition

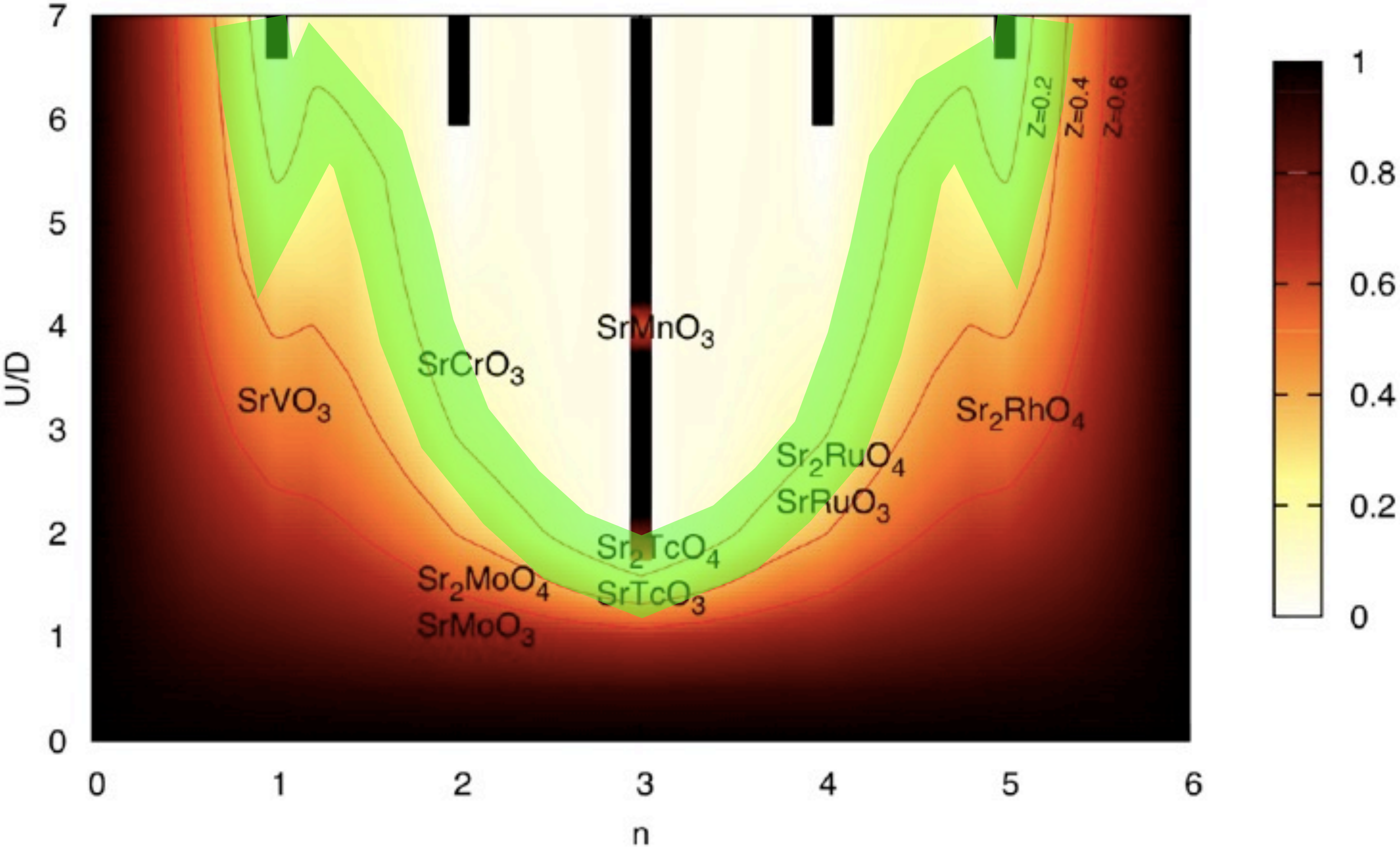
3-orbital model

Werner, Gull, Troyer & Millis, PRL (2008)

- “quasi-particle weight” z

from De’ Medici, Mravlje & Georges, PRL (2011)

large local moment fluctuations

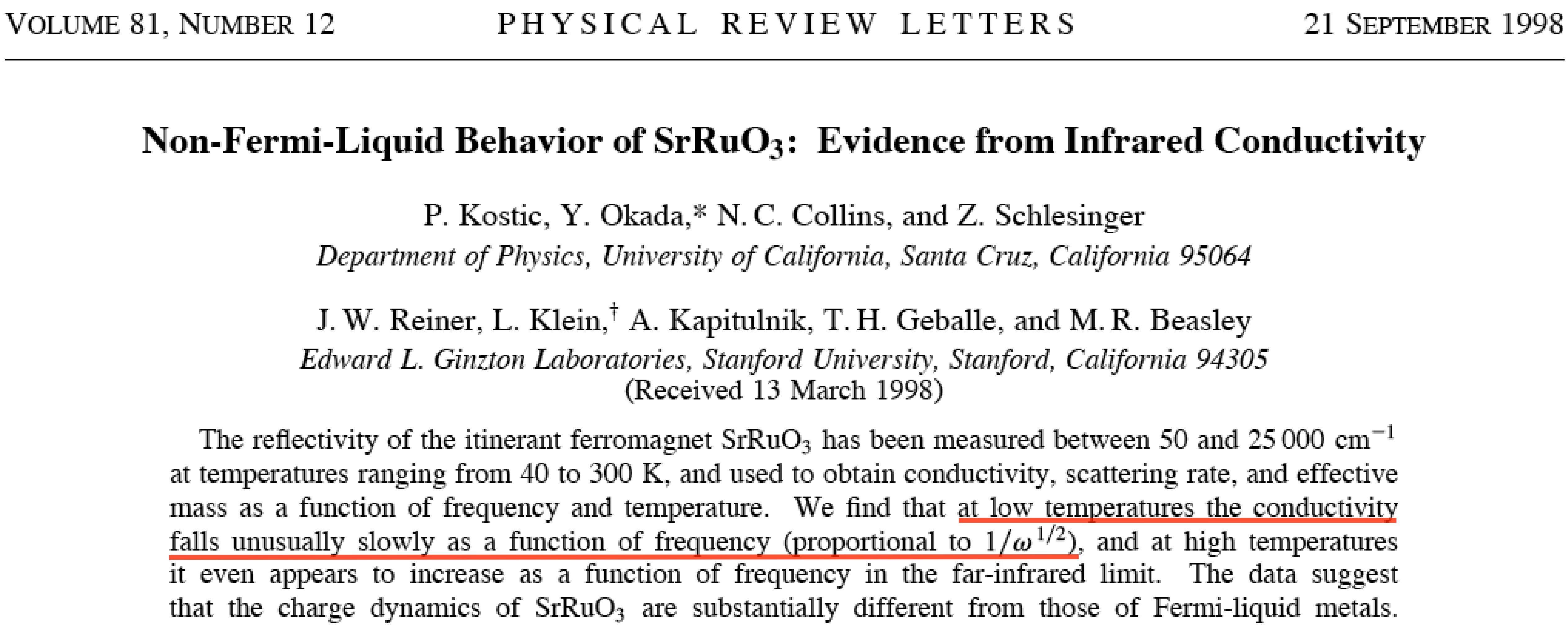


- Hund coupling J: Strongly correlated metal far from the Mott transition

Strontium Ruthenates

Werner, Gull, Troyer & Millis, PRL (2008)

- A self-energy with frequency dependence $\Sigma(\omega) \sim \omega^{1/2}$ implies an optical conductivity $\sigma(\omega) \sim 1/\omega^{1/2}$



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Non-Fermi-Liquid Behavior of $SrRuO_3$: Evidence from Infrared Conductivity

P. Kostic, Y. Okada,* N. C. Collins, and Z. Schlesinger
Department of Physics, University of California, Santa Cruz, California 95064

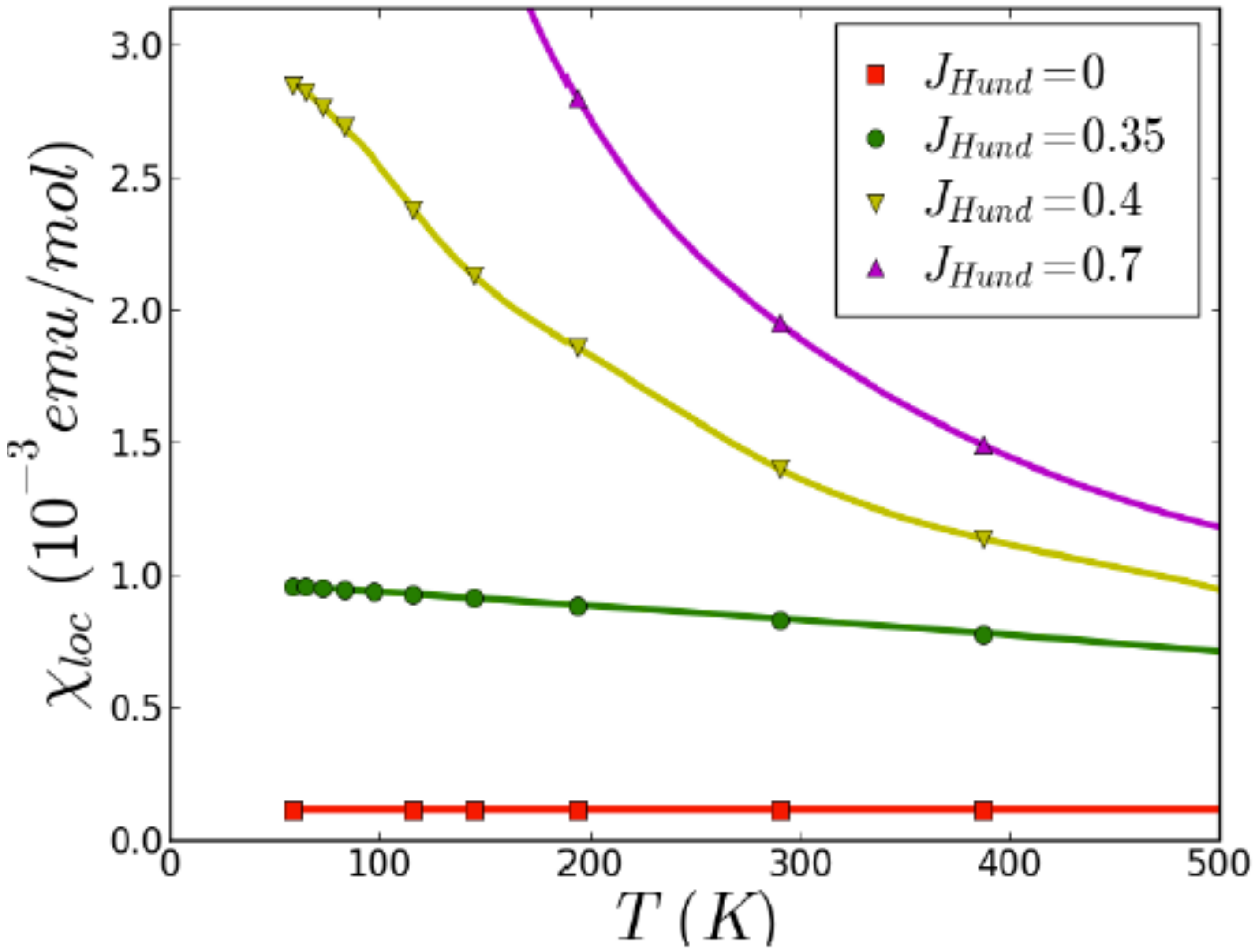
J. W. Reiner, L. Klein,† A. Kapitulnik, T. H. Geballe, and M. R. Beasley
Edward L. Ginzton Laboratories, Stanford University, Stanford, California 94305

(Received 13 March 1998)

The reflectivity of the itinerant ferromagnet $SrRuO_3$ has been measured between 50 and 25 000 cm^{-1} at temperatures ranging from 40 to 300 K, and used to obtain conductivity, scattering rate, and effective mass as a function of frequency and temperature. We find that at low temperatures the conductivity falls unusually slowly as a function of frequency (proportional to $1/\omega^{1/2}$), and at high temperatures it even appears to increase as a function of frequency in the far-infrared limit. The data suggest that the charge dynamics of $SrRuO_3$ are substantially different from those of Fermi-liquid metals.

Pnictides

- Strongly correlated despite moderate *U*

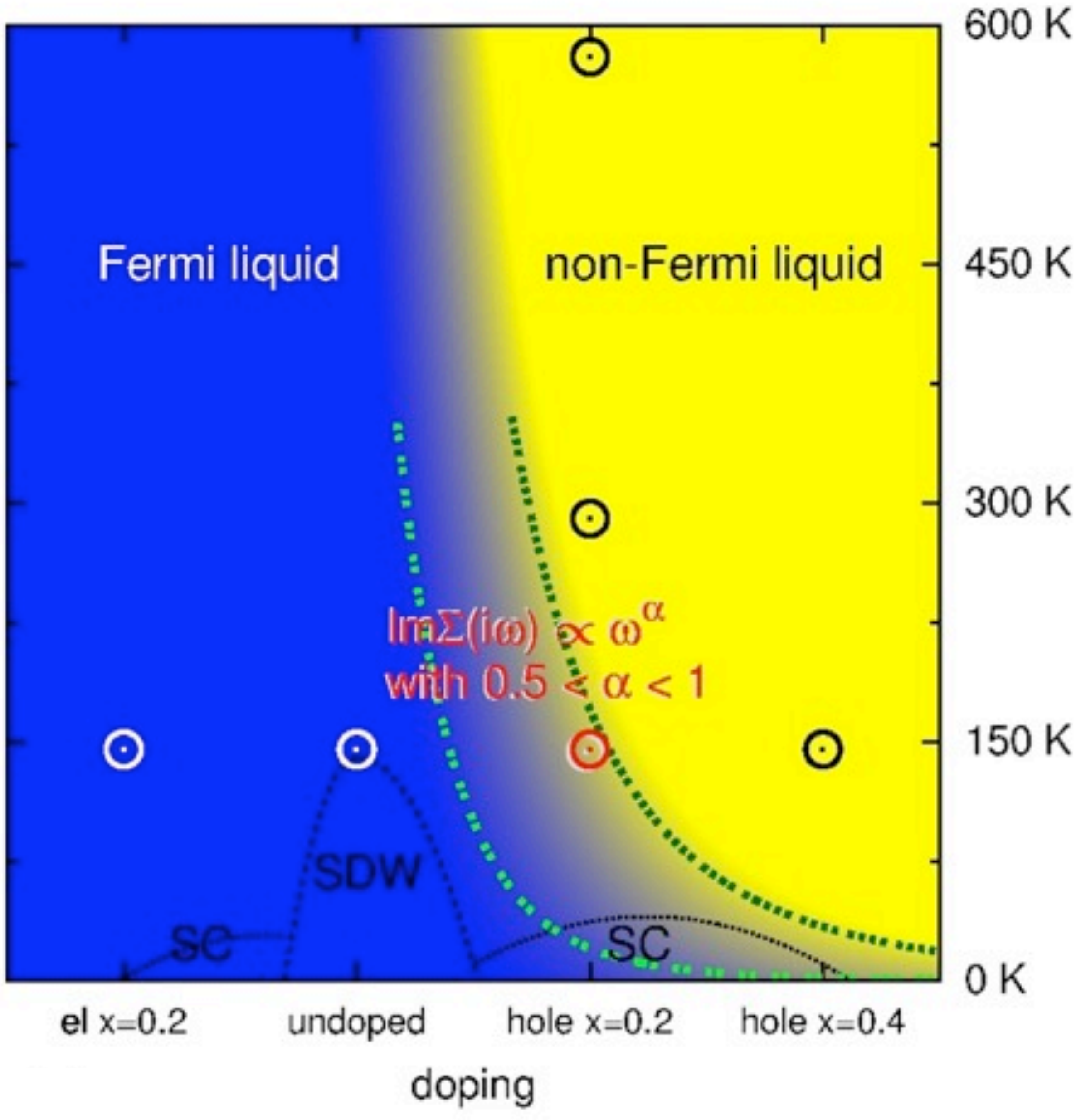


incoherent metal state resulting from Hund's coupling

Haule & Kotliar, NJP (2009)

