Connecting real materials to low-energy effective Hamiltonian: applications of symmetry-respecting Wannier functions

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Outlines

Simple illustrations:
- Orbital order in LaMnO$_3$
- Gapless CDW in TaSe$_2$
- Ferro-orbital order in Fe-SCs

Interacting problems:
- Local excitations
- Propagation and decay of local excitations

Effects of disordered impurities in Fe-SCs:
- Substitution of Fe: doping or not?
- Fe vacancy: “violation” of Luttinger theorem
- Ru substitution: realization of superdiffusion
RG perspective: bifurcation of many-body systems

Energy

$H_{\text{eff}}$ $H_{\text{eff}}$ $H_{\text{eff}}$ $H_{\text{eff}}$

$H_{\text{bare}}$

Constraint: no double occupation

Other Constraint?

- Lagrangian: integrate out high-energy sectors

vs.

- Hamiltonian: block diagonalize low-energy sectors
Direct mapping to effective Hamiltonian

DFT : LDA+$U$

Wannier Function

calculate $t,U,V,J$

$ab\ initio$ full $H$

$H^{LDA+U}$

tight-binding

effective $H_{\text{eff}}$

canonical transformation

few-band $H_{\text{eff}}$

Team 2

Applications:

- CMR manganites
- HTSC cuprates


LDA+$U+WF=H_{\text{HF}}$

Orbital Ordering in LaMnO$_3$

**e-e vs. e-l interactions & effective many-body $H^{\text{eff}}$**

- Large O component $\rightarrow$ nature of charge transfer gap
- Systematic study of e-e vs. e-l interactions in ordering orbitals

**Equation:**

$$H = \sum_{\langle ij \rangle \gamma \gamma'} t_{ij}^{\gamma \gamma'} d_{ij}^{\gamma'} d_{ij}^{\gamma} - g \sum_i \vec{T}_i \cdot \vec{Q}_i + \frac{K}{2} \sum_i \vec{Q}_i \cdot \vec{Q}_i + U_{\text{eff}} \sum_i n_{i\uparrow} n_{i\downarrow} - E_z \sum_i T_i^z.$$

**pseudo-spin:**

$$T_i^z = \frac{1}{2}(d_{i\uparrow}^\dagger d_{i\uparrow} - d_{i\downarrow}^\dagger d_{i\downarrow}), \quad T_i^x = \frac{1}{2}(d_{i\uparrow}^\dagger d_{i\downarrow} + d_{i\downarrow}^\dagger d_{i\uparrow}).$$

Gapless charge density wave in TaSe$_2$

R. L. Barnett, A. P., E. Demler, W.-G. Yin, and Wei Ku

Gapless Charge-Density Wave in TaSe$_2$

- commensurate CDW
- gapless excitations throughout the whole Fermi surface in CDW phase
- Fermi surface Nesting vector too large for $q^{\text{CDW}}$?

Conventional gapped CDW picture

- Fermi surface instability
  → divergent \( \chi(q^{\text{CDW}}, \omega \to 0) \)
  → nesting preferred
- gap → energy gain
Local Picture: Low-Energy Wannier Function

- $3z^2 - r^2 \ (a_g)$ symmetry near EF, noticeable hybridization with $e_g'$
- WS in one site contains complete information of the full k-space
- center: $a_g$ symmetry; tail: $e_g'$ symmetry
Surprising Hopping Path

\[
t^{DFT}_{Rn,R'n'} = \langle Rn | h^{DFT} | R'n' \rangle
\]

- \( t_1 = 38 \text{(meV)} \)
- \( t_2 = 115 \text{(meV)} \)
- \( t_{\perp,1} = 29 \text{(meV)} \)
- \( t_{\perp,2} = 23 \text{(meV)} \)

\[
t_2 \gg t_1
\]

- perfect tight-binding “fit” to the band structure
- surprising hopping strength to 2nd nearest neighbors
Why $t_2 >> t_1$?
Why $t_2 \gg t_1$?

