# Current-Control of Structural and Physical Properties in Spin-Orbit-Coupled Insulators

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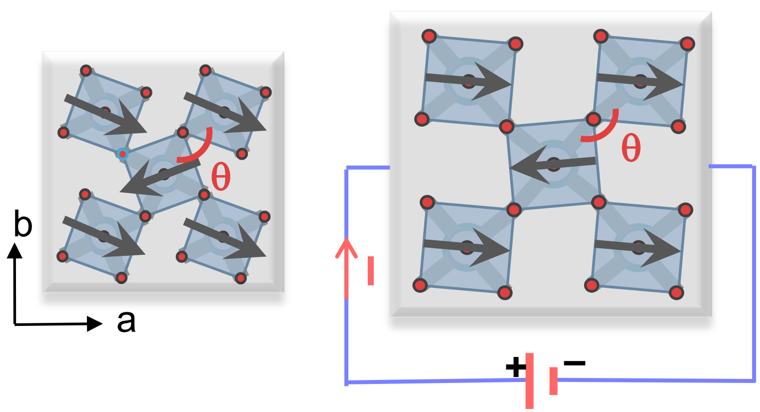
- Overview
- Two Model Systems: $Sr_2IrO_4$ and $Ca_2RuO_4$
- Outlook






*Quantum Matter: Computation Meets Experiments, March 12, 2020*

# Extended 4d/5d-Orbitals, Competing Energies

- **Strong Spin-Orbit Interactions** $\lambda_{so}$

$\lambda_{so}$ **~** $\mathbf{Z^2}$ **L·S,** e.g., Z = 29 for Cu and 77 for Ir

0.4 - 1 eV (5d)

0.1 - 0.3 eV (4d)

0.01 - 0.1 eV (3d)

- **Reduced Coulomb Interaction U**

0.4 - 2.0 eV (5d)

2 - 3 eV (4d)

5 - 7eV (3d)

- **Reduced Hund's Rule Coupling** $\mathbf{J_H}$

~ 0.5 eV (5d)

0.5 - 0.6 eV (4d)

0.7 - 0.9 eV (3d)

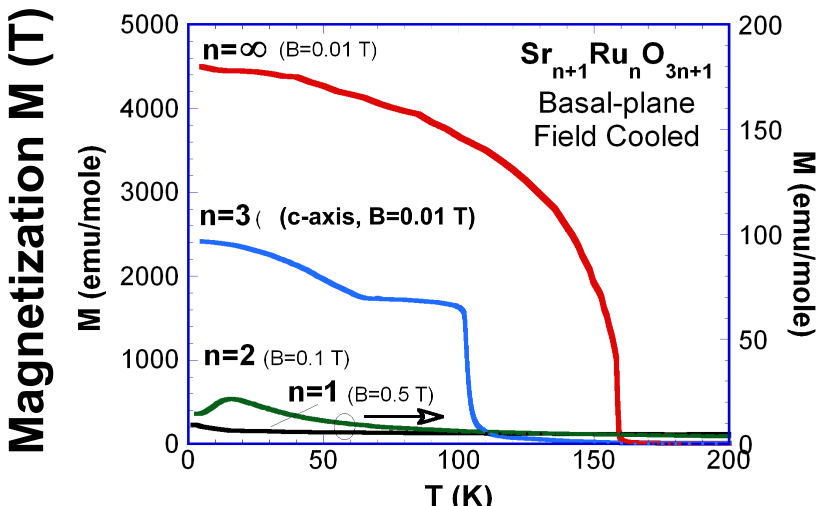
# Comparison between 3d and 4d/5d electrons

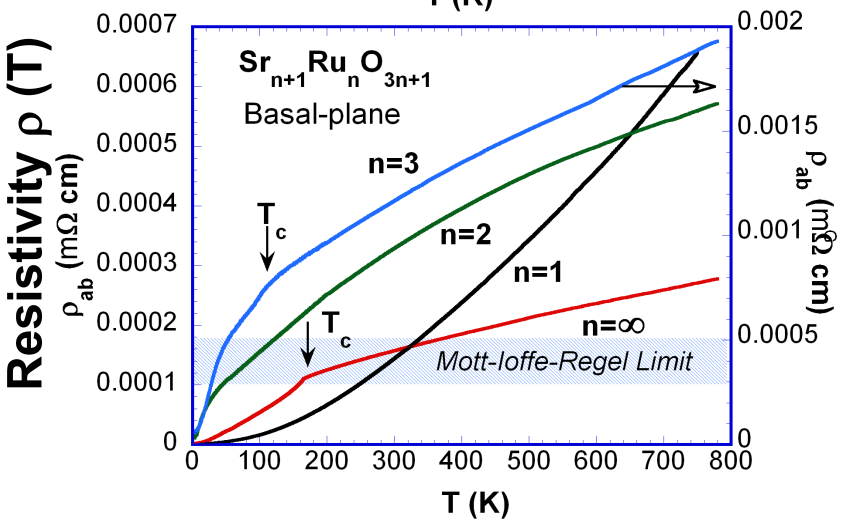
| Electron Type | U (eV) | $\lambda_{so}$ (eV) | $J_H$ (eV) | Key Interactions | Exemplary Phenomena |
|---|---|---|---|---|---|
| ***3d*** | 5-7 | 0.01-0.1 | 0.7-0.9 | $U >> J_H > \lambda_{so}$ | HTSC/CMR |
| ***4d*** | 2-3 | 0.1-0.3 | 0.5-0.6 | $U > J_H > \lambda_{so}$ | Orbital order/ Novel SC |
| ***5d*** | **0.4-2.0** | **0.4-1** | **~ 0.5** | $\mathbf{U \sim J_H \sim \lambda_{so}}$ | Novel insulating state |

A new balance between relevant energies drives new physics introduction

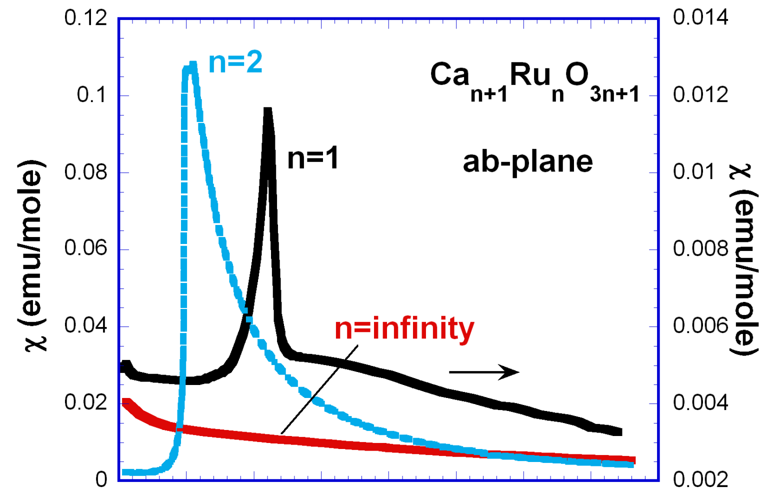
# Lattice-Driven Ground States
# Magnetization and Resistivity for Layered Ruthenates

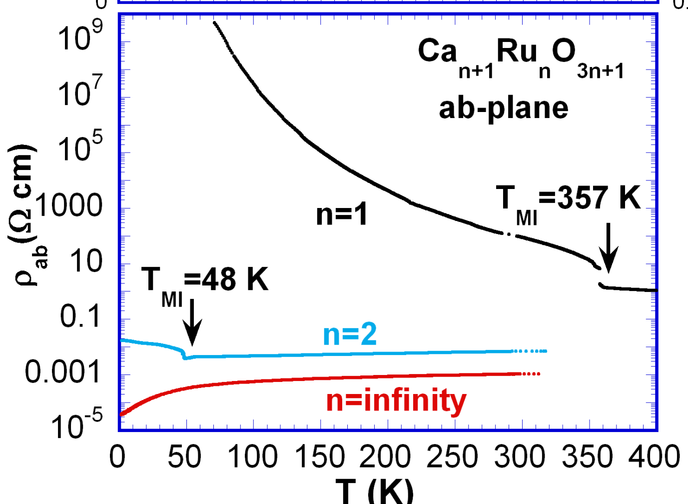
## *Ferromagnetic Metals*
## *$Sr_{n+1}Ru_nO_{3n+1}$*




## *Antiferromagnetic Insulators*
## *$Ca_{n+1}Ru_nO_{3n+1}$*






*Frontiers of 4d- and 5d-Transition Metal Oxides*, GC and DeLong, 2013 introduction

# Empirical Trends in Iridates

- Iridates tend to be magnetic insulators
- Unusual correlation between magnetic and insulating states
- No metallization at high pressure (e.g., $Sr_2IrO_4$ remains insulating up to 185 GPa)
- A metallic state can be readily realized upon chemical doping
- Yet, superconductivity remains unconfirmed despite similarities to cuprates.
- *There is a growing list of theoretical predictions or proposals (spin liquids, various topological states, superconducting state, etc.), but they have met limited experimental confirmation*
- **Extremely sensitive to disorder and structural distortions**



*GC & P. Schlottmann, Reports on Progress in Physics, A Key Issues Review (2018)*