Pnictides

- Strong doping and temperature dependence of electronic structure



$BaFe_2As_2$:

conventional FL metal in the underdoped regime

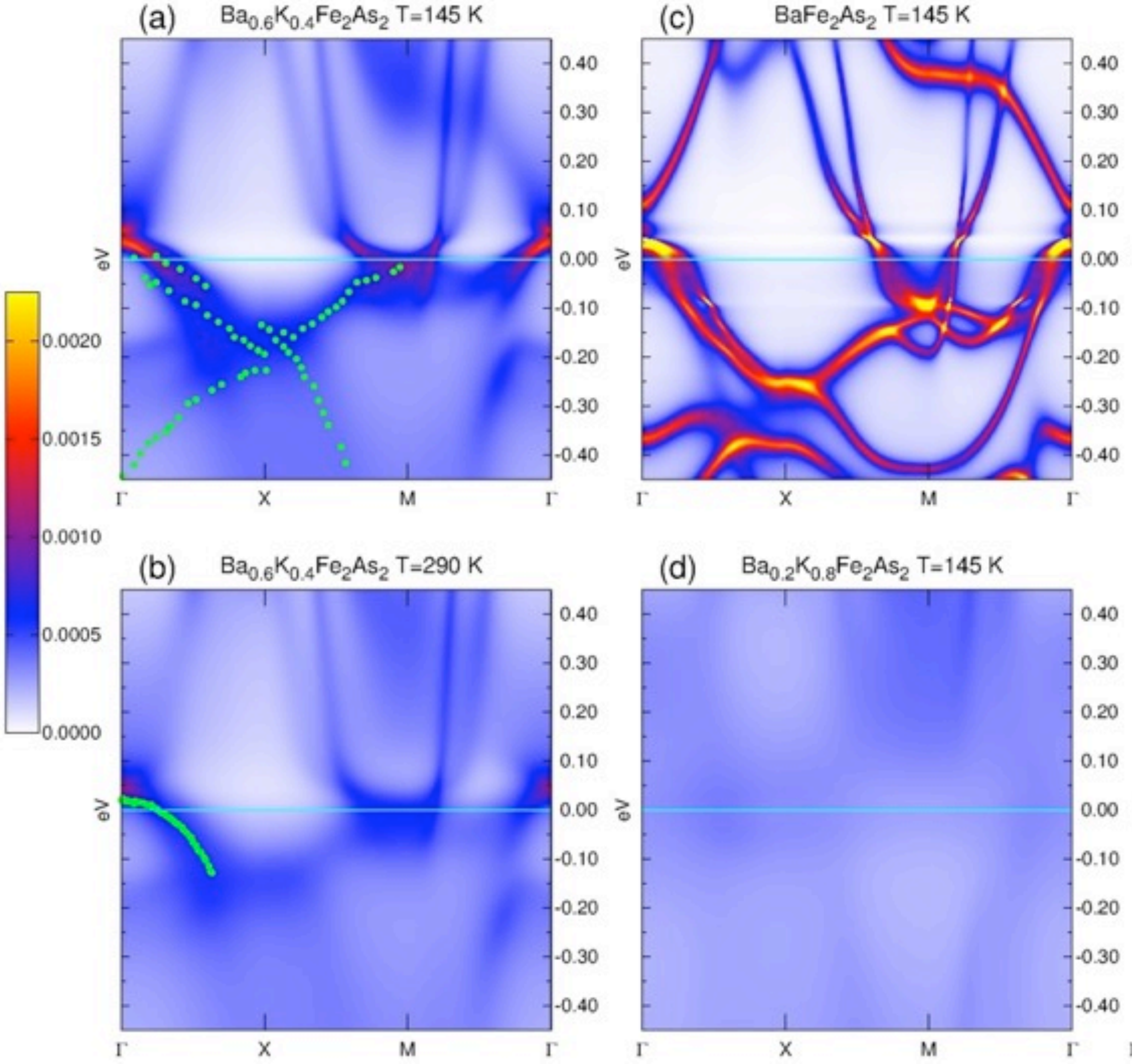
non-FL properties near optimal doping

incoherent metal in the overdoped regime

Werner et al., Nat. Phys. (2012)

Pnictides

- Strong doping and temperature dependence of electronic structure



Long-range order

Hoshino & Werner, PRL (2015)

- Identify ordering instabilities by divergent lattice susceptibilities
  - Calculate local vertex from impurity problem
  - Approximate vertex of the lattice problem by this local vertex
  - Solve Bethe-Salpeter equation to obtain lattice susceptibility

- The following orders (staggered and uniform) are considered:
  - diagonal orders:

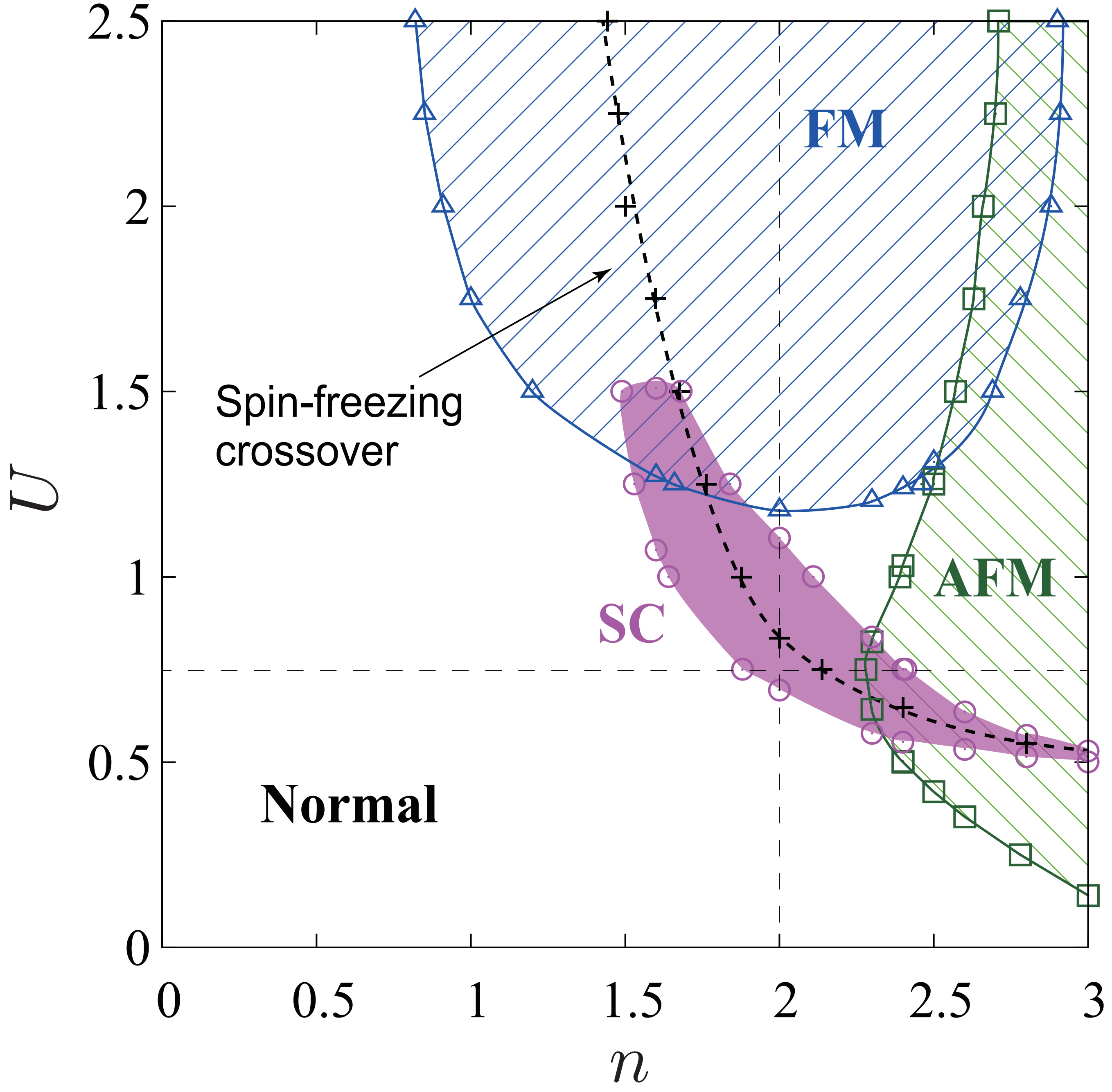
    charge, spin, orbital, spin-orbital
  - off-diagonal orders:

    orbital-singlet-spin-triplet SC, orbital-triplet-spin-singlet SC

Long-range order

Hoshino & Werner, PRL (2015)

- 3-orbital model, Ising interactions



AFM near half-filling

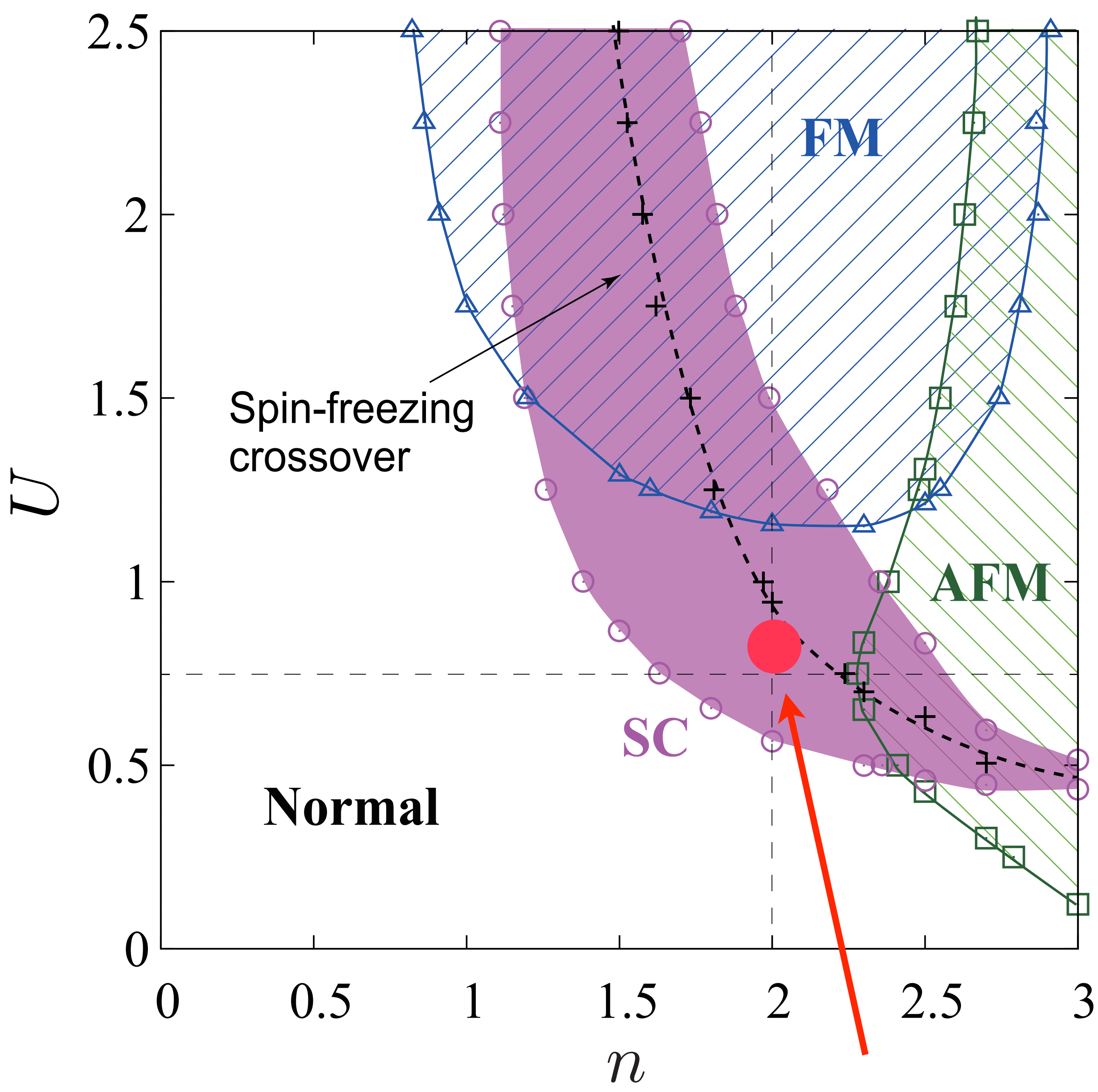
FM at large U away from half-filling

spin-triplet superconductivity in the spin-freezing crossover region

Long-range order

Hoshino & Werner, PRL (2015)

- 3-orbital model, Ising interactions (lower temperature)



AFM near half-filling

FM at large U away from half-filling

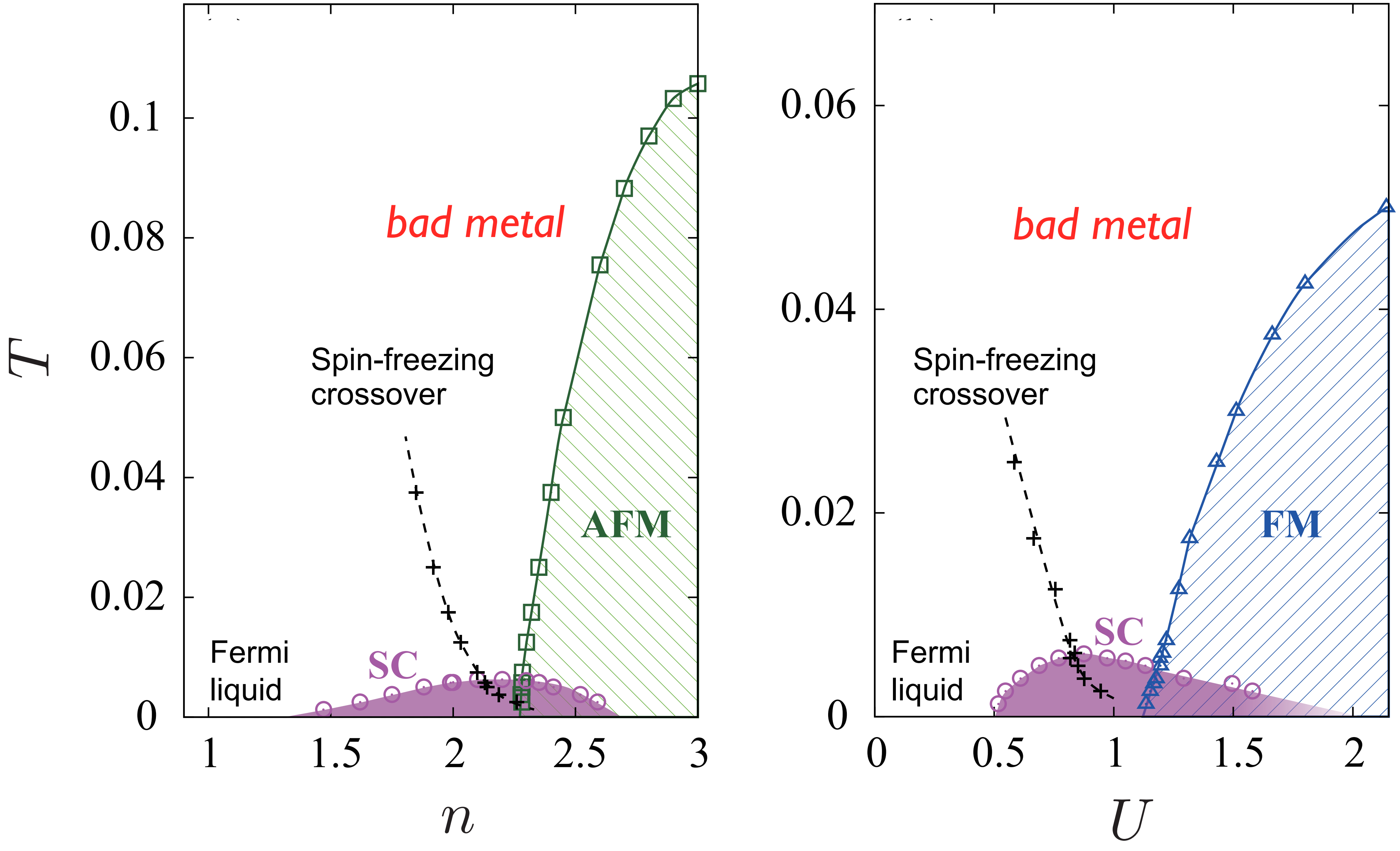
spin-triplet superconductivity in the spin-freezing crossover region

parameter regime relevant for Sr_2RuO_4

Long-range order

Hoshino & Werner, PRL (2015)

- T_c dome and non-FL metal phase next to magnetic order

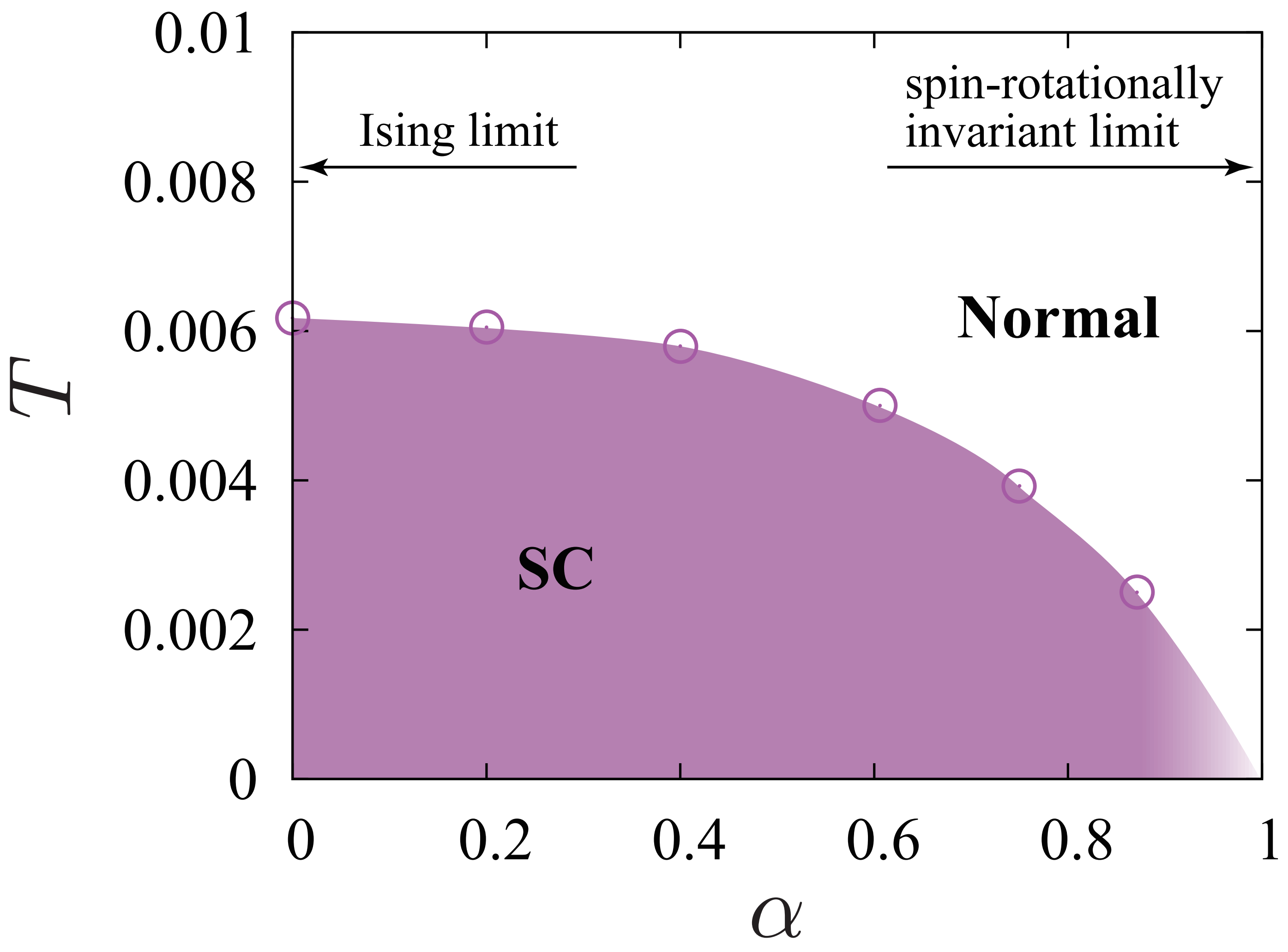


- Generic phasediagram of unconventional SC without QCP!

