

# MOTT CRITICALITY AS THE CONFINEMENT TRANSITION OF A PSEUDOGAP-MOTT METAL

## PRESENTATION FOR POSTDOCTORAL POSITION AT ICTS-TIFR

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JANUARY 7, 2026



# A BRIEF INTRODUCTION

University of  
Calcutta  
**(B.Sc. Physics)**

IISER Kolkata  
**(M.S.)**

IISER Kolkata  
**(Ph.D.,  
Prof. S Lal)**



2018



2021



2026\*

## Research Interests

- Mott transition and criticality
- Unconventional superconductivity
- Kondo breakdown in heavy-fermion systems
- Various forms of quantum matter

## Skills and Techniques

- Field theory (RG methods) and effective theory-based techniques
- **Numerical computation** of correlation functions, entanglement measures, dynamical correlations (spectral function, self-energy, etc). Developed Python and **Julia libraries** for the same.
- Developed **new method** for using impurity models to analyse interacting lattice models

## LIST OF PROJECTS

- ✓ **Mott Criticality as the Confinement Transition of a Pseudogap-Mott Metal.**  
arXiv:2507.17201 (2025)
- Revealing the magnetic dimensional crossover in the Heisenberg ferromagnet CrSiTe<sub>3</sub> through picosecond strain pulses. Phys. Rev. B 111, L140414 (2025)
- **Holographic entanglement renormalisation for fermionic quantum matter.** J. Phys. A: Math. Theor. 57 275401 (2024)
- **Kondo frustration via charge fluctuations: a route to Mott localisation.** New J. Phys. 25 113011 (2023)
- Frustration shapes multi-channel Kondo physics: a star graph perspective. J. Phys.: Condens. Matter 35 315601 (2023)
- Unveiling the Kondo cloud: Unitary renormalization-group study of the Kondo model. Phys. Rev. B 105, 085119 (2022)

# A NOTE OF THANKS



Prof. Siddhartha  
Lal (IISER K)



Prof. N. S.  
Vidhyadhiraja  
(JNCASR)



Prof. A.  
Taraphder (IIT  
KGP)



Prof. A.  
Mukherjee  
(NISER)



Prof. S. R. Hassan  
(IMSc)



- SERB & IISER Kolkata for funding
- PARAM RUDRA HPC facility for compute time

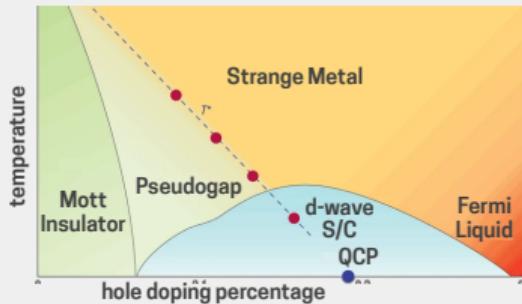


# **MOTT CRITICALITY AS THE CONFINEMENT TRANSITION OF A PSEUDOGAP-MOTT METAL**

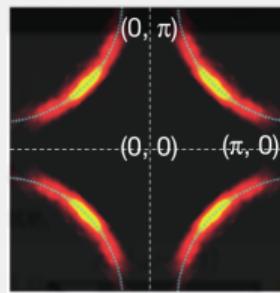
**ARXIV:2507.17201 (2025). ABHIRUP MUKHERJEE, S R. HASSAN, A MUKHERJEE, N S. VIDHYADHIRAJA, A TARAPHDER, S LAL**

# THE CHALLENGE: THE PSEUDOGAP AND STRANGE METAL

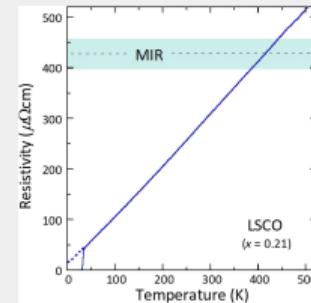
- Nature & origin of pseudogap and strange metal phases of hole-doped Mott insulators
- Difficulty in understanding several puzzling experimental observations.



Schematic **High-T<sub>c</sub> SC** Phase Diagram



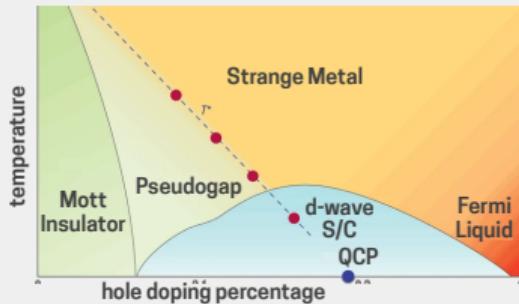
ARPES plot of **Pseudogap**:  
gapped and gapless regions coexist



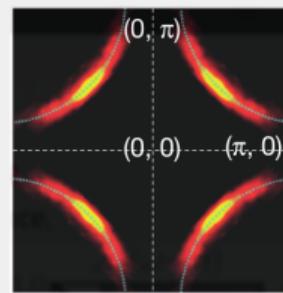
Linear Resistivity in **Strange Metal** crosses MIR bound

# THE CHALLENGE: THE PSEUDOGAP AND STRANGE METAL

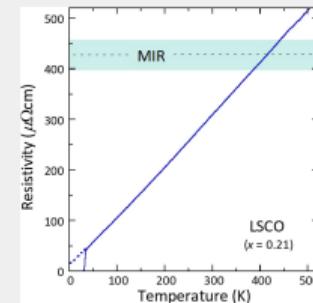
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Schematic **High-Tc SC** Phase Diagram



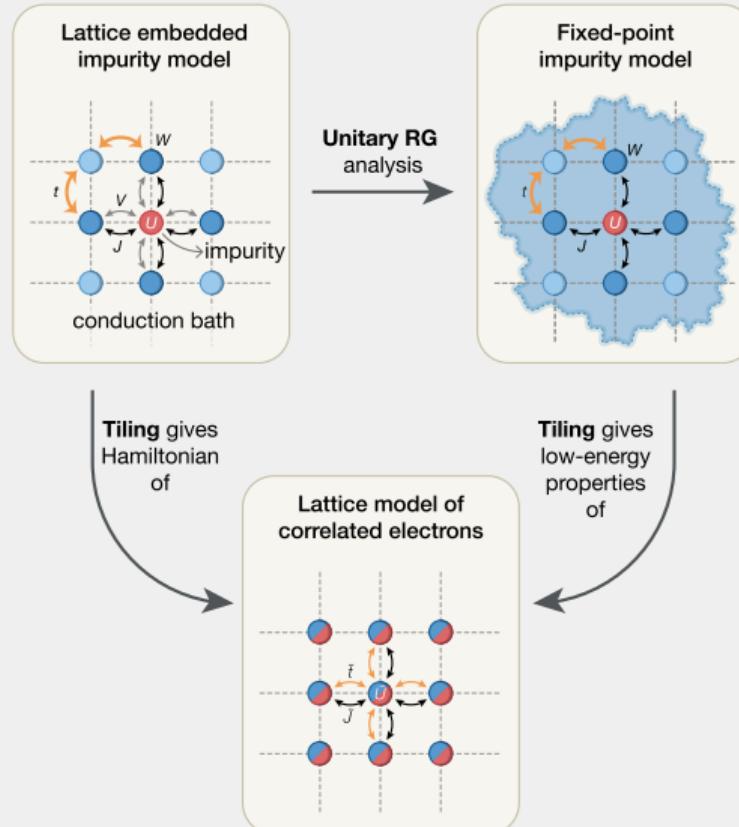
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Linear Resistivity in **Strange Metal** crosses MIR bound

- Pseudogap a **precursor** to Mott insulator/ Superconductor? Nature of its excitations?
- Which metal is **parent phase** of Mott insulator, **at 1/2-filling** ?
- Is strange metal a new scale-invariant **long-range entangled** strongly interacting phase?

# OUR APPROACH, IN A NUTSHELL



Solve an **impurity model**  $H_{\text{imp}}$  with certain properties:

- Lattice symmetry
- Localisation transition

**Construct correlated lattice** model by applying many-body translation operators (“tiling”)

- Restore discrete translation invariance

Analyse impurity mode using **unitary RG** \* method.

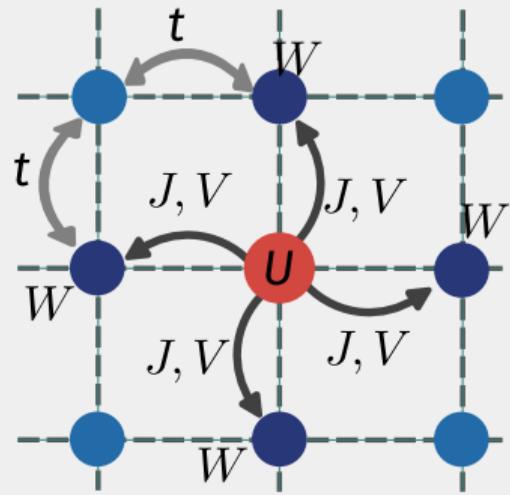
Tile with **fixed-point impurity model** for low-energy properties of lattice model.