# Key Characteristics

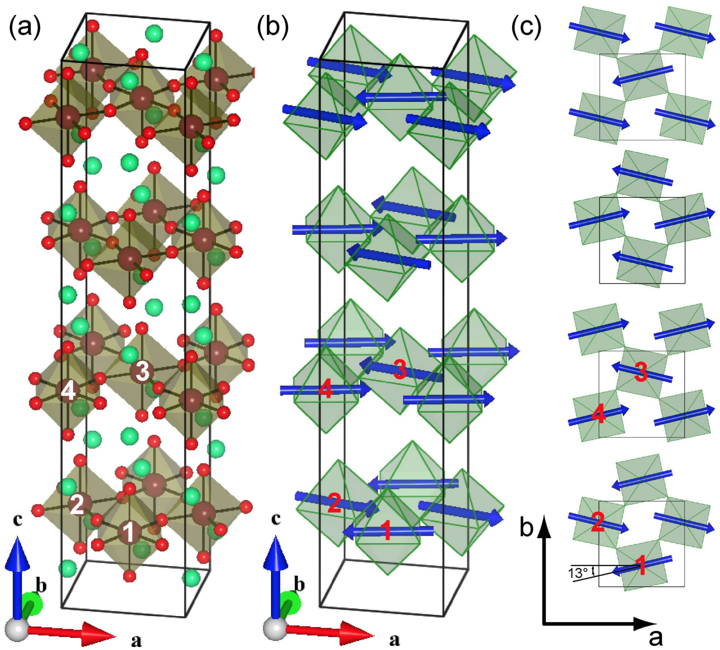
- ***Lattice-driven physical properties***
- ***Strong interlocking between the lattice and magnetic moments***

example 1 – $Sr_2IrO_4$

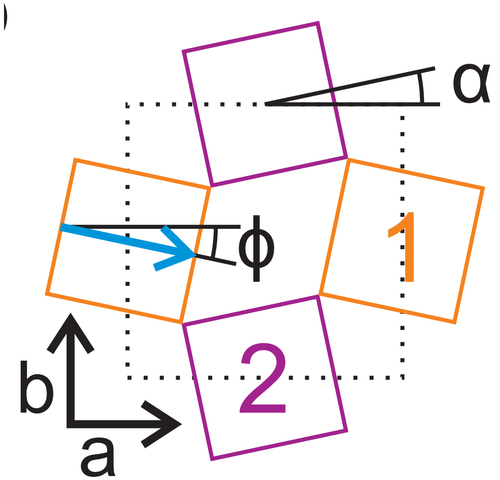
# Model System: $Sr_2IrO_4$

**Key Point: Strong interlocking between the lattice and moments**

The moments rigidly track the octahedral rotation about c-axis ($\alpha$=11.8°) by deviating from the a-axis by $\phi$=13.2° at 4 K.



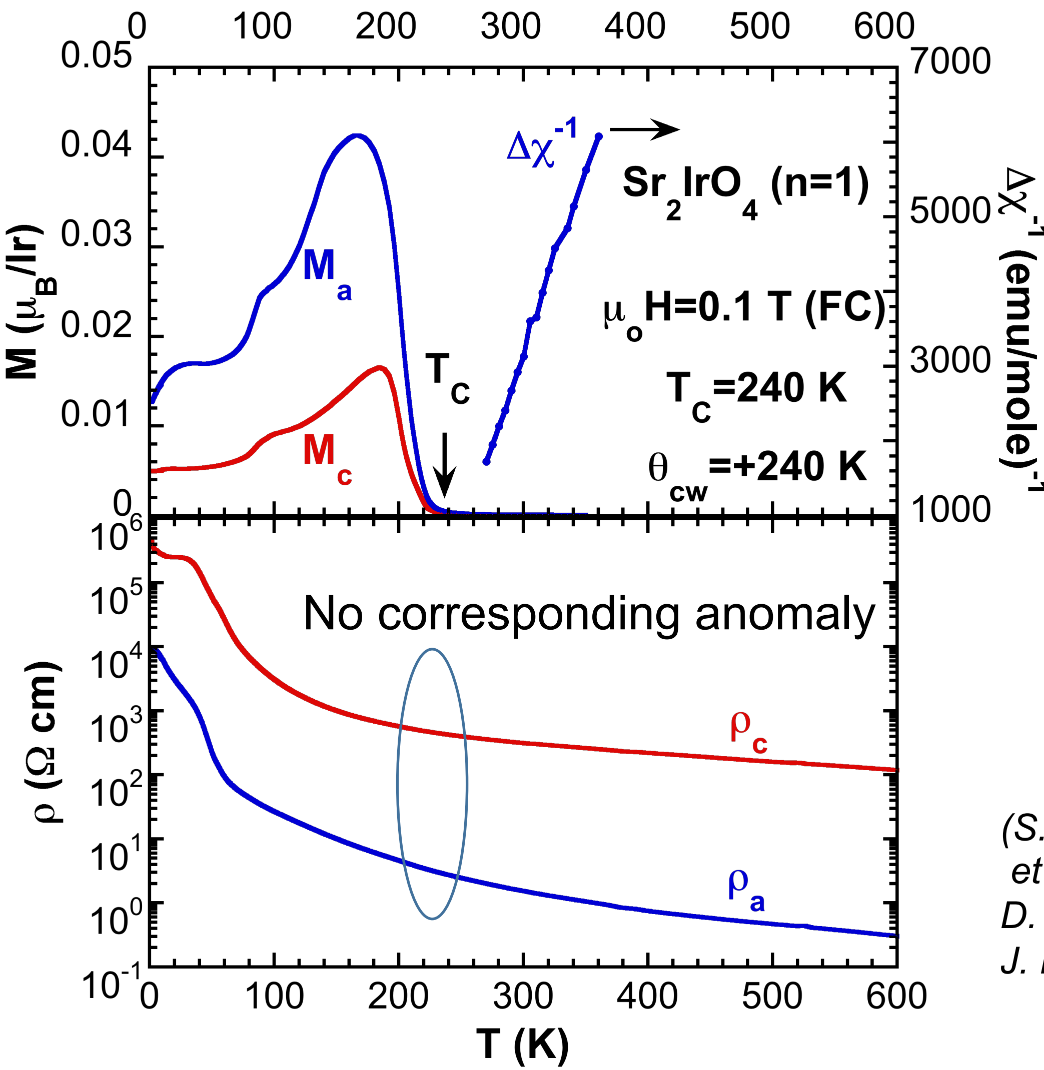

*Feng Ye et al. Phys. Rev. B 2013*




*D. Torchinsky et al. Phys. Rev. Lett.2015*

example 1 – $Sr_2IrO_4$

# Magnetization and Resistivity of $Sr_2IrO_4$



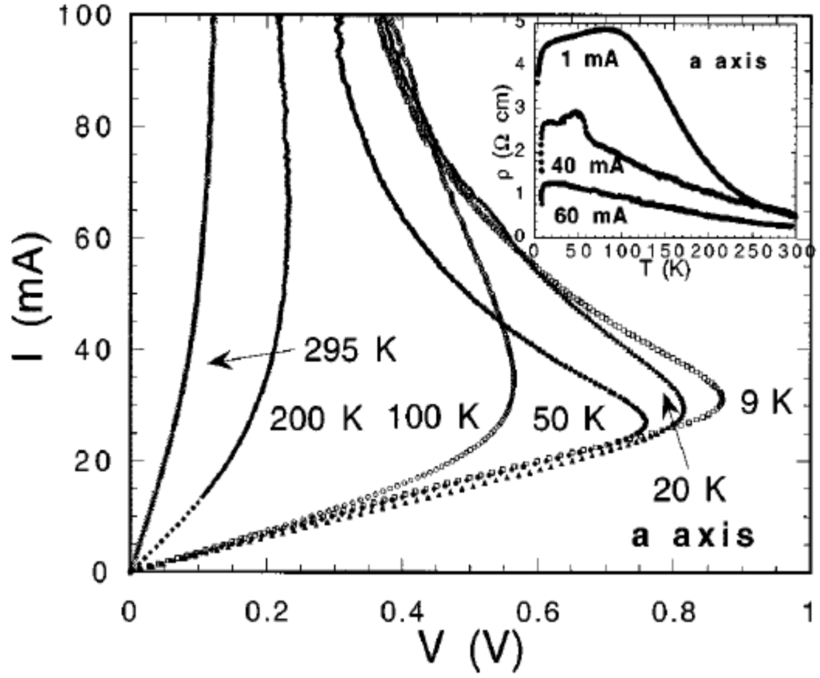

- Neel temperature
  **$T_N$=240 K**
- Charge gap
  **$\Delta \leq$ 620 meV**

*(S.J. Moon, et al., 2008; B.J. Kim, et al., 2011; Q. Wang, et al., 2012; D. Hsieh et al, 2012; J. Dai, et al., 2013; J. Nichols, et al., 2013)*

*GC et al, 1998*

example 1 – $Sr_2IrO_4$

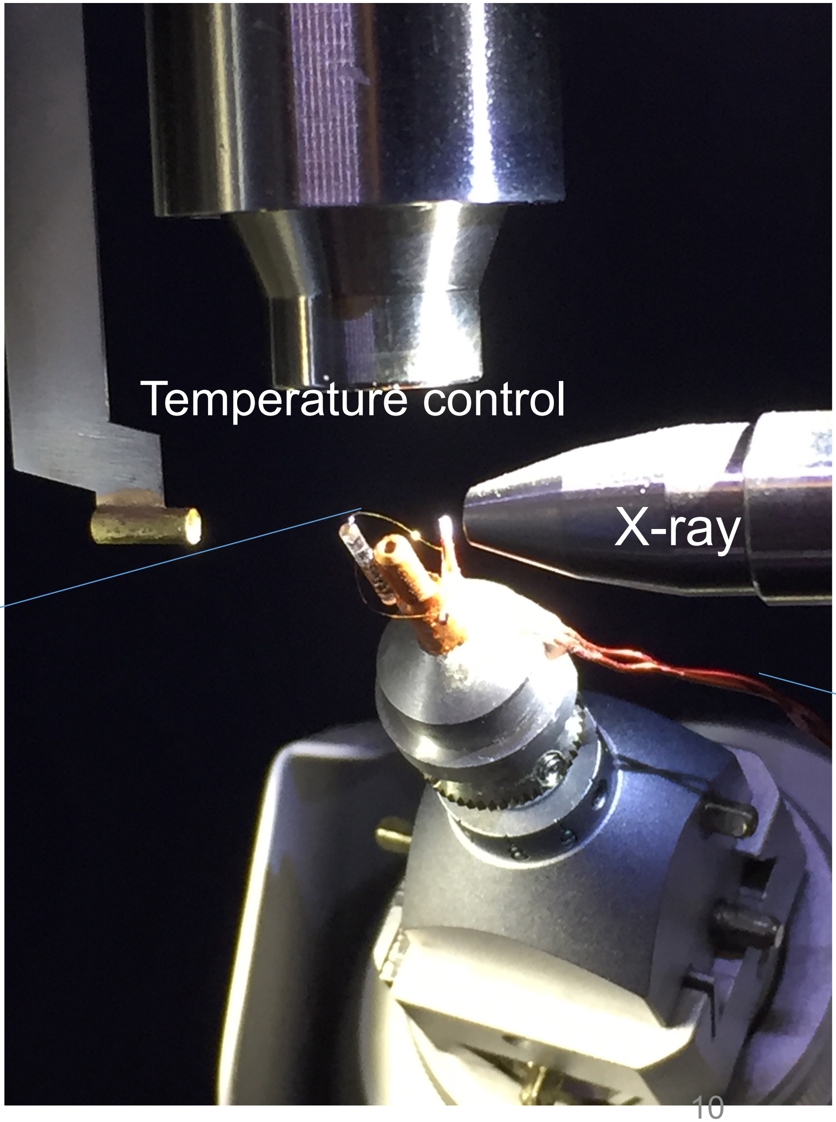
# Early Observation of Unusual I-V Characteristics in $Sr_2IrO_4$




*GC et al, 1998*

example 1 – $Sr_2IrO_4$

# Single-Crystal X-ray Diffraction as a Function of Current

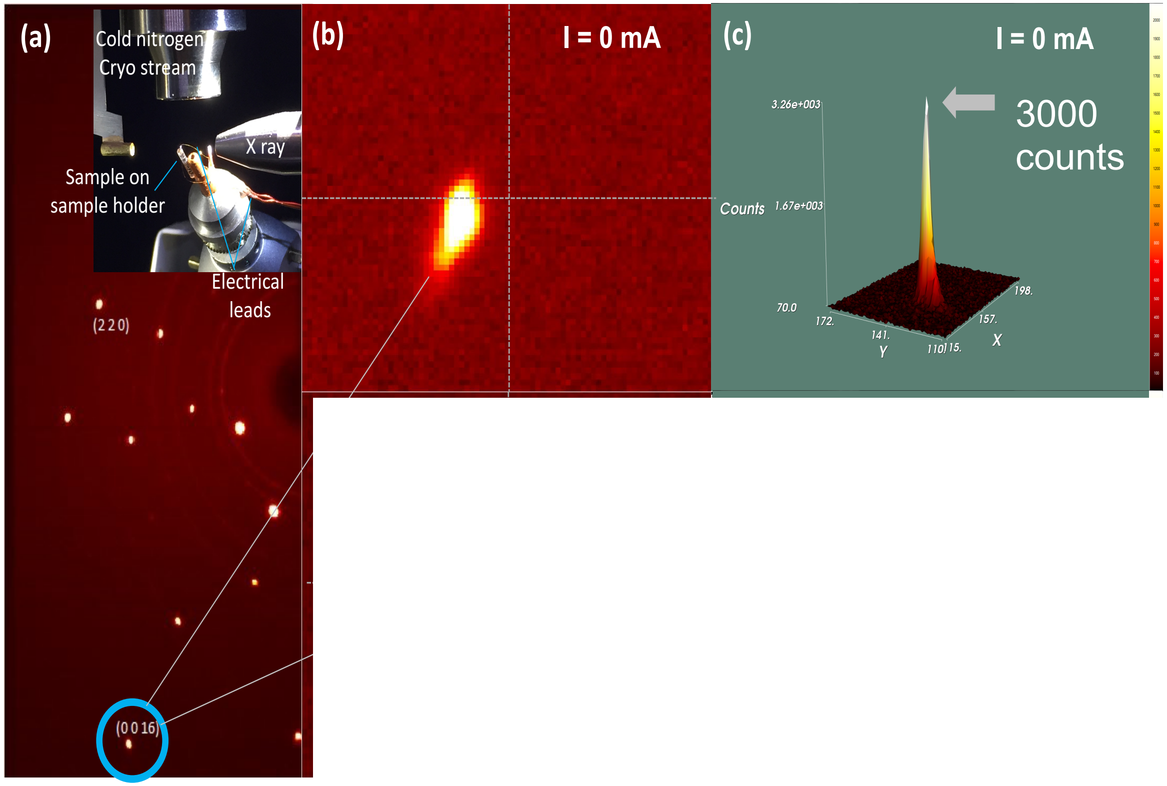



Gold electrical leads and single-crystal sample

Copper electrical wires to current source

example 1 – $Sr_2IrO_4$

# Bragg peaks as a Function of Current

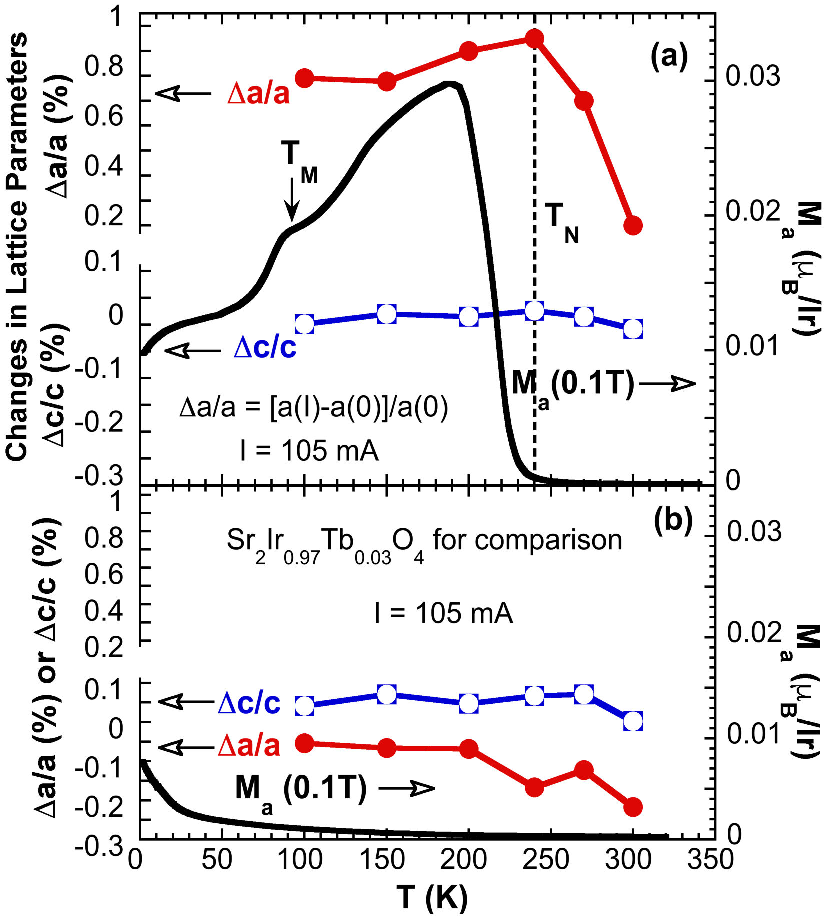



GC et al, Phys. Rev. Lett., 2018

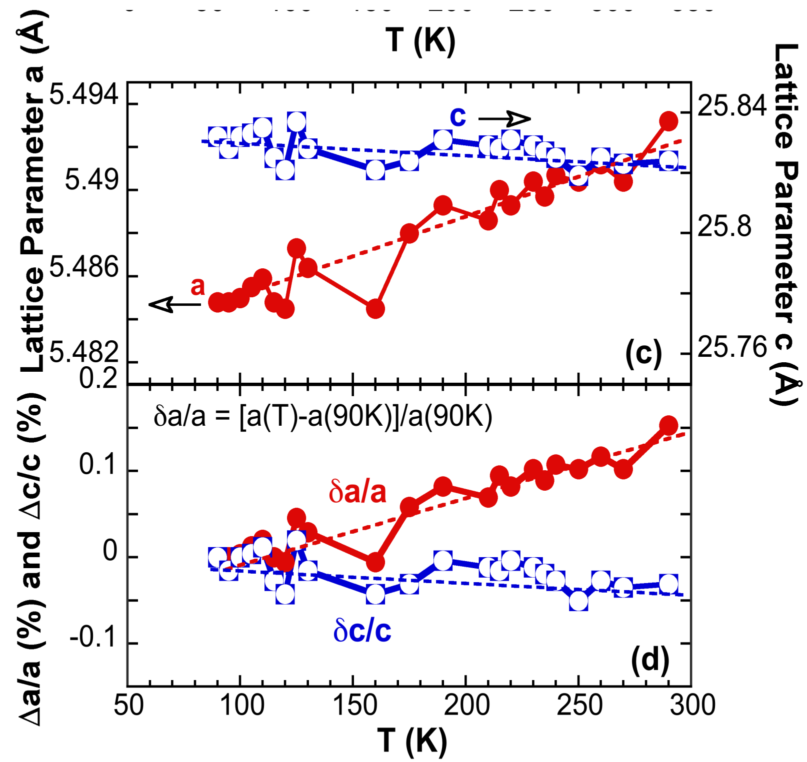
example 1 – $Sr_2IrO_4$

# Coupling of the Lattice and Magnetic Order

**Electrical-current controlled lattice change:**
**The a-axis grows by about 1% !**




Pure thermal expansion <u>without current:</u>
The a-axis expands by only 0.1%
from 90 K to 300 K

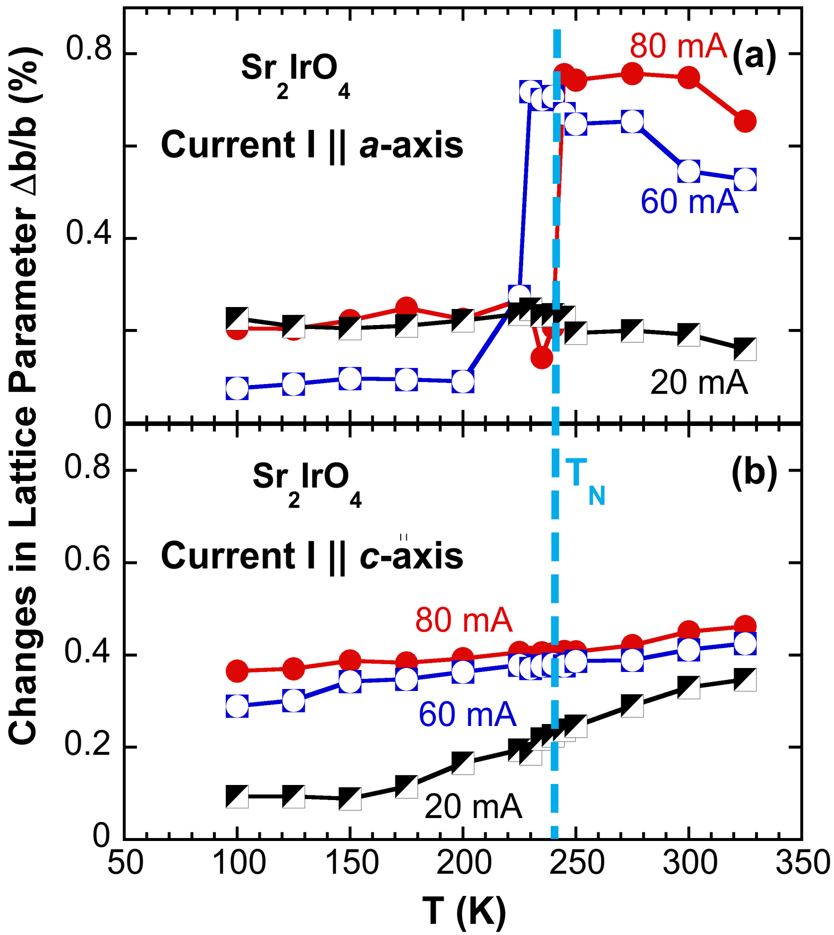





*GC et al, Phys. Rev. Lett., 2018*

example 1 – $Sr_2IrO_4$

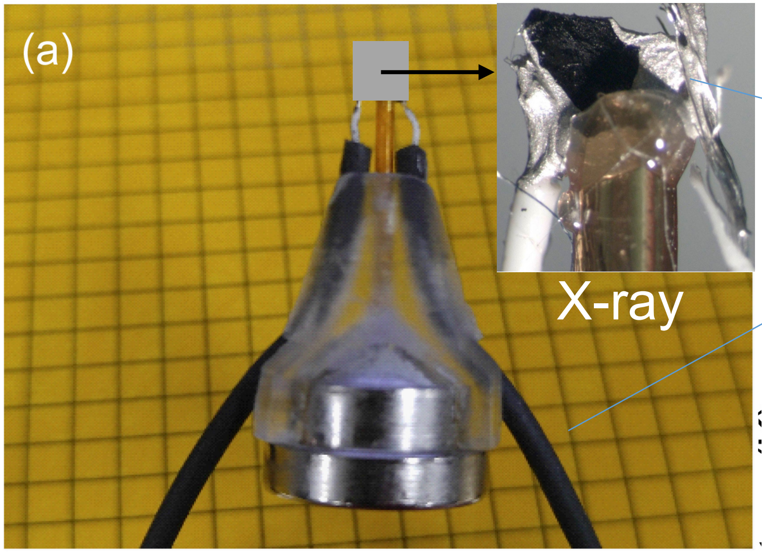
# Anisotropic response of the lattice to current I applied to the a-axis and c-axis




Recall the moments within the basal plane

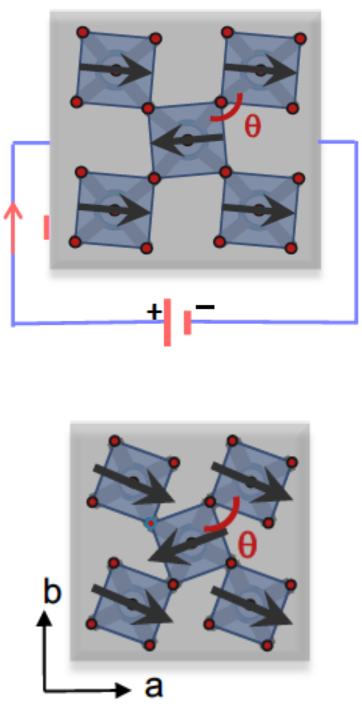
example 1 – $Sr_2IrO_4$

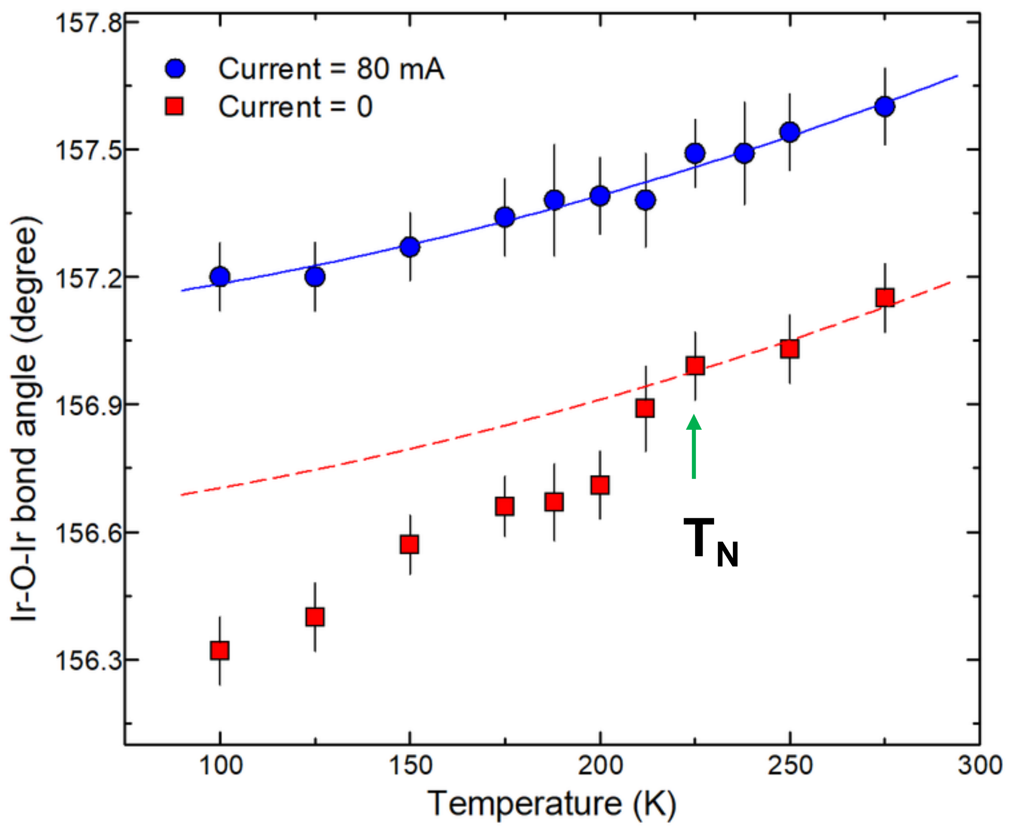
## Single-Crystal Neutron Diffraction as a Function of Current



example 1 – $Sr_2IrO_4$

# Ir-O-Ir Bond Angle Increases with Applied Current I (Neutron Diffraction)



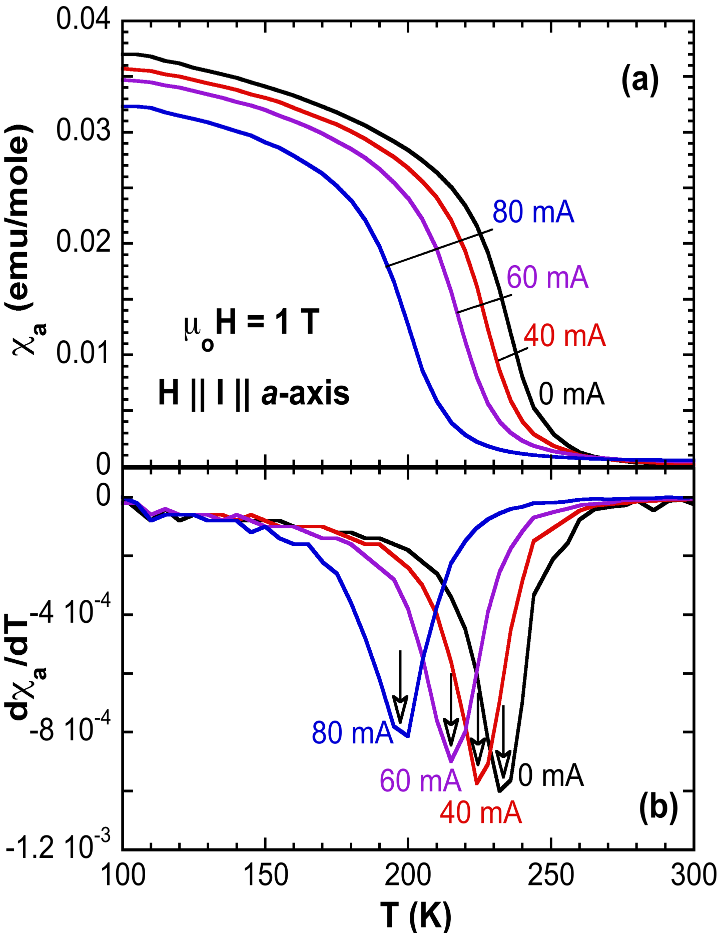




**Δθ ~ 1 degree at 100 K due to I = 80 mA**

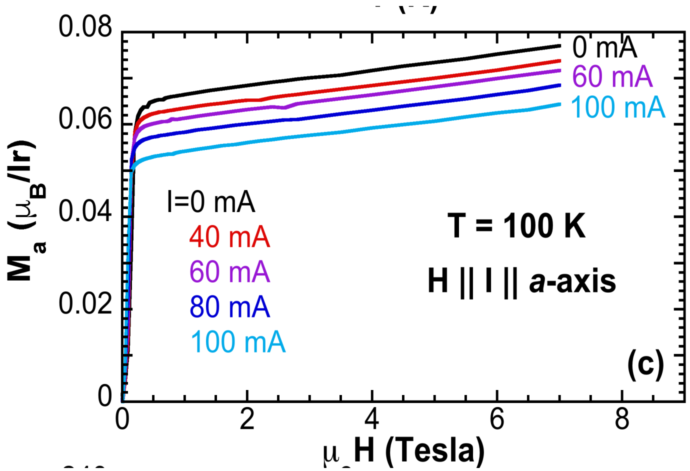
example 1 – $Sr_2IrO_4$

# Electrical-Current Controlled Magnetic Properties

## $T_N$ drops by 40 K



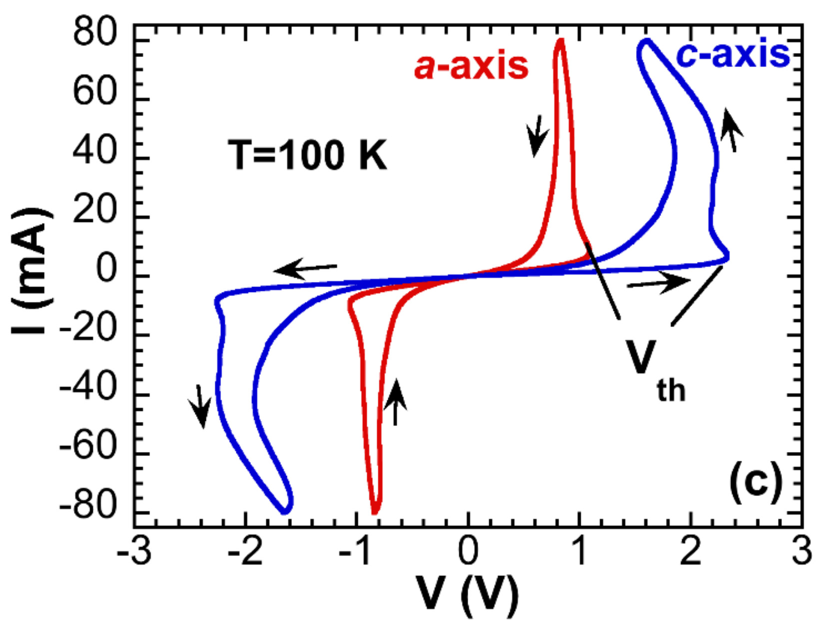

## M (H) drops by 16 %



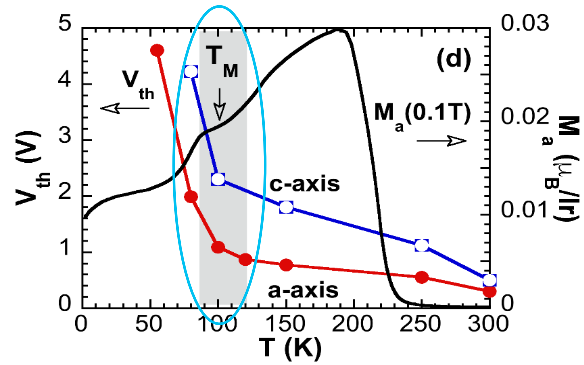
example 1 – $Sr_2IrO_4$

# Electrical-Current Controlled Transport Properties

## I-V Characteristics & Switching




## Coupling of $V_{th}$ and M; dI/DV

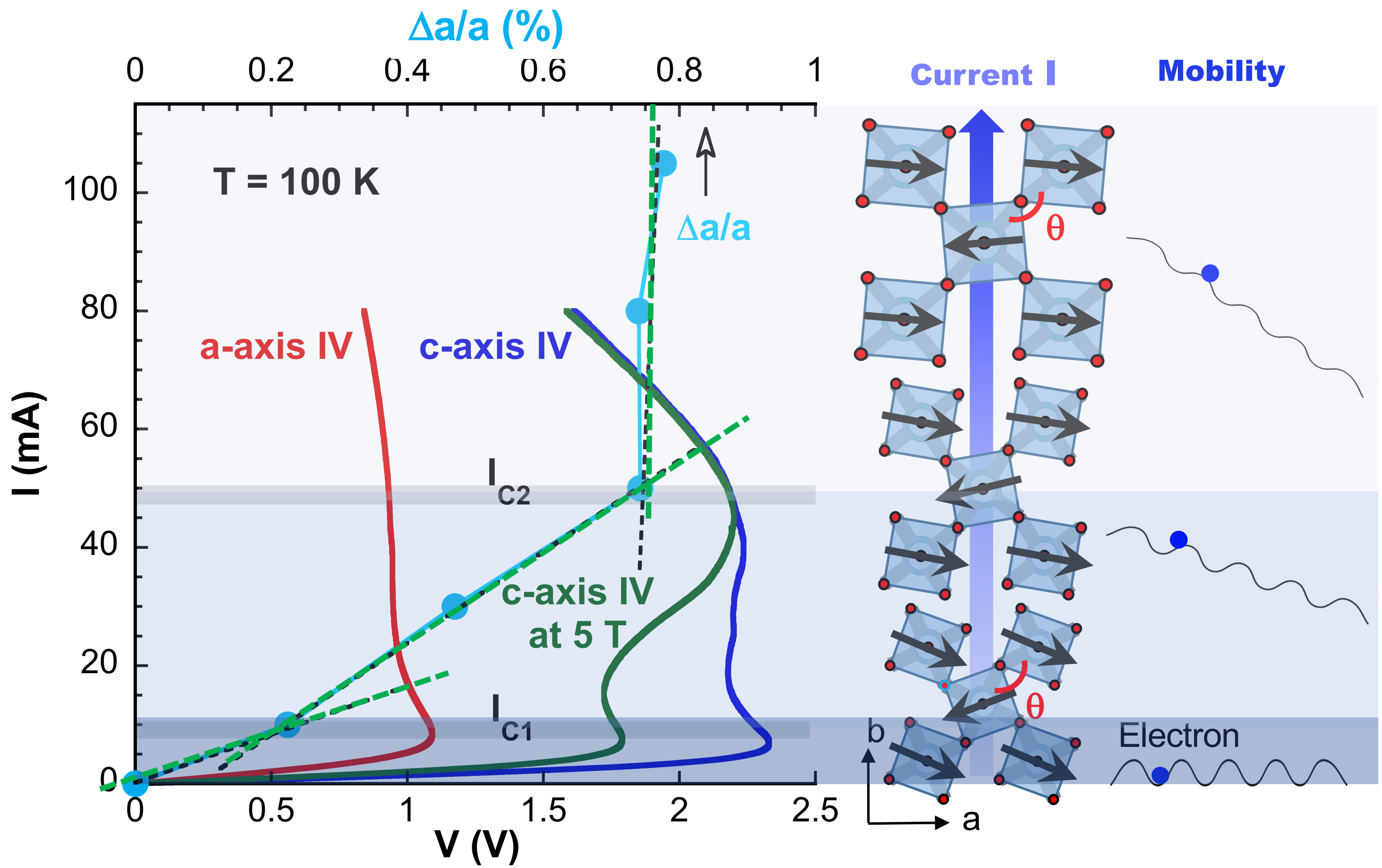



*GC et al, Phys. Rev. Lett., 2018*

example 1 – $Sr_2IrO_4$

# Electrical-Current Controlled Resistive Switching

## I-V curves closely track the current-controlled *a*-axis expansion

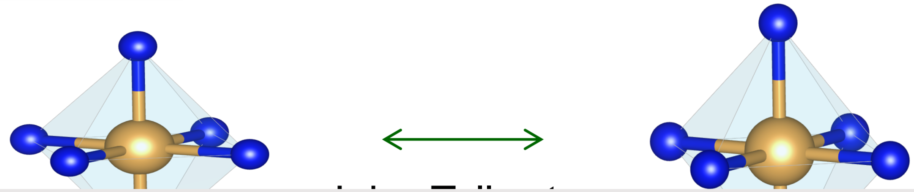





GC et al, *Phys. Rev. Lett.*, 2018

example 2 – $Ca_2RuO_4$

# Model System: $Ca_2RuO_4$



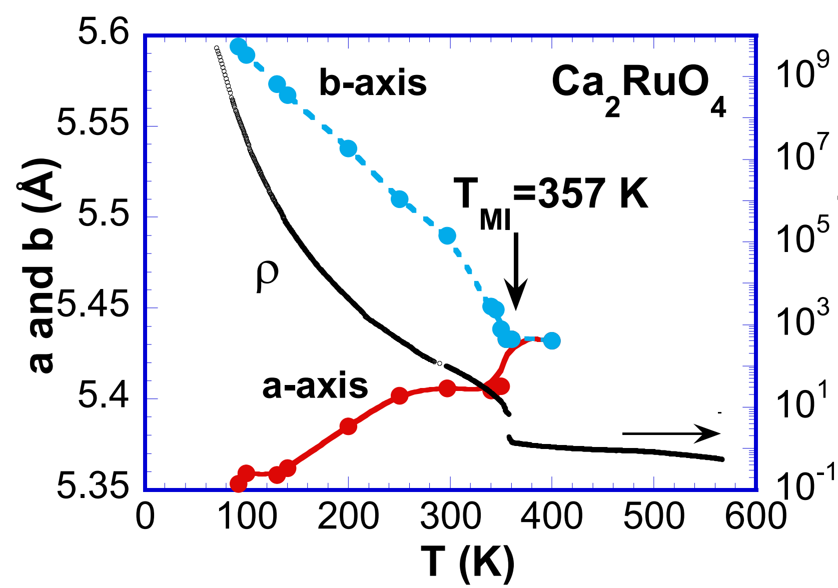
$Ru^{4+}$ ($4d^4$)

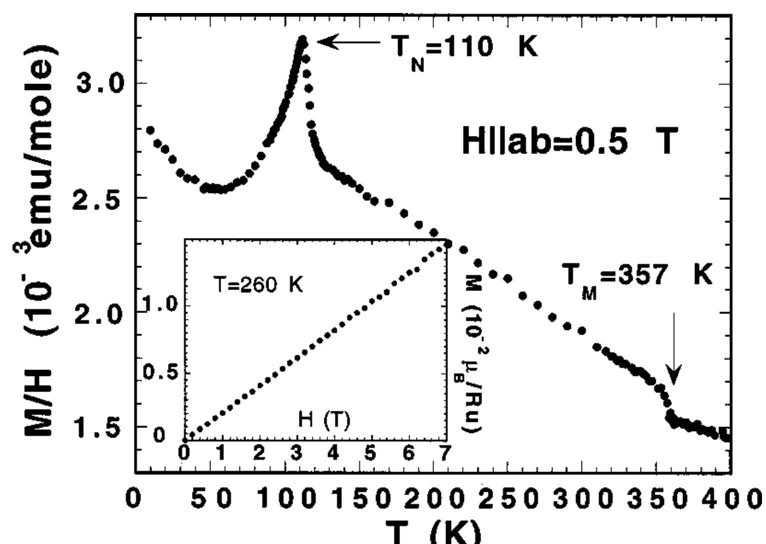
First-order structural/orbital transition

$T_{MI}$ = 357 K

Antiferromagnetic transition

$T_N$ = 110 K



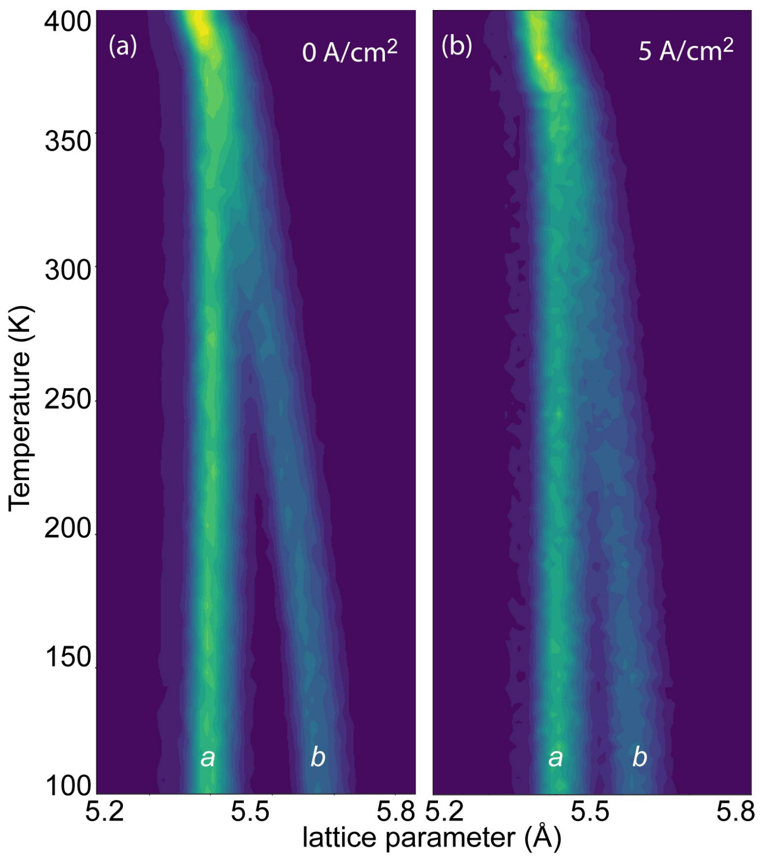



*GC et al PRB 1997-1999*

example 2 – $Ca_2RuO_4$

# Current suppresses the structural distortions in 3% Mn doped $Ca_2RuO_4$

Neutron diffraction as a function of current density



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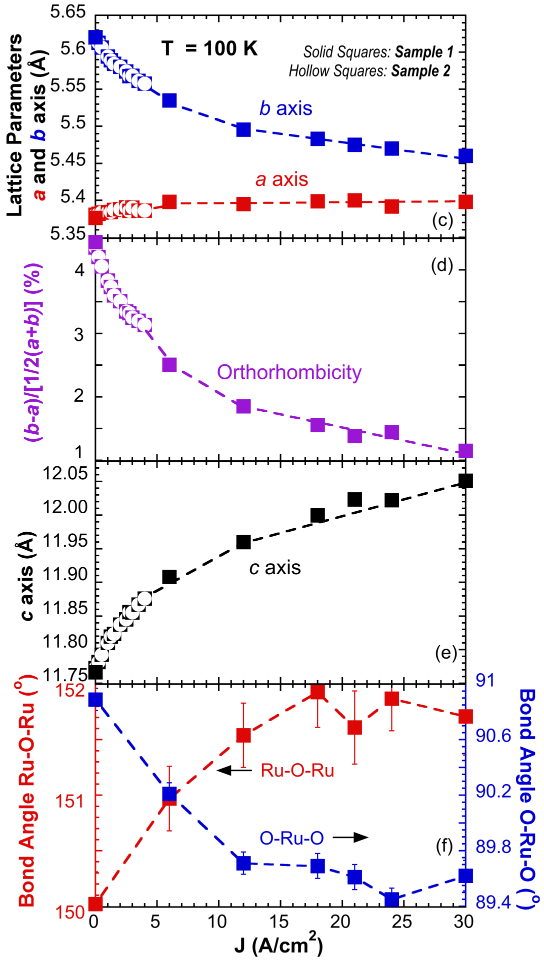
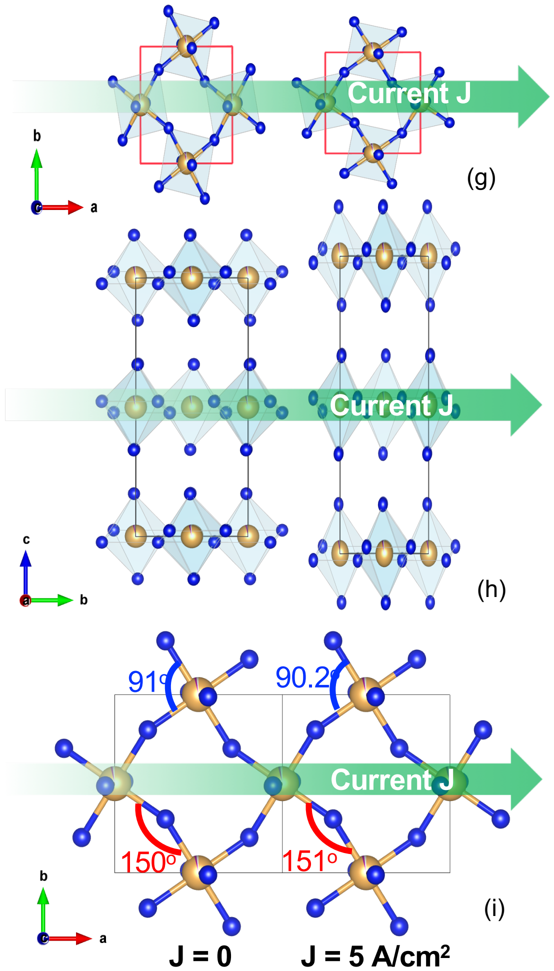


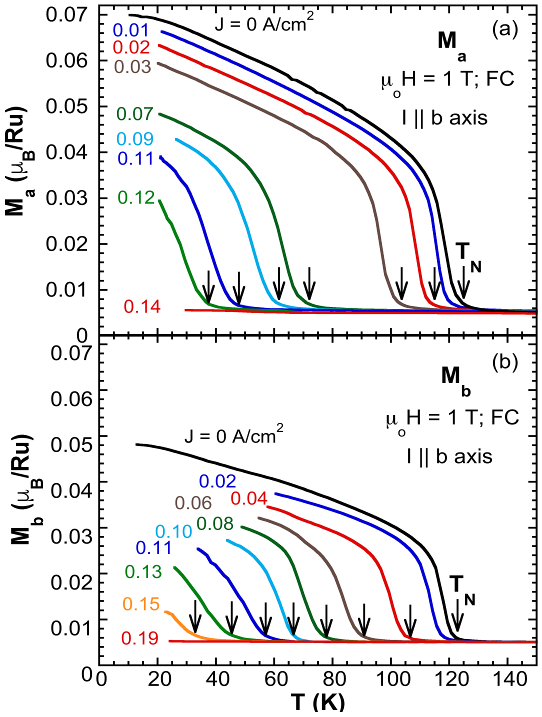




*H. D. Zhao et al PRB Rapid, 2019*

example 2 – $Ca_2RuO_4$

# Current suppresses the antiferromagnetic order and insulating state

Simultaneously measured magnetization and resistivity

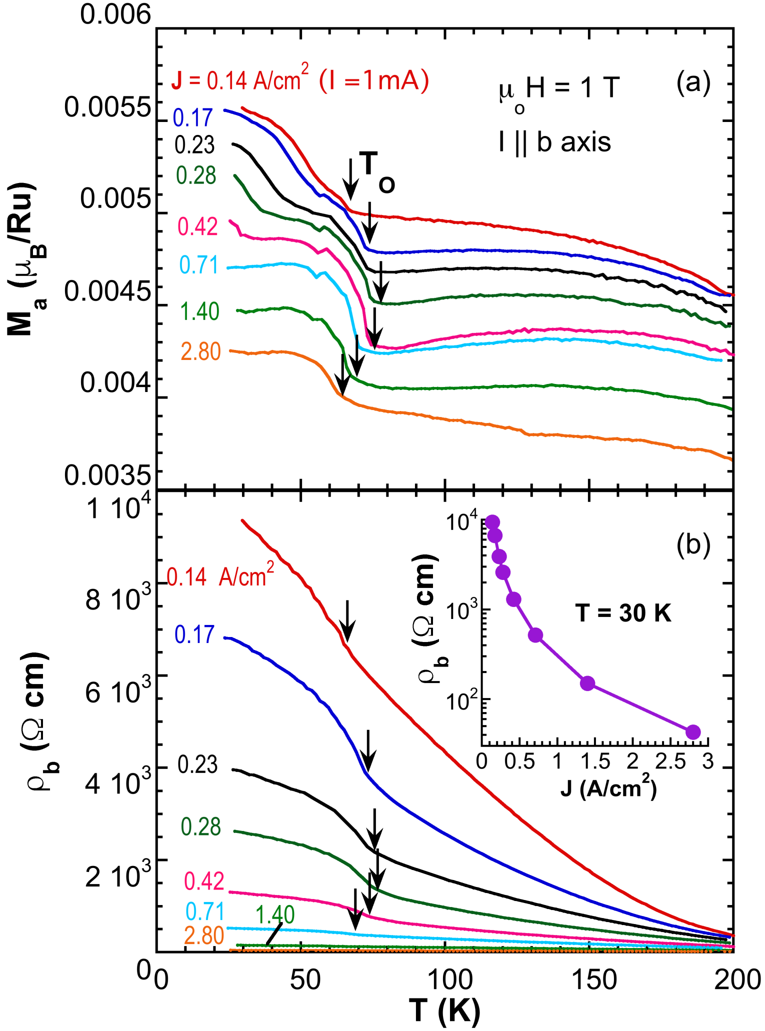


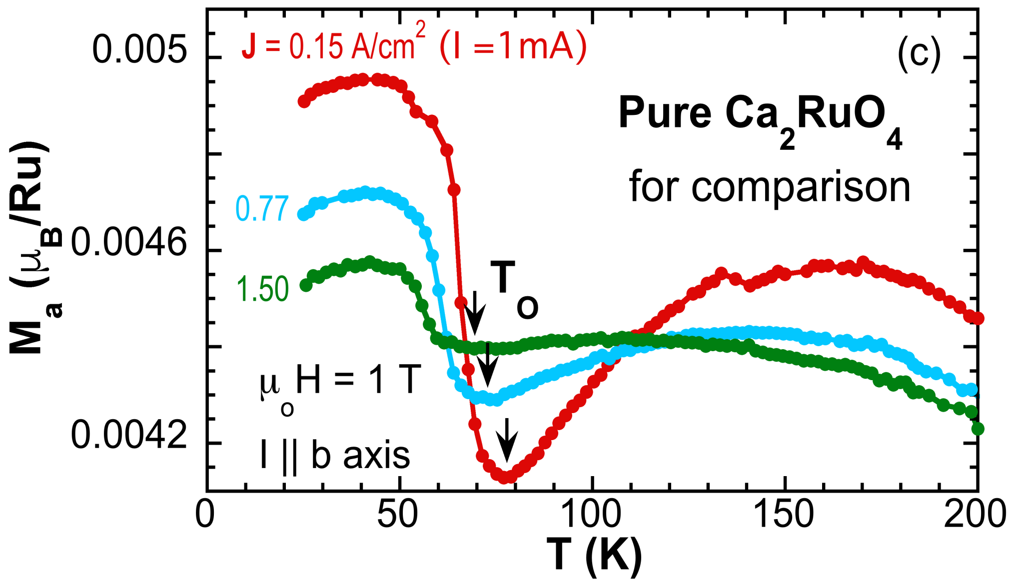

*H. D. Zhao et al PRB Rapid, 2019*

example 2 – $Ca_2RuO_4$

# Current induces a new orbital order below 80 K

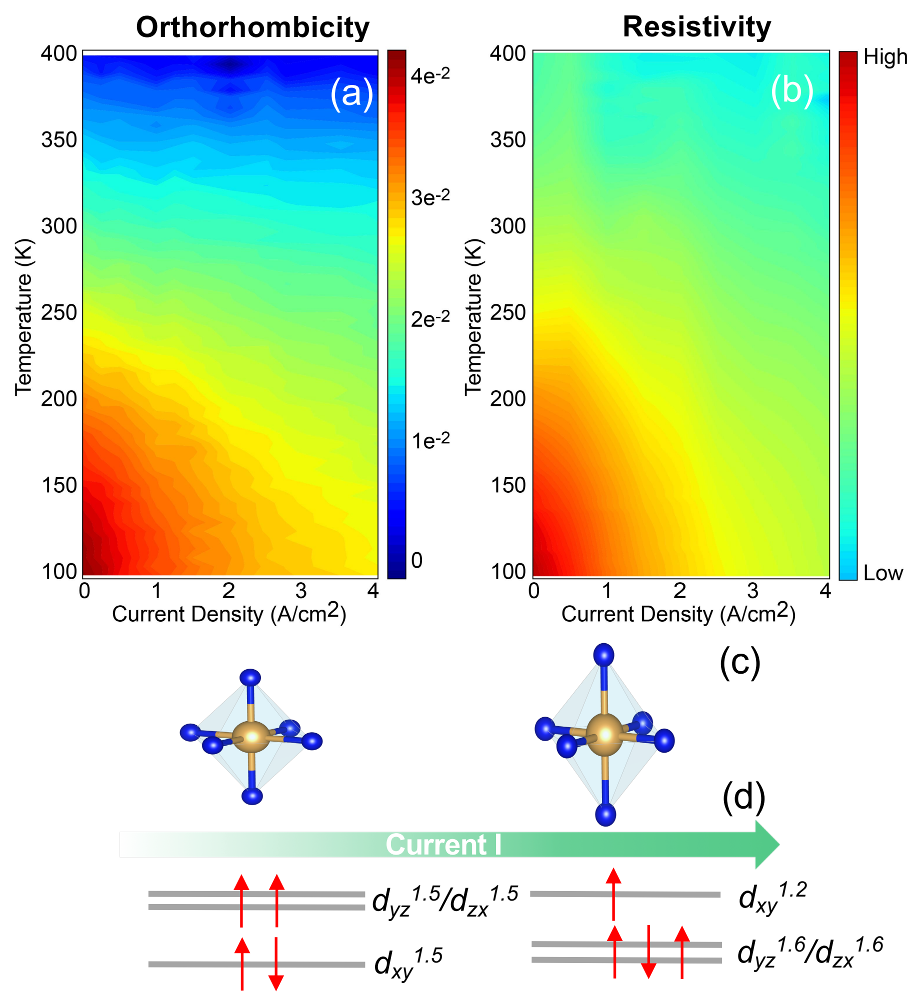
Magnetization and resistivity as a function of temperature



example 2 – $Ca_2RuO_4$

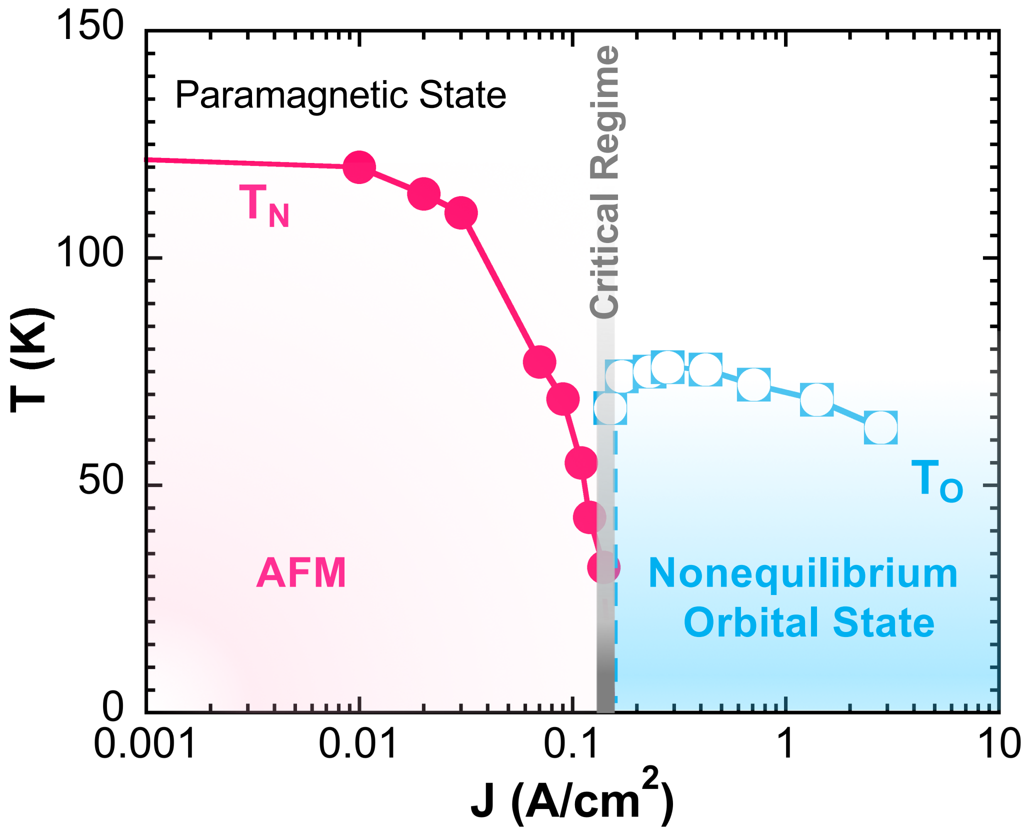
# Direct correlation between orthorhombicity and resistivity




*H. D. Zhao et al PRB Rapid, 2019*

## example 2 – $Ca_2RuO_4$

# Phase Diagram




*The applied current drives the system from the native AFM state (purple) through the critical regime near 0.15 A/cm² (gray) to the current-induced, nonequilibrium orbital state (blue).*

*H. D. Zhao et al PRB Rapid, 2019*

# Empirical Trends in Current-Controlled Materials

- Magnetic and insulating ground state, which is not enabled by large U but is driven by subtle interactions assisted by spin-orbit interactions (SOI).
- Susceptible to slight lattice changes.
- Distorted crystal structures/canted moments, which allow current to tune rotations/tilts via a strong magnetoelastic coupling.
- Current-controlled phenomena are seen a range of high-Z materials *where the role of SOI is significant and electron orbitals are extended* – SOI lock magnetic moments to $MO_6$-octahedra and the extended orbitals facilitate a strong coupling of current and electron orbitals.

## Challenges

- *How can we adequately describe the coupling of current and the lattice/orbitals*?
- *How can we better understand nonequilibrium states?*
- *How can we adequately describe current-controlled changes in band structures responsible for non-Ohmic I-V characteristics?*
- *……..*

# Collaborators

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*Florida State University*

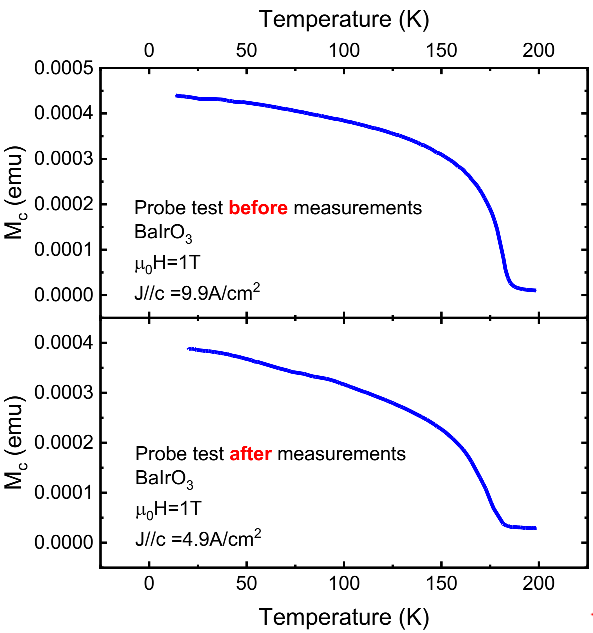
**Jasminka Terzic**
*National High Magnetic Field Laboratory*

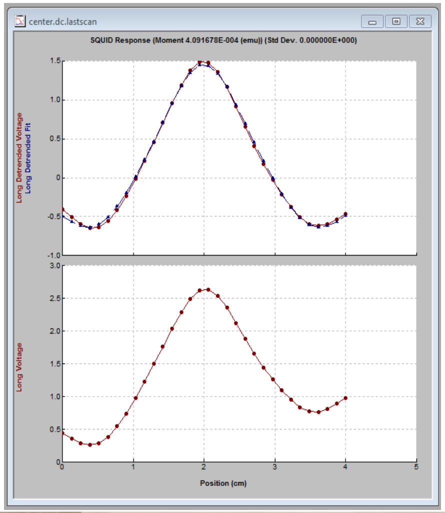
**Peter Riseborough**
*Temple University*

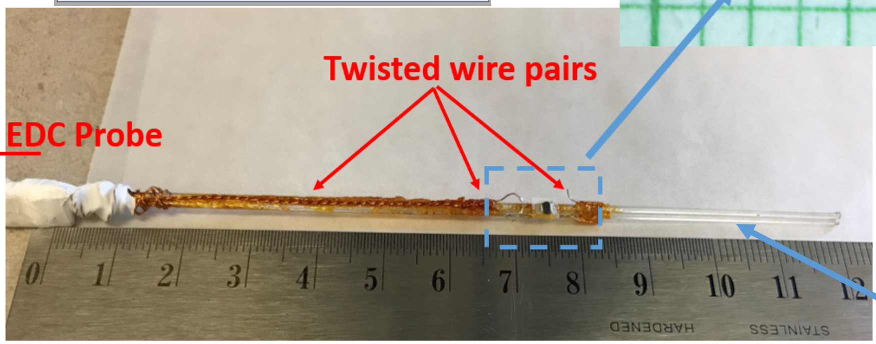
# Current suppresses the antiferromagnetic order and insulating state

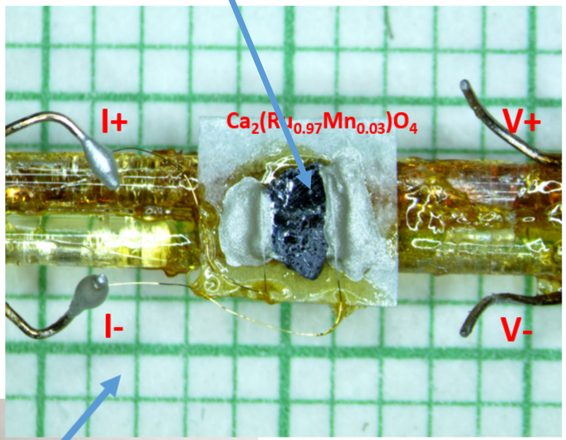
The home-made probe for simultaneous measurements of magnetization and resistivity

## $BaIrO_3$



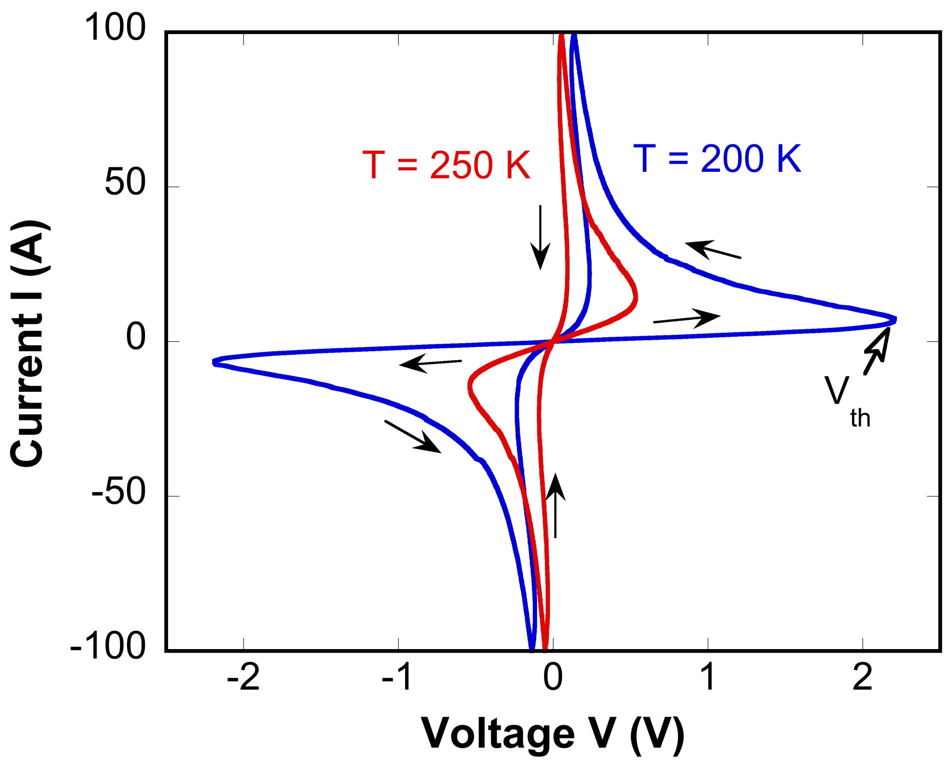




Quartz rode

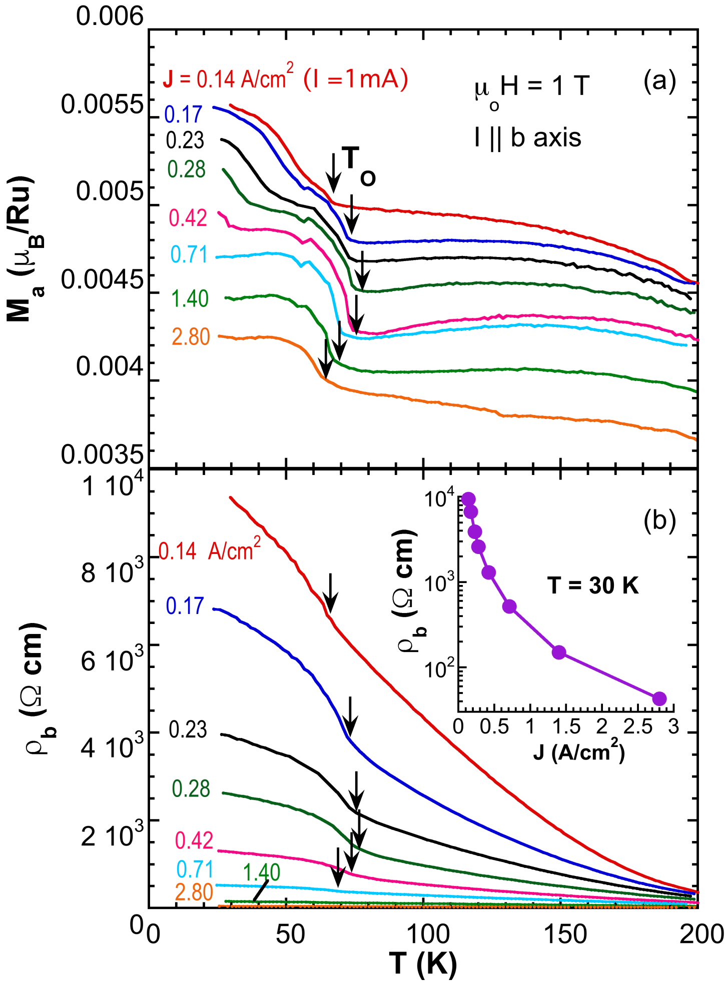
# I-V characteristics for $Ca_2Ru_{0.97}Mn_{0.03}O_4$ at T = 200 and 250 K

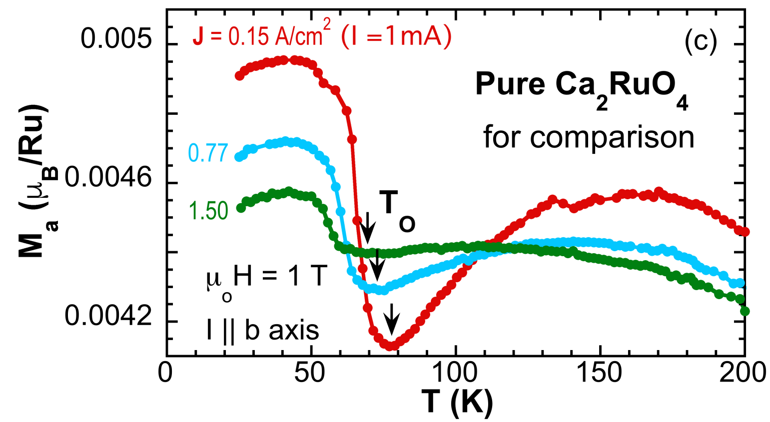


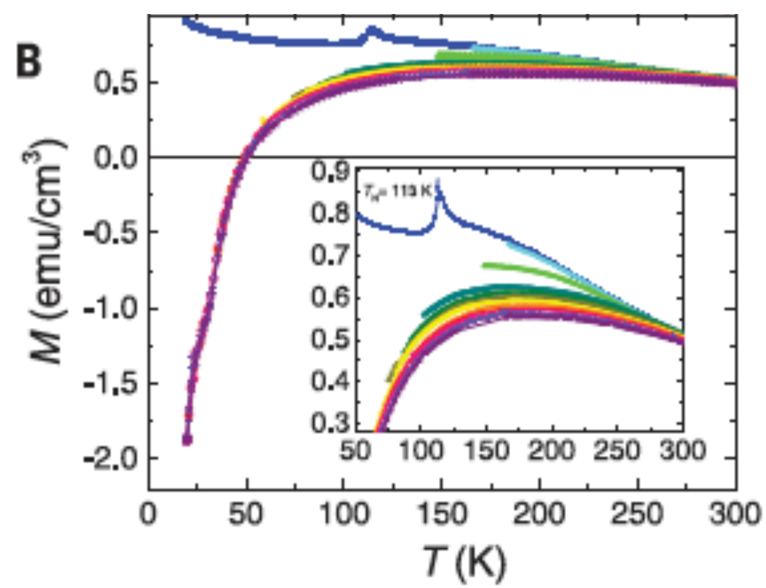
current-control of quantum states

# Current induces a new orbital order below 80 K

## Magnetization and resistivity as a function of temperature




*C. Sow…Y. Maeno et al Science 358, 1084 (2017)*

# Current induces a glassy state above 80 K

Magnetization and resistivity as a function of temperature