- phase interference from $e_g$ hybridization tail $\rightarrow t_2 \gg t_1$
Gapless Charge-Density Wave in TaSe$_2$

- $x^2-y^2$ hybridization $\rightarrow$ $t_2 \gg t_1$ $\rightarrow$ decoupling of 3 sublattices
- minimization of tight-binding $H$ against distortion
  $\rightarrow$ one sublattice undistorted
  $\rightarrow$ gapless band structure

Ferro-orbital order & anisotropic magnetic structure in 1111 (&122)

Chi-Cheng Lee, Wei-Guo Yin & Wei Ku

Energy resolved, symmetry respecting Wannier function

\[ |\bar{R}n\rangle = \sum_{km} |\bar{k}m\rangle \langle \bar{k}m|\bar{R}n\rangle \]
\[ = \frac{1}{\sqrt{N_{cell}}} \sum_{km} |\bar{k}m\rangle e^{-ik \cdot \bar{R}} U^{(k)}_{mn} \]
\[ = \frac{1}{\sqrt{N_{cell}}} \left( \sum_{m} U^{(k)}_{mn} |\bar{k}m\rangle \right) e^{-ik \cdot \bar{R}} \]

 NM onsite energy (eV)
\[ z^2 \quad -0.03 \]
\[ x^2-y^2 \quad -0.20 \]
\[ yz \quad 0.10 \]
\[ xz \quad 0.10 \]
\[ xy \quad 0.34 \]

- small crystal field splitting
- degenerate \( xz \) and \( yz \)
→ orbital freedom!

methods see: W. Ku et al., PRL 89, 167204 (2002); W. Yin et al., PRL 96, 116405 (2006)
Comparing LDA band structures

- in NM 1\textsuperscript{st}-BZ
- \(d_{xz}\) & \(d_{yz}\) most relevant to the low-\(E\)
- Only \(d_{yz}\) splits strongly near \(E_F\)
- \(d_{yz}\) more spin polarized \(\sim 0.34 \mu_B\) than \(d_{xz}\) \(\sim 0.15 \mu_B\)
- more different with \(U=2\text{eV}\)
  - 0.58 vs. 0.23 \(\mu_B\)
  - \(\rightarrow\) orbital symmetry broken
- \(\Delta \sim W\)
- \(\rightarrow\) large \((\omega,k)\)-space involved
- \(\rightarrow\) local picture more suitable
- \(\rightarrow\) Fermi surface nesting not essential
- \(\rightarrow\) SDW less convenient

unfolding methods see:
Wei Ku \textit{et al.}, PRL \textbf{104}, 216401 (2010)
### Anti-intuitive hopping parameters

| $<\text{WFs}|H|\text{WFs}>$ | Fe1 $z^2$ | $x^2-y^2$ | $yz$ | $xz$ | $xy$ |
|--------------------------|---------|---------|-----|-----|-----|
| Fe2 (Fe4) $z^2$         | 0.13    | 0.31 (-0.31) | -0.10 (0.00) | 0.00 (0.10) | 0.00 |
| $x^2-y^2$                | 0.31 (-0.31) | -0.32 | 0.42 (0.00) | 0.00 (0.42) | 0.00 |
| $yz$                     | -0.10 (0.00) | 0.42 (0.00) | -0.40 (-0.13) | 0.00 | 0.00 (0.23) |
| $xz$                     | 0.00 (0.10) | 0.00 (0.42) | 0.00 | -0.13 (-0.40) | -0.23 (0.00) |
| $xy$                     | 0.00 | 0.00 | 0.00 (0.23) | -0.23 (0.00) | -0.30 |
| Fe3 $z^2$                | 0.06    | 0.00    | -0.08 | 0.08 | 0.26 |
| $x^2-y^2$                | 0.00    | -0.10   | 0.12 | 0.12 | 0.00 |
| $yz$                     | 0.08    | -0.12   | 0.25 | -0.07 | -0.05 |
| $xz$                     | -0.08   | -0.12   | -0.07 | 0.25 | 0.05 |
| $xy$                     | 0.26    | 0.00    | 0.05 | -0.05 | 0.16 |

- Unusual coupling direction
- Cubic symmetry broken seriously by As
- Perpendicular hopping direction!

