Novel Quantum Phenomena in Layered Ruthenates

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Collaborators

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- W. Bao (Los Alamos) and J.W. Lynn (NIST)
- Z. Islam (Argonne)
- L. Spinu (University of New Orleans)

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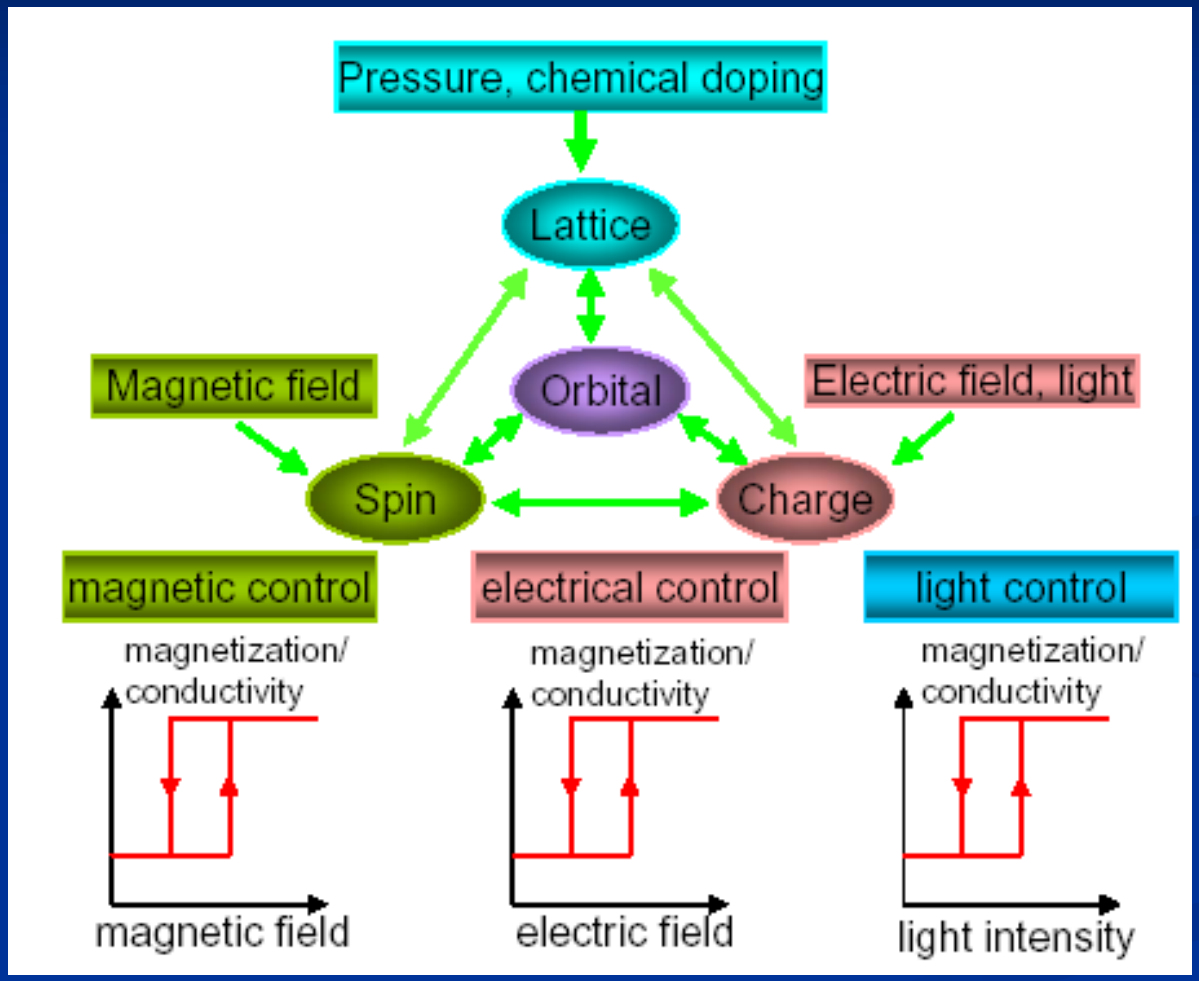
Outline