Long-range order

Hoshino & Werner, PRL (2015)

- T_c dome and non-FL metal phase next to magnetic order



- Need spin-anisotropy (SO coupling) for high T_c

  probably relevant for: Sr_2RuO_4, UGe_2, URhGe, UCoGe, ...

Long-range order

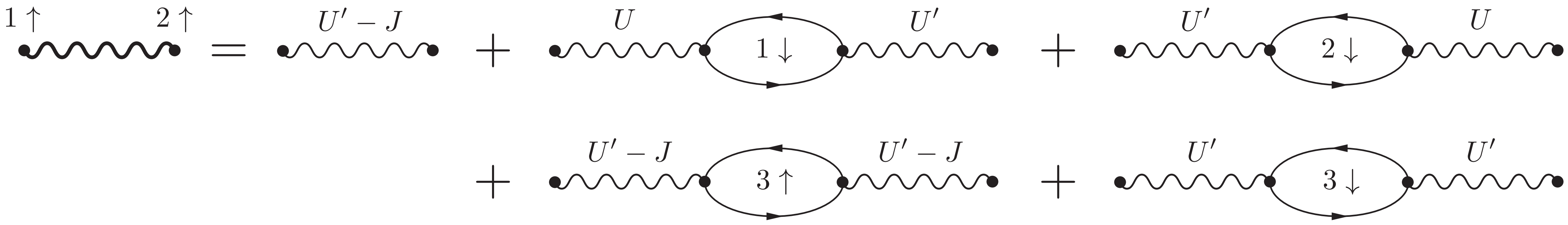
Hoshino & Werner, PRL (2015)

- Pairing induced by local spin fluctuations

  Weak-coupling argument inspired by Inaba & Suga, PRL (2012)

- Effective interaction which includes bubble diagrams:

$$\tilde{U}_{\alpha\beta}(q) = U_{\alpha\beta} - \sum_{\gamma} U_{\alpha\gamma}\chi_{\gamma}(q)\tilde{U}_{\gamma\beta}(q)$$



- Effective inter-orbital same-spin interaction

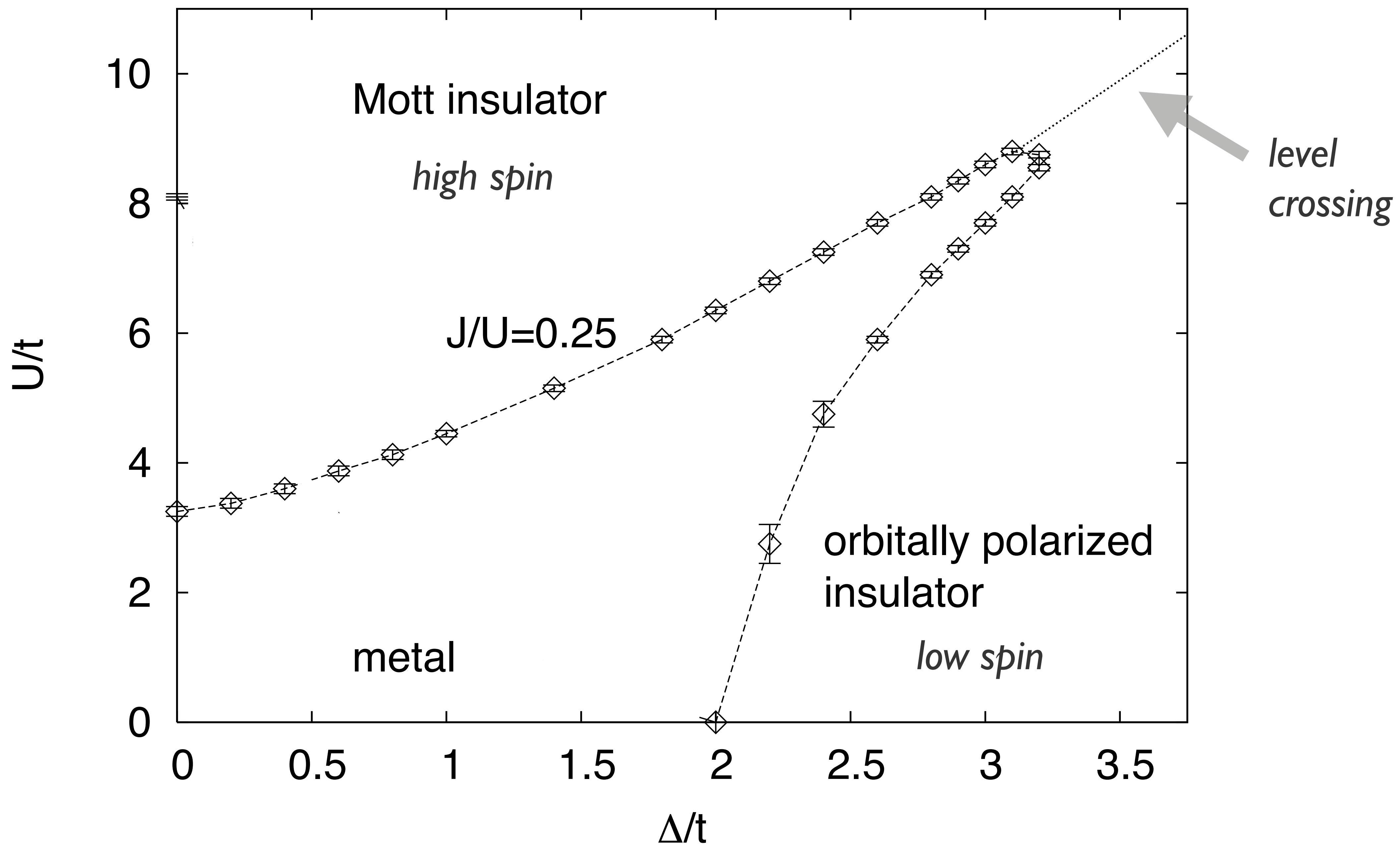
$$\tilde{U}_{1\uparrow,2\uparrow}(0) = U' - J - [2UU' + (U'-J)^2 + U'^2]\chi_{\mathrm{loc}}$$

in the weak-coupling regime: $\chi_{\mathrm{loc}} = \Delta\chi_{\mathrm{loc}}$

Crystal field splitting

Hoshino & Werner, PRB (2016)

- Complicated phase diagrams, even in the two-orbital case
  - High-spin/low-spin transitions *Werner & Millis, PRL (2007)*



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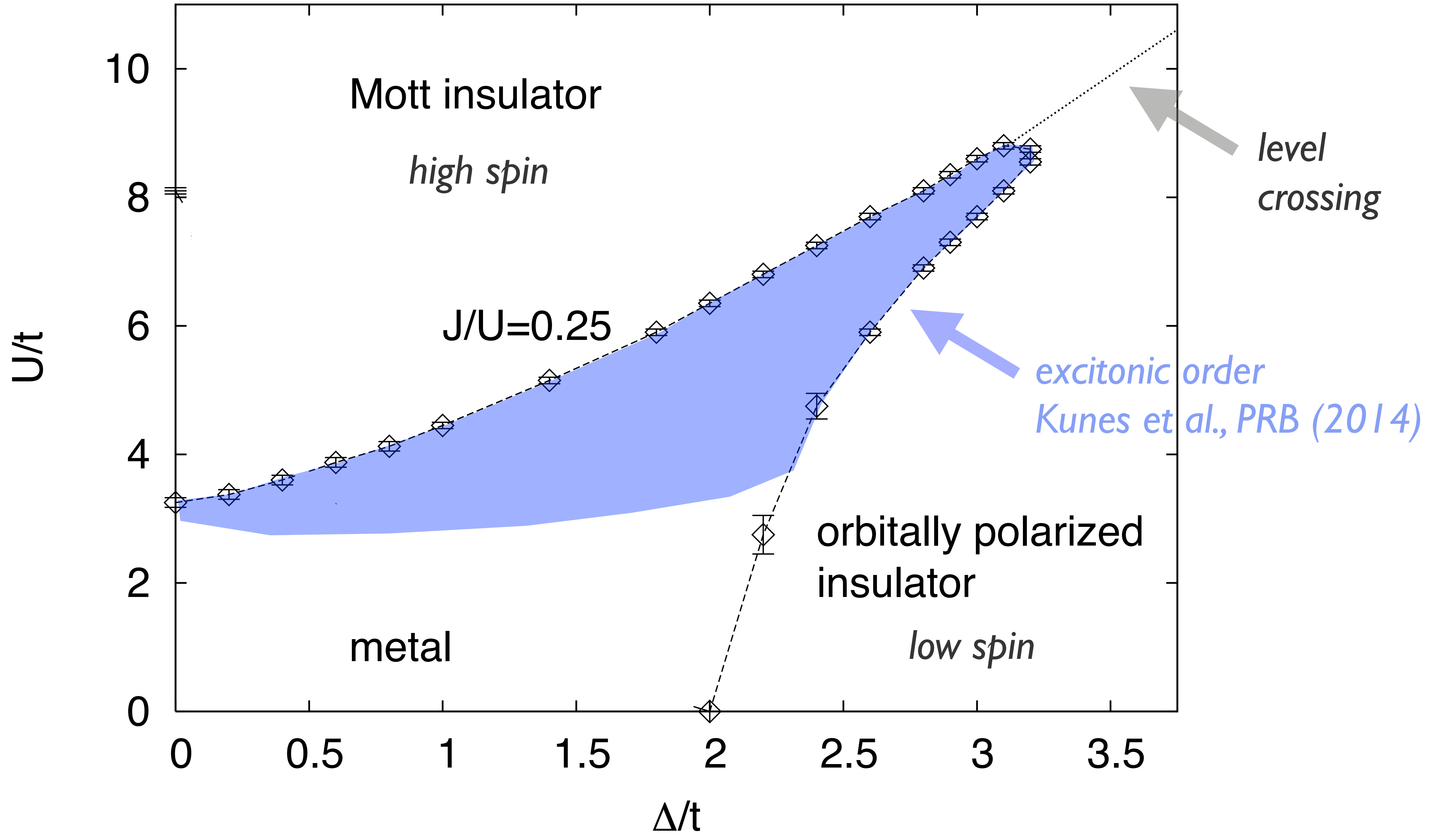

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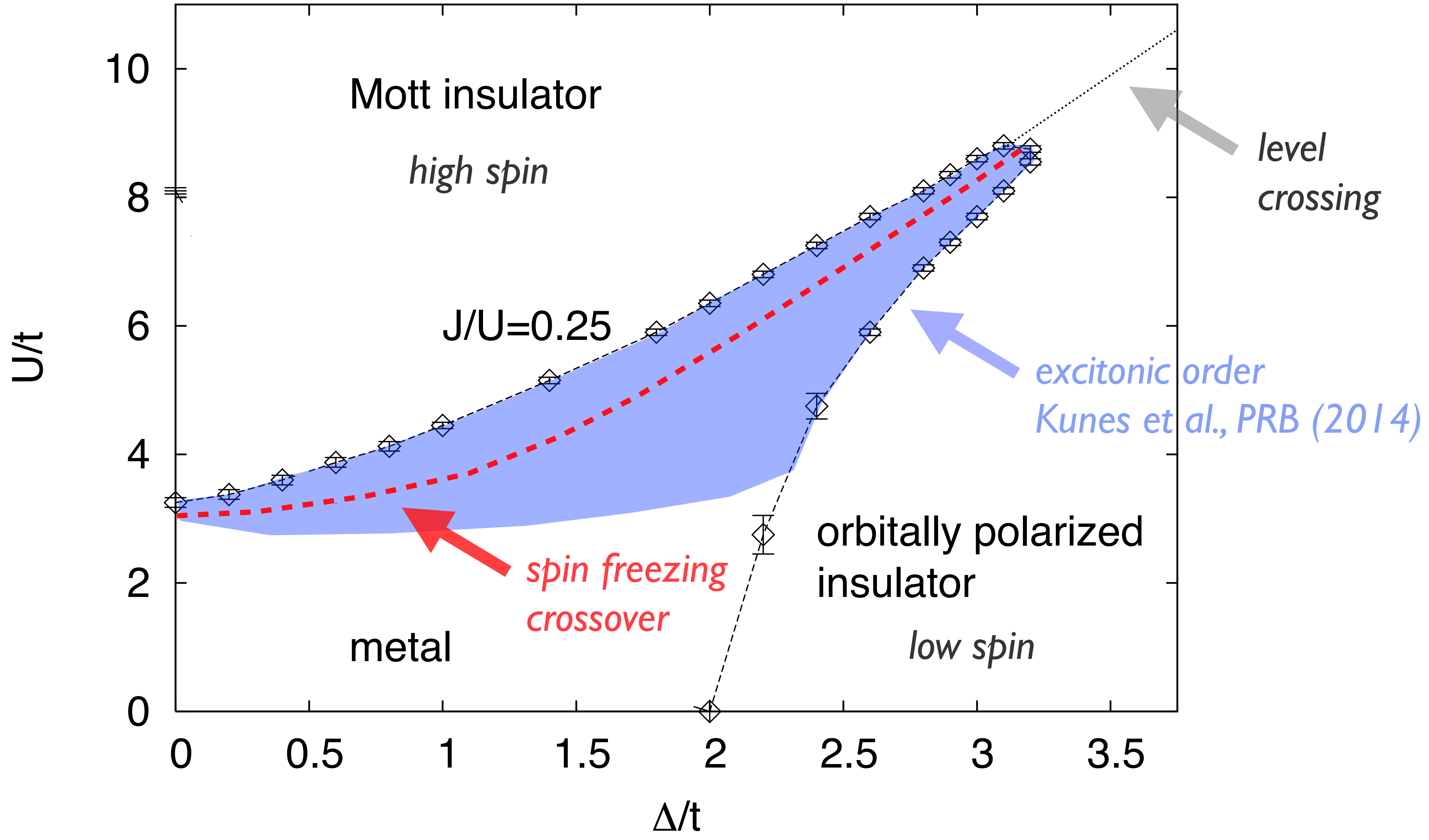


level crossing

excitonic order
Kunes et al., PRB (2014)

spin freezing crossover

Crystal field splitting

Hoshino & Werner, PRB (2016)

- Complicated phase diagrams, even in the two-orbital case
  - High-spin/low-spin transitions *Werner & Millis, PRL (2007)*
  - Excitonic (spin-orbital) order *Kunes et al., PRB (2014)*

- Exact mapping: $c_{i2\sigma} \to \sum_{\sigma'} \sigma^x_{\sigma\sigma'} c^\dagger_{i2\sigma'} e^{\mathrm{i}Q\cdot R_i}$

$$\begin{array}{rcl} \text{half-filled model with } \Delta > 0 & \to & \text{doped model with } \Delta = 0 \\ \text{crystal field splitting } \Delta & \to & \text{chemical potential shift } \mu \\ \text{spin-orbital order} & \to & \text{spin-triplet SC} \end{array}$$

- Spin-orbital order and spin-triplet SC instabilities driven by fluctuating local moments