Chi-Cheng Lee et al., PRL 103, 267001 (2009)
C-AF magnetic structure and ferro-orbital order

- Strongly anisotropic super-exchange: $J_{1x} > J_2 >> J_{1y}$
  - no competition with G-AF at all! $J_1 \sim 2J_2$ irrelevant!
  - single orbital Heisenberg model inadequate
- Orbital polarization and ferro-orbital correlation important
  - rotational symmetry breaking
- Unusual coupling direction and strong anisotropic hoppings!
  - common staggered OO loses
  - $a > b$: AF across long bond (rare)

$\Delta E = -2t^2/U$

$\Delta E = -t^2/(U' - J_H)$

Chi-Cheng Lee et al., PRL 103, 267001 (2009)
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- Fe vacancy: “violation” of Luttinger theorem
- Ru substitution: realization of superdiffusion
Propagation of local excitations in strongly correlated materials

P. Abbamonte et. al., PNAS 105, 12159 (2008)
Acknowledgement

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Theoretical works
- Chi-Cheng Lee (BNL & Tamkang Univ.)
- Chen-Lin Yeh (BNL & Tamkang Univ.)
- Hung-Chung Hsueh (Tamkang Univ.)

Experiments
- Peter Abbamonte, Xiaoqian M. Chen, Yu Gan (UIUC)
- Ben C. Larson (ORNL)
- John Hill (BNL), Young Cai (BNL)
- Andrivo Rusydi (NUS, Singapore)
- Young-June Kim (U. Toronto, Canada)
- Nozomu Hiraoka (Sping-8, Japan)
Local Picture for Strongly Correlated Systems

\[ H = H_0 + V = H_{\text{local}} + H_{\text{nonlocal}} \]

- \( V \) too big for perturbation
- Maximize the terms in the “local” part
  \( \rightarrow \) symmetric Wannier Representation \( \rightarrow \) defines “local”
- Treat local part “accurately”
- Add non-local part as modification
How to define “local” in CT-insulators?

- Periodic symmetry
- Point group symmetry
- Simultaneously keep both? How to split the Hilbert space?
Symmetric Wannier Functions for CT-Insulators


- O-p orbitals $\rightarrow$ additional Ni-d orbitals (no double counting of O orbitals)
- “local” is now defined by this “super-atom”
Super Atom for Charge Transfer Insulator

\[ H = H_{\text{local}} + H_{\text{nonlocal}} \]

Maximize the contributions of “local atom”
Density response for super atom

(antibonding-type)

NiO

\[ q = 3.5 \text{ Å}^{-1} \]

[111]

\[ q = 7 \text{ Å}^{-1} \]
\[ q = 4 \]
\[ q = 3 \]
\[ q = 2 \]

\[ \alpha = 36^\circ \]
\[ \alpha = 46^\circ \]
\[ \alpha = 90^\circ \]

Uncert

\[ s(q, \omega) (\text{eV}^{-1} \text{nm}^3) \]

\[ -\ln X (1/\text{eV} \text{nm}^3) \]

\[ q = [111] 7.11 \text{ Å}^{-1} \]
\[ q = [111] 3.55 \text{ Å}^{-1} \]
\[ q = [113] 10.20 \text{ Å}^{-1} \]
\[ q = [113] 6.80 \text{ Å}^{-1} \]

\[ \Delta E (\text{eV}) \]

\[ w (\text{eV}) \]
Multiplet Splitting Made Possible with MB Hilbert Space

GS

\( \begin{align*}
\text{Ni } eg & \quad \text{Ni } t_2g \\
\text{Ni } eg & \quad \text{Ni } t_2g \\
\text{O } eg & \quad \text{O } s \\
\text{O } s & \quad \text{O } s
\end{align*} \)

\( \begin{align*}
(0.9484) & \quad (-0.2226) & \quad (0.2226)
\end{align*} \)

\( T_{1g} \)
1eV excitation found!
very different q-dependence of spectral weight

