Calculating model parameters: The case of Heisenberg $J_{ex}$
First-principles Approach to SCES

- Strongly correlated electron materials & many outstanding problems (e.g., high-temperature superconductivity)
- ‘Solving’ those problem often means more than just to describe the ‘correct’ electronic structure.
- Many different experimental and theoretical methods & techniques (traditionally this has not been a main field of research for first-principles calculations.)

Constructing the bridges
Having/providing the unique facets
First-principles Approach (1)

Calculating the correct/realistic electronic structure

- Conventional approximations (e.g., LDA and GGA) are known to fail for the case of correlated electron systems.
- LDA+U, +DMFT (dynamical mean-field theory), GW, etc.
First-principles Approach (2)

Direct estimation of key quantity or parameter

Strength of correlations in electron- and hole-doped cuprates

Cédric Weber *, Kristjan Haule and Gabriel Kotliar

Optical conductivity and the correlation strength of high-temperature copper-oxide superconductors

ARMIN COMANAC¹, LUCA DE’ MEDICI², MASSIMO CAPONE³,⁴ AND A. J. MILLIS¹∗
First-principles Approach (2) – cont’d

\[ \chi^t = \chi - \chi^r \]

From Dyson’s equation for \( W \) in RPA, \( W = v + v\chi_0 W \)

fully screened Coulomb, \( W \)

\[ W = [1 - v\chi_0]^{-1}v = [1 - v\chi_0^r - v\chi_0^t]^{-1}v 
= \left[\left(1 - (1 - v\chi_0^r)^{-1}v\chi_0^t\right)(1 - v\chi_0^r)\right]^{-1}v 
= [1 - W^r\chi_0^t]^{-1}W^r \]

partially screened Coulomb, \( W^r \)
First-principles Approach (2) – cont’d

Calculation by S. W. Jang
First-principles Approach (3)

Calculating new concept and/or quantity?!

- Effective charge
- Charge transfer energy
- Chern number

Boeri et al., PRL (2008)
Heisenberg Exchange Parameter

\[ H = -J_{12} S_1 \cdot S_2 \]

✓ The sign and the strength of two-spin interaction represented by a constant \( J_{12} \)

✓ (With the minus sign in front of it,) the positive and negative \( J_{12} \) corresponds to the parallel (ferro-) and antiparallel (antiferro) spin alignment, respectively.
Not always straightforward to determine it from experiments (sample quality, fitting, …etc)

Model-based theories often just assume Js.

Independent method or way to estimate J

Extract some more (hopefully unique) information

The range of experimental values

Calculation by H. K. Yoon and T. J. Kim
Calculating J from Total Energies

\[
H = -\frac{1}{2} \sum_{ij} J_{ij} \hat{S}_i \cdot \hat{S}_j \quad \rightarrow \quad E = -\frac{1}{2} J_{ij} S_i S_j
\]

Metastable spin structure (not the ground state) can usually be stabilized (from the self-consistent) by choosing the desired spin orientation as the input.

→ Total energies corresponding to different spin orders can be estimated and compared.
Calculating $J$ as a Response Function

\[ J^{\alpha \beta}(R_{ij}) = \sum_{\mathbf{q}} J^{\alpha,\beta}_{R_{ij}}(\mathbf{q}) e^{-i \mathbf{R}_{ij} \cdot \mathbf{q}} \]

\[ J^{\forall \text{orbitals}}_{R_{ij}}(\mathbf{q}) = \sum_{\alpha, \beta} J^{\alpha,\beta}_{R_{ij}}(\mathbf{q}) \]

\[ J^{\alpha,\beta}_{R_{ij}}(\mathbf{q}) = \sum_{n,n'} \sum_{k} \frac{f_{\uparrow n,k} - f_{\downarrow n',k+q}}{\epsilon_{\downarrow n,k} - \epsilon_{\uparrow n',k+q} - i \eta} M(n\alpha, n'\beta) \]

\[ M(n\alpha, n'\beta) = \langle \psi_{\uparrow \mathbf{k},\alpha} | V_{\mathbf{i}} | \psi_{\downarrow \mathbf{k}+\mathbf{q},\beta} \rangle \times \langle \psi_{\downarrow \mathbf{k}+\mathbf{q},\beta} | V_{\mathbf{j}} | \psi_{\uparrow \mathbf{k},\alpha} \rangle \]

Orbital index: $\alpha, \beta$
Two spin site: $R_{i} = R + r$ and $R_{j} = R' + r'$
$R_{ij} = R_{j} - R_{i}$

CF: Neutron scattering measurement

Liechtenstein et al. PRL (2009)
MJH et al. PRB (2004)
Advantage

✓ No need to perform the multiple large-supercell calculations
✓ Being able to describe the long range behavior
✓ More similar with neutron scattering

Example 1: Fe-pnictides Superconductors

Frustration and quantum criticality?

<table>
<thead>
<tr>
<th>System</th>
<th>Moment</th>
<th>$J_{1a}$</th>
<th>$J_2$</th>
<th>$J_{1b}$</th>
<th>$J_{1a}/2J_2$</th>
<th>$J_{1a} + 2J_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>LaFeAsO</td>
<td>1.69</td>
<td>47.4</td>
<td>22.4</td>
<td>−6.9</td>
<td>1.06</td>
<td>92.2</td>
</tr>
<tr>
<td>CeFeAsO</td>
<td>1.79</td>
<td>31.6</td>
<td>15.4</td>
<td>2.0</td>
<td>1.03</td>
<td>62.4</td>
</tr>
<tr>
<td>PrFeAsO</td>
<td>1.76</td>
<td>57.2</td>
<td>18.2</td>
<td>3.4</td>
<td>1.57</td>
<td>93.6</td>
</tr>
<tr>
<td>NdFeAsO</td>
<td>1.49</td>
<td>42.1</td>
<td>15.2</td>
<td>−1.7</td>
<td>1.38</td>
<td>72.5</td>
</tr>
<tr>
<td>CaFe$_2$As$_2$</td>
<td>1.51</td>
<td>36.6</td>
<td>19.4</td>
<td>−2.8</td>
<td>0.95</td>
<td>75.4</td>
</tr>
<tr>
<td>SrFe$_2$As$_2$</td>
<td>1.69</td>
<td>42.0</td>
<td>16.0</td>
<td>2.6</td>
<td>1.31</td>
<td>74.0</td>
</tr>
<tr>
<td>BaFe$_2$As$_2$</td>
<td>1.68</td>
<td>43.0</td>
<td>14.3</td>
<td>−3.1</td>
<td>1.51</td>
<td>71.5</td>
</tr>
<tr>
<td>KFe$_2$As$_2$</td>
<td>1.58</td>
<td>42.5</td>
<td>15.0</td>
<td>−2.9</td>
<td>1.42</td>
<td>72.5</td>
</tr>
<tr>
<td>LiFeAs</td>
<td>1.69</td>
<td>43.4</td>
<td>22.9</td>
<td>−2.5</td>
<td>0.95</td>
<td>89.2</td>
</tr>
</tbody>
</table>

MJH et al., PRL (2009)

Si and Abrahams, PRL (2008)
Fang et al., PRB (2008)
Xu et al., PRB (2008)
Example 2: Superconducting FeTe

Spin fluctuation theory: Yes or No?

Li et al., PRB (2009)

Picture from Mazin Nature (2010)

Subedi et al., PRB (2008)
Example 2: FeTe – cont’d

<table>
<thead>
<tr>
<th>System</th>
<th>Moment</th>
<th>$J_{1a}$</th>
<th>$J_{1b}$</th>
<th>$J_{2a}$</th>
<th>$J_{2b}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Double-stripe Fe$_{1.068}$Te</td>
<td>2.09 (1.97$^b$)</td>
<td>-7.6</td>
<td>-26.5</td>
<td>46.5</td>
<td>-34.9</td>
</tr>
<tr>
<td>FeTe</td>
<td>2.16</td>
<td>-4.2</td>
<td>12.9</td>
<td>-6.2</td>
<td>-15.3</td>
</tr>
<tr>
<td>Single-stripe FeTe</td>
<td>2.09</td>
<td>38.6</td>
<td>21.7</td>
<td>5.0</td>
<td>…</td>
</tr>
<tr>
<td>LaFeAsO$^a$</td>
<td>1.69 (0.36$^c$)</td>
<td>47.4</td>
<td>-6.9</td>
<td>22.4</td>
<td>…</td>
</tr>
</tbody>
</table>

Han and Savrasov PRL (2009)
Han and Savrasov PRL (2010)
Further Extension

- Additional resolution
  - Calculation by Hongki Yoon

<table>
<thead>
<tr>
<th>J (meV)</th>
<th>$D_{xy}$</th>
<th>$d_{yz}$</th>
<th>$d_{xz}$</th>
<th>$d_{x^2-y^2}$</th>
<th>$d_z^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_{xy}$</td>
<td>-0.5</td>
<td>-0.5</td>
<td>-0.5</td>
<td>0.0</td>
<td>1.7</td>
</tr>
<tr>
<td>$d_{yz}$</td>
<td>-0.5</td>
<td>-0.5</td>
<td>1.2</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td>$d_{xz}$</td>
<td>-0.5</td>
<td>1.2</td>
<td>0.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$d_{x^2-y^2}$</td>
<td>2.7</td>
<td></td>
<td></td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>$d_z^2$</td>
<td></td>
<td></td>
<td></td>
<td>2.7</td>
<td></td>
</tr>
</tbody>
</table>

- Spin-orbit coupling and other coupling parameters

Iron ferromagnet $T_c \sim 1000K$

Kvashnin et al., PRL (2016)