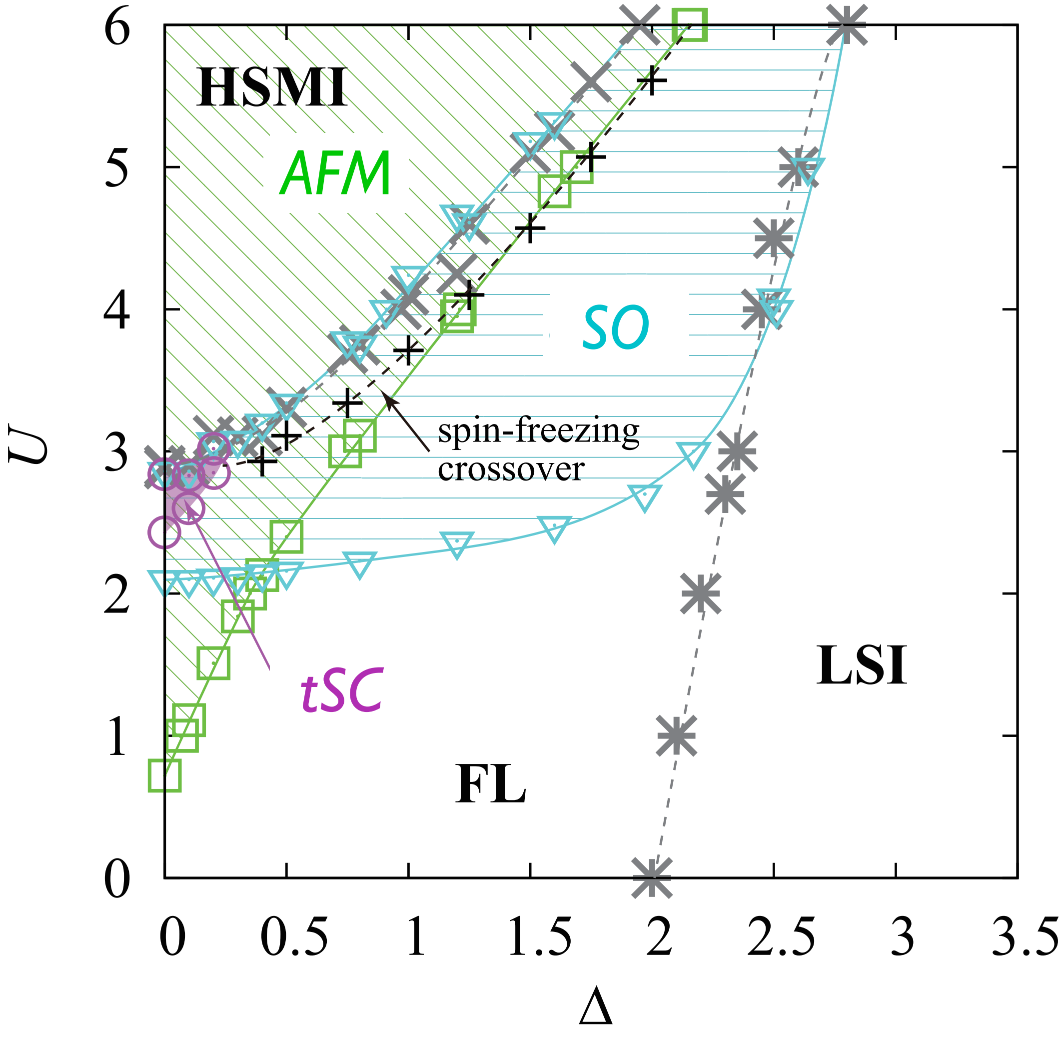
Hoshino & Werner, PRB (2016)

Crystal field splitting

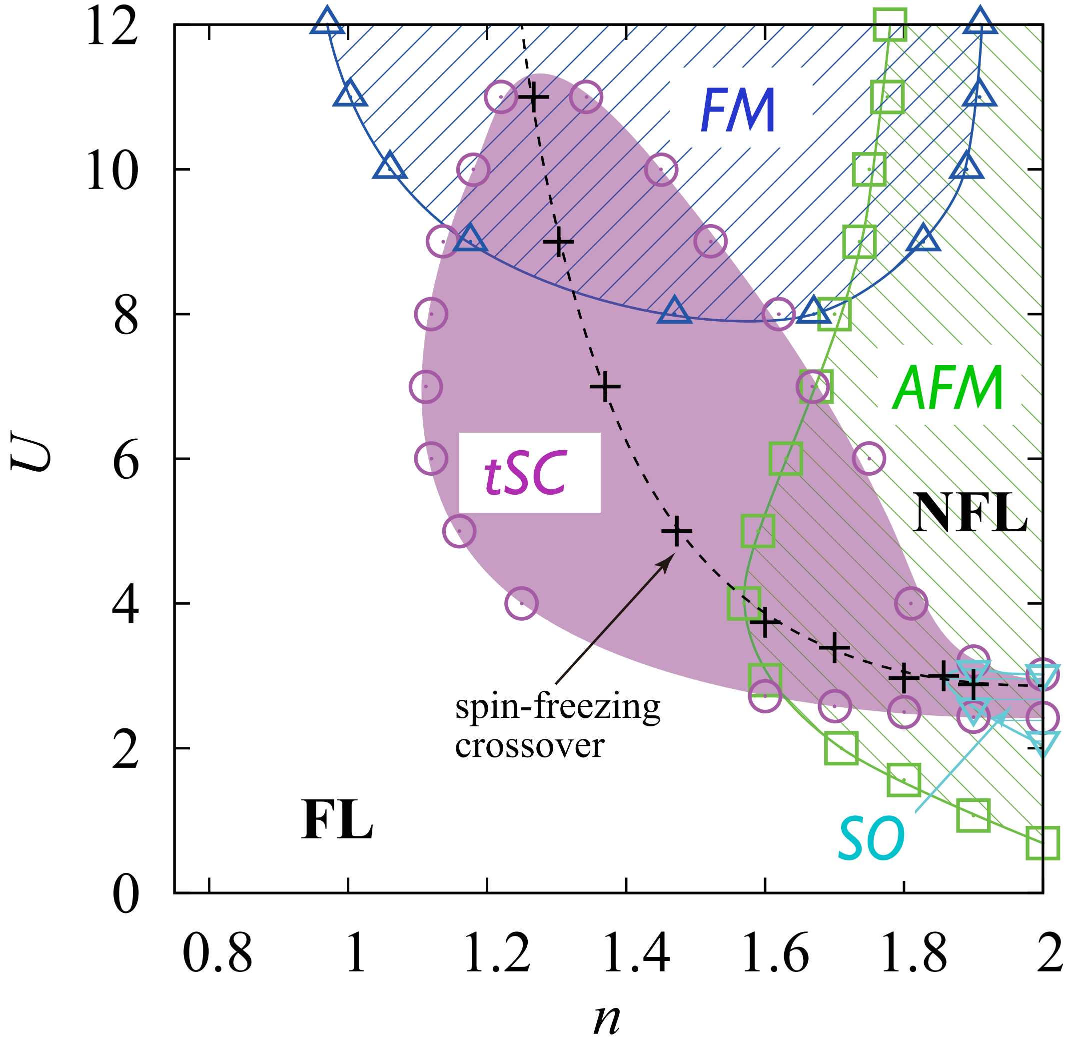
Hoshino & Werner, PRB (2016)

- Complicated phase diagrams, even in the two-orbital case
  - High-spin/low-spin transitions *Werner & Millis, PRL (2007)*
  - Spin-orbital order (excitonic insulator phases) *Kunes et al., PRB (2014)*

$\Delta > 0, n = 2$



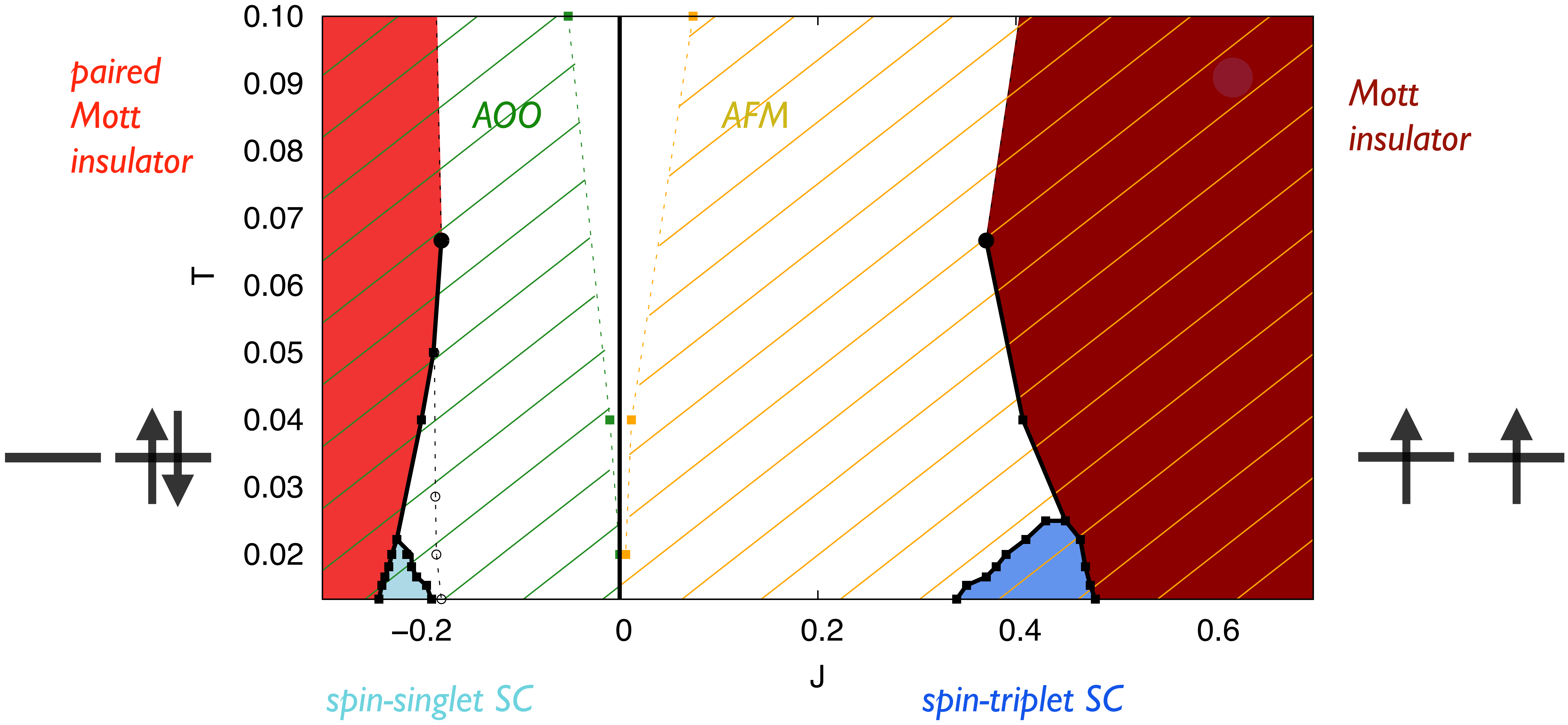
$\Delta = 0, n < 2$



Negative J and orbital freezing

Steiner et al., PRB (2016)

- 2-orbital model (*U*=bandwidth=4)



Negative J and orbital freezing

Steiner et al., PRB (2016)

- 2-orbital model (U=bandwidth=4)
- Mapping between J<0 and J>0:

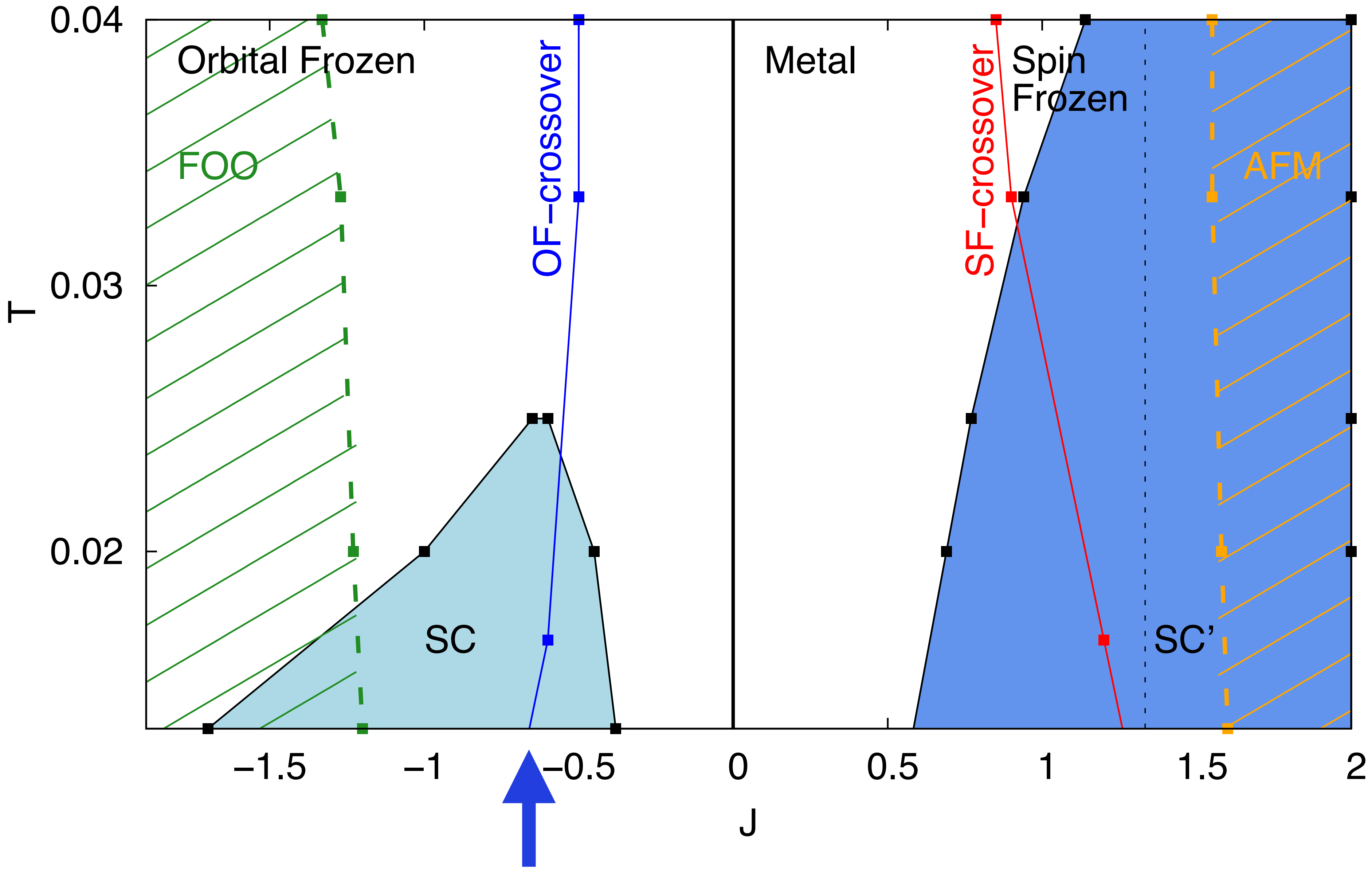
$$\begin{pmatrix} d_{i,1\downarrow} \\ d_{i,2\uparrow} \end{pmatrix} \longrightarrow \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} d_{i,1\downarrow} \\ d_{i,2\uparrow} \end{pmatrix}$$

J<0:		*J*>0:
spin-singlet SC	→	spin-triplet SC
antiferro OO	→	AFM
ferro OO	→	FM
orbital freezing	→	spin freezing

Negative J and orbital freezing

Steiner et al., PRB (2016)