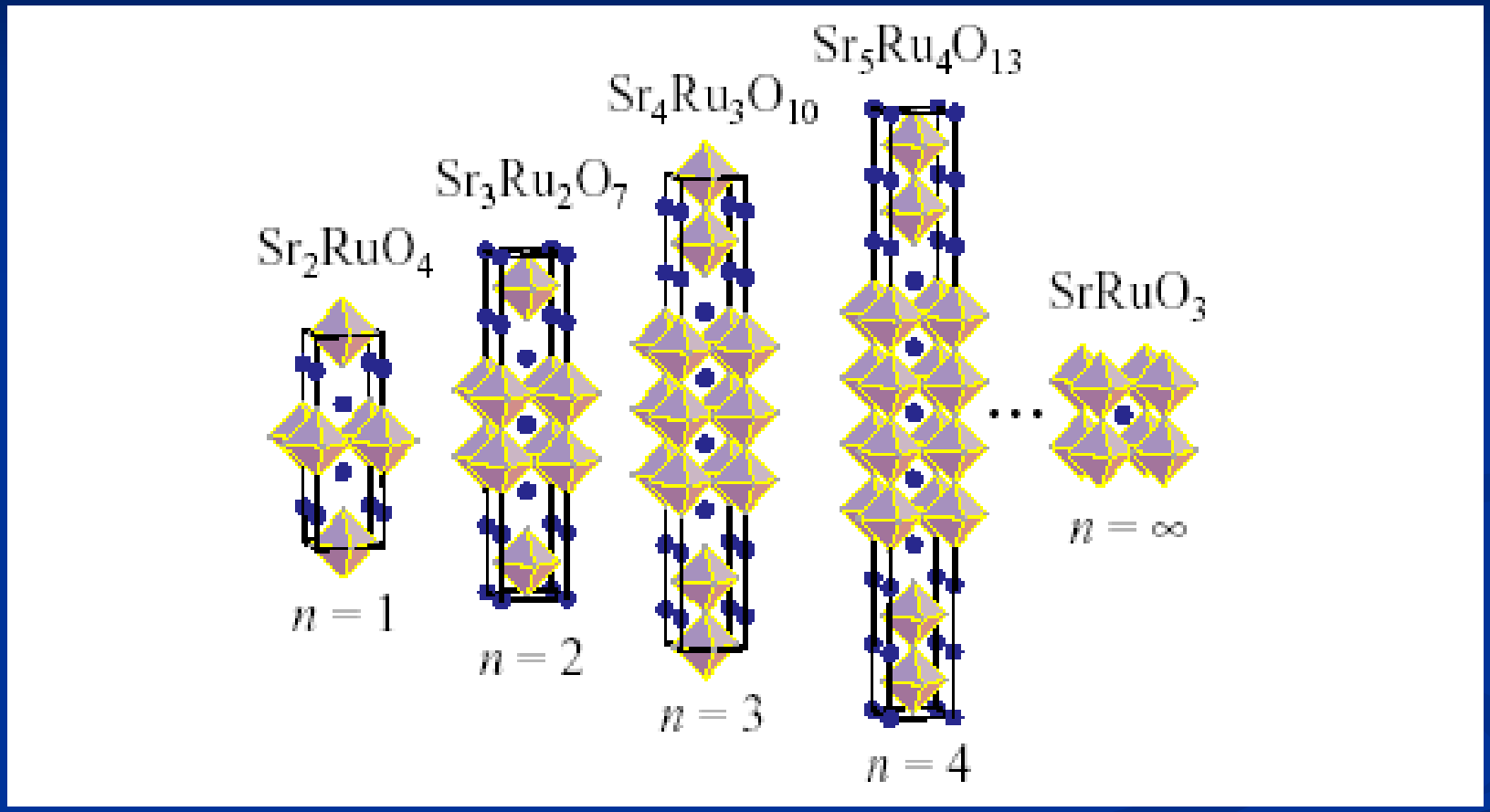
- Brief overview of ruthenate physics
- Novel quantum phenomena in double-layered ruthenates
  - **Bulk spin-valve effect in $Ca_3Ru_2O_7$**
  - **Complex electronic and magnetic states in $(Sr_{1-x}Ca_x)_3Ru_2O_7$**
- Conclusion

Strongly correlated electron systems



- Coexistence of physical interactions of comparable strength between spin, charge, lattice, and orbital degrees of freedom
- Competing ground states, tunable by external parameters.
- Phase transitions tuned by external parameters usually accompanied with switching behaviors in conductivity or magnetization.

Ruddlesden-Popper series $(Sr,Ca)_{n+1}Ru_nO_{3n+1}$



S.N. Ruddlesden and P. Popper, Acta Crys. 11, 549 (57)

Diverse Ground States of $(Sr,Ca)_{n+1}Ru_nO_{3n+1}$

Sr_2RuO_4: Spin-triplet superconductivity

A.P. Mackenzie and Y. Maeno, Rev. Mod. Phys. 75, 657 (03)

$Sr_3Ru_2O_7$: **Metamagnetic quantum critical end point**

S.A. Grigera *et al.*, Science 294, 329 (2001).

$SrRuO_3$: itinerant ferromagnetic state

P.B. Allen *et al.*, Phys. Rev. B 53, 4393 (1996)

Ca_2RuO_4: AFM Mott insulator/orbital ordering

S. Nakatsuji and Y. Maeno, Phys. Rev. Lett. 84, 2666 (00).

$Ca_3Ru_2O_7$: Giant MR / bulk spin valve

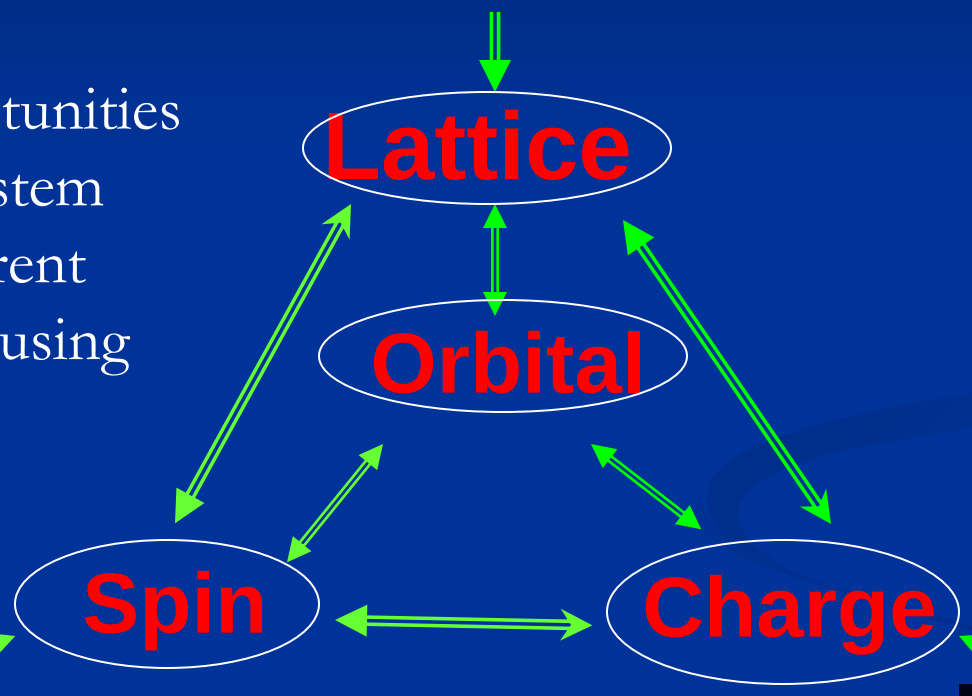
G. Cao *et al.*, Phys. Rev. B 67, 060406 (03)

D.J. Singh and S. Auluck, Phys. Rev. Lett. 96, 097203 (06)

Origin of diverse ground states

Pressure, chemical doping

Unique opportunities to tune the system between different ground states using non-thermal parameters.



Such tuning often leads to quantum criticality and/or new exotic phenomena.

Magnetic field

Electrical field

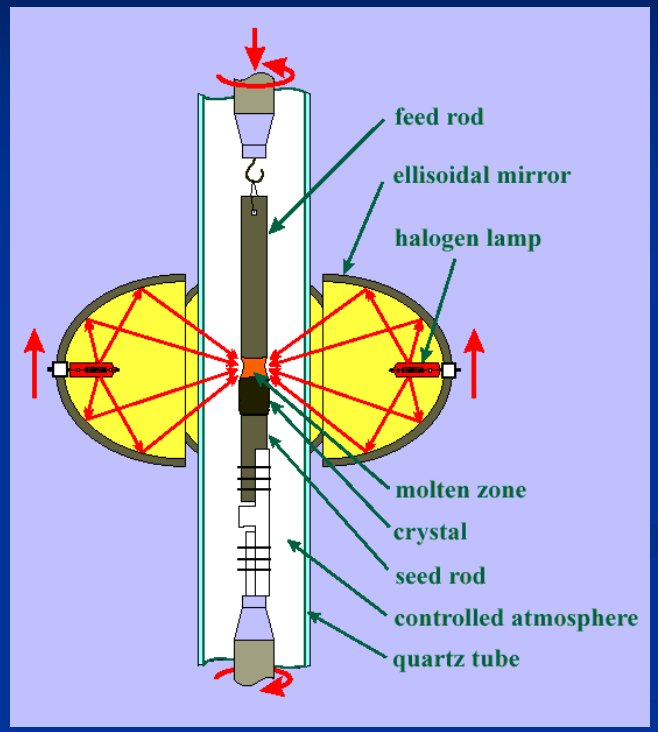
Goal: Search for novel quantum phenomena in ruthenates, investigate their underlying physics, and explore their applications.

Crystal Growth of Ruthenates using a Floating-zone method

Two distinct advantages:

(1) High-purity

Important for ruthenates, their ground state properties extremely sensitive to impurities.



(2) Large size

Useful for neutron scattering measurement

Low temperature measurements

Physical Property Measurement system (PPMS) equipped with EverCool dewar

He3 cryostat with the base temperature of 0.3 K

Resistivity, Hall effect, Magnetization, and specific heat

Research collaboration

- Neutron scattering

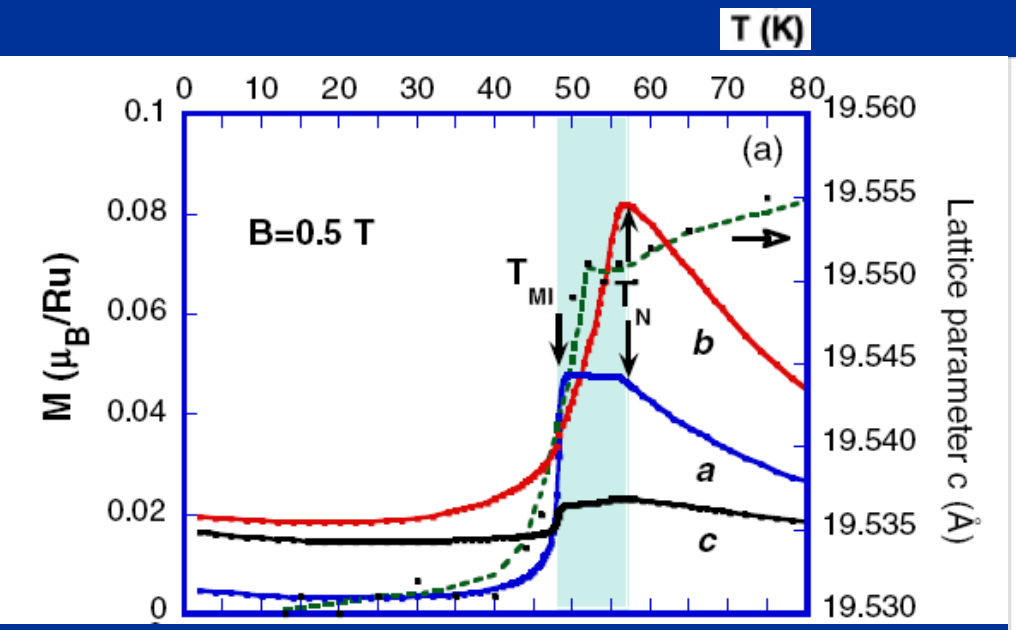
  Los Alamos (W. Bao)

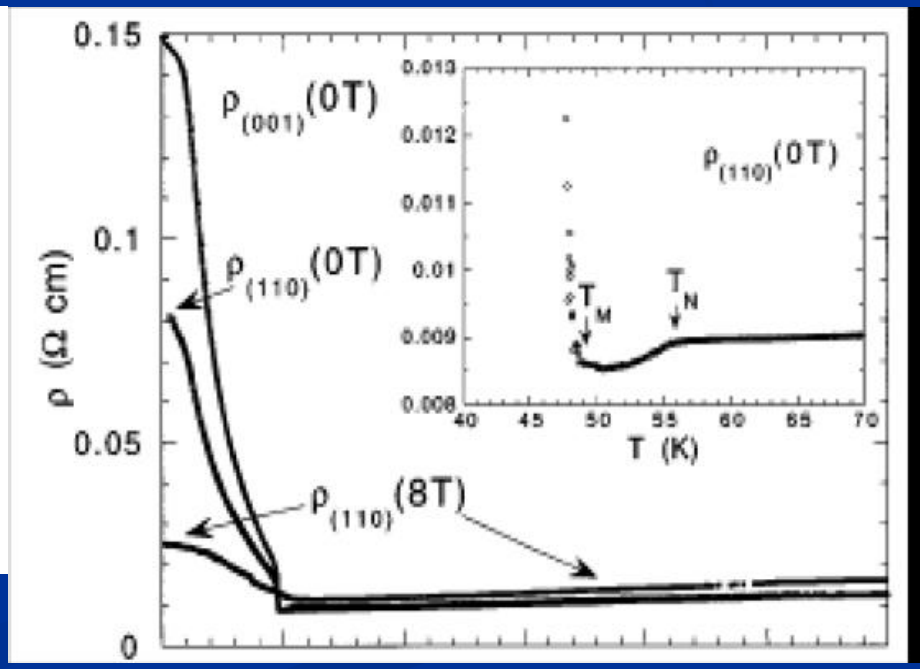
  NIST (J.W. Lynn)
- High-energy x-ray diffraction

  Argonne National Lab (Z. Islam)

Giant magnetoresistance and bulk spin valve in $Ca_3Ru_2O_7$

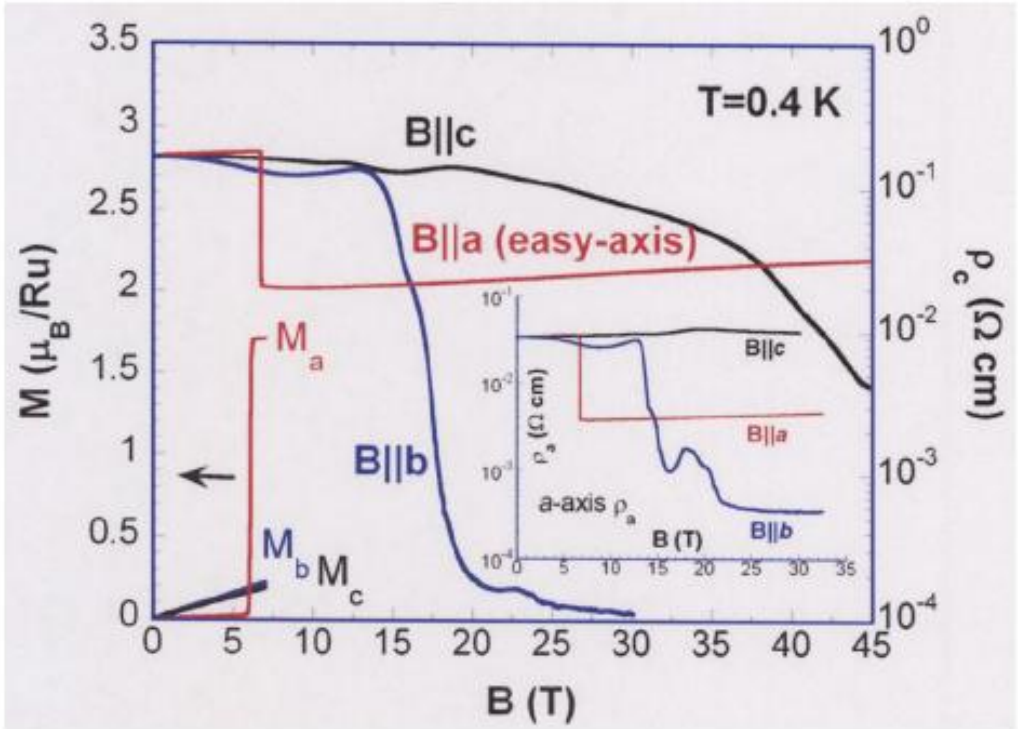
Antiferromagnetic transition at 56 K
Mott-Insulator transition at 48 K





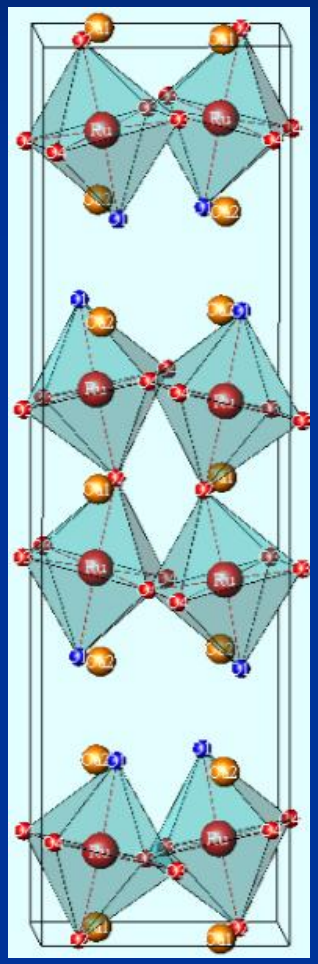
Cao *et al*. PRB 69, 014404 (2004)
Cao *et al*., PRL **78**, 1751 (1997).

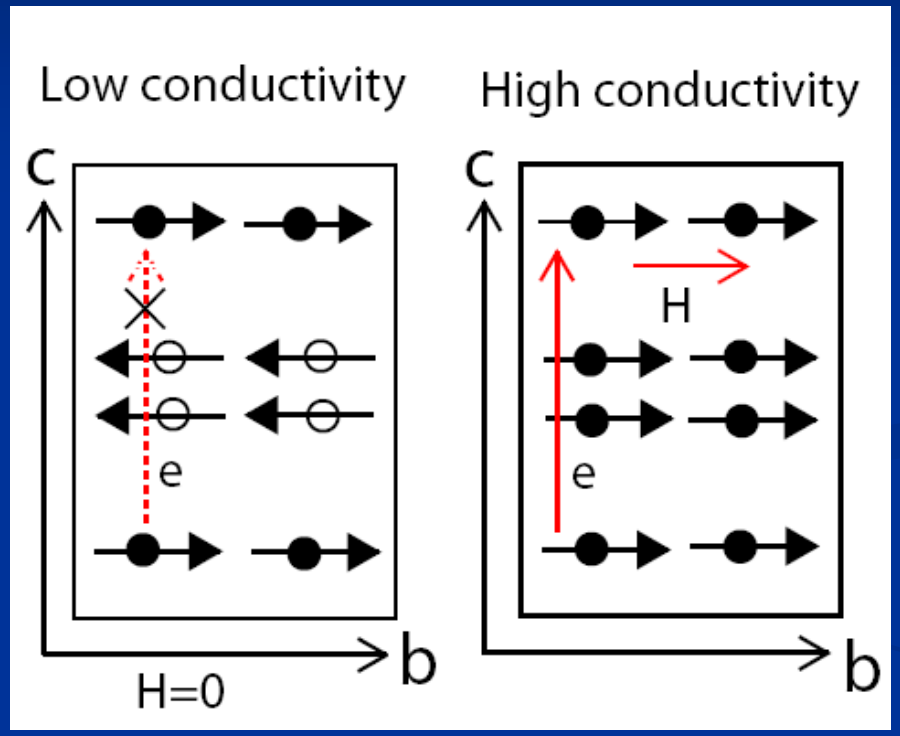
$Ca_3Ru_2O_7$: GMR for H// the easy axis
CMR for H//the hard axis



X.N. Lin *et al*. PRL **95**, 017203 (2005)

Bulk Spin Valve Bahavior ?





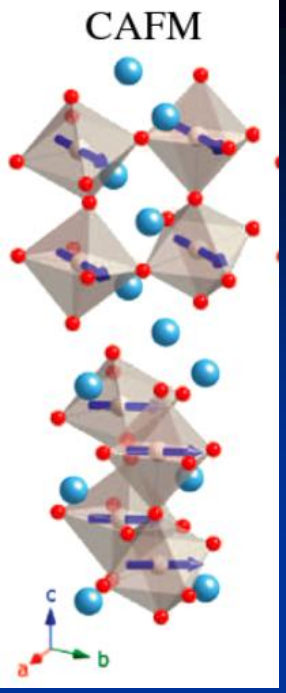
Difficulty:

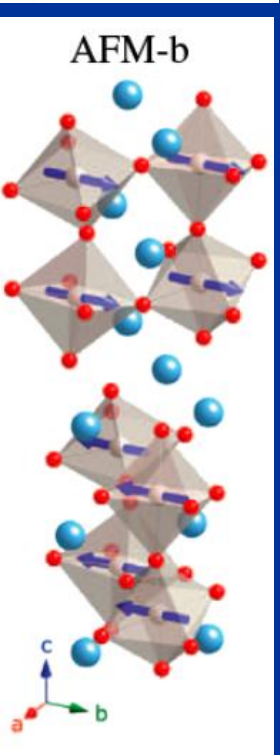
The fully spin-polarized phase seems the least favorable for electronic conduction.

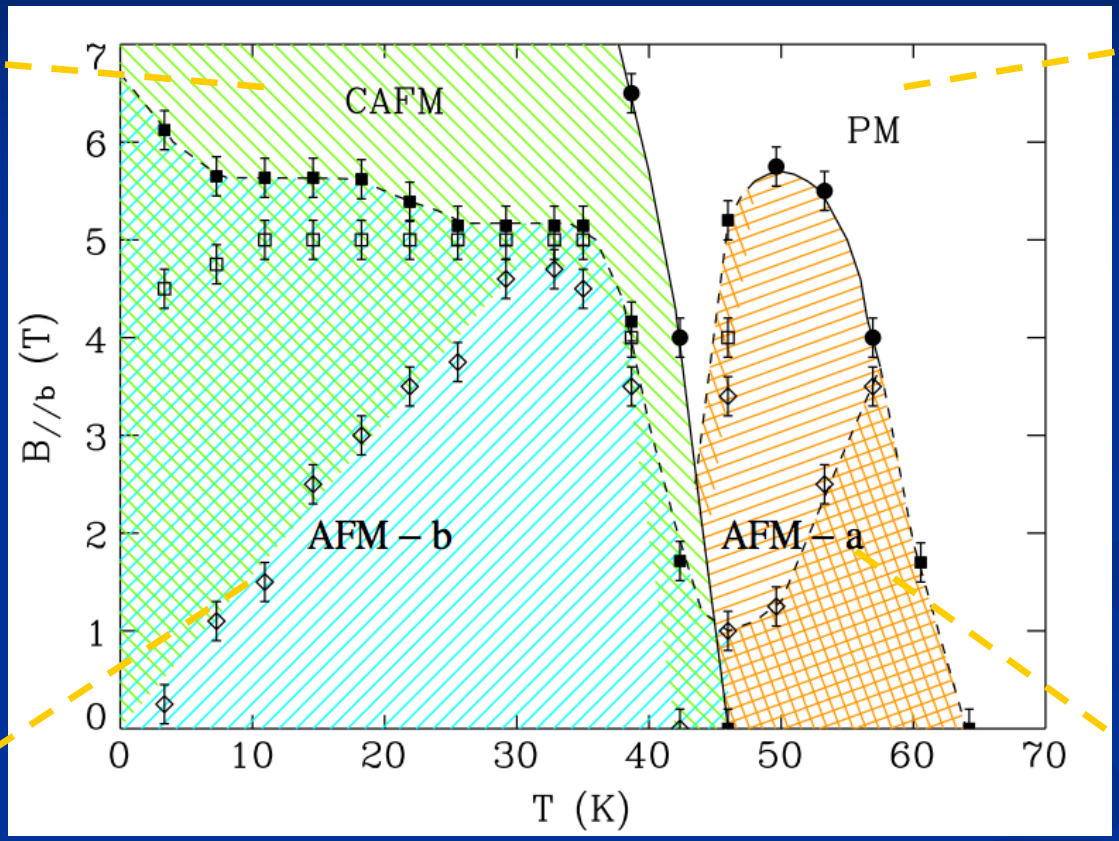
D.J. Singh *et al.* PRL 96, 097203 (2006)
Y. Yoshida *et al.*, Phys. Rev. B 72, 054412 (2005).
G. Cao *et al*. PRB 67, 060406(R) (2003)

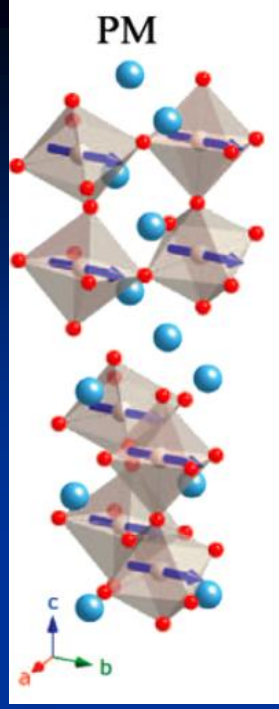
Magnetic structures of $Ca_3Ru_2O_7$

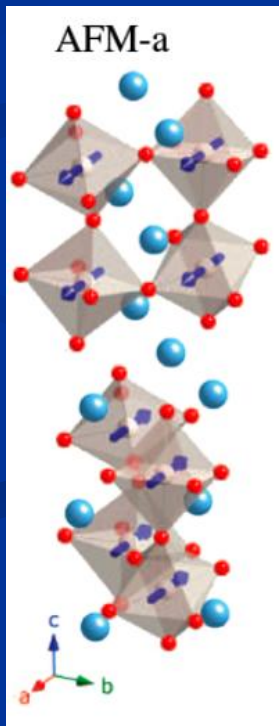
Bao, Mao, Qu and Lynn, PRL **100**, 247203 (08)







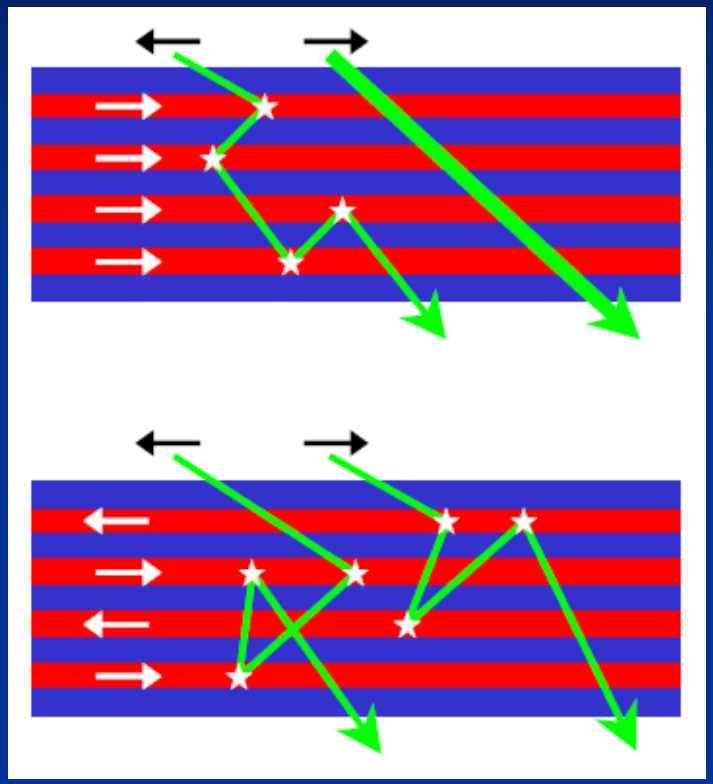




These results confirm bulk-spin-valve as the origin of GMR, and clarify the smaller MR for H//the easy axis.

Spin valve behavior in magnetic multilayer thin films (GMR effect)

Co/Cu/Co,
Fe/Cr etc.



GMR first discovered in magnetic multilayer thin films, extensively used in information technology

GMR's mechanism:
spin valve effect — the resistance shows a significant decrease when the magnetic state switches from a anti-parallel spin alignment to a parallel spin alignment.

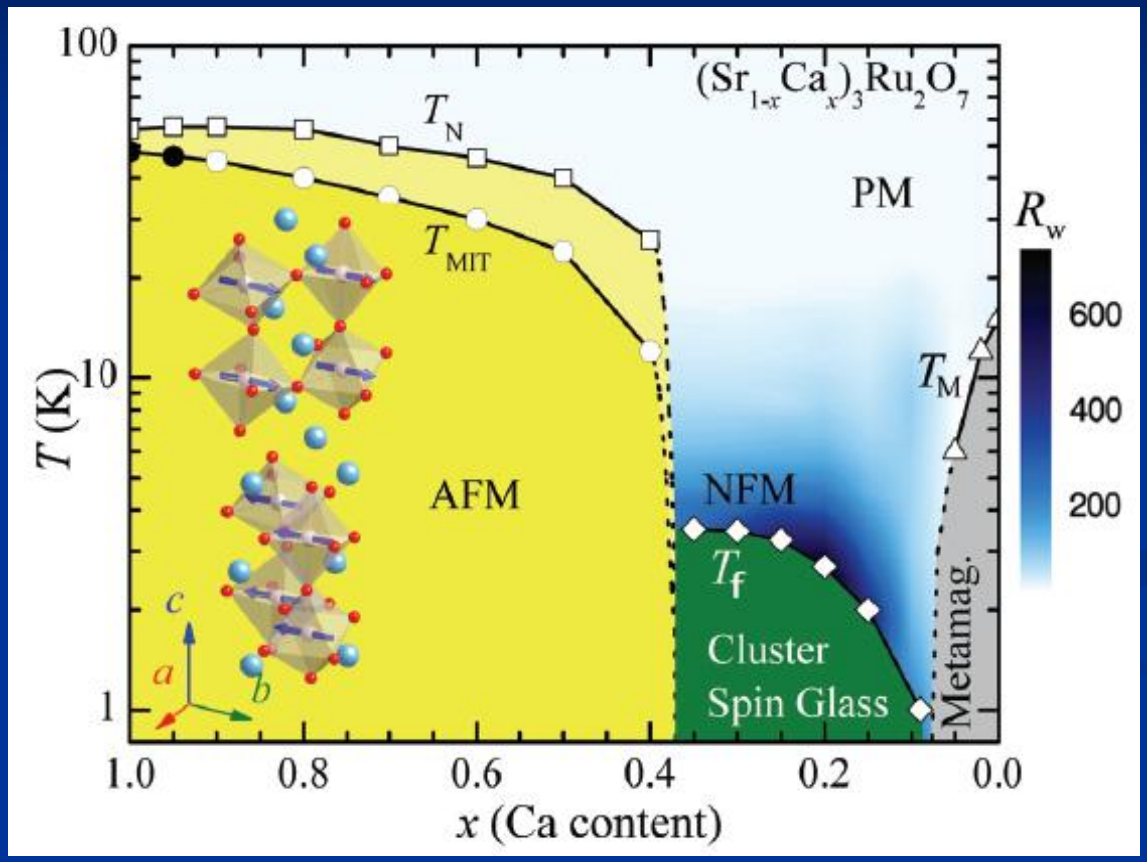
M. N. Baibich *et al.* PRL 61, 2472 (1988)
P. Grünberg *et al*. PRL 57, 2442 (1986)

Albert Fert and Peter Grunberg:

2007 Nobel Prize in physics

Complex magnetic states of $(Sr_{1-x}Ca_x)_3Ru_2O_7$

Qu *et al* Phys. Rev. B **78**, 180407 (Rap. Comm.) (2008)



- $x \geq 0.4$:
  AFM ground state
  G. Cao *et al.* PRB **56**, 5387 (97)
  S. Ikeda *et al.* PRB **57** 978 (98)

- $0.4 > x \geq 0.1$:
  Heavy-mass nearly FM state

$x < 0.1$:
Itinerant metamagnet

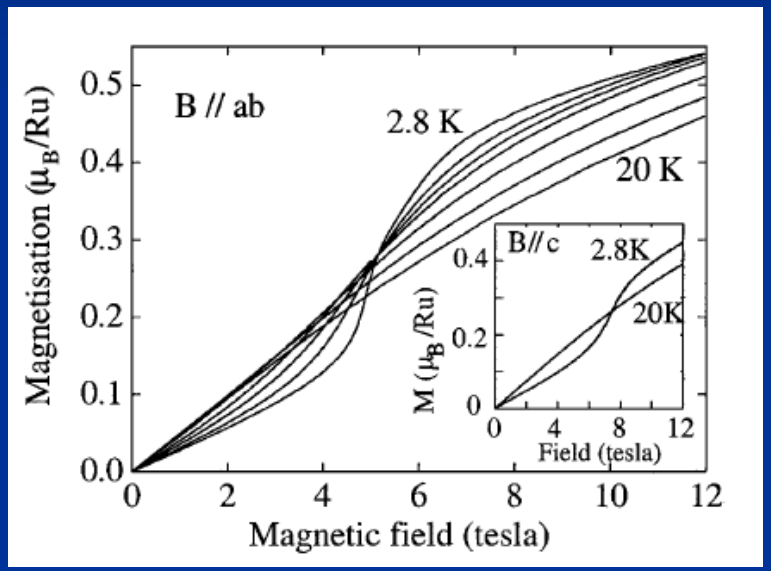
Wilson ratio: $R_W = (\pi^2 k_B^2 / 3\mu_B^2)(\chi/\gamma_e)$

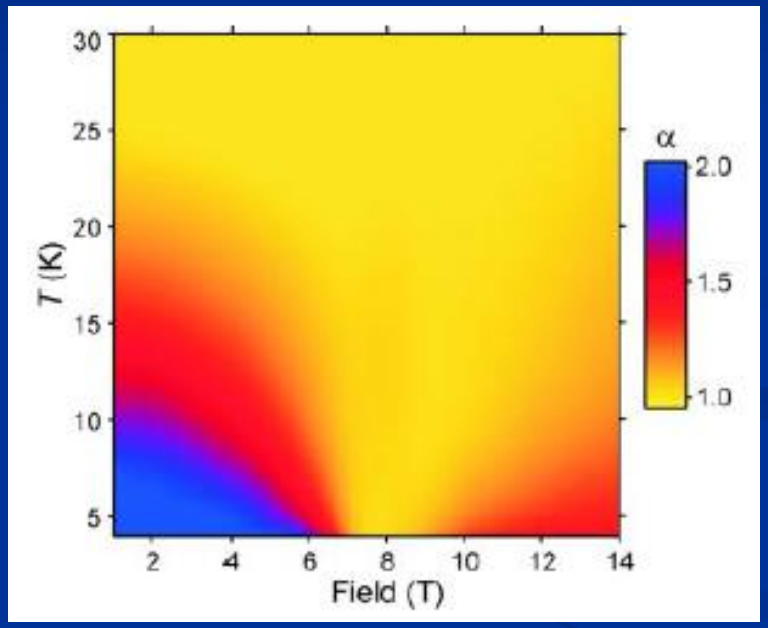
$0.4 > x \geq 0.1$: In close proximity to a 2D FM with $T_c = 0$ K

$Sr_3Ru_2O_7$

Perry *et al*. PRL **86**, 2661 (2001)

Grigera *et al*. Science **294**, 329 (01).
Borzi *et al*. Science **315**, 214 (07).

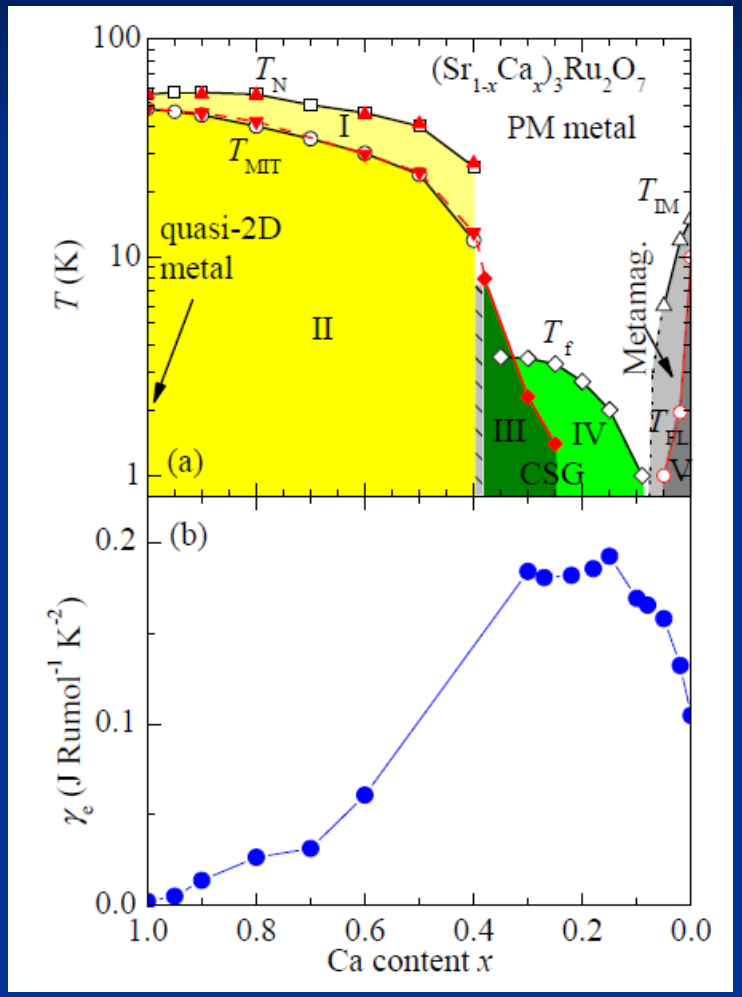




Metamagnetic transition driven by a Stoner instability

Metamagnetic quantum critical end point
Non Fermi liquid behaviors,
Nematic electron liquid phase.

Complex electronic states in $(Sr_{1-x}Ca_x)_3Ru_2O_7$



(Qu, Mao, et al., submitted to PRB)

$Ca_3Ru_2O_7$: quasi-2D metal
(Yoshida et al., PRB **69**, 220411 (04))

I: AFM metal

II: Anderson localized state

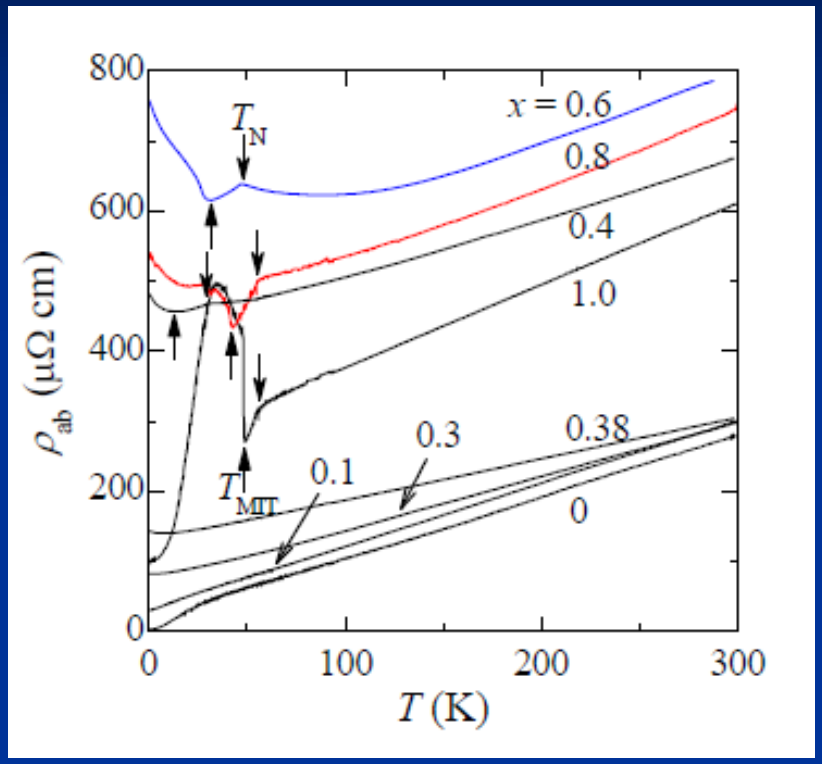
III: Weakly localized state induced by magnetic scattering

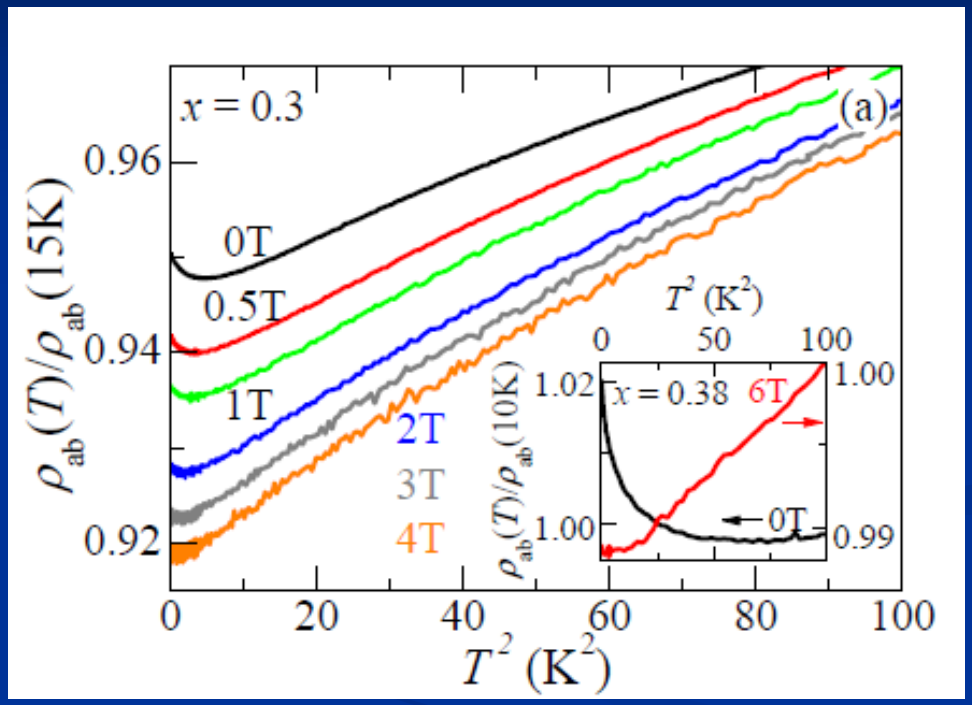
IV: Magnetic metallic state with $\rho \propto T^n$, $2<n<3$.

V: Fermi liquid (FL)

Non FL behaviors near $x \sim 0.08$

In-plane resistivity of $(Sr_{1-x}Ca_x)_3Ru_2O_7$