# THE CORE INGREDIENT: A LATTICE-EMBEDDED IMPURITY MODEL

Lattice-variant of extended Anderson impurity model

- **Red site:** correlated impurity site (strong local  $U$ )
- **Rest of the sites:** conduction bath (hopping  $t$ )
- Impurity-bath hybridisation: **Kondo**  $J$ , hopping  $V$
- Weak **local interaction**  $W$  on N.N. bath sites

For  $U \gg V$ , Mott transition driven by  $J$  and  $W^{**}$



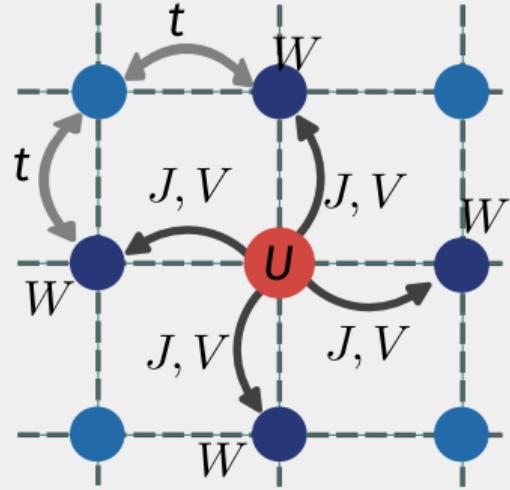
\*\*Observed in  $\infty$ -dimensional problem: Mukherjee et al., NJP (2023)

# THE CORE INGREDIENT: A LATTICE-EMBEDDED IMPURITY MODEL

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$$H_{\text{aux}} = H_{\text{coup}} + H_{\text{cbath}}$$

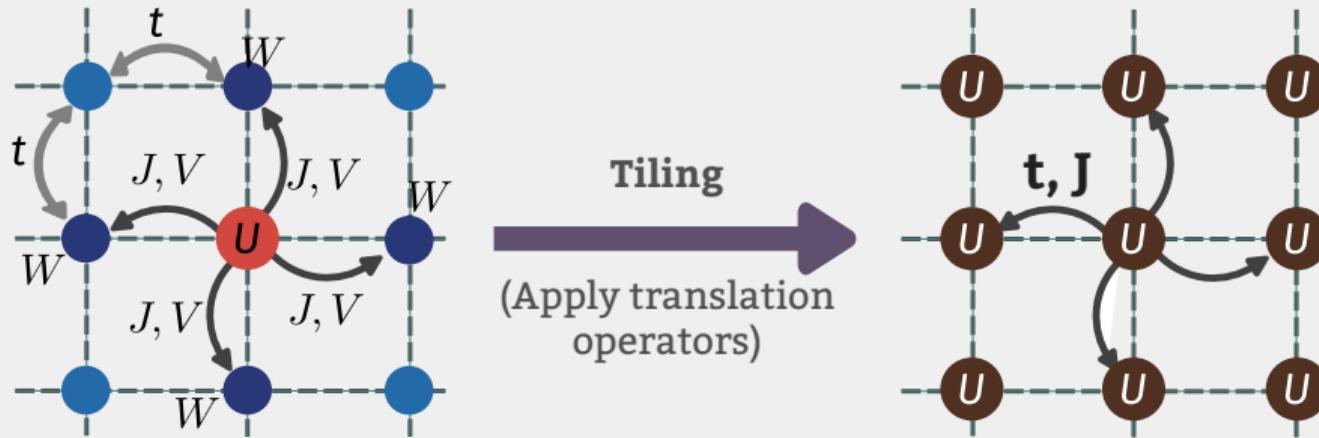
$$H_{\text{cbath}} = \sum_{k,\sigma} \epsilon_k n_{k,\sigma} - \frac{W}{2} \sum_Z (n_{Z\uparrow} - n_{Z\downarrow})^2, \quad Z = \text{N.N}$$

$$H_{\text{coup}} = J \sum_Z \mathbf{S}_d \cdot \mathbf{S}_Z, \quad J_{k,k'} = \frac{J}{2} [\cos(k_x - k'_x) + \cos(k_y - k'_y)]$$

\*\*Observed in  $\infty$ -dimensional problem: Mukherjee et al., NJP (2023)

# TILING FROM IMPURITY MODEL TO TWO DIMENSIONS

“Tiling” allows us to translate physics obtained by from the impurity model into that of the 2D extended Hubbard model



**Reconstructed lattice Hamiltonian:**

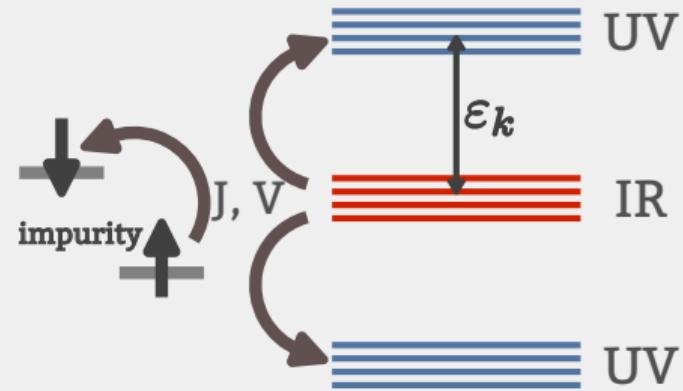
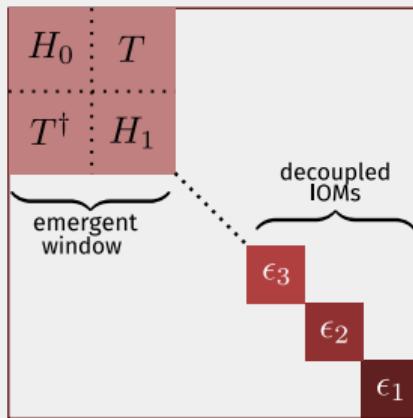
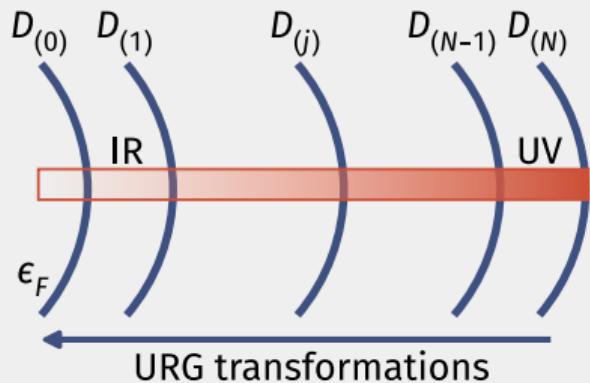
$$-\frac{\tilde{t}}{\sqrt{Z}} \sum_{\langle \mathbf{r}_i, \mathbf{r}_j \rangle; \sigma} (c_{\mathbf{r}_i, \sigma}^\dagger c_{\mathbf{r}_j, \sigma} + \text{h.c.}) + \frac{\tilde{J}}{Z} \sum_{\langle \mathbf{r}_i, \mathbf{r}_j \rangle} \mathbf{s}_{\mathbf{r}_i} \cdot \mathbf{s}_{\mathbf{r}_j} - \frac{1}{2} \tilde{U} \sum_{\mathbf{r}} (\hat{n}_{\mathbf{r}, \uparrow} - \hat{n}_{\mathbf{r}, \downarrow})^2$$

$$\tilde{t} = 2V/Z$$

$$\tilde{U} = U + W$$

$$\tilde{J} = 2J/Z$$

# UNITARY RENORMALISATION SCHEME

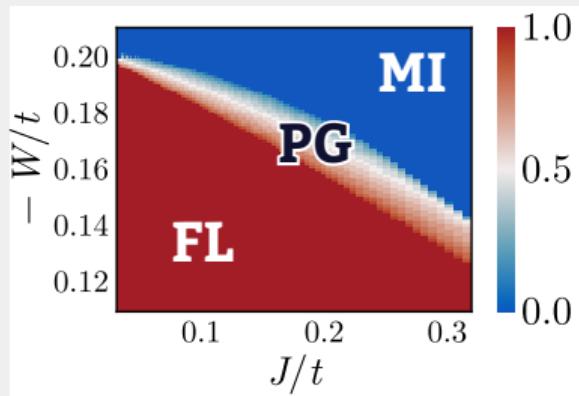
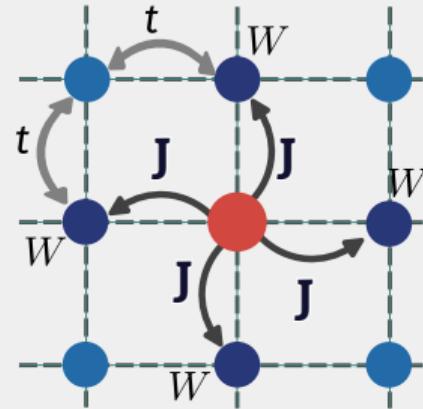


- Wanted: **low energy theory**
- RG procedure accounts for non-trivial scattering into various energy sectors
- RG proceeds via decoupling of high energy degrees of freedom via many-body **unitary** transformations