- Away from half-filling: SC dome peaks near orbital freezing line



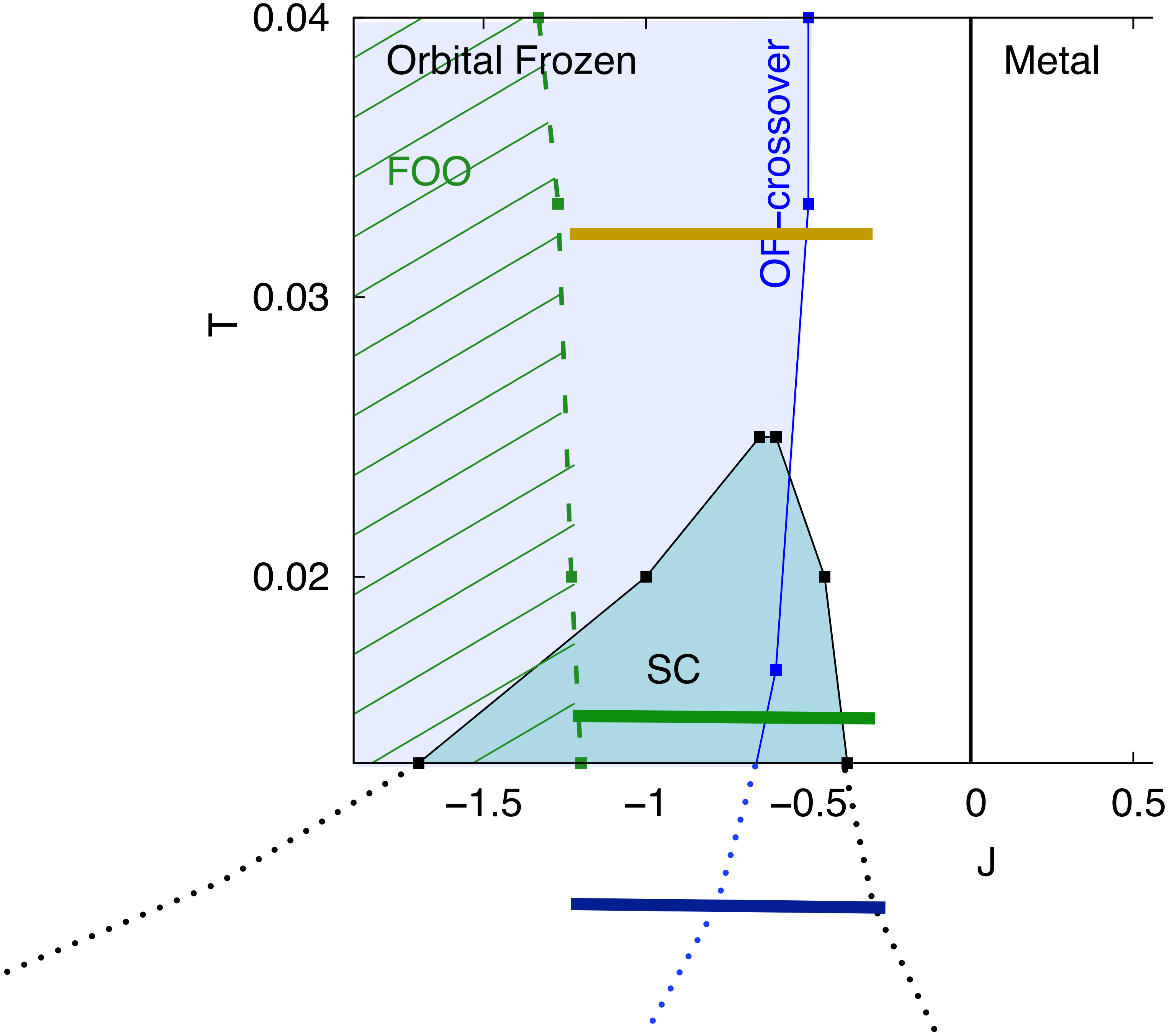
line of maximum orbital fluctuations

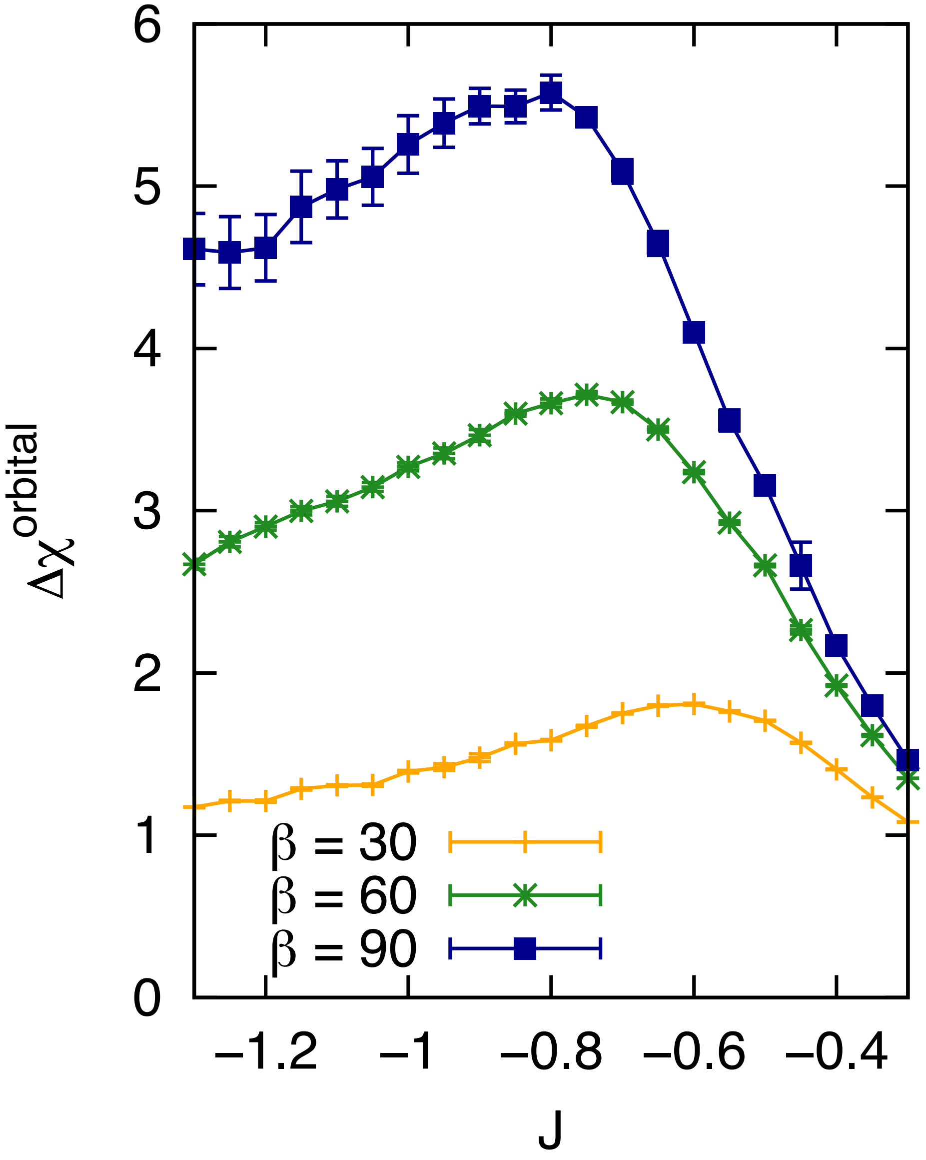
Negative J and orbital freezing

Steiner et al., PRB (2016)

- Away from half-filling: SC dome peaks near *orbital freezing* line

1.5 electrons in 2 orbitals





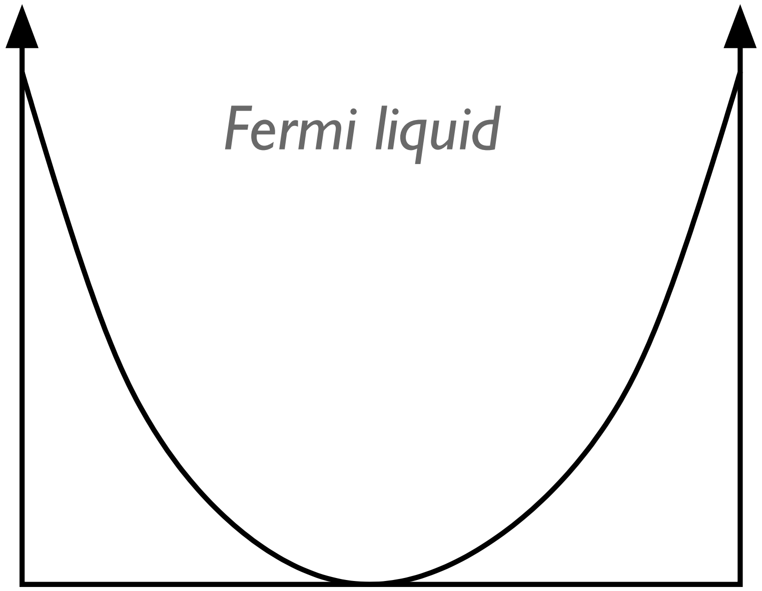
T-dependence of orbital fluctuations

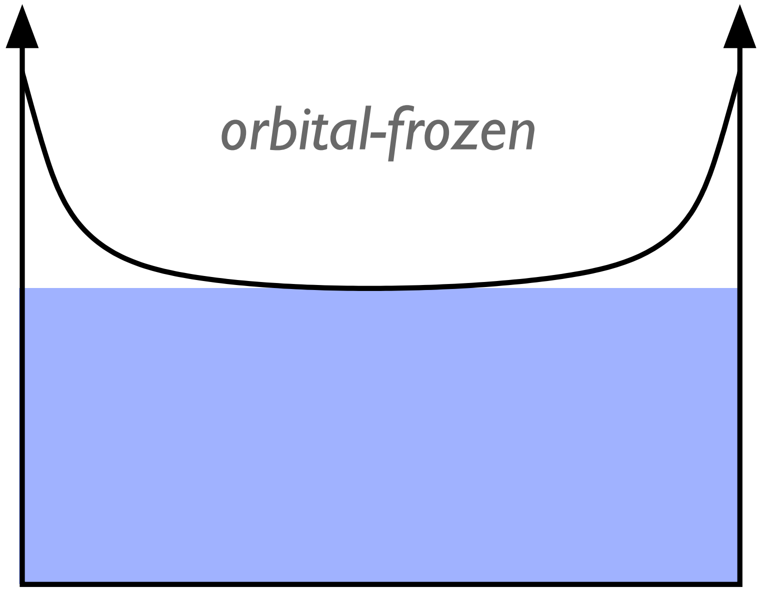
Negative J and orbital freezing

Steiner et al., PRB (2016)

- Orbital freezing seen in the decay of the (imaginary-time) orbital-orbital correlation function $\langle o(\tau) o(0) \rangle, \ o = n_1 - n_2$
  - fermi liquid metal: $\langle o(\tau) o(0) \rangle \sim 1/\tau^2 \ \ (\tau \text{ large})$
  - orbital-frozen metal: $\langle o(\tau) o(0) \rangle \sim \text{const} > 0$
- Orbital freezing crossover line: maximum of orbital fluctuations

$$\Delta\chi_{\text{orb}} \equiv \int_0^\beta d\tau [\langle o(\tau) o(0) \rangle - \langle o(\beta/2) o(0) \rangle]$$





Negative J and orbital freezing

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  - orbital-frozen metal: $\langle o(\tau)o(0)\rangle \sim \text{const} > 0$
- Orbital freezing crossover line: maximum of orbital fluctuations

$$\Delta\chi_{\text{orb}} \equiv \int_0^\beta d\tau[\langle o(\tau)o(0)\rangle - \langle o(\beta/2)o(0)\rangle]$$

- Orbital fluctuations induce attractive interaction for on-site pairs
  - Effective interaction which includes bubble diagrams:

analogous to: Inaba & Suga, PRL (2012)

$$\tilde{U}_{\alpha\beta}(q) = U_{\alpha\beta} - \sum_\gamma U_{\alpha\gamma}\chi_\gamma(q)\tilde{U}_{\gamma\beta}(q)$$

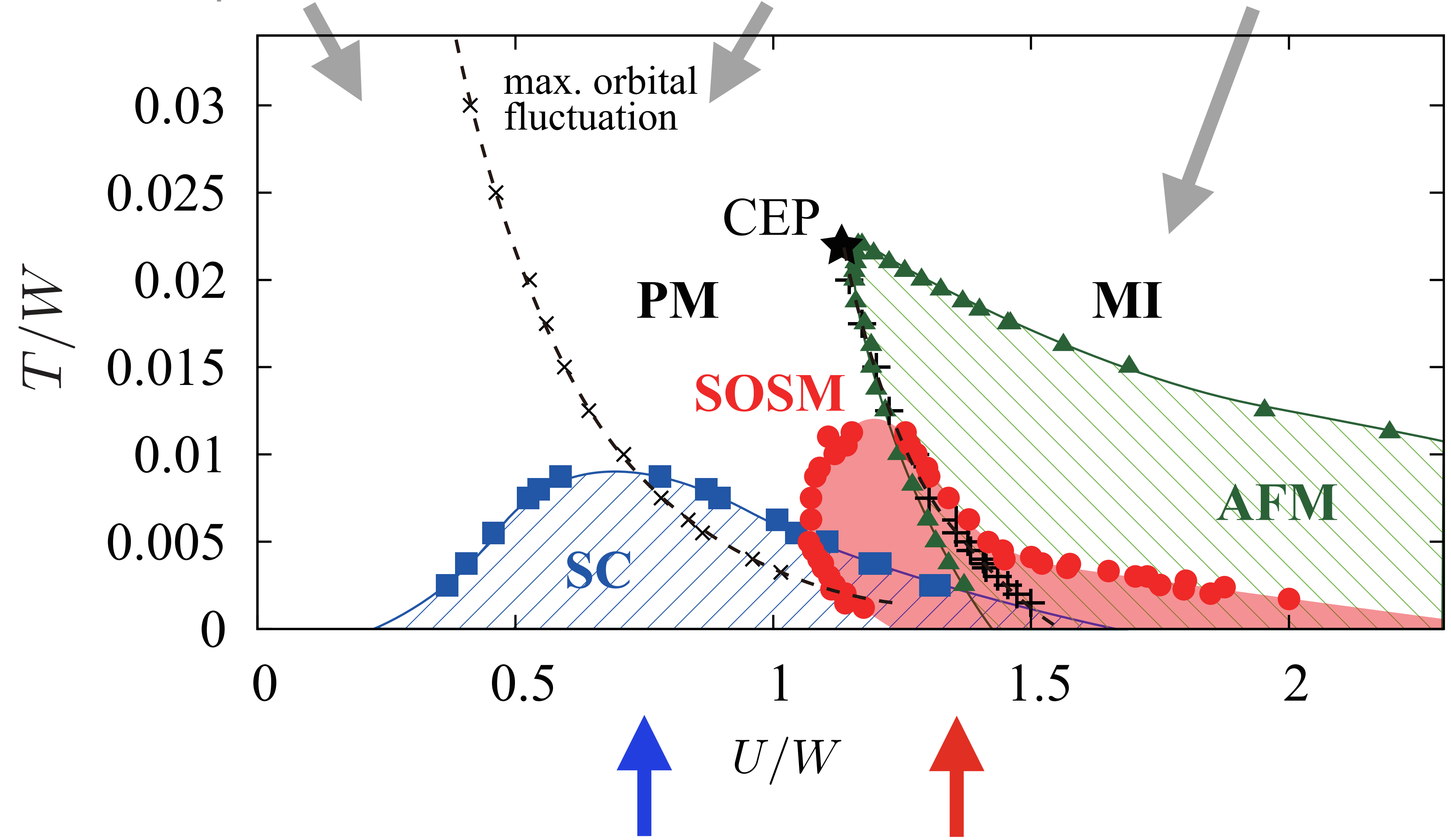
$$\Rightarrow \quad \tilde{U} = U - 4U'[U' + |J|]\Delta\chi_{\text{orb}} + O(U^3)$$

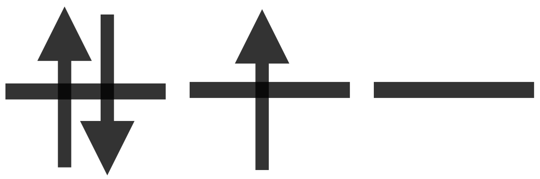
Negative J and orbital freezing

Hoshino & Werner, PRL (2016)

- Half-filled 3-orbital model (A_3C_{60})

Fermi liquid metal *orbital frozen metal* *Mott insulator*





SC dome peaks in the region of maximum orbital fluctuations

spontaneous symmetry breaking into an orbital selective Mott phase (“Jahn-Teller metal”)

Cuprates

Werner, Hoshino & Shinaoka, PRB (2016)

- Unconventional SC in the spin-freezing regime
  - Strontium ruthenates
  - Uranium-based SC
  - Pnictides
  - CrAs
  - ...

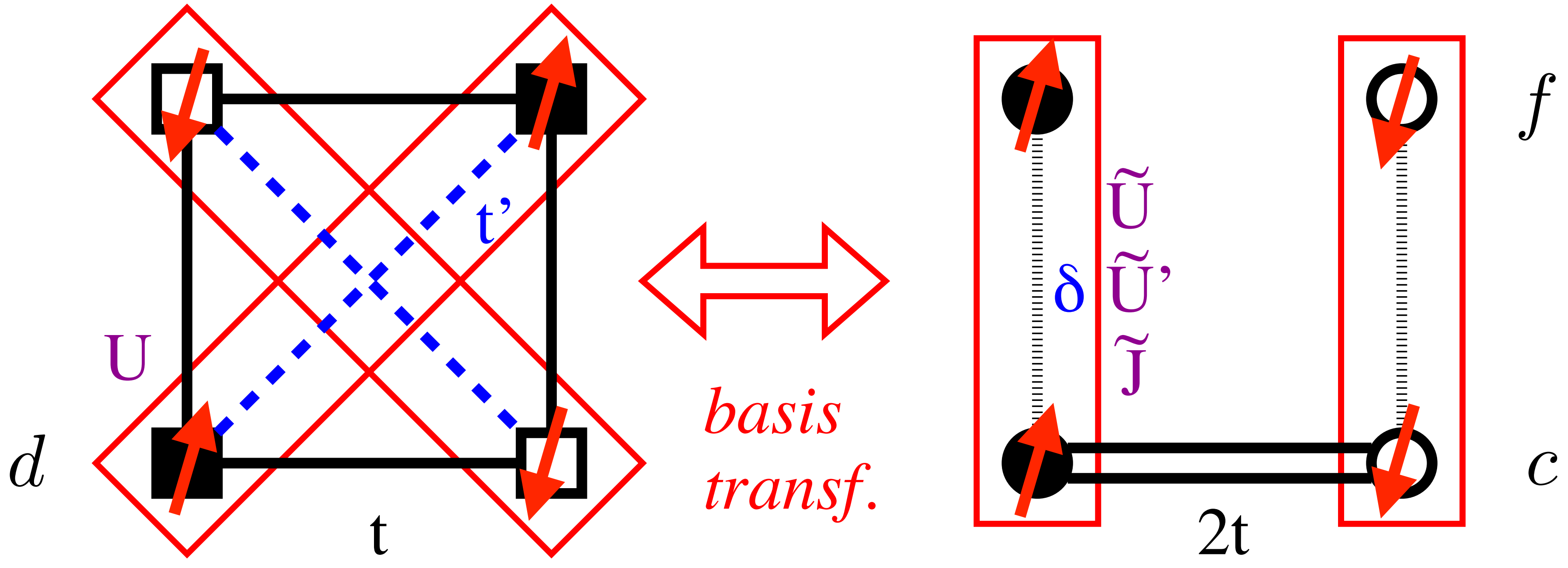
- Unconventional SC in the orbital-freezing regime
  - Alkali-doped fullerides

- What about cuprates? Can spin-freezing play any role in a single-band 2D Hubbard model?
  - naive answer: NO, correct answer: YES

Cuprates

Werner, Hoshino & Shinaoka, PRB (2016)

- Mapping to an effective two-orbital model:



$$c_1 = \frac{1}{\sqrt{2}}(d_1 + d_3) \quad c_2 = \frac{1}{\sqrt{2}}(d_2 + d_4)$$

