Magnetism, Electronic Transport and Magneto-Structural Coupling in $\text{Sr}_2\text{IrO}_4$

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JNU, Frustrated Magnetism, Feb 13, 2015
Our Group at JNU

- Work in experimental condensed matter physics.
- Most work in low temperature regime.
- Do prepare materials, characterize them and study physical properties.
- At present group of 7 people: 3 Ph.D students and 3 M.Sc students.
- Transition metal oxides (4d & 5d) based materials.
Sr$_2$IrO$_4$ (214) : Overview

- Sr$_2$$^{2+}$Ir$^{4+}$O$_4$$^{2-}$ is 5$d$ based Transition Metal Oxide

- Electronic configuration: Ir$^{4+}$ → $4f^{14}5d^5$

- Significant Crystal Field effect Low spin state

- Half filled $t_{2g}$ band.

- Extended nature of 5$d$ orbitals.

- Reduced electronic correlation effect than 3$d$ and 4$d$ counterpart.

- Therefore, 5d oxides expected to be metallic.

- Heavy character of Ir (77) atom.

- Significant spin-orbit coupling (SOC) effect ($\propto z^4$, atomic number)

- Comparable energy scale of Coulomb interaction and SOC $\sim$ 0.5 eV
Sr$_2$IrO$_4$ (214) : Structural Overview

- Member of Ruddlesden-Popper series
  \[ \text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1} \text{ with } n = 1. \]
- Layered (K$_2$NiF$_4$) structure.
- Crystalizes in tetragonal structure
  Space group : \( I4_1/\text{acd} \).
- Alternate tilting of IrO$_6$ octahedra
  along \( c \)-axis (\( \theta_{\text{Oct}} \sim 11^\circ \)).
- Iso-structural with
  La$_2$CuO$_4$ and Sr$_2$RuO$_4$
- Possible superconductivity !!!
  Wang, PRL, 106, 136402 (2011)
  Yang, PRB 89, 094518 (2014)
Sr$_2$IrO$_4$ (214) : Magnetic Overview

- Canted Antiferromagnet with $T_N \sim 240$ K.
- Structural distortion induced Dzyaloshinsky-Moriya (DM) interaction.
- Weak ferromagnetism with much lower moment than spin-only value ($1 \mu_B$/f.u) for $S = \frac{1}{2}$
Sr$_2$IrO$_4$ (214) : Electronic Transport

Sr$_2$IrO$_4$
Korneta, PRB, 82, 115117 (2010)

Sr$_2$RhO$_4$
Perry, JPCM, 8, 175 (2006)
Sr$_2$IrO$_4$ (214) : Insulating behaviour

- $J_{\text{eff}}$ Mott Insulator : Electronic correlation driven

- Interplay between SOC, W and U gives novel ground state.
- SOC splits $t_{2g}$ band onto $J_{\text{eff}} = 1/2$ and $J_{\text{eff}} = 3/2$ band.
- $J_{\text{eff}} = 1/2$ band is half-filled and narrow.
- Moderate $U$ can lead to Mott gap.

B. J. Kim, PRL 101, 076402 (2008); Science, 323, 1329 (2009)
Sr$_2$IrO$_4$ (214) : Insulating behaviour

Slater Insulator : Magnetic Order driven

*Ab initio* Studies on the Interplay between Spin-Orbit Interaction and Coulomb Correlation in Sr$_2$IrO$_4$ and Ba$_2$IrO$_4$

R. Arita,$^{1,2,3}$ J. Kuneš,$^4$ A. V. Kozhevnikov,$^5$ A. G. Eguiluz,$^6$ and M. Imada$^{1,3}$

Time resolved optical study
Hsieh, PRB, 86, 035128(2012)

STM/STS investigation.
Li, Scientific Reports, 3, 3073 (2013)
Sr$_2$IrO$_4$ (214) : Material Synthesis

- Single-phase polycrystalline material prepared using solid state method.
- Materials are characterized using XRD and allied Rietveld analysis.
- Sample crystallizes in tetragonal structure with $I4/\text{acd}$ symmetry.
- Lattice parameters: $a = 5.4980(2)$ Å and $c = 25.779(1)$ Å.

Bhatti, AKP, JPCM, 27, 016005 (2014)
- Weak FM transition around 238 K (dM/dT).
- Steep decrease in $M_{ZFC}(T)$ below $\sim 95$ K.
- Curie-Weiss behaviour in limited temperature.
- Estimated $\theta_p = 233$ K and $\mu_{\text{eff}} = 0.56 \mu_B$/f.u.
- Expected $\mu_{\text{eff}} = 0.57$ and $\mu_H = 0.33 \mu_B$/f.u ($g_J = 2/3$).

Ge, PRB, 84, 100402 (2011).
Sr$_2$IrO$_4$ (214) : Temperature Dependent Structure

- Representative XRD data in PM, FM and low temperature state.
- No structural phase transition down to 20 K.
**Sr₂IrO₄ (214) : Temperature Dependent Structure**

Temperature dependent changes in structural parameters.

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**Unit Cell**

- **a (Å)**
- **c (Å)**
- **c/a**
- **V (Å³)**

**IrO₆ Octahedra**

- **<Ir-O₂-Ir> (Deg)**
- **θ_{Oo} (Deg)**
- **d_{Ir-O₂} (Å)**
- **d_{Ir-O₁} (Å)**
Sr$_2$IrO$_4$ (214) : Prediction for Magneto-Structural correlation

- Magnetism is linked to IrO$_6$ octahedra distorsion.
- $\alpha$ - IrO$_6$ octahedra distortion angel.
- $\Phi$ - spin canting angle.
- $\theta$ - tetragonal distortion parameter.

Jackeli, PRL, 102, 017205 (2009)
Sr$_2$IrO$_4$ (214) : Electronic Transport

- Resistivity shows insulating behaviour ($d\rho/dT < 0$).
- Resistivity increases by five orders at low temperature.
- Electronic transport can be understood using 2D Mott variable range hopping model.

\[
\rho = \rho_0 \exp\left(\frac{T_0}{T}\right)^{1/3}
\]

<table>
<thead>
<tr>
<th>Temperature range (K)</th>
<th>$T_0$ (K)</th>
<th>$\xi$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>300–240</td>
<td>$1.44 \times 10^5$</td>
<td>3.04</td>
</tr>
<tr>
<td>240–70</td>
<td>$4.68 \times 10^4$</td>
<td>4.42</td>
</tr>
<tr>
<td>40–5</td>
<td>$4.82 \times 10^3$</td>
<td>9.44</td>
</tr>
</tbody>
</table>
**Sr$_2$IrO$_4$ (214) : Magneto-transport**

Magnetoresistance (MR) = \[
\frac{\Delta \rho}{\rho(0)} = \frac{\rho(H) - \rho(0)}{\rho(0)}
\]

- **Positive MR** (weak antilocalization) in strong SOC systems, Bi$_2$Se$_3$, Bi$_2$Te$_3$, Na$_2$IrO$_3$ films.
- **Negative MR** at low T in VRH regime – weak localization – quantum interference effect – Quadratic field dependence.

![Graphs showing magnetoresistance vs. magnetic field and field squared vs. field]
Sr$_2$IrO$_4$ (214) : Critical Analysis

- Magnetic isotherms across FM transition

\[ M_s(T) = M_0(-\epsilon)^\beta, \quad \epsilon < 0, \]

\[ \chi_0^{-1}(T) = \Gamma(\epsilon)^\gamma, \quad \epsilon > 0, \]

\[ M = XH^{1/\delta}, \quad \epsilon = 0, \]

\[ \epsilon = (T - T_C) / T_C \]
$\textbf{Sr}_2\textbf{IrO}_4$ (214) : Critical Analysis

3D Heisenberg Model
$\beta = 0.365$
$\gamma = 1.386$

3D Ising Model
$\beta = 0.325$
$\gamma = 1.24$

Mean Field Model
$\beta = 0.5$
$\gamma = 1.0$
**Sr$_2$IrO$_4$ (214) : Critical Analysis**

Modified Arrott plot:

\[
\left( \frac{H}{M} \right)^{1/\gamma} = a \frac{(T - T_C)}{T} + b M^{1/\beta}
\]

- $\beta = 0.55$
- $\gamma = 1.15$
**Sr$_2$IrO$_4$ (214) : Critical Analysis**

**Critical Plot**

- $T_c = 225$ K
- $\beta = 0.57(2)$
- $\gamma = 1.147(1)$

**Kouvel-Fisher Plot**

- $T_c = 225$ K
- $\beta = 0.5517(1)$
- $\gamma = 1.14(3)$

**Critical isotherm:** $M \propto H^{1/\delta}$
Sr$_2$IrO$_4$ (214) : Critical Analysis

Scaling analysis

$$M(H, \epsilon) = \epsilon^\beta f \left( \frac{H}{\epsilon^{\beta+\gamma}} \right)$$

### Exponents

<table>
<thead>
<tr>
<th></th>
<th>$\beta$</th>
<th>$\gamma$</th>
<th>$\delta$</th>
</tr>
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<tbody>
<tr>
<td>This Work</td>
<td>0.55</td>
<td>1.15</td>
<td>3.03</td>
</tr>
<tr>
<td>Mean Field</td>
<td>0.5</td>
<td>1.0</td>
<td>3.0</td>
</tr>
<tr>
<td>3D Heisenberg</td>
<td>0.365</td>
<td>1.386</td>
<td>4.8</td>
</tr>
<tr>
<td>3D Ising</td>
<td>0.325</td>
<td>1.241</td>
<td>4.82</td>
</tr>
</tbody>
</table>
**Sr$_2$IrO$_4$ (214) : Thermal Demagnetization**

Thermal Demagnetization $\Delta M$:

Spin-wave (SW) excitation (Bloch):

$$\frac{\Delta M}{M(0)} = \frac{M(T) - M(0)}{M(0)} = BT^{3/2}$$

Stoner single-particle (SP) excitation:

$$\frac{\Delta M}{M(0)} = \frac{M(T) - M(0)}{M(0)} = CT^{3/2} \exp\left(-\frac{\Delta}{k_B T}\right)$$

$$\Delta M_{\text{Total}} = \Delta M_{\text{SW}} + \Delta M_{\text{SP}}$$
Sr$_2$IrO$_4$ (214) : Magnetocaloric Effect (MCE)

- Magnetic Entropy change: $\Delta S_M(T,H) = S_M(T,H) - S_M(T,0) = \int_0^H \left( \frac{\delta M(T,H)}{\delta T} \right)_H dH$

- Relative cooling power: $\Delta S_M(T,H) \times \delta T_{FWHM}$
Sr$_2$IrO$_4$ (214) : Field dependence of MCE

Nice linear field dependence
For both $\Delta S_M$ and RCP.

Linear dependence of
$\Delta S_M$ and $(H/T_C)^{2/3}$ shows mean
Field nature.
Sr$_2$IrO$_4$ (214) : Resistivity derivative

Mott OR Slater OR Both !!!
Acknowledgement

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Rajeev Rawat, UGC-DAE CSR

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UGC, Govt of India  
DST, Govt of India
!!! Thank you !!!

- Point of explosive (or silent) giving up
- Intercede ("scaffold") here
- Watch out for this rise!