# UNITARY RG PHASE DIAGRAM AND PSEUDOGAPPING TRANSITION

**Competition** between Kondo coupling and local interaction on bath sites:

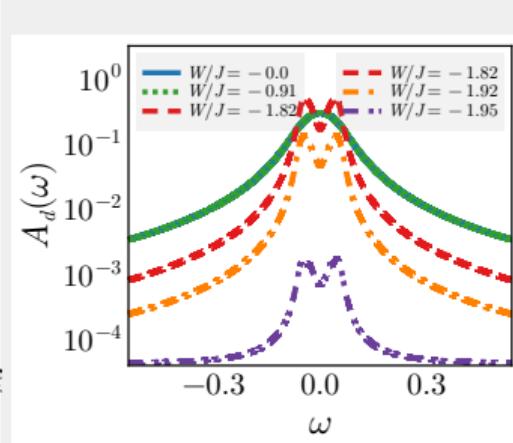
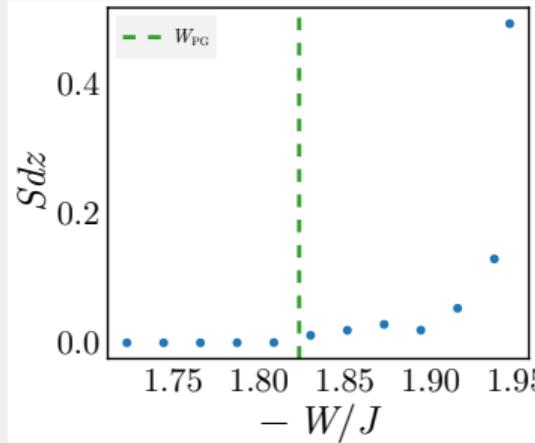
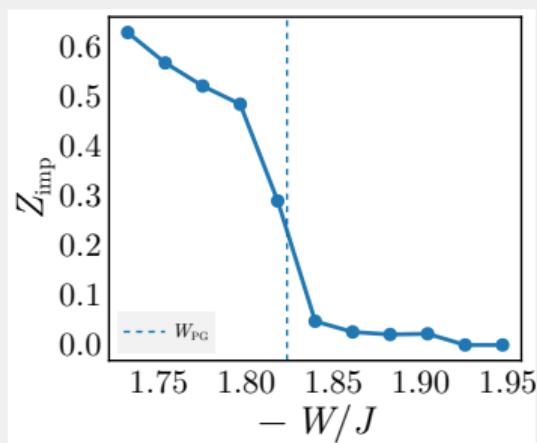
$$\Delta J_{k_1, k_2}^{(j)} = -2 \sum_{\mathbf{q}} \frac{J_{k_2, \mathbf{q}}^{(j)} J_{\mathbf{q}, k_1}^{(j)} + 4 J_{\mathbf{q}, \bar{\mathbf{q}}}^{(j)} W_{\mathbf{q}, k_2, k_1, \mathbf{q}}}{\omega - \frac{1}{2} |\varepsilon_j| + J_{\mathbf{q}}^{(j)}/4 + W_{\mathbf{q}}/2}$$



- Competition leads to **Kondo breakdown** for  $W < 0$
- Phase diagram shows **pseudogap** phase lying between Fermi liquid (FL) and Mott insulator (MI).
- PG possesses non-Fermi liquid excitations – a **Mott Metal**

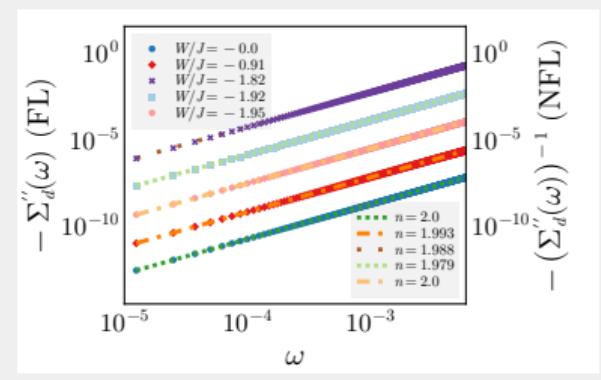
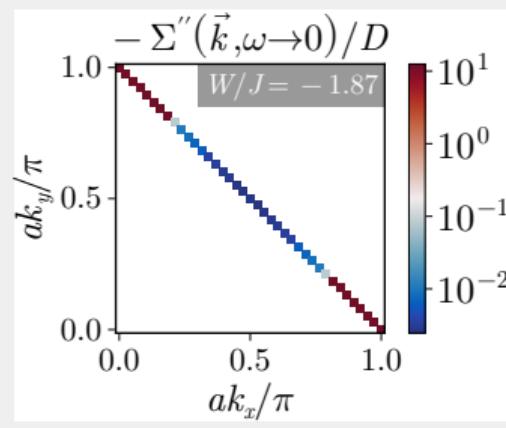
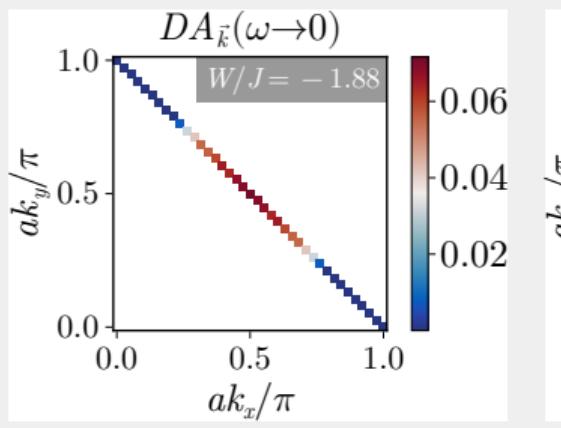
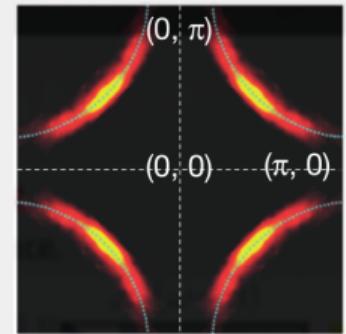
# JOURNEY INTO THE PSEUDOGAP

- Strength of Landau quasiparticle excitations of FL (**QP residue**  $Z$ ) vanishes upon entering PG.
- Impurity magnetisation  $\langle S_d^z \rangle$  grows dramatically in PG: **breakdown** of Kondo screening.
- Impurity spectral function shows **pseudogap** at  $\omega = 0$ !



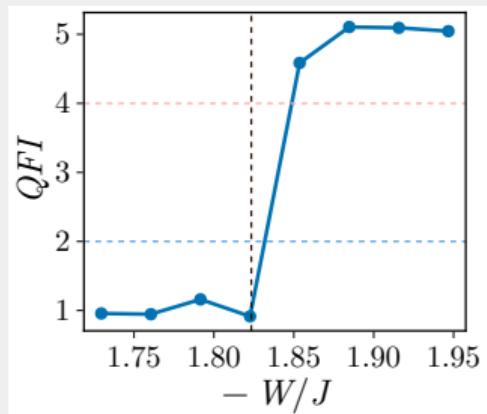
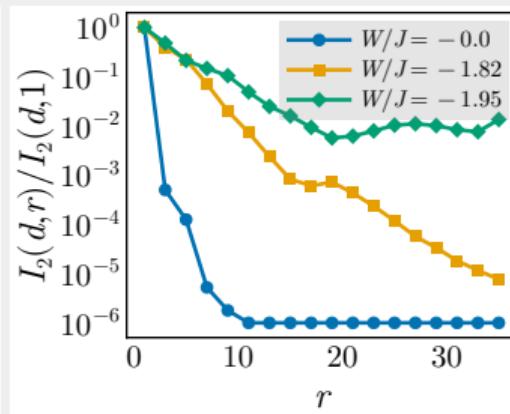
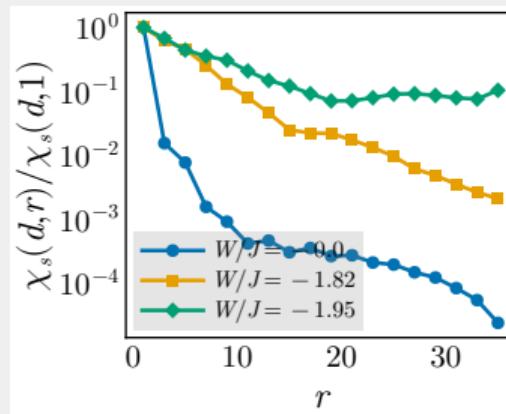
# LUTTINGER SURFACES IN THE PSEUDOGAP

- PG shows **electronic differentiation** in lattice spectral function: gapped antinodal regions (**Luttinger surfaces**), gapless excitations in nodal regions.
- Electron **scattering rate** shows divergences in gapped antinodal regions, while it is analytic in gapless nodal regions.
- $1/\Sigma'' \sim 1/\Sigma_0'' + \omega^2$ . Appearance of power-law exponents such as 2 signals **universality**.



# LONG-RANGED AND MULTIPARTITE ENTANGLEMENT IN THE PG

- real-space correlations and entanglement undergo a crossover within the pseudogap from short-ranged to **long-ranged** behaviour
- Quantum Fisher information for  $O = \sum_{\text{odd } i} (S_i^+ S_{i+1}^- + \text{h.c.})$  shows a jump in **multipartite entanglement** of 2 in FL to 5 within PG.
- Densely entangled **Quantum Soup** !



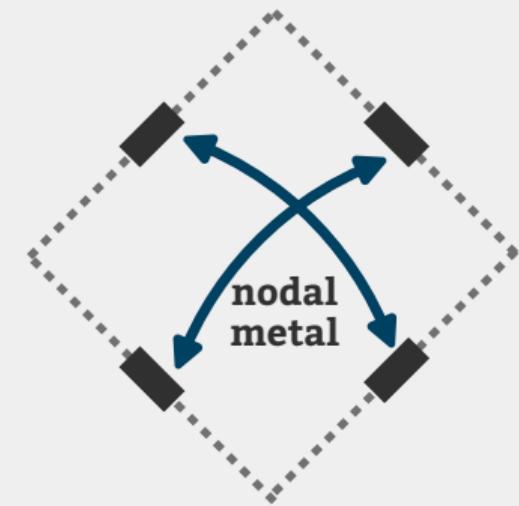
# SINGULAR NODAL METAL AT CRITICAL POINT

- Mott critical point has **nodal non-Fermi liquids**. Theory can be obtained in the form of exactly solvable **Hatsugai-Kohmoto model**!

$$H_{\text{eff}} = \sum_{q,\sigma} \epsilon_q r_{q,\sigma} + U \sum_{q,\sigma} r_{q\sigma} r_{q\bar{\sigma}}$$

$r_{q\sigma}$  : nested combination across FS

- Nodal metal is singular, i.e., has a Mott pole in self-energy at Fermi energy, but is still gapless. Gapless excitations are **holons & doublons**.



