IRG-1

Spin-Orbit Coupling in Correlated Materials: Search for Novel Phases and Phenomena

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Big Science Drivers for IRG-1

Topological Band Insulators
6s/6p

Metals & Band Insulators

Our focus:
4d/5d materials

Heavy Fermions
5f/6f

High $T_c$ Superconductors
3d

Coulomb interactions $U/W$

$W$ = Band width
Big Science Drivers for IRG-1

Why is band theory inadequate for large U/W?

Electrons interact → strong correlations → many body problem

- Movement of one electron requires adjustment of all electrons

- Gapless surface states protected by topology and symmetry

- Hasan & Kane, RMP (2010)
- Qi & Zhang, RMP (2010)

- Problem still unsolved after 25 years
4d/5d materials: The next frontier

- Topological Band Insulators
  - 6s/6p
- Metals & Band Insulators
- Heavy Fermions
  - 5f/6f
- High T_c Superconductors
  - 3d

\[ \lambda \approx U \approx W \]
comparable energy scales

New phases and new phenomena

3 June 2014
What will 4d/5d oxides enable that is not achievable from other materials

Why is topology important?
- Robust dissipationless current
- Spin-momentum locking \( \rightarrow \) spin filters
- Precise quantization 1 part in \( 10^9 \) in QHE

(1) Topology + magnetism
(1) Magneto-electric response
(3) Tunable room temperature magnetic heterostructures
\( \rightarrow \) Fault tolerant spin based quantum computing

Why are Coulomb interactions important?

Emergent phases
- Magnetism
  - Data storage
  - Generators
  - High \( T_c \) SC
  - Transmission cables
  - Wind turbines
Interplay of Spin-Orbit coupling & Correlations

\[ \mathcal{H}_{\text{SOC}} = \lambda \mathbf{L} \cdot \mathbf{S} \]

\[ \mathcal{H}_J = -J \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j \]
4d/5d materials, bandwidth & interactions

<table>
<thead>
<tr>
<th>3d</th>
<th>Cr</th>
<th>Mn</th>
<th>Fe</th>
<th>Co</th>
</tr>
</thead>
<tbody>
<tr>
<td>4d</td>
<td>Mo</td>
<td>Tc</td>
<td>Ru</td>
<td>Rh</td>
</tr>
<tr>
<td>5d</td>
<td>W</td>
<td>Re</td>
<td>Os</td>
<td>Ir</td>
</tr>
</tbody>
</table>

Examples of electron count

- \( \text{Cd}_2\text{Os}_2\text{O}_7 \rightarrow \text{Os}^{5+} \) with \( 5d^3 \)
- \( \text{Y}_2\text{Os}_2\text{O}_7 \rightarrow \text{Os}^{4+} \) with \( 5d^4 \)

Tuning correlations \( U \) & SOC \( \lambda \)

Beyond 5d\(^5\) Iridates

Using structure & strain to tune orbital degeneracy & bandwidth \( W \)

Perovskite  Double Perovskite  Pyrochlore
Goals of IRG-1: 4d/5d materials

Lay Foundations and Establish New Paradigms

Topological phases
*Discovery*

Novel magnetism
*Understanding*

Tunable magnetic heterostructures
*Control*

CREATE:

- New magnetic materials: 4d and 5d oxides
- New principles: SOC + U
- New tunable heterostructures

\[ | \Psi \rangle = c^\dagger | 0 \rangle \]
Our Expertise

Epitaxial Films
Single Crystals
New Materials
(Woodward, Yan, Yang)

Theory & Computation
(Randeria, Trivedi, Windl)

Spin-Orbit coupling + Correlations

Advanced Characterization
ARPES, THz, TEM, Synchrotron &
neutron scattering, XRD,
Magnetometry, Transport
(Kaminski, McComb, Valdés Aguilar,
Woodward, Yan, Yang)

Woodward co-lead (Chemistry)
Yang (Physics)
McComb (Materials Science)
Yan (Materials Science, Tennessee)
Kaminski (Physics, Iowa State)

Windl (Materials Science)
Randeria (Physics)
Trivedi co-lead (Physics)
Valdes-Aguilar (Physics)

Over 50 joint publications
8 with over 200 citations
Materials are key

**Design & Synthesis of New Materials**

*Woodward*

Proven experience with 5d oxides, including Os

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**Single Crystal Growth**

*Yan*

- Vapor Transport
- Flux Growth
- High pressure floating zone

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**Epitaxial Films**

*Yang*

- Off-axis sputtering
- High quality and uniformity
- Stoichiometric & chemically ordered complex oxides

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**Sr\textsubscript{2}CoOsO\textsubscript{6}**

Grown 10 new compounds with Ru or Os

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**Prominent Laue oscillations on strained films**

First single crystal
Materials are key

- Design, Synthesis,
  and Characterization
  of New Materials

- Single Crystal
  Growth

- Frontiers in
  Crystalline Matter

- Proven experience with
  5d oxides, including:
  - Vapor Transport
  - Flux Growth
  - High pressure floating zone
  - Off-axis sputtering

- First single crystal
  growth of Sr$_2$CoOsO$_6$

- Grown 10 new compounds
  with Ru or Os

- Crisis in the US about
  availability of high
  quality samples

- Concerns about
  competitiveness and
  exploitation of new
  materials

- Prominent Laue oscillations
  on strained films
Theory: Insights → Predictions → Guide Experiments

Experiments
New materials
Characterization

Model

Theory beyond DFT
Strong interactions + Disorder + \( T \neq 0 \)
Effective field theory: Randeria
Quantum Monte Carlo simulations: Trivedi
Topological invariants
Exact diagonalization

Predictions

Finite temperature magnetic properties
Raising \( T_c \)
Doping and disorder
ARPES and Optical conductivity
Finite temperature band structure
Grand challenges requiring a team effort

Topological phases

Novel magnetism

Tunable magnetic heterostructures
What is a Weyl semi-metal?

Bulk Dirac dispersion

Surface Fermi Arcs

$H^{\pm} = \pm c \ p \cdot \sigma$

3D analog of graphene; Weyl point robust

Broken time reversal symmetry or inversion symmetry

Unusual magneto-electric effects

Wan, Turner, Vishwanath, Savrasov
PRB 83, 205101 (2011)
Weyl semi-metal in Pyrochlore Iridates

Theory Prediction for $A_2\text{Ir}_2\text{O}_7$


Our Strategy:

Epitaxial films of Pyrochlore Iridates → Tuning with both rare-earth `A’ and strain

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First epitaxial film of any rare-earth iridate pyrochlore

Scanning transmission electron microscopy (STEM) (McComb)

Nd$_2$Ir$_2$O$_7$: ideally placed for observing Weyl semi-metal

Eu$_2$Ir$_2$O$_7$: Eu is non-magnetic; all magnetism from Ir

Growth and XRD (Yang)
Advanced Spectroscopic capabilities to probe topological states like Weyl semi-metal

**ARPES: FERMI ARCS**

![ARPES Arrows]

*Bi$_2$Sr$_2$CaCu$_2$O$_8*

Kondo, ... & Kaminski, PRL 111, 157003 (2013)

First ARPES observation of Fermi Arcs in cuprates:
~ 1000 citations

**THz: KERR ROTATION**

- 10 T
- 5K
- Bi$_2$Se$_3$
- $\theta = 65 \pm 3^\circ$
- colossal Kerr rotation!

**NEUTRONS:**

*Ho$_2$Ti$_2$O$_7*

Dynamical spin fluctuations
Dunsiger, Yan, Woodward

Clancy, Ruff, Dunsiger et. al, PRB 79, 014408 2009
Grand challenges requiring a team effort

- Topological phases
- Novel magnetism
- Tunable magnetic heterostructures
Novel Magnetism & Exchange Pathways

\( \text{Sr}_2\text{CoOsO}_6 \)
Os spins order at \( T_{N1} = 110 \text{ K} \)
Co spins order at \( T_{N2} = 70 \text{ K} \)

Coupling constants (DFT)

\[
J_1^{\text{eff}} = -1.3 \text{ meV}
\]
\[
J_2^{\text{eff}} = -47 \text{ meV}
\]
\[
J_3^{\text{eff}} = +20 \text{ meV}
\]
\[
J_4^{\text{eff}} = -13 \text{ meV}
\]

Questions

Why do Goodenough-Kanamori rules fail?
Why are longer range \( J_2 \) and \( J_3 \) so large?
Why do Os and Co order independently?

Proposed:

Single xtals, other 3d/5d compounds
Inelastic neutron scattering, XMCD
Derive Hamiltonian w/ SOC →
anisotropic directional magnetism;
Competing, frustrated states

Novel Magnetism & Quantum Phase Transitions

5d\(^2\)  \quad \leftrightarrow \quad 5d\(^3\)  \quad \leftrightarrow \quad 5d\(^4\)  \quad \leftrightarrow \quad 5d\(^5\)

\(\text{Cd}_2\text{Re}_2\text{O}_7\)
- single xtal 2-3mm
- Superconductor
- \(T_c = 1\) K

\(\text{Cd}_2\text{Os}_2\text{O}_7\)
- single xtal < 0.2 mm
- Slater insulator
- \(T_N = 227\) K

\(\text{Y}_2\text{Os}_2\text{O}_7\)
- polycrystal
- Orbitally entangled ferromagnet

\(\text{RE}_2\text{Ir}_2\text{O}_7\)
- thin film
- Weyl semimetal

\(\text{Cd}_2\text{Re}_{(2-x)}\text{Os}_{(x)}\text{O}_7\)

Proposed:
- Metal-insulator transition
- AF-paramagnet transition
- Role of SOC
- Tools: synthesis, xtal growth, THz, ARPES, neutrons, theory

Prediction of novel magnetic \(d^4\) Insulator
(Resonant X-ray scattering)

Meetei, Cole, Randeria, Trivedi, arXiv:1311.2823
Grand challenges requiring a team effort

- Topological phases
- Novel magnetism
- Tunable magnetic heterostructures
Magnetic materials for oxide heterostructures

 masih

Oxide epitaxy

Strain tuning

Room temperature magnetism

High Tc and Exquisite strain tunable magneto-crystalline anisotropy ~ 10s of Tesla

Strain engineering on piezoelectric substrates

Hauser, ..., Woodward & Yang, PRB (2011) & PRB (2012);
Sr$_2$CrReO$_6$ XMCD: Hauser, ..., Windl, ..., Woodward & Yang, PRB (2014)
SFMO anisotropy: Du, ..., Yang & Hammel, PRL (2013)

3d/5d magnets
Sr$_2$CrOsO$_6$ (3d$^3$-5d$^3$) Ferrimagnet $T_C=720K$

Meetei, Ertten, Randeria, Trivedi, & Woodward, PRL 110, 087203 (2013);
Osmates LDA + DMFT: Meetei, Mravlje, Biermann, Georges, Randeria & Trivedi (preprint)
IRG-1: Correlations + SOC in 4d & 5d materials: Why is our discovery program important?

Topological phases
*Discovery*

Novel magnetism
*Understanding*

Tunable magnetic heterostructures
*Control*

Beyond d⁵ Iridates
New Paradigms

Piezoelectric control of magnetism