Nozomu Hiraoka et al, Europhys. Lett. 96, 37007 (2011)
3D Map of Local Excitations in the Mott Gap in NiO

- Detailed comparison: exp vs. TD-LDA+U & cluster model
  - good general agreement with some discrepancies in details
How does the local excitations propagate and decay?
Propagation of Tightly-Bound Excitons: case study of LiF

- Tightly bound exciton
- Charge transfer insulator
  → p-h in different atoms
- Frenkel or Wannier exciton?
- Dispersion
  → propagation in space/time

Inelastic X-ray scattering
- Structured spectral weight
- Clear dispersion at large $q$!
- observe $fs$ dynamics

P. Abbamonte et. al., PNAS 2008
Excitons in LiF as a Frenkel Exciton in a “Super Atom”
Matrix Element and Structure in $q$-space

$q = 0 \sim 1.5$ Intensity divided by 2.6

P. Abbamonte et al., PNAS 2008
Propagation of tightly bound excitons

- Treat tightly bound excitons (and other local excitations) as a composite boson. Define its propagation kinetic kernel $T$ via local and full propagator $D[H_L]$ and $D[H]$: 

$$D[H] = D[H_L] + D[H_L]T D[H]$$

- $T$ integrates out all the pair fluctuation in space and encapsulates propagation and decay processes.

- We then approximate $T$ using unbound exciton propagator 

$$T = D^{-1}[H_L] - D^{-1}[H] \sim D_0^{-1}[G[H_L]] - D_0^{-1}[G[H]]$$

- Separation of local many-body problem from non-local propagation

- Many orders of magnitudes cheaper than Bethe-Salpeter equation

Effective Two-Particle Hopping

Define effective two particle kinetic kernel $T$ via

$$D[H] = D[H_L] + D[H_L]TD[H]$$

in the basis of local bound pair $b^+_{RN} \equiv c^+_R c^-_h$ and simplify

$$T = D^{-1}[H_L] - D^{-1}[H]$$
$$= \left( D_0^{-1}[G[H_L]] - I[H_L] \right) - \left( D_0^{-1}[G[H]] - I[H] \right)$$
$$\simeq D_0^{-1}[G[H_L]] - D_0^{-1}[G[H]]$$

using the empty bubble

$$D_0(RN, R'N'; t, t') = G(Rp, R'p'; t, t')G(R'h', Rh; t', t)$$

$T$ gives hopping of p-h pair in real space $\rightarrow$ dispersion in $q$-space

Effective Two-Particle Hopping in LiF

$T(\omega)$ is complex and strongly $\omega$-dependent to fully account for
1. Landau continuum (integrating out virtual pair breaking processes) $\rightarrow$ exact for $E_b = 0$
2. Lower mobility with stronger p-h binding $\rightarrow$ correct $t^2/E_b$ behavior
3. Renormalization of on-site energy from kinetic energy
Effective Two-Particle Hopping

Define effective two particle kinetic kernel $T$ via

$$D[H] = D[H_L] + D[H_L]TD[H]$$

in the basis of local bound pair $b_{RN}^+ \equiv c_{Rp}^+ c_{Rh}$ and simplify

$$T = D^{-1}[H_L] - D^{-1}[H_L]$$

$$= (D_0^{-1}[G[H_L]] - I[H_L]) - (D_0^{-1}[G[H]] - I[H])$$

$$\approx D_0^{-1}[G[H_L]] - D_0^{-1}[G[H]]$$

using the empty bubble

$$D_0(RN, R'N'; t, t') = G(Rp, R'p'; t, t')G(R'h', Rh; t', t)$$

$T$ gives hopping of p-h pair in real space $\Rightarrow$ dispersion in $q$-space

1. Full results similar to the diagonalization of $\omega_{\text{exciton}} + \text{Re} T(\tilde{\omega}_{\text{exciton}})$