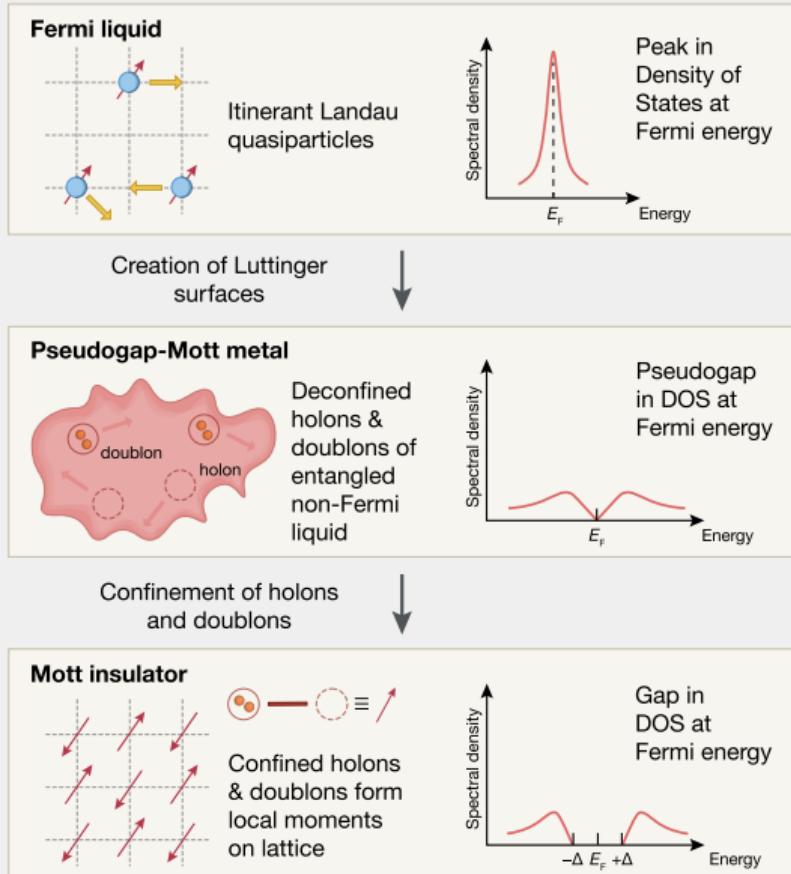
# CONCLUSIONS: MAIN TAKEAWAYS

Realisation of **Mott's original vision** (1949) with deconfined holes & doubles

- new, (likely) universal phase of **strongly interacting** quantum matter,
- **noisy**, incoherent environment for electron-like excitations,
- a long-ranged and **multipartite** entangled “quantum soup”,
- scale invariant at Mott critical point & described by exactly solvable model

## Open Questions:

Fate at non-zero temperatures, doping, other geometries?



## BRIEF MENTIONS OF OTHER PROJECTS

### **Kondo frustration via charge fluctuations: a route to Mott localisation**

**New J. Phys.** 25 113011 (2023). **Abhirup Mukherjee**, N S. Vidhyadhiraja, A Taraphder, S Lal  
Precursor to the Mott metal work. Demonstrated how an extended Anderson impurity model captures the  $d = \infty$  Mott MIT on the Bethe lattice.

### **Holographic entanglement renormalisation for fermionic quantum matter**

**J. Phys. A: Math. Theor.** 57 275401 (2024). **Abhirup Mukherjee**, S Patra, S Lal  
Demonstration of the holographic principle by showing how entanglement renormalisation in a free fermion system leads to a holographic dimension.

### **Revealing the magnetic dimensional crossover in the Heisenberg ferromagnet CrSiTe<sub>3</sub> through picosecond strain pulses**

**Phys. Rev. B** 111, L140414 (2025). A Kumar N M, S Mukherjee, **Abhirup Mukherjee**, A Punjal, S Purwar, T Setti, S Prabhu S., S Lal, N Kamaraju  
Investigated the two-step magnetic dimensional crossover in CrSiTe<sub>3</sub>. We came up with a simple Ginzburg-Landau model of phonons interacting with the lattice spin fluctuations to explain the softening/gapping of various phonon modes observed from a pump-probe experiment.

## BRIEF MENTIONS OF OTHER PROJECTS

### **Frustration shapes multi-channel Kondo physics: a star graph perspective**

**J. Phys.: Condens. Matter** 35 315601 (2023). S Patra, **Abhirup Mukherjee**, A Mukherjee, N S Vidhyadhiraja, A Taraphder, S Lal

We investigated the single-channel Kondo model and demonstrated the presence of two-particle correlations and entanglement within the Kondo cloud in the form of an effective Hamiltonian; we also calculated how they evolved during the high to low-temperature crossover.

### **Unveiling the Kondo cloud: Unitary RG study of the Kondo model**

**Phys. Rev. B** 105, 085119 (2022). A Mukherjee, **Abhirup Mukherjee**, N S. Vidhyadhiraja, A Taraphder, S Lal

Shed light on the role played by the ground state degeneracy in the non-Fermi liquid physics - how it leads to an orthogonality catastrophe in the low-energy excitations and how it modified the various correlations into anomalous forms.

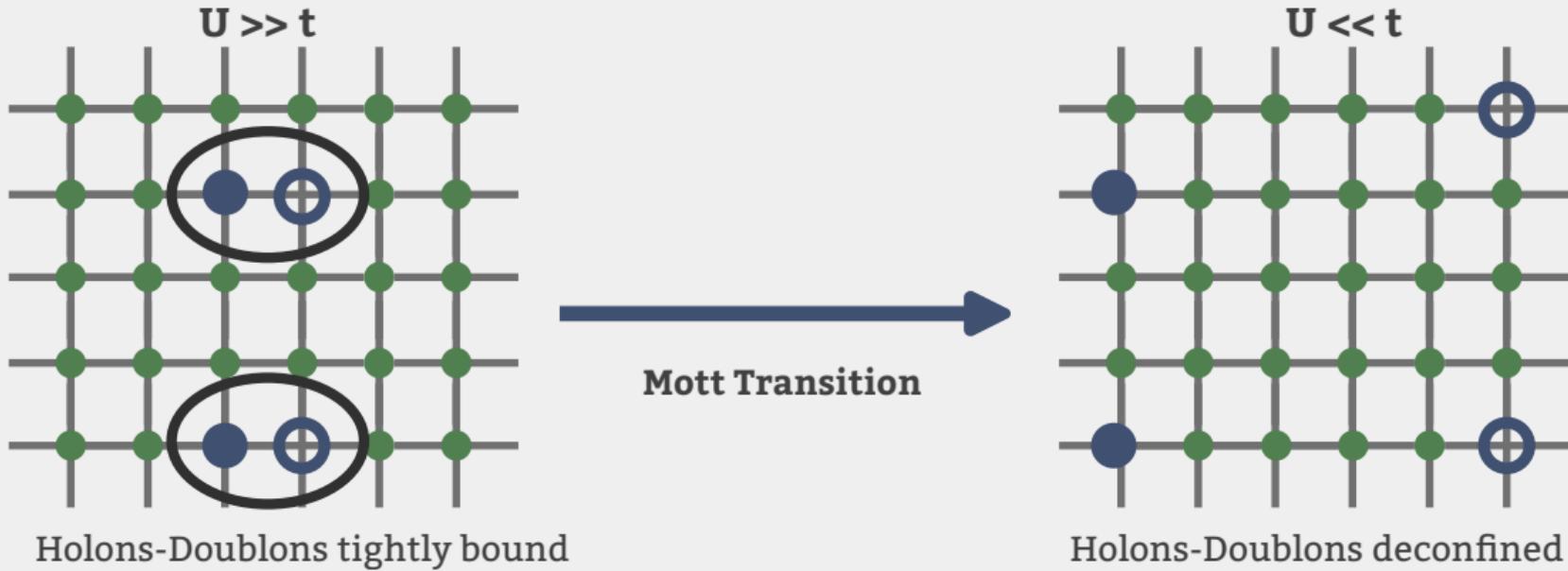
## FUTURE INTERESTS

- Investigations into **quantum critical/non-Fermi liquid** behaviour in correlated models (randomness, heavy-fermions, etc)
  - **Spin liquids** : Phase transitions, nature of entanglement, etc
  - Topological phases of matter
- 

**Thank You**

# MOTT MIT AS HOLON-DOUBLON DECONFINEMENT

The Mott MIT is essentially a holon-doublon **binding-unbinding** transition



- Delocalised **gas of holons & doublons** form metal: Fermi liquid?
- Nature and mechanism of transition?