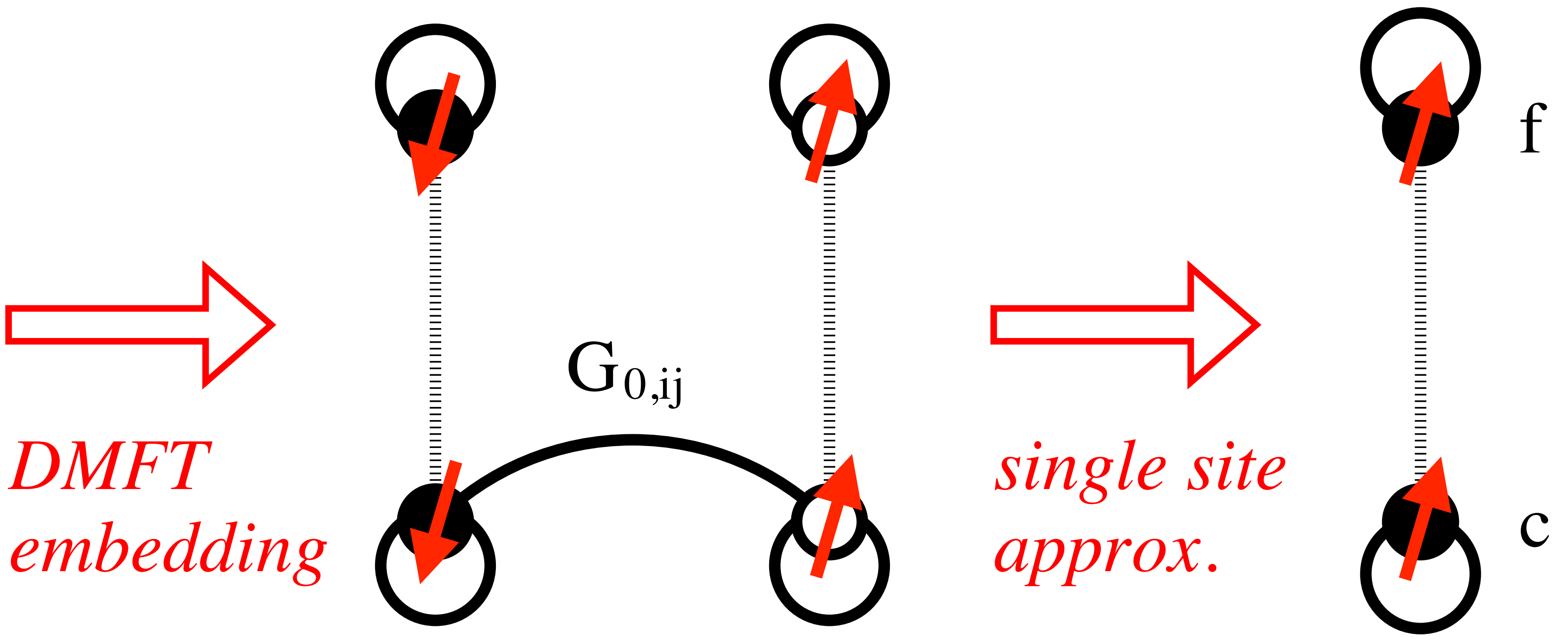
$$f_1 = \frac{1}{\sqrt{2}}(d_1 - d_3) \quad f_2 = \frac{1}{\sqrt{2}}(d_2 - d_4)$$

- Slater-Kanamori interaction with $\tilde{U} = \tilde{U}' = \tilde{J} = U/2$
  nnn hopping translates into a crystal-field splitting $\delta = 2t'$

Cuprates

Werner, Hoshino & Shinaoka, PRB (2016)

- Mapping to an effective two-orbital model:



$$c_1 = \frac{1}{\sqrt{2}}(d_1 + d_3) \quad c_2 = \frac{1}{\sqrt{2}}(d_2 + d_4)$$

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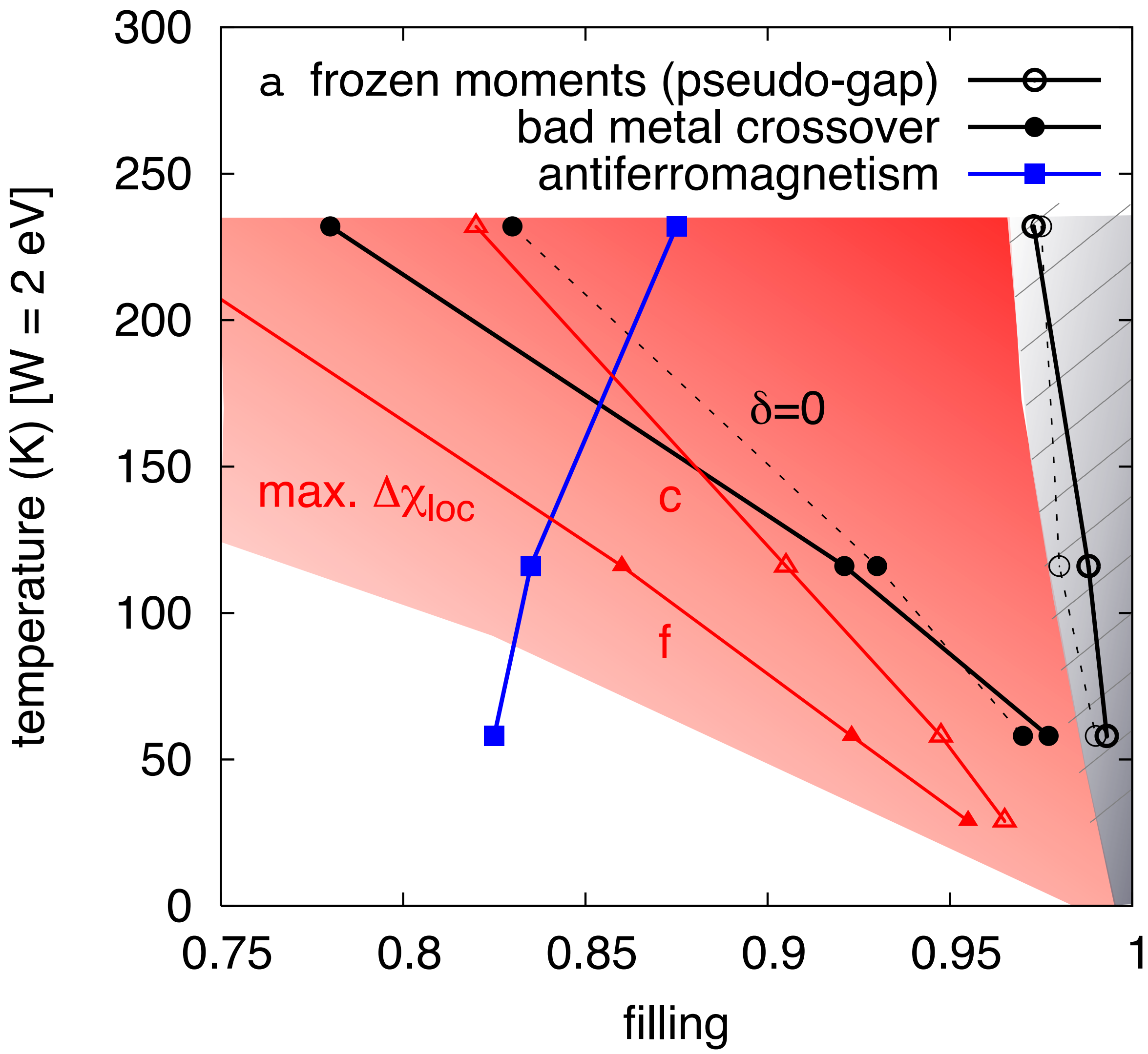
- Slater-Kanamori interaction with $\tilde{U} = \tilde{U}' = \tilde{J} = U/2$
  nnn hopping translates into a crystal-field splitting $\delta = 2t'$

Cuprates

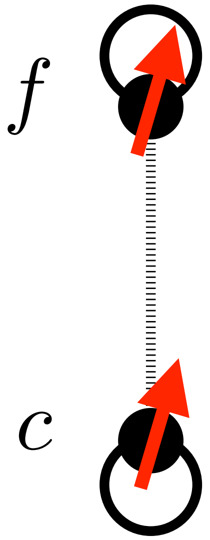
Werner, Hoshino & Shinaoka, PRB (2016)

- Phasediagram (1-site/2-orbital DMFT)

emerging (fluctuating)
local moments
= bad metal regime



frozen moments
=pseudo-gap phase

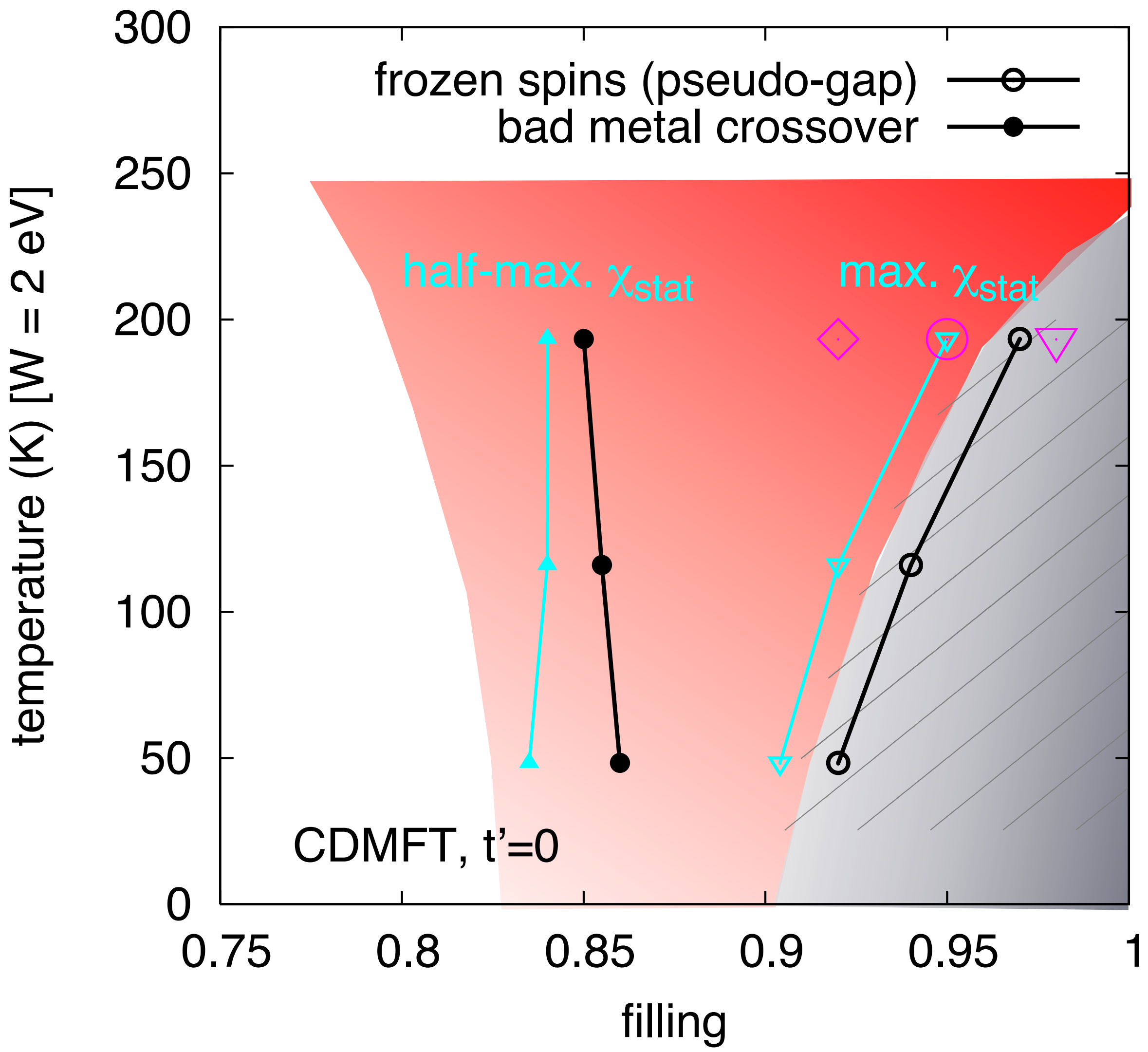


Cuprates

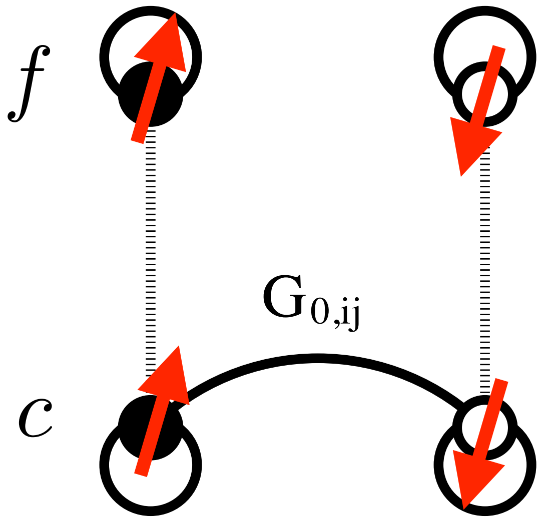
Werner, Hoshino & Shinaoka, PRB (2016)

- Phasediagram (2-site/2-orbital cluster DMFT)

emerging (fluctuating)
local moments
= bad metal regime



frozen moments
=pseudo-gap phase



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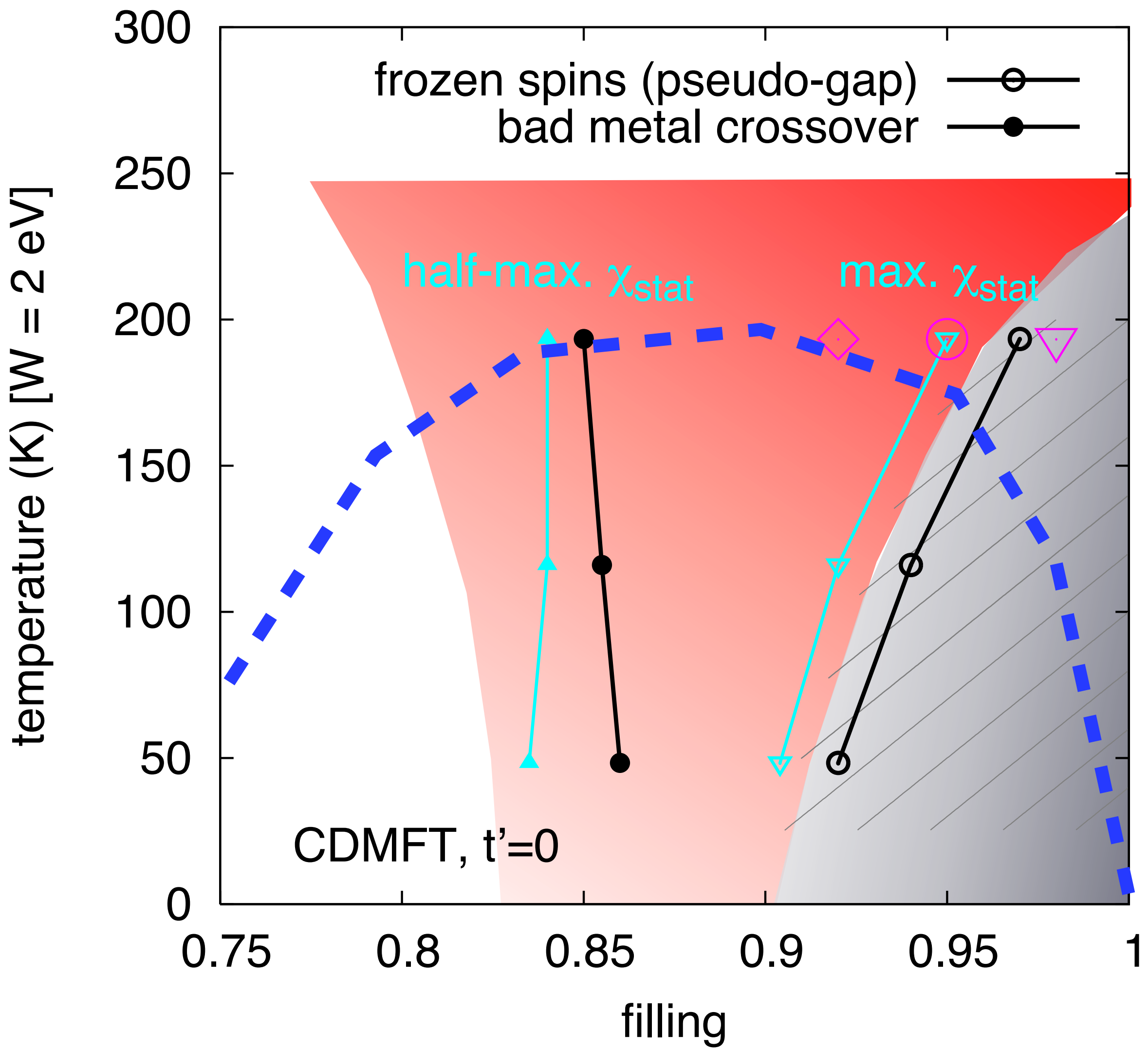


frozen moments =pseudo-gap phase

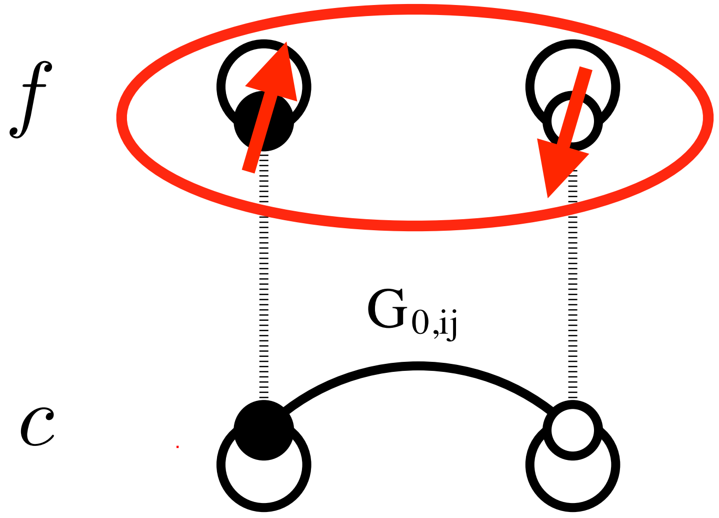
SC dome [4-site cluster DMFT, Maier et al, (2005)] induced by fluctuating local moments?

