Multi-Orbital Physics in the Oxypnictides: Are they important?

F. Kruger

D. Argyriou

J. Wu, W. C. Lee (not shown)
What are the pnictides Really?

Cuprates

manganites
What are the pnictides Really?

Cuprates

manganites
What are the pnictides Really?

Cuprates

manganites

multi-orbitals, orbital order, lattice distortions magnetism, local/ itinerant electrons
What are the pnictides Really?

Cuprates

manganites

multi-orbitals,
orbital order,
lattice distortions
magnetism, local/
itinerant electrons

\[ J_H S_c \ll \infty \]
To explain??
To explain??

(Tet->Ortho/resistivity anomaly)

Resistivity

\[ T_{RA} = T_{SPT} \]
To explain??

(Tet -> Ortho/resistivity anomaly)

$T_{SDW}$

Resistivity

$T_{RA} = T_{SPT}$

(a)
To explain??

Resistivity

\[ T_{RA} = T_{SPT} \]

(Tet\textendash{}Ortho/resistivity anomaly)

\[ T_{SDW} \]

\[ T_c \]
localized moments

spins

orbitals

itinerant electrons
localized moments

spins + orbitals

localized electrons

+ itinerant electrons
how far can one get with local moments?
How far can one get with local moments?

$J_2 > J_1/2$
how far can one get with local moments?

Two interpenetrating AF sublattices

$J_2 > J_1 / 2$
how far can one get with local moments?

$J_2 > J_1 / 2$

$\varphi = \hat{n}_1 \cdot \hat{n}_2$

$\langle \varphi \rangle \neq 0$

$\langle n_1 \rangle = \langle n_2 \rangle \neq 0$

Two interpenetrating AF sublattices
how far can one get with local moments?

\[
\psi = \hat{n}_1 \cdot \hat{n}_2
\]

\[
\langle \varphi \rangle \neq 0 \quad \langle n_1 \rangle = \langle n_2 \rangle \neq 0
\]

`nematic'

\[
\langle \varphi \rangle \neq 0 \quad \langle n_1 \rangle = \langle n_2 \rangle = 0
\]

Two interpenetrating AF sublattices
- Inelastic neutron scattering

Inelastic neutron scattering

Isotropic $J_{1a} \approx J_{1b}$

$SJ_1^a = 27$ meV

$SJ_1^b = 25$ meV

$SJ_2 = 36$ meV

• Inelastic neutron scattering

Isotropic $J_{1a} \approx J_{1b}$

- $SJ_1^a = 27$ meV
- $SJ_1^b = 25$ meV
- $SJ_2 = 36$ meV

Anisotropic $J_{1a} \gg J_{1b}$

- $SJ_1^a = 49.9 \pm 9.9$ meV
- $SJ_1^b = -5.7 \pm 4.5$ meV
- $SJ_2 = 18.9 \pm 3.4$ meV

Inelastic neutron scattering


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$SJ_{2} = 18.9 \pm 3.4 \text{ meV}$
Problem?

*itinerant model*

local picture
(different signs of $J$ along x and y axes???)
Problem?

Inconsistent model

Local picture (different signs of J along x and y axes???)
Problem?

- Inherent model
- Local picture
- Different signs of $J$ along x and y axes.
Is there a simple model that captures this physics?

localized/extended electrons+Hund’s coupling

unfrustrated magnetism, SPT, RA

orbital ordering
Coulomb Repulsion:

\[ U = e^2 \int |\psi_{i_1 m_1}(r)|^2 \frac{e^{-|r-r'|/r_0}}{|r-r'|} |\psi_{i_2 m_2}(r')|^2 \]

Energy Difference:

\[ \Delta(\delta) = \frac{U_b(\delta) - U_a}{U_a} \]
SPT in Ising Universality Class

\[ H_{\text{SPT}} = -J_{\text{SPT}} \sum_{\langle i,j \rangle} M_i M_j \]

\[ M_i = \pm 1, \ i = d_{yz}, d_{xz} \]
SPT-induced Collinear AF
SPT-induced Collinear AF

$T > T_{\text{SPT}}$

a)

$dxz/dyz$

spin disordered
SPT-induced Collinear AF

\[ T > T_{SPT} \]

\[ T < T_{SPT} \]

a) \[ d_{xz}/d_{yz} \]

b) \[ d_{yz} \]

spin disordered
SPT-induced Collinear AF

\[ T > T_{SPT} \]

\[ T < T_{SPT} \]