2. Similar dispersion to the F $p$-bands (same symmetry)

Observation of Multiple Exciton Bands

1. Similar weight in momentum space
2. Similar dispersion
3. Switching of bands (breaks in intensity)
exp. 2D map of exciton propagation is in preparation
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  - Fe vacancy: “violation” of Luttinger theorem
  - Ru substitution: realization of superdiffusion
Treating materials with disordered impurities

T. Berlijn, D. Volja, and Wei Ku, PRL 106, 077005 (2011)

For various applications, see


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Collaborators
P. Hirschfeld, & H.-P. Chen, U of Florida
First-principles methods for disorder

Relevance
- Defects, vacancy, chemical substitution, doping & intercalation
- Thermal/quantum fluctuation induced spatial inhomogeneity
- Paramagnetic phase with disordered local moment

Different approaches
- Mean-field potential: virtual crystal approximation
- Mean-field scattering: coherent potential approximation
- Perturbation: diagrammatic summation
- Non-perturbed direct method: configuration average over super cells
Mean-field approaches

**VCA: Virtual Crystal Approximation**

\[ V_{\text{virtual crystal}} = (1-x)V_0 + xV_1 \]

no scattering

**CPA: Coherent Potential Approximation**

\[ =(1-x)0 + x1 \]

lack cluster-scattering

A. Gonis, “Green functions for ordered and disordered systems” (1992)
Non-local physics: $k$-dependent broadening

$\omega$ vs $k$

same $\omega$ different broadening
Non-local physics: large-sized impurity states

Anderson localization, multiple scattering required
Non-local physics: short-range order
Configuration average over super cell

\[ \langle G \rangle \approx P_1 G_1 + \ldots + P_N G_N \]

Challenge: solving many Hamiltonians with big super cells
Configuration average over super cell

\[ \langle G \rangle \approx P_1 G_1 + \ldots + P_N G_N \]

Challenge: solving many Hamiltonians with big super cells
Constructing small and accurate effective $H$

Consider $H$ from influence of impurities at $x_1, \ldots, x_N$

$$H(x_1, \ldots, x_N) = H^0 + \sum_{i=1}^{N} \Delta(x_i) + \sum_{i > j = 1}^{N} \Delta(x_i, x_j) + \ldots$$

1. Calculate $H^0, H(x_i)$ & $H(x_i, x_j)$ in same Wannier basis from DFT
2. Calculate $\Delta$ by taking difference of $H^\text{DFT}$ in Wannier basis
   
   $$\Delta(x_i) = H(x_i) - H^0; \quad \Delta(x_i, x_j) = H(x_i, x_j) - H^0 - \Delta(x_i) - \Delta(x_i)$$
3. Assemble $H_c$ for each disorder configuration
   
   $$\langle r' n' | H((r_1, m_1), \ldots, (r_N, m_N)) | r'' n'' \rangle =$$
   
   $$\langle r' - r'', n' | H^0 | 0 n'' \rangle + \sum_{i=1}^{N} \langle r' - r_i, n' | \Delta^{(m_i)} | r'' - r_i, n'' \rangle + \ldots$$
4. Solve $H$ and unfold $A(k, \omega)$ to the normal cell unit
5. Average over configurations with different size & shape

$$\langle A(k, w) \rangle = \sum_c P_c A_c(k, w)$$

T. Berlijn, D. Volja, and Wei Ku, PRL 106, 077005 (2011)
Including Effects in Density Functional Theory

two DFT Calculations
undoped normal cell

1 impurity in large super cell
Reducing Hilbert Space via Wannier Transformation

DFT CoO$_2$

Wannier functions

Energy (eV)

Reducing Hilbert Space via Wannier Transformation
Obtain Effects of Impurities on $H^{\text{DFT}}$

2 Wannier transformations

$$|R_n> = \sum_{k,j} e^{-ik \cdot R} U_{nj}(k)|k_j>$$

Reduced Hilbert space

2 Tight Binding Hamiltonians

$H_{\text{DFT}}^0$ undoped

$H_{\text{DFT}}^1$ 1 impurity

$$\Delta^{(x_i)} = H^{(x_i)} - H^0$$
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   $$\langle r' - r'', n' \vert H^0 \vert 0 n'' \rangle + \sum_{i=1}^{N} \langle r' - r_i, n' \vert \Delta(m_i) \vert r'' - r_i, n'' \rangle + \ldots$$
4. Solve $H$ and unfold $A(k, \omega)$ to the normal cell unit
5. Average over configurations with different size & shape
   $$\langle A(k, w) \rangle = \sum_c P_c A_c(k, w)$$