Cuprates

Werner, Hoshino & Shinaoka, PRB (2016)

- d-wave SC induced by local spin fluctuations
- Transformation of the d-wave order parameter:

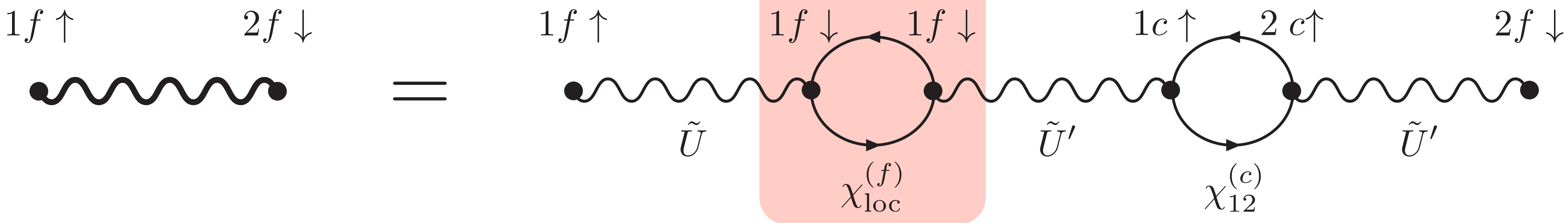
$$\begin{aligned}&(d^\dagger_{1\uparrow}d^\dagger_{2\downarrow}-d^\dagger_{1\downarrow}d^\dagger_{2\uparrow})-(d^\dagger_{2\uparrow}d^\dagger_{3\downarrow}-d^\dagger_{2\downarrow}d^\dagger_{3\uparrow})\\+&(d^\dagger_{3\uparrow}d^\dagger_{4\downarrow}-d^\dagger_{3\downarrow}d^\dagger_{4\uparrow})-(d^\dagger_{4\uparrow}d^\dagger_{1\downarrow}-d^\dagger_{4\downarrow}d^\dagger_{1\uparrow})\end{aligned}\quad\longrightarrow\quad 2(f^\dagger_{1\uparrow}f^\dagger_{2\downarrow}-f^\dagger_{1\downarrow}f^\dagger_{2\uparrow})$$



- Effective attractive interaction:

$$\tilde{U}^{\mathrm{eff}}_{(1,f,\uparrow),(2,f,\downarrow)}=2\tilde{U}^3\chi^{(f)}_{\mathrm{loc}}\chi^{(c)}_{12}+O(\tilde{U}^5)$$

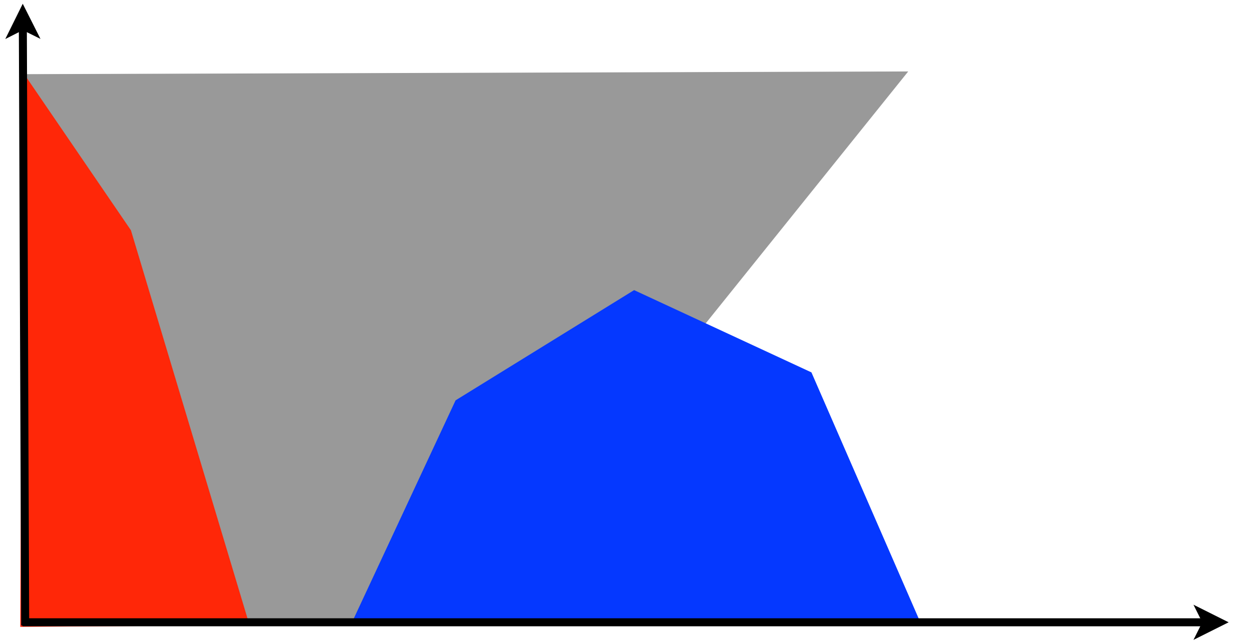
- Leading contribution:



local spin fluctuations (needed because U'-J=0)

Summary I

- Spin/orbital freezing as a universal phenomenon in unconventional superconductors
  - Strontium ruthenates
  - Uranium-based SC
  - Pnictides
  - Fulleride compounds
  - Cuprates
  - ...

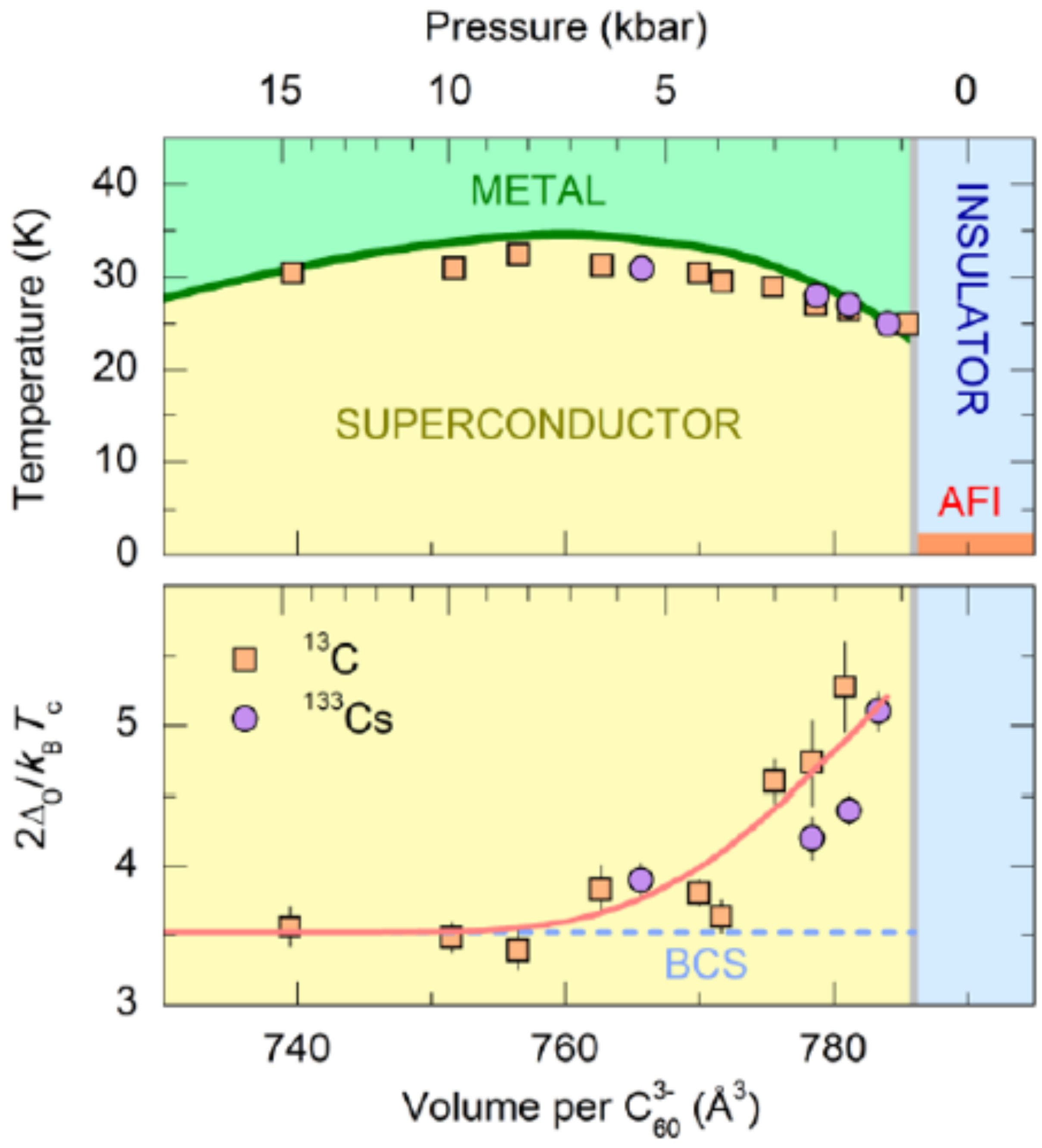


- Pairing induced by <u>local</u> spin or orbital fluctuations

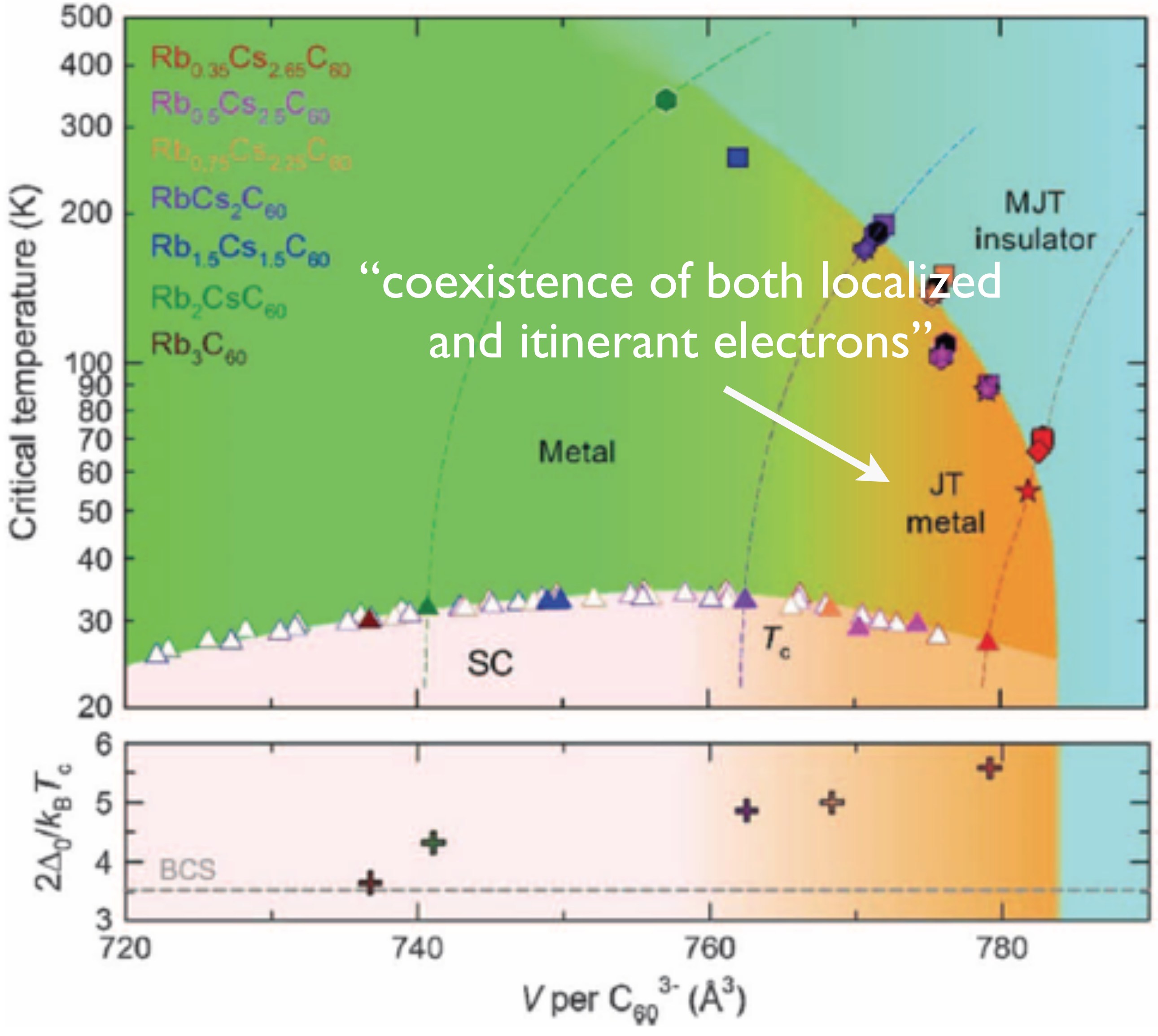
- Bad metal physics originates from fluctuating/frozen moments

Jahn-Teller metal

- Experimental results for Cs_3C_{60} and $Rb_xCs_{3-x}C_{60}$



Potocnik et al., Sci. Rep. (2014)

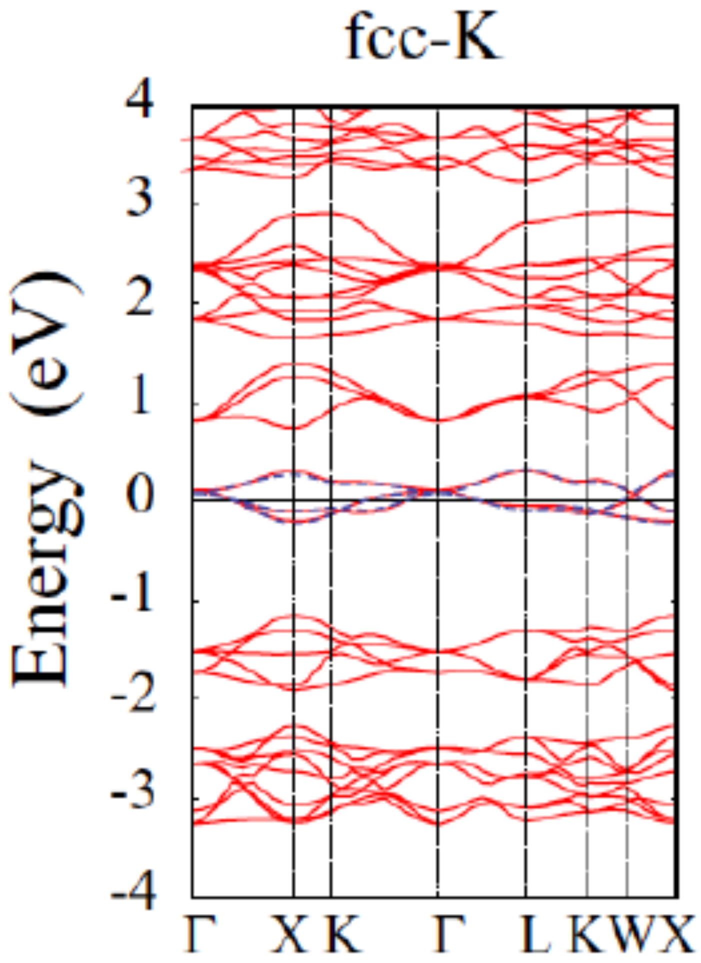


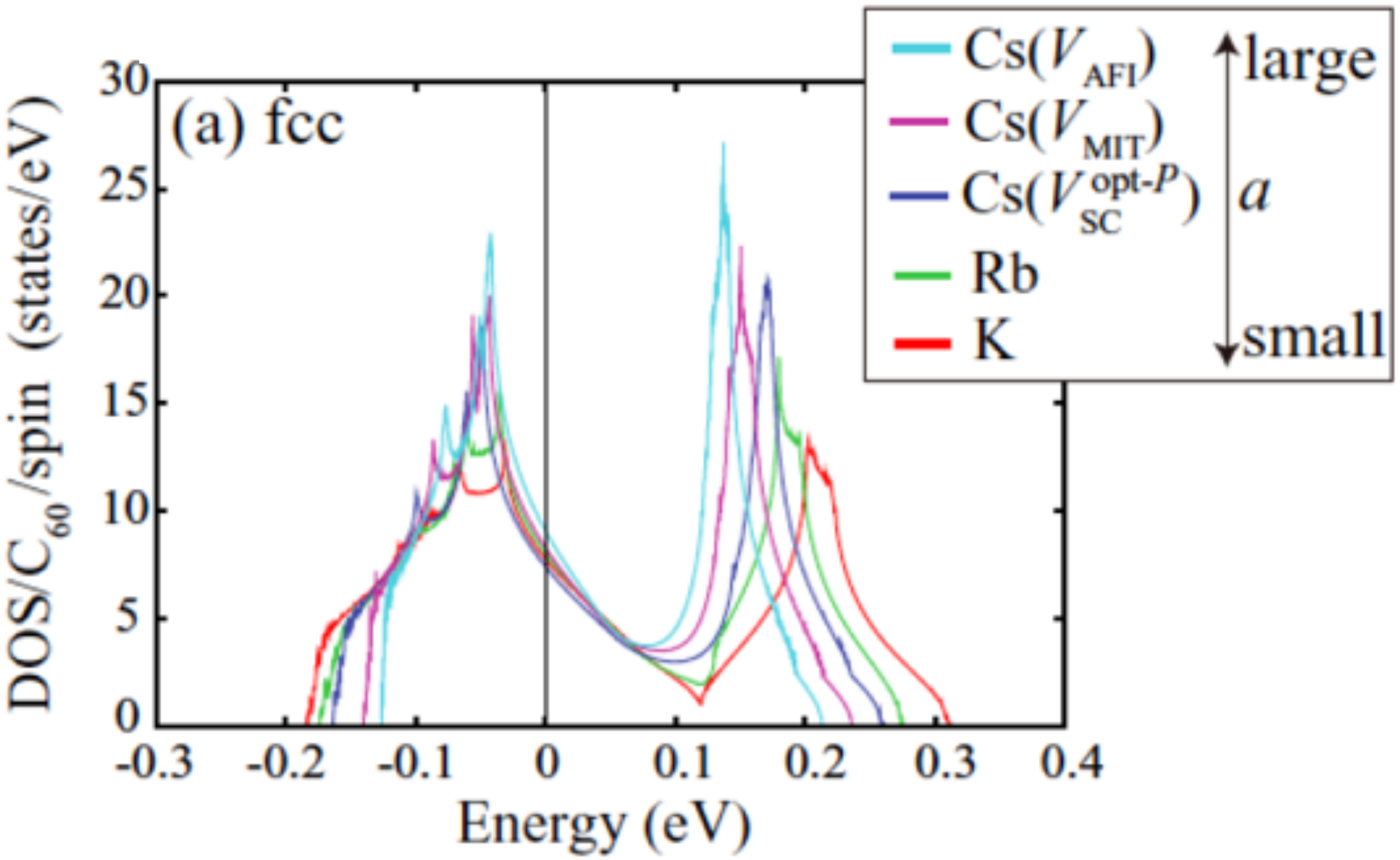
"coexistence of both localized and itinerant electrons"