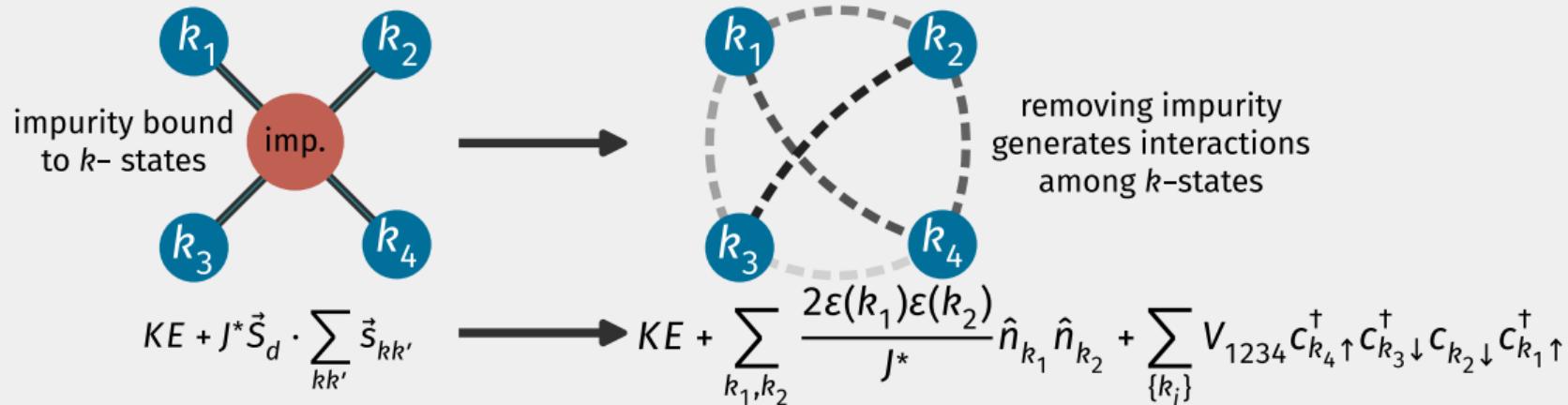
# **THE SINGLE-CHANNEL KONDO PROBLEM: ANATOMY OF THE KONDO CLOUD**

**Anirban Mukherjee et al., Phys. Rev. B 105, 085119**

# EFFECTIVE HAMILTONIAN FOR THE KONDO CLOUD

We first applied the **unitary RG** to obtain a low energy fixed point theory.

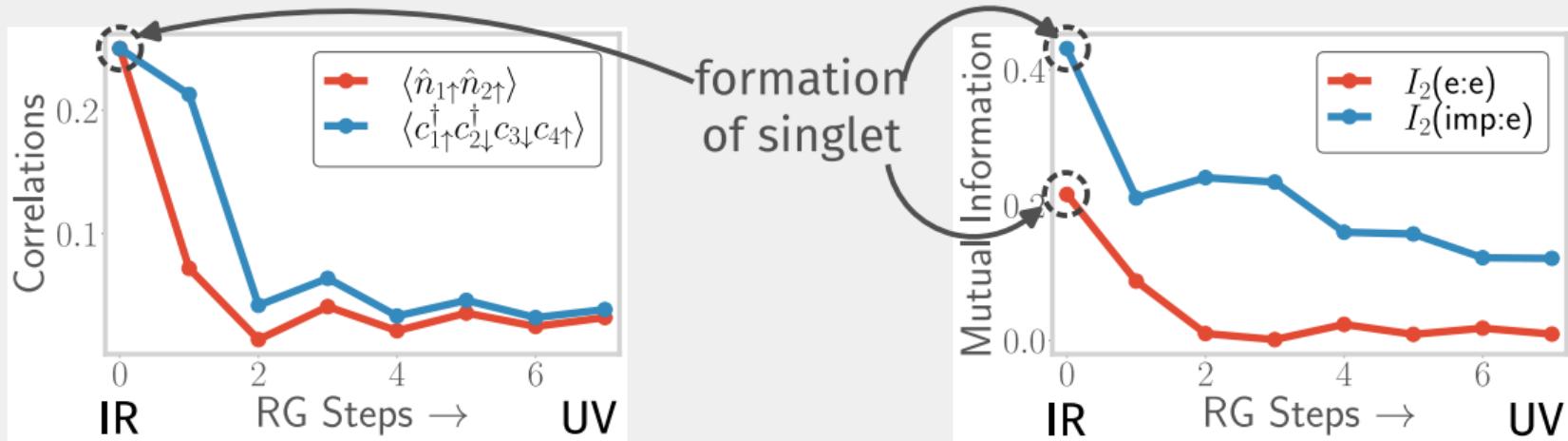
To obtain a theory for the Kondo cloud, we **trace out impurity** from fixed point Hamiltonian.



- all-to-all interactions between momentum states, **large entanglement**
- 2-particle interaction terms **not** present in Fermi liquid, are **responsible for screening**

# QUANTIFYING ENTANGLEMENT WITHIN THE KONDO CLOUD

In order to demonstrate formation of Kondo cloud, we study the **variation of entanglement** and correlations under RG transformations.



- Both entanglement and  $k$ -space correlations **increase** as RG proceeds from UV to IR.
- This shows the formation of the **Kondo singlet** and the growth of two-particle correlations in the **Kondo cloud**.

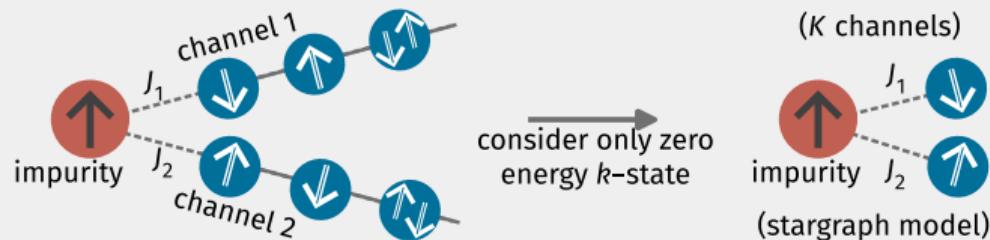
# DISTORTING THE KONDO SINGLET: THE MULTI-CHANNEL KONDO PROBLEM

Siddhartha Patra et al., 2023 J. Phys.: Condens. Matter 35 315601

# WHAT IS THE MULTICHANNEL KONDO PROBLEM?

Single impurity interacting with **multiple channels** in the bath

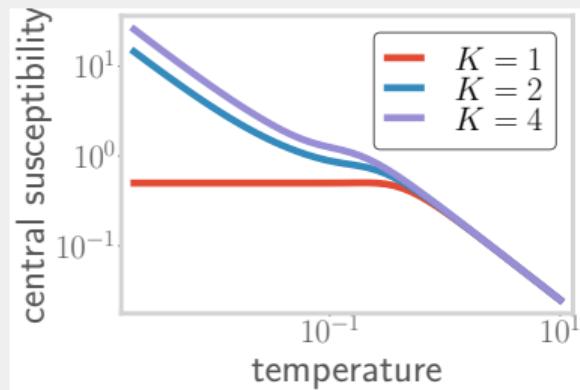
$$H_{\text{Kondo}} = KE_{\text{bath}} + \sum_l J_l \vec{S}_{\text{imp}} \cdot \vec{S}^{(l)}$$



Known to display divergent  $T = 0$  impurity susceptibility (incomplete screening), and orthogonality catastrophe, **non-Fermi liquid** excitations.

Zero bandwidth limit is (analytically) solvable:  $\{ |S_{\text{tot}}^z \rangle \}$

- Ground state degeneracy for  $K > 1$  explains **orthogonality catastrophe**
- $S_{\text{tot}}^z \neq 0$  in ground states shows incomplete screening
- Excitations shows **non-Fermi liquid** physics in the form of inter-channel scattering.



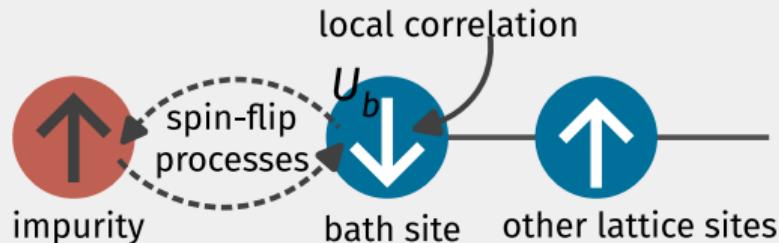
# **HOW TO DESTROY THE KONDO CLOUD: EFFECT OF LOCAL INTERACTIONS IN THE BATH**

**Abhirup Mukherjee et al 2023., New J. Phys. 25 113011**

# WHAT IS THE NEW PHYSICS INGREDIENT?

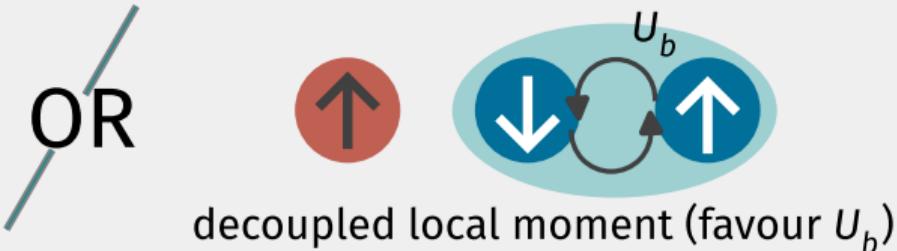
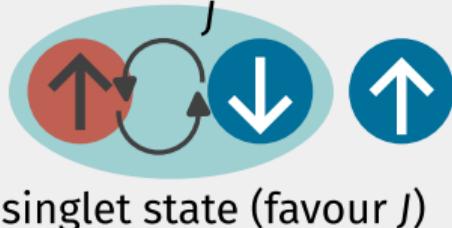
Add **local correlation** on bath (zeroth) site coupled to impurity

$$KE_{\text{bath}} + J \vec{S}_{\text{imp}} \cdot \vec{S}_{\text{bath}} - U_b (\vec{S}_{\text{bath}})^2$$



URG equations show that an **attractive**  $U_b$  frustrates the zeroth site.

$$\Delta J \sim J^2 + 4U_b J \implies \text{phase transition at } J = -4U_b$$



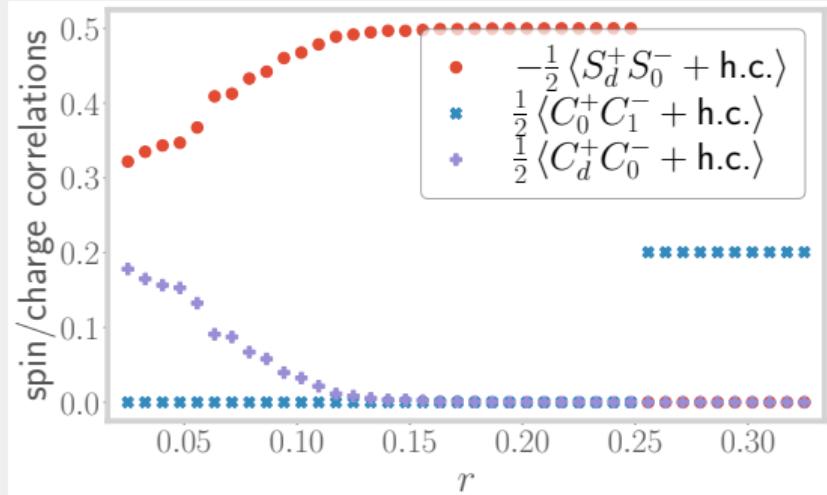
Such a model sheds light on the Mott MIT in  $\infty$ -dimensions (as seen from DMFT).