T. Berlijn, D. Volja, and Wei Ku, PRL 106, 077005 (2011)
Substitution & vacancies: $\text{Zn}_{1-x}\text{Cu}_x\text{O}_{1-y}$

Q1) Where do the doped electrons reside?
Q2) What is the role of oxygen vacancy?

A1) Doped electrons reside in Cu orbitals
A2) \(V_O\) mediates Cu moments

Do transition metal substitution dope carriers in Fe-based superconductors

T. Berlijn & Wei Ku

PRL 108, 207003 (2012)
Do TM substitutions dope carriers in Fe-SC?

more valence electrons than Fe → expect electron doping
Additional Charge Residing in the Substituted Atoms in Fe-SC

Theory
DFT (density)
Co:122

Experiment
XANES
Co:122

H. Wadati *et al*, PRL **105** (2010)
E. M. Bittar *et al*, PRL **107** (2011)

- Substitution does not dope the system (?)
12.5% Co Substituted BaFe$_2$As$_2$

- **Change of density in substituted atoms** \(\rightarrow\) not doping?
- **Enlarged Fermi surfaces & shift of chemical potential** \(\rightarrow\) doping?

<table>
<thead>
<tr>
<th>$\alpha$</th>
<th>Substituted Atoms</th>
<th>Density</th>
</tr>
</thead>
<tbody>
<tr>
<td>122</td>
<td>Fe</td>
<td>7.22</td>
</tr>
<tr>
<td>Co:122</td>
<td>Fe</td>
<td>7.19 x 7/8</td>
</tr>
<tr>
<td>Co:122</td>
<td>Co</td>
<td>8.40 x 1/8</td>
</tr>
</tbody>
</table>
Example: Does Co/Zn substitution dope Fe-SC?

Most definitely yes, in the simplest manner, but some carriers lose coherence!

Fe vacancy in K2Fe4Se5

T. Berlijn, P. Hirschfeld, & Wei Ku

PRL 109, 147003 (2012)
A heavily electron doped system?
Appears to be heavily doped ~ 0.5 e / Fe with disordered Fe vacancy.

Tom Berlijn, Peter Hirschfeld, and Wei Ku, 109, 147003 (2012)
Origin of the effective “doping” with Fe vacancy

- Need both disorder and the impurity potential

Tom Berlijn, Peter Hirschfeld, and Wei Ku, 109, 147003 (2012)
Effects of disordered impurity potential

Tom Berlijn, Peter Hirschfeld, and Wei Ku, 109, 147003 (2012)
Ru substitution of BaFe2As2

Limin Wang, Tom Berlijn, Chia-Hui Lin, Yan Wang, Peter Hirschfeld, Wei Ku

PRL 110, 037001 (2013)
Ru/Co atoms replace the most essential Fe atom. Superconductivity can survive even with 40% Ru substitution.
Ru substituted samples exhibit a residual resistivity comparable to 8% Co substituted system at a much higher 35% substitution level
Experimental (ARPES) controversy

- Nearly substitution-independent FS

[R. S. Dhaka et al, PRL 107, 267002(2011)]
Experimental (ARPES) controversy

- A crossover from two-dimensional to three dimensional structure
  [N. Xu et al, PRB 86, 064505]
The bands are heavily smeared. (-0.8 eV)
x = 0.55 is the most disordered one.
Fe bands around the Fermi level remain very sharp.
Origin of the superdiffusion around the Fermi level

- diagonal disorder only: strong smearing of the entire $d$-band
- off-diagonal disorder only: scattering diminishes near the center of the $d$-band.
- combined $\rightarrow$ weak impurity scattering near $E_F$
Origin of the superdiffusion around the Fermi level

(a) $\langle \Delta(\Gamma,\omega) \rangle$ (eV$^{-1}$)

(b) $x=0.38$

(c) diagonal disorder

(d) off-diagonal disorder

(e) The insensitivity originates from a coherent interference between on-site and off-site impurity effects.

| $k$ | $j$ | $\langle kj|\Delta|kj'\rangle$ | $\langle kj|\Delta_d|kj'\rangle$ | $\langle kj|\Delta_{od}|kj'\rangle$ |
|-----|-----|------------------------------|------------------------------|------------------------------|
| $\Gamma$ | 9 0.3$\Gamma$M | 10 | -0.0578 | -0.3023 | 0.2445 |
| $\Gamma$ | 8 0.3$\Gamma$M | 7  | -0.2938 | -0.3316 | 0.0378 |
Superdiffusion in 1D model

Single-particle Hamiltonian

\[
H = H_{\text{lattice}} + \sum_{k} \varepsilon_{0} a_{k}^{\dagger} a_{k} + \sum_{k,\mu} 2V_{\mu} \cos(\phi_{\mu} - k_{\mu}) a_{k}^{\dagger} a_{k} \\
+ \sum_{k, q, \mu} 2i [\gamma_{\mu} \sin(k_{\mu} - \theta_{\mu}) - \gamma_{\mu} \sin(k_{\mu} - q_{\mu} - \theta_{\mu}) \\
- G_{\mu} \sin(q_{\mu}) ] \chi_{q}^{\mu} a_{k}^{\dagger} a_{q-k-q_{\mu}} ,
\]

1. correlated disorder $\gamma = G$

2. site-diagonal disorder.

3. site-off diagonal disorder.

Correlations between site and off-diagonal disorder give rise to superdiffusion transport.
Fermi surface (2Fe) for different Ru substitution

- The hole pockets become more three dimensional
- The electron pockets show a slight substitution dependence.
Resolution of the ARPES controversy

(a) $x=0.02$  (b) $x=0.21$  (c) $x=0.36$  (d) $x=0.55$

- This photon polarization couples mostly to the $yz$ orbital.
- The $yz$ character of our unfolded BS also show little change.

[R. S. Dhaka et al, PRL 107, 267002 ]
### Suppression of the long-range order

<table>
<thead>
<tr>
<th>( x )</th>
<th>0</th>
<th>0.21</th>
<th>0.38</th>
<th>0.55</th>
<th>0.75</th>
<th>1.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>( e/Fe = h/Fe )</td>
<td>0.162</td>
<td>0.151</td>
<td>0.142</td>
<td>0.127</td>
<td>0.123</td>
<td>0.120</td>
</tr>
<tr>
<td>DOS(( E_F ))</td>
<td>2.393</td>
<td>2.024</td>
<td>1.775</td>
<td>1.458</td>
<td>1.299</td>
<td>0.868</td>
</tr>
</tbody>
</table>

- Larger splitting between the conduction and valence bands \( \rightarrow \) carrier reduction
- The reduced DOS & enhanced 3D nature \( \rightarrow \) suppress the long-range orbital/magnetic order

[R. S. Dhaka et al, PRL 107, 267002]
Conclusions

- Symmetry-respecting Wannier function
  - Simple, flexible and efficient basis to describe physics
  - Intuitive microscopic picture + quantitative realism
- Unfolding 1\textsuperscript{st}-principles band structures
  - Visualizing each band’s coupling to the order parameters and other symmetry breakers
  - Useful for the study of extrinsic (doping, intercalation, vacancy, substitution, & defects) and intrinsic (CDW, SDW, OO, & structure distortion) effects on the electronic structure
  - Direct comparison with ARPES spectrum, built-in matrix elements effects
- A non-perturbative beyond-mean-field first-principles method for materials with disordered impurities
  - Defects, vacancy, chemical substitution, doping & intercalation
  - Thermal/quantum fluctuation induced spatial inhomogeneity
  - Paramagnetic phase with disordered local moments