Zadik et al., Sci. Express (2015)

3-band model of A_3C_{60}

- Bandstructure
  - 3 bands near Fermi level
  - half-filling
  - bandwidth ~ 0.4 eV, increasing correlations from A=K to Cs



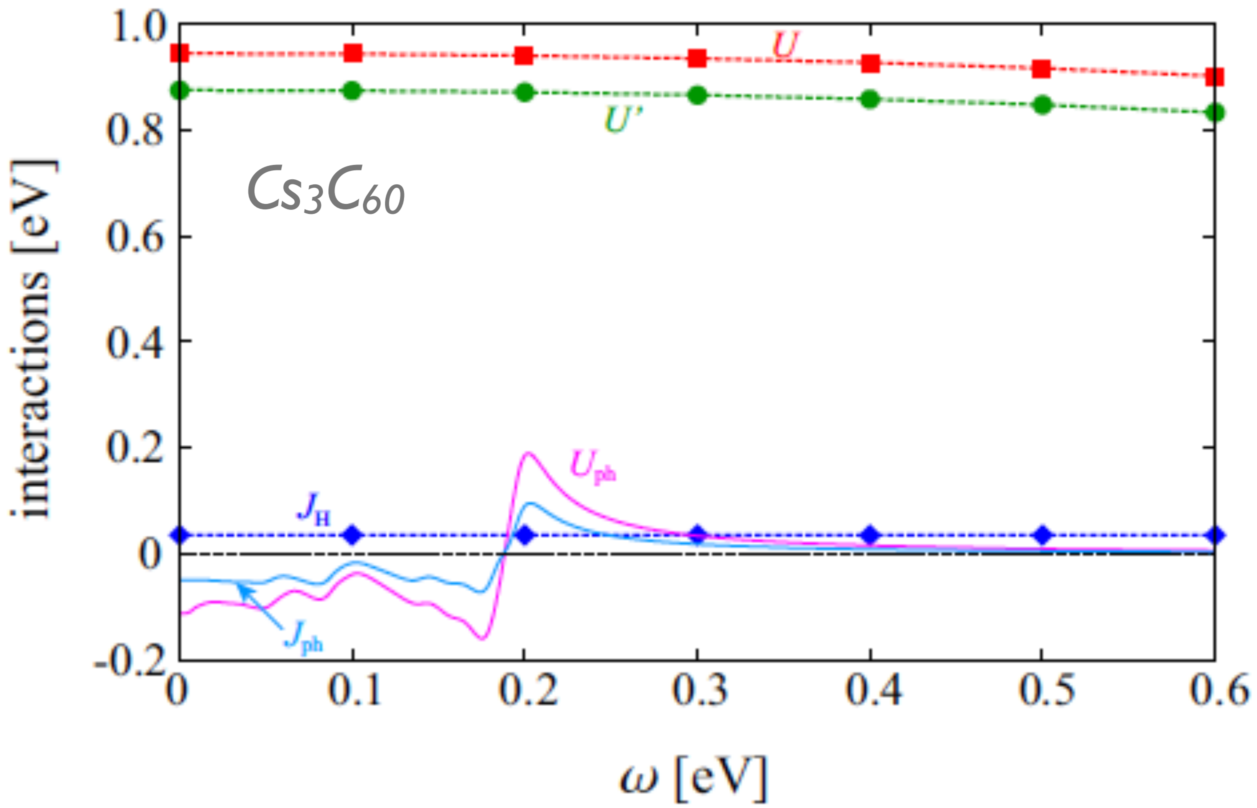


Nomura et al. (2012)

3-band model of A_3C_{60}

- Inverted Hund coupling
  - *U*~1eV > bandwidth ⟶ strongly correlated
  - Extended molecular orbitals ⟶ small bare *J* (~0.035 eV)
  - Reduction of *J* by 0.05 eV due to Jahn-Teller phonons:

$$J_{\text{eff}} = J_H(0) - J_{\text{ph}}(0) \approx -0.02 \text{ eV}$$

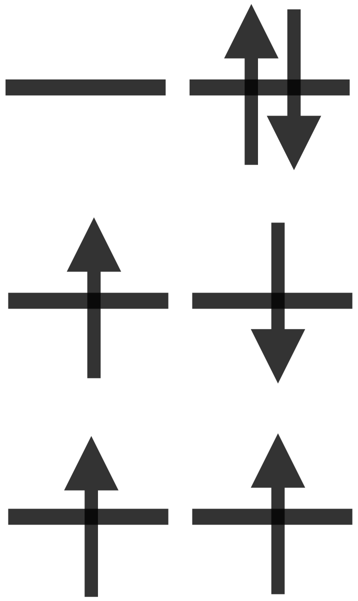


Nomura et al. (2012)

3-band model of A_3C_{60}

Capone et al., Science (2002)
Nomura et al., Science Expr. (2015)

- Inverted Hund coupling
  - Lowest energy atomic state has paired electrons ("seed" for SC)



$$U$$

$$U - 2J > U$$

$$U - 3J > U$$

  - $|J|$ small compared to bandwidth, but E_{kin} strongly reduced by large *U*: cooperation between correlation and phonon effects

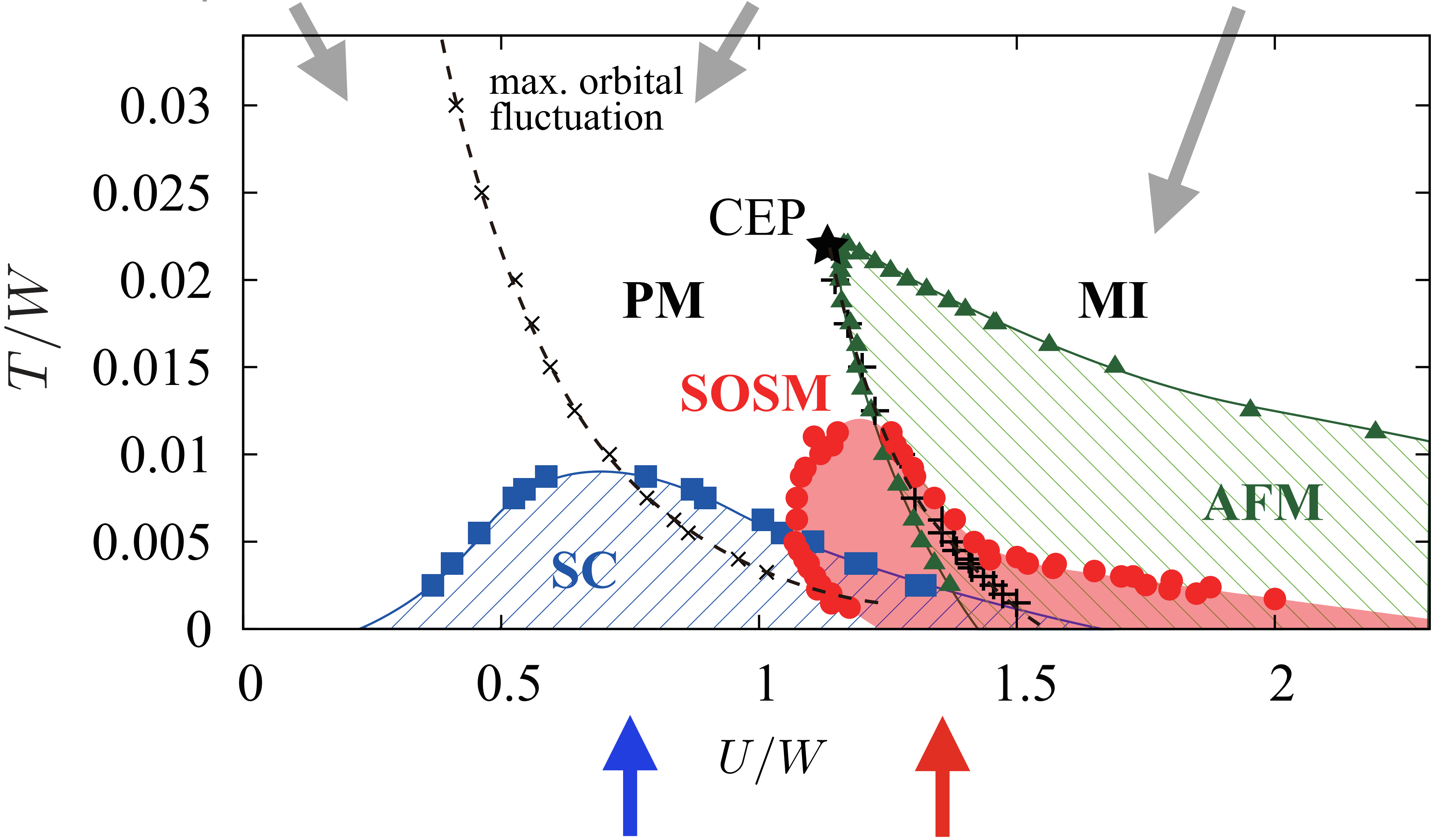
  - For superconductivity, pairs have to be mobile: important role of pair-hopping term

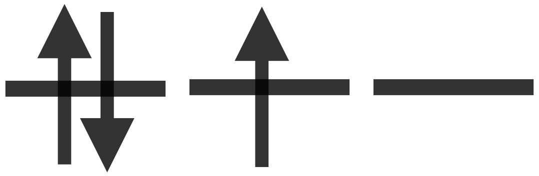
Jahn-Teller metal

Hoshino & Werner, PRL (2017)

- Half-filled 3-orbital model (A_3C_{60})

Fermi liquid metal *orbital frozen metal* *Mott insulator*





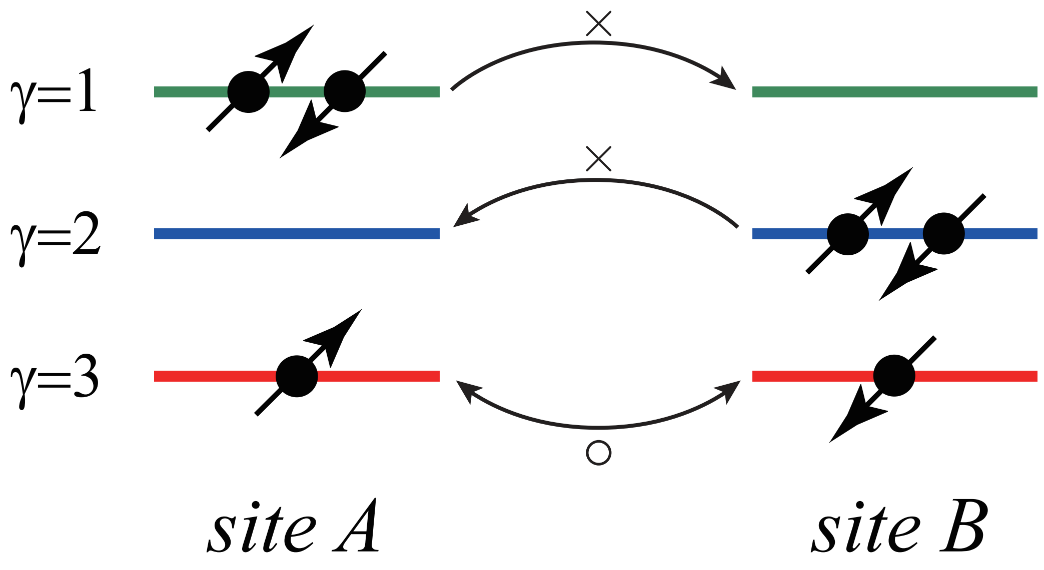
SC dome peaks in the region of maximum orbital fluctuations

spontaneous symmetry breaking into an orbital selective Mott phase ("Jahn-Teller metal")

Jahn-Teller metal

Hoshino & Werner, PRL (2017)

- Half-filled 3-orbital models with negative J exhibit a symmetry-broken phase characterized by a composite order parameter
  - completely degenerate bands
  - no ordinary orbital moment (all orbitals half-filled)
  - but: orbital-dependent double-occupation



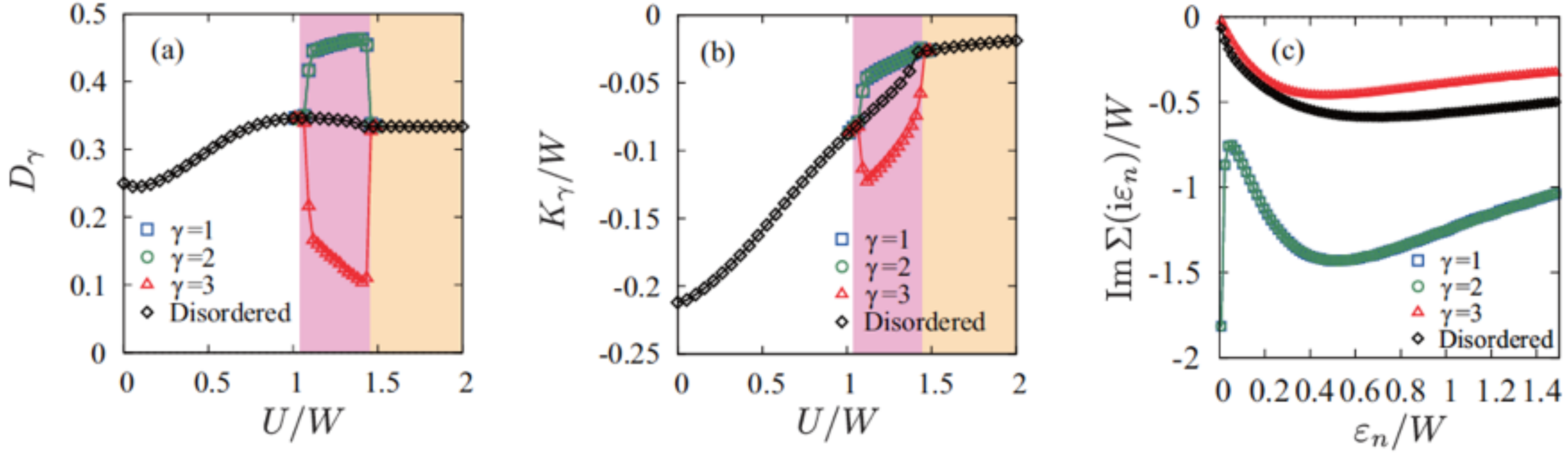
  - coexistence of Mott insulating and metallic orbitals

Jahn-Teller metal

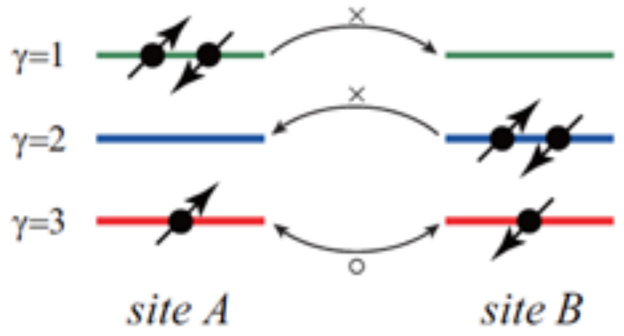
Hoshino & Werner, PRL (2017)

- DMFT results for $J = -U/4$, density-density interaction
  - orbital-dependent double occupation (a), kinetic energy (b), and self-energy (c)

orbital-selective Mott insulator *metal* *paired Mott insulator*



enhanced D in the (paired) Mott insulator



Odd-frequency order

Hoshino & Werner, PRL (2017)

- Define the time-dependent orbital moment

$$T^{\eta}(\tau) = \sum_{i\gamma\gamma'\sigma} \langle c^{\dagger}_{i\gamma\sigma} \lambda^{\eta}_{\gamma\gamma'} c_{i\gamma'\sigma}(\tau) \rangle = T^{\eta}_{\text{even}} + T^{\eta}_{\text{odd}}\tau + O(\tau^2)$$

Gell-Mann matrix

ordinary orbital moment (=0)

- Odd time/frequency component characterizes the SOSM state

$$T^{8}_{\text{odd}} = \sum_{\gamma} \lambda^{8}_{\gamma\gamma}(K_{\gamma} + 2UD_{\gamma}) + \text{terms depending on } U', J$$

oribtal-dependent E_{kin} and double occupation

- "diagonal order" version of odd-frequency superconductivity

Berezinskii (1974), Kirkpatrick & Belitz (1991)

Summary II

- A_3C_{60}: 3-band system with strong U and inverted J

- T_c dome: enhanced pairing in the orbital-freezing crossover region
    - Analogous to unconventional superconductivity induced by spin-freezing in systems with J>0

- Jahn-Teller metal: symmetry-broken state with a composite order parameter (orbital-dependent double occupation)
    - Coexistence of 2 Mott insulating and 1 metallic orbital
    - Diagonal-order analogue of odd-frequency superconductivity