# NATURE OF THE TRANSITION

Across the transition,

- impurity correlations vanish
- bath correlations become non-zero

Shows that **pairing correlations** in the bath are responsible for the transition.



The state **precisely at the transition** is special:

- non-Fermi liquid excitations
- **fractional** impurity magnetisation and occupancy

# **HOLOGRAPHY OF ENTANGLEMENT IN 2D FREE FERMIONS**

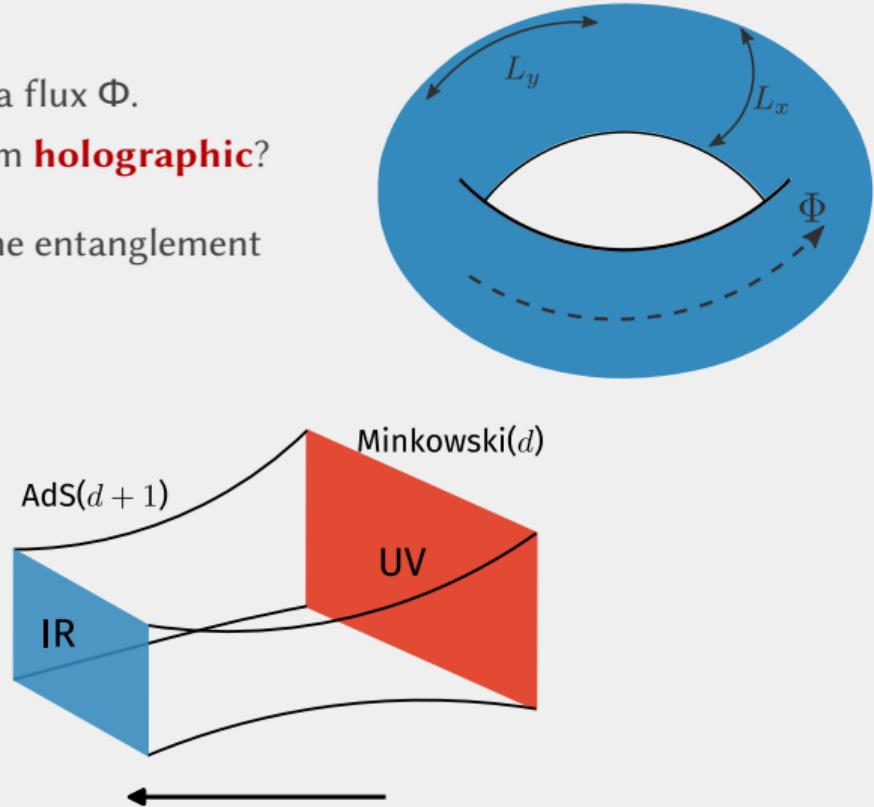
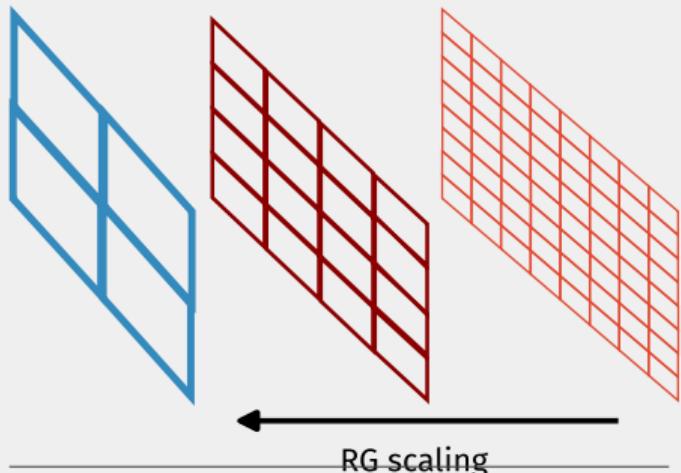
**ABHIRUP MUKHERJEE, SIDDHARTHA PATRA, SIDDHARTHA LAL**

**J. PHYS. A: MATH. THEOR. 57 275401 (2023)**

## SOME BROAD QUESTIONS

We consider 2D electrons placed on a torus in a flux  $\Phi$ .

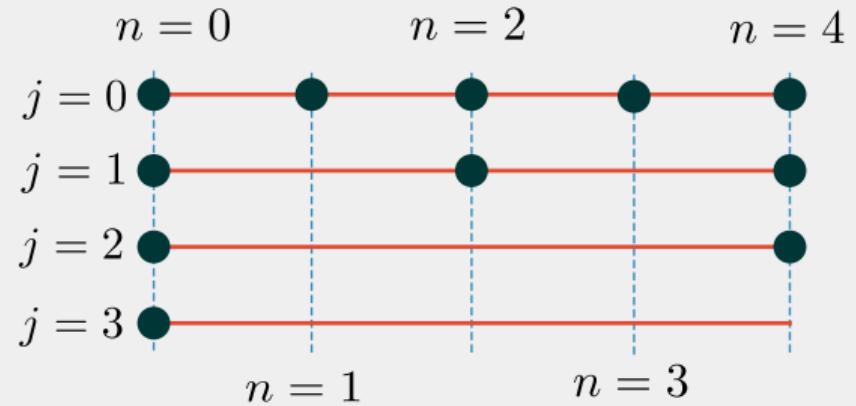
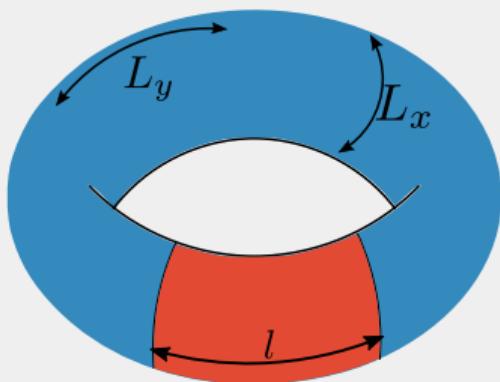
- Is the entanglement content of this system **holographic**?
- Is there any **topological** notion within the entanglement measures?



# RESULTS

- Choose subsystem in real space (red region)
- Apply **coarse-graining transformations** in  $k$ -space

Evolution of subspace entanglement shows interesting properties.

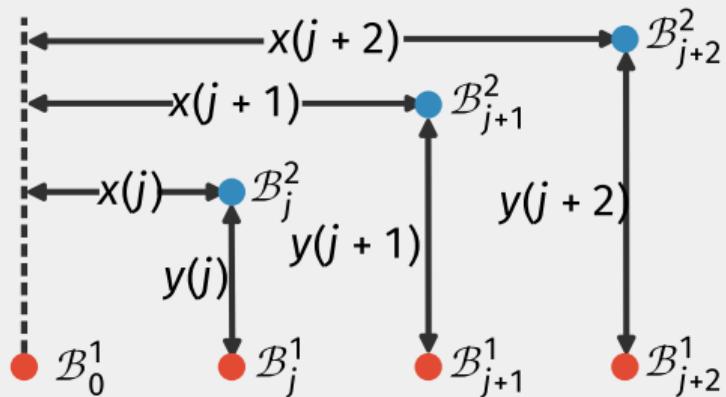


# RESULTS

Use mutual information  $I_2$  to define **distance**.

- Larger  $I_2 \implies$  smaller distance
- Allows notion of **curvature** as well.

Coarse-graining transformations lead to **emergent** spatial dimension

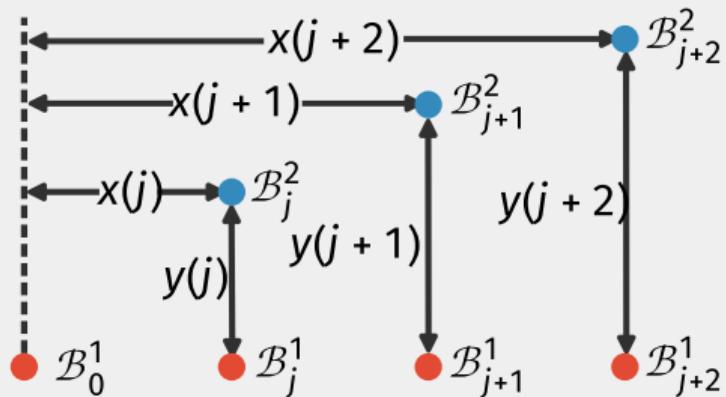


# RESULTS

Use mutual information  $I_2$  to define **distance**.