(a) \[ d_{xz/d_{yz}} \]

spin disordered

(b)

\[ t_a \quad t_b \]

\[ J \sim \frac{t^2}{U} \]

\[ J_b > J_a \]
SPT-induced Collinear AF

\[ T > T_{SPT} \]

\[ T < T_{SPT} \]

a) \( d_{xz}/d_{yz} \)

b) \( d_{yz} \)

spin disordered

\[
\begin{align*}
\frac{t_b}{t_a} &> 1 \\
J &\sim \frac{t^2}{U} \\
J_b &> J_a
\end{align*}
\]
SPT-induced Collinear AF

\[ T > T_{SPT} \]

\[ T < T_{SPT} \]

a) \( d_{xz}/d_{yz} \)

b) \( d_{yz} \)

spin disordered

\[
\begin{align*}
    t_b & > t_a \\
    J & \sim \frac{t^2}{U} \\
    J_b & > J_a
\end{align*}
\]

AF1)

\( E_1 = -2J_a + 4J' \)
SPT-induced Collinear AF

\[ T > T_{SPT} \]

\[ T < T_{SPT} \]

\( d_{xz}/d_{yz} \)  \( d_{yz} \)

spin disordered

\[ t_b > t_a \]

\[ J \sim \frac{t^2}{U} \]

\[ J_b > J_a \]

\[ E_1 = -2J_a + 4J' \]

\[ E_2 = +2J_a - 4J' \]
SPT-induced Collinear AF

\[ T > T_{SPT} \]

\[ T < T_{SPT} \]

\[ d_{xz/dyz} \]

\[ t_a \]

\[ t_b \]

\[ J \sim \frac{t^2}{U} \]

\[ J_b > J_a \]

AF1)

\[ E_1 = -2J_a + 4J' \]

AF2)

\[ E_2 = +2J_a - 4J' \]

spin disordered

\[ J' > \frac{J_a}{2} \]
SPT-induced Collinear AF

\[ T > T_{SPT} \]

\[ T < T_{SPT} \]

a) \( d_{xz}/d_{yz} \) spin disordered

b) \( d_{yz} \)

\[ t_b > t_a \]
\[ J \sim \frac{t^2}{U} \]
\[ J_b > J_a \]

AF1) 
\[ E_1 = -2J_a + 4J' \]

AF2) 
\[ E_2 = +2J_a - 4J' \]

\[ J' > \frac{J_a}{2} \]
SPT-induced magnetism

\[ H_{SO} = J_{SPT} \sum_{\langle i,j \rangle} M_i M_j + \sum_{\langle\langle i,j \rangle\rangle} J_2 (M_i, M_j) S_i \cdot S_j \]

\[ + \sum_i J_{1x} (M_i, M_{i+\hat{x}}) S_i \cdot S_{i+\hat{x}} \]

\[ + \sum_i J_{1y} (M_i, M_{i+\hat{y}}) S_i \cdot S_{i+\hat{y}} \]

\[ J_{1x} (M_i, M_j) = \delta_{M_i, M_j} (J_{1b} \delta_{M_i,1} + J_{1a} \delta_{M_i,-1}) \]

\[ J_{1y} (M_i, M_j) = \delta_{M_i, M_j} (J_{1a} \delta_{M_i,1} + J_{1b} \delta_{M_i,-1}) \]

\[ J_2 (M_i, M_j) = \delta_{M_i, M_j} J_2 \]

122 Fe-Fe is shorter: \( J_{-1b} \) is enhanced
orbital ordering

transport anisotropies
Orbital-polarized Fermi surface in antiferromagnetic state of

\[ \text{BaFe}_2\text{As}_2 \]

- Polarized ARPES
Orbital-polarized Fermi surface in antiferromagnetic state of BaFe$_2$As$_2$

Shimojima, et al.

arXiv:0904.1632

• Polarized ARPES

(b) Domain A

(b) Domain B

Crystal surface

$d_{xz}$ in domain A

$d_{xz}$ in domain B
(a) PM state:
1. $d_{xz}$, $d_{yz}$ degenerate
2. Fermi surface is composed by multiple orbitals

(b) AF state:
1. Localized moment is formed by $d_{xz}$
2. Fermi surface is orbital-polarized
Resistivity Anomaly

![Graph showing resistivity anomaly](image)
spin-wave spectrum

origin of ferromagnetic coupling?
spin-wave spectrum

origin of ferromagnetic coupling?

\[ SJ_1^a = 49.9 \pm 9.9 \text{ meV} \]
\[ SJ_1^b = -5.7 \pm 4.5 \text{ meV} \]
\[ SJ_2 = 18.9 \pm 3.4 \text{ meV} \]
spin-wave spectrum

\begin{align*}
S J_1^a &= 49.9 \pm 9.9 \text{ meV} \\
S J_1^b &= -5.7 \pm 4.5 \text{ meV} \\
S J_2 &= 18.9 \pm 3.4 \text{ meV}
\end{align*}

origin of ferromagnetic coupling?
A local-itinerant model

• A local-itinerant model

• A local-itinerant model


Isotropic strongly frustrated $J_1$-$J_2$ model

Doubly-degenerate $d_{xz}$ and $d_{yz}$ type orbitals:

$t_{\pi} > t_{\sigma}$
• A local-itinerant model

Isotropic strongly frustrated \( J_1-J_2 \) model


Doubly-degenerate \( d_{xz} \) and \( d_{yz} \) type orbitals:

Ferromagnetic Hund’s couplings \( J_H \)

\( t_\pi \quad t_\sigma \quad t_\sigma \quad t_\pi \)

\( t_\sigma > t_\pi \)

\( S = 1/2 \quad J_2 \gtrsim J_1/2 \)
• A local-itinerant model

Isotropic strongly frustrated $J_1$-$J_2$ model


Doubly-degenerate $d_{xz}$ and $d_{yz}$ type orbitals:

Ferromagnetic Hund’s couplings $J_H$

$S = 1/2$  $J_2 \gtrsim J_1/2$

consistent with the local multiplet structure:
Double-Exchange Model

\[ \mathcal{H}_{\text{loc}} = \frac{J_1}{S^2} \sum_{\langle i,j \rangle} S_i \cdot S_j + \frac{J_2}{S^2} \sum_{\langle\langle i,j \rangle\rangle} S_i \cdot S_j \]

unlike manganites

\[ \nu J_H \ll \infty \]
Double-Exchange Model

\[ \mathcal{H}_{\text{loc}} = \frac{J_1}{S^2} \sum_{\langle i,j \rangle} S_i \cdot S_j + \frac{J_2}{S^2} \sum_{\langle\langle i,j \rangle\rangle} S_i \cdot S_j \]

\[ \mathcal{H}_{\text{it}} = -\sum_{ij,\alpha\beta,\nu} t_{ij}^{\alpha\beta} c_{i\alpha\nu}^{\dagger} c_{j\beta\nu} + \frac{V}{2} \sum_{i,\alpha \neq \beta,\nu\nu'} \hat{n}_{i\alpha\nu} \hat{n}_{i\beta\nu'} \]

\[ \mathcal{H}^{(0)}_{\text{H}} = -\frac{J_H}{2} \sum_{k,\alpha,\nu} \nu \tilde{c}_{k\alpha\nu}^{\dagger} \tilde{c}_{k\alpha\nu} \]

unlike manganites

\[ \nu J_H \ll \infty \]
• Orbital ordering

\[ n_o = \sum_\nu (\rho_{yz,\nu} - \rho_{xz,\nu}) \]
\[ n_s = \sum_\alpha (\rho_{\alpha,\uparrow} - \rho_{\alpha,\downarrow}) \]
Spin-wave dispersion with both super- and double-exchange

\[ J_1 = 0.04t_\sigma, \quad J_2 = 0.6J_1 \]

\[ n = 0.1, \quad t_\pi = 0.1t_\sigma \]
Comparison with experiments:

\[ J_H \sim t_\sigma \sim 1 \text{ eV} \quad \tilde{J} \sim 0.01 \ t_\sigma \sim 10 \text{ meV} \]

emergent ferro-orbital order
OO-induced anisotropies

T.-M. Chuang, et al.
Science (2010)

AF is easy axis!

FM easy axis!

- Resistivity
  arXiv:1002.3364
**OO-induced anisotropies**

- **STM**
  - FM easy axis!

- **Resistivity**
  - AF is easy axis!
    - arXiv:1002.3364
    - Science (2010)
Are the pnictides important?
pnictides $d^6$
bad metals
multi-orbital
low-moment
moderate
$T_c$
Cuprates $d^9$
(one-unpaired electron),
single-orbital
Mott-system,
low-moment,
high $T_c$

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pnictides $d^6$
bad metals
multi-orbital
low-moment
moderate $T_c$
Cuprates $d^9$ (one-unpaired electron), single-orbital Mott-system, low-moment, high $T_c$

Manganites multi-orbital high-moment no $T_c$

Pnictides $d^6$ bad metals multi-orbital low-moment moderate $T_c$
Cuprates $d^9$
(one-unpaired electron),
single-orbital Mott-system,
low-moment, high $T_c$
Cuprates $d^9$
(one-unpaired electron),
single-orbital Mott-system, low-moment, high $T_c$

pnictides $d^6$
bad metals multi-orbital low-moment moderate $T_c$

Multi-orbital Mott system, low-moment ??? $T_c$
EFRC Center
Does multi-orbital physics (>1 e/unit cell) increase $T_c$?

Cuprates $d^9$
- (one-unpaired electron)
- single-orbital Mott-system
- low-moment, high $T_c$

Multi-orbital Mott system, low-moment
- $T_c$

EFRC Center

Pnictides $d^6$
- bad metals
- multi-orbital
- low-moment
- moderate $T_c$
Possible systems (multi-orbital Mott systems)?

**Simple**

$NiO_{1-x}F_x$

hole-doping a $d^8$ system.

Does it superconduct?

**Complex** (oxychalchogenides)

$A_2O_3Fe_2M_2$

$A=La, Y$

$M=Se, S, Te$

**Co and Cu-based**

$La_2Co_2Se_2O_3$

$Na_2OCu_4OSe_2$
La$_2$Co$_3$Se$_2$O$_3$: A Quasi-Two-Dimensional Mott Insulator with Unusual Cobalt Spin State and Possible Orbital Ordering

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Abstract: The new oxyselenide La$_2$Co$_3$Se$_2$O$_3$ containing CoO square-planar layers, has been successfully synthesized using solid-state reactions under vacuum. The compound crystallizes in space group $I4/mmm$ with lattice parameters $a = 4.0697(8)$ Å and $c = 18.419(4)$ Å. Magnetic susceptibility measurements indicate an antiferromagnetic transition at $\sim 220$ K. The magnetic entropy associated with the transition is close to $R \ln 2$, suggesting an unusual low-spin state for the Co$^{3+}$ ions. The as-prepared sample shows insulating behavior with room-temperature resistivity of $\sim 10^7$ Ω cm, which decreases by 4 orders of magnitude under a pressure of $\sim 5$ GPa. Band structure calculations using the LSDA+$U$ approach reproduce the insulating ground state with low spin for Co and suggest strong orbital polarization for the valence electron near the Fermi level. It is also revealed that the spin and orbital degrees of freedom in the antiferromagnetic checkerboard spin–lattice are mutually coupled.

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Electronic structure, spin state, and magnetism of the square-lattice Mott insulator La$_2$Co$_3$Se$_2$O$_3$ from first principles

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Figure 2. Temperature dependence of resistivity for the La$_2$Co$_3$Se$_2$O$_3$ polycrystalline sample. The inset shows the pressure dependence of resistance at room temperature. Note that the resistance at ambient pressure is too high to be measured accurately.

Figure 3. Temperature dependence of the magnetic susceptibility for La$_2$Co$_3$Se$_2$O$_3$. The applied field is 1000 Oe. The data fittings in the insets are based on Curie–Weiss law.
Low-temperature nuclear and magnetic structures of $La_2O_3Fe_2OSe_2$ from x-ray and neutron diffraction measurements

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Band Narrowing and Mott Localization in Iron Oxychalogenides $La_2O_3Fe_2O(Se, S)_2$

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Layered Oxychalcogenides and Oxypnictides

For example:

- Replace apical oxides by larger anion
- Intercalate Na under kinetic control
- Fill tetrahedral holes

Various compounds shown including:
- Sr₂Ti₂O₇
- Na₂Sr₂Ti₂O₇
- Sr₂MnO₂Cu₂S₂
- Sr₂Fe₂O₂Cu₂S₂
- Na₅Cu₅O₅Se₅
- LaFe₂O₅Se₂
- Bi₅O₂Cu₂Se₂

Related compounds:
- PbO
- BaO
- LaOCl
- LaOCuS
- LaOCuSe
- LaOFsAs
$T_N \sim 100\text{K}$ as reference

$x=0.00$

$H=1\text{kOe}$

D. Argyrioiu, HMI, Berlin
T_N \sim 100K as reference

what about the doped system? superconductivity?
$T_N \sim 100K$ as reference

$x=0.00$

$H=1\text{kOe}$
$T_N \sim 100K$ as reference

$x=0.00$
$H=1kOe$

$x=0.05$
$H=50Oe$

$x=0.1$
$H=50Oe$

ZFC
FC
Do the doped oxychalcogenide superconduct? multi-orbital Mott systems?

Thanks to D. Argyriou, W-C Lee, F. Kruger, W-C. Lv, J. Wu
Superconductors come of age

A South Korean company has placed by far the biggest commercial order for superconducting wires.

Superconducting wires could soon help to light up Seoul.

iStockphoto.com / Min-Gyu Seong

American Superconductor have not disclosed the value of the deal. But Jason Fredette, managing director of corporate communications at the company, says that LS Cable will use the wire to make about 20 circuit kilometres of cable as part of a programme to modernize the South Korean electricity network starting in the capital, Seoul.