- Larger  $I_2 \implies$  smaller distance
- Allows notion of **curvature** as well.

Coarse-graining transformations lead to **emergent** spatial dimension

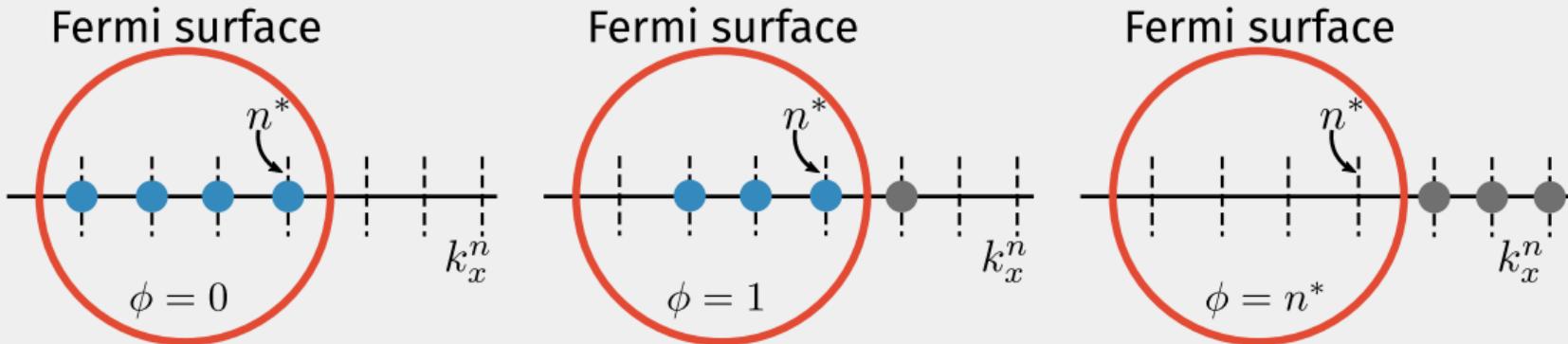
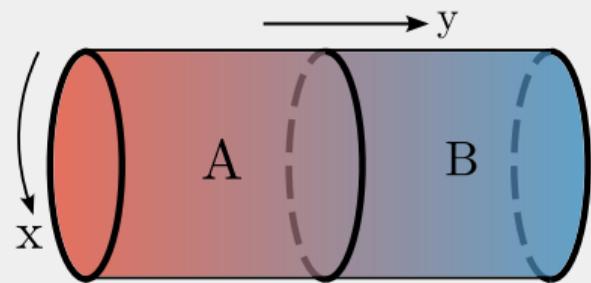


Other consequences:

- **hierarchy** of entanglement exists along the RG
- hierarchy also present in **multipartite entanglement**

# RESULTS

- By tuning flux, we relate Luttinger's volume to functions of entanglement
- Entanglement spectral flow is also related to Chern numbers in presence of magnetic field

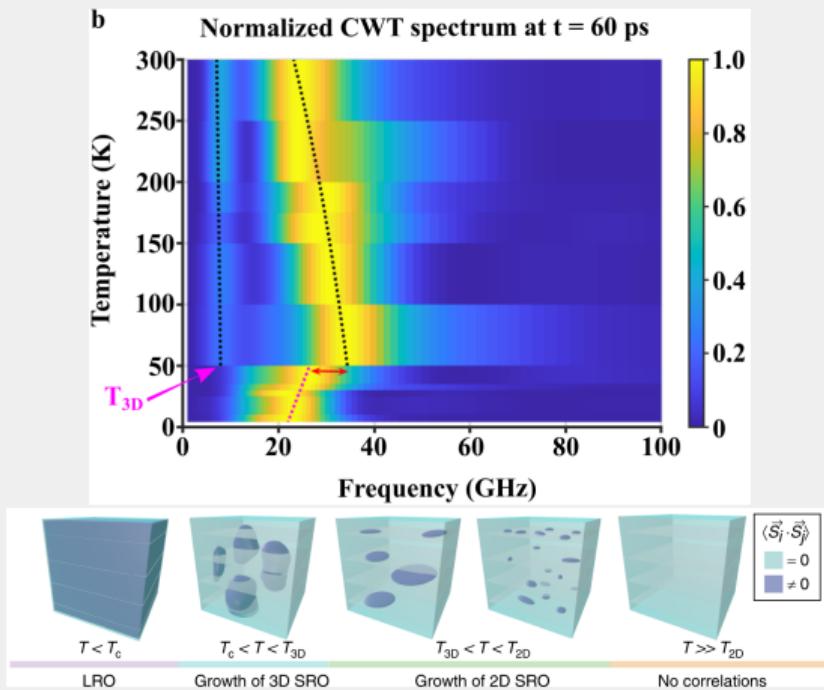


# **REVEALING THE MAGNETIC DIMENSIONAL CROSSOVER IN THE HEISENBERG FERROMAGNET CrSiTe<sub>3</sub> THROUGH PI- COSECOND STRAIN PULSES**

**A KUMAR N M, S MUKHERJEE, Abhirup Mukherjee, A PUNJAL, S PURWAR, T  
SETTI, S PRABHU S., S LAL, N KAMARAJU  
PHYS. REV. B 111, L140414 (2025)**

# GL THEORY FOR SPIN-PHONON INTERACTION

Prof. Kamaraju's group investigated the **magnetic dimensional crossover** (paramagnet → 2D short-range order → 3D long-range order) in  $\text{CrSiTe}_3$  using **pump-probe** spectroscopy.



- The acoustic strain pulses show a red-shift (**softening**) of the high-frequency phonons and a **gapping** out of the low-frequency phonon modes.

- We came up with a **Ginzburg-Landau model** of phonons interacting with lattice spin fluctuations to explain these features.

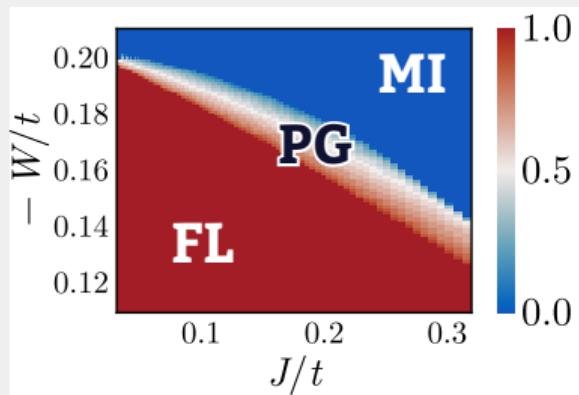
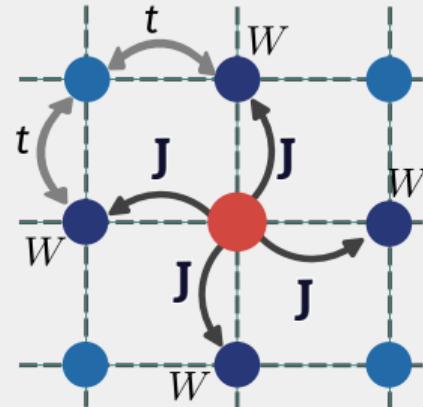
$$L = \sum_j [\dot{Q}_j^2 - (\lambda - M^2\chi)(Q_{j+1} - Q_j)^2 - M^2\xi Q_j^2]$$

- Renormalisation of **phonon dispersion and spring constant** due to interactions explains the softening and gapping.

# UNITARY RG PHASE DIAGRAM AND PSEUDOGAPPING TRANSITION

**Competition** between Kondo coupling and local interaction on bath sites:

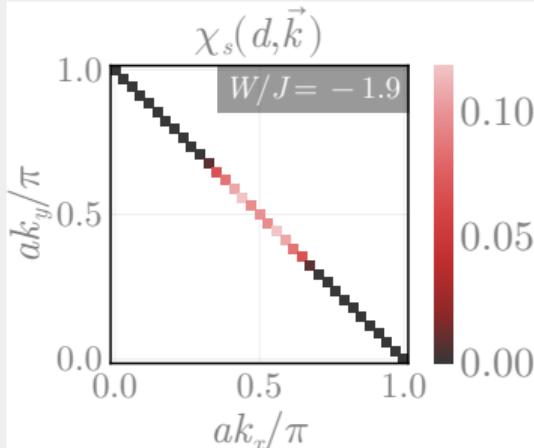
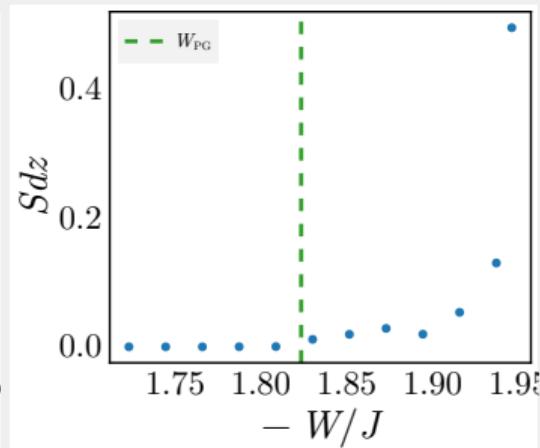
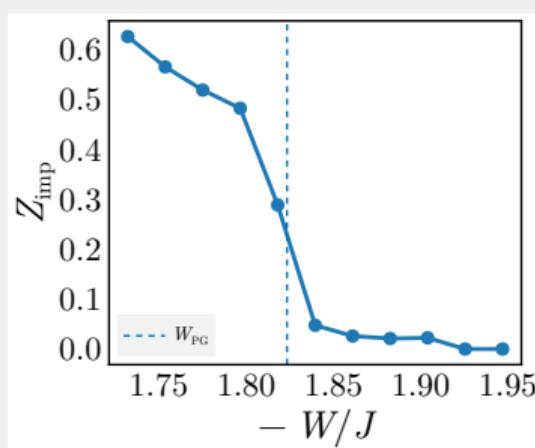
$$\Delta J_{k_1, k_2}^{(j)} = -2 \sum_{\mathbf{q}} \frac{J_{k_2, \mathbf{q}}^{(j)} J_{\mathbf{q}, k_1}^{(j)} + 4 J_{\mathbf{q}, \bar{\mathbf{q}}}^{(j)} W_{\mathbf{q}, k_2, k_1, \mathbf{q}}}{\omega - \frac{1}{2} |\varepsilon_j| + J_{\mathbf{q}}^{(j)}/4 + W_{\mathbf{q}}/2}$$



- Competition leads to **Kondo breakdown** for  $W < 0$
- Phase diagram shows **pseudogap** phase lying between Fermi liquid (FL) and Mott insulator (MI).
- PG possesses non-Fermi liquid excitations – a **Mott Metal**

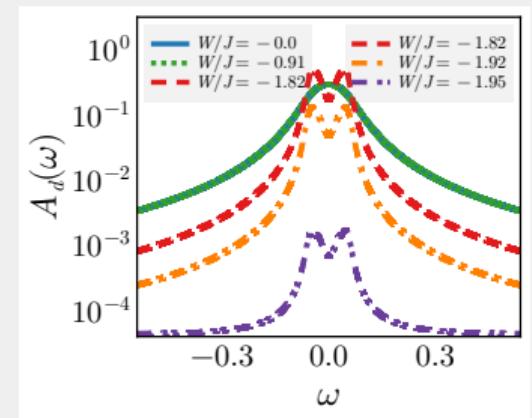
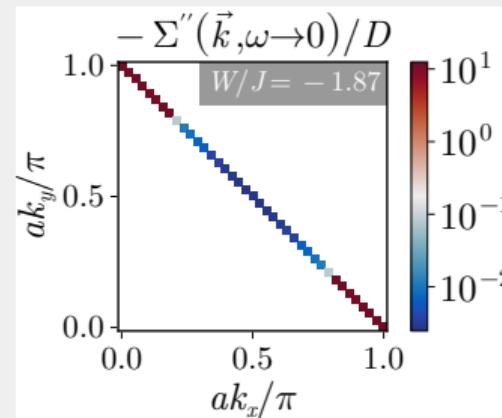
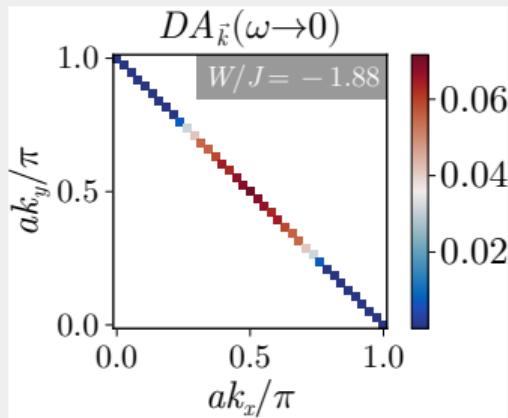
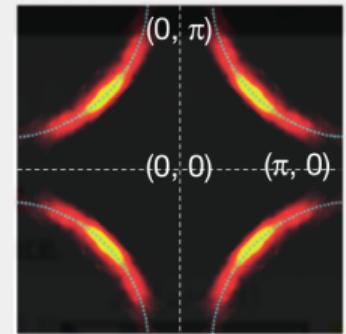
# JOURNEY INTO THE PSEUDOGAP

- Strength of Landau quasiparticle excitations of FL (**QP residue**  $Z$ ) vanishes upon entering PG.
- Impurity magnetisation  $\langle S_d^z \rangle$  grows dramatically in PG: **breakdown** of Kondo screening.
- Impurity-bath spin correlations vanish around antinode: signature of **pseudogap**



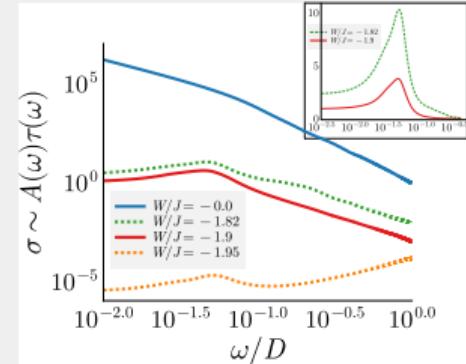
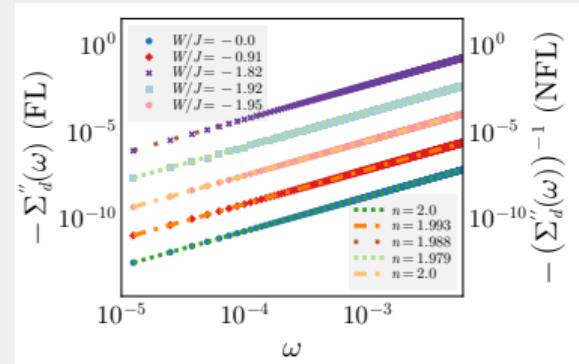
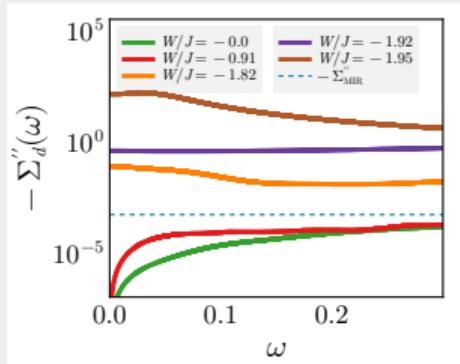
# LUTTINGER SURFACES IN THE PSEUDOGAP

- PG shows **electronic differentiation** in lattice spectral function: gapped antinodal regions (**Luttinger surfaces**), gapless excitations in nodal regions.
- Electron **scattering rate** shows divergences in gapped antinodal regions, while it is analytic in gapless nodal regions.
- Impurity spectral function shows **pseudogap** of Fermi arcs!



# UNIVERSAL SCALING OF SPECTRAL FEATURES

- Electron Scattering Rate of NFLs cross **Mott-Ioffe-Regel (MIR) bound** (no electron-like excitations!), while FLs are within it.
- $1/\Sigma'' \sim 1/\Sigma_0'' + \omega^2$ . Appearance of power-law exponents such as 2 signals **universality**.
- Optical Conductivity  $\sigma \sim A(\omega)\tau(\omega)$  shows a **shifted “Drude” peak**



## TILING DETAILS: GREENS FUNCTION

$$G(k, \sigma; \omega) = \sum_{v,n} C_{v,k} \left[ \frac{\langle \psi_{\text{gs}}(r_d) | T_{v,\sigma} | \psi_n(r_d) \rangle \langle \psi_n(r_d) | T_{v,\sigma}^\dagger | \psi_{\text{gs}}(r_d) \rangle}{\omega - (E_n - E_0 + \epsilon_v)} \right. \\ \left. + \frac{\langle \psi_{\text{gs}}(r_d) | T_{v,\sigma}^\dagger | \psi_n(r_d) \rangle \langle \psi_n(r_d) | T_{v,\sigma} | \psi_{\text{gs}}(r_d) \rangle}{\omega + (E_n - E_0 - \epsilon_v)} \right]. \quad (1)$$

$$G_{\text{loc}}(\omega) = G(d\sigma; \omega - \epsilon_{\text{loc}}) + \sum_v C_v \left[ G(T_{v,\sigma}, d\sigma; \omega - \epsilon_v) + \sum_k G(T_{v,\sigma}, T_{k,\sigma}; \omega - \epsilon_v) \right] \\ + \sum_k G(d\sigma, T_{k,\sigma}; \omega), \quad (2)$$

## TILING DETAILS: STATIC CORRELATION

$$\begin{aligned}
& \sum_{r_d} \langle \Psi_{\text{gs}} | c_{r_d+r,\sigma}(t) c_{r_d,\sigma}^\dagger | \Psi_{\text{gs}} \rangle \\
&= \sum_n F_n^*(r) \langle \psi_{\text{gs}}(r_d) | c_{r_d,\sigma}(t) | \psi_n(r_d) \rangle \langle \psi_n(r_d) | c_{r_d,\sigma}^\dagger | \psi_{\text{gs}}(r_d) \rangle \\
&+ \sum_{k,n} F'_n(k) F_n^*(r) \langle \psi_{\text{gs}}(r_d) | T_{k,\sigma}(t) | \psi_n(r_d) \rangle \langle \psi_n(r_d) | c_{r_d,\sigma}^\dagger | \psi_{\text{gs}}(r_d) \rangle \\
&+ \sum_{k,n} F'_n(k)^* F_n^*(r) \langle \psi_{\text{gs}}(r_d) | c_{r_d,\sigma}(t) | \psi_n(r_d) \rangle \langle \psi_n(r_d) | T_{k,\sigma}^\dagger | \psi_{\text{gs}}(r_d) \rangle \\
&+ \sum_{k_1,k_2} F'_n(k_1) F'_n(k_2)^* F_n^*(r) \langle \psi_{\text{gs}}(r_d) | T_{k_1,\sigma}(t) | \psi_n(r_d) \rangle \langle \psi_n(r_d) | T_{k_2,\sigma}^\dagger | \psi_{\text{gs}}(r_d) \rangle , \tag{3}
\end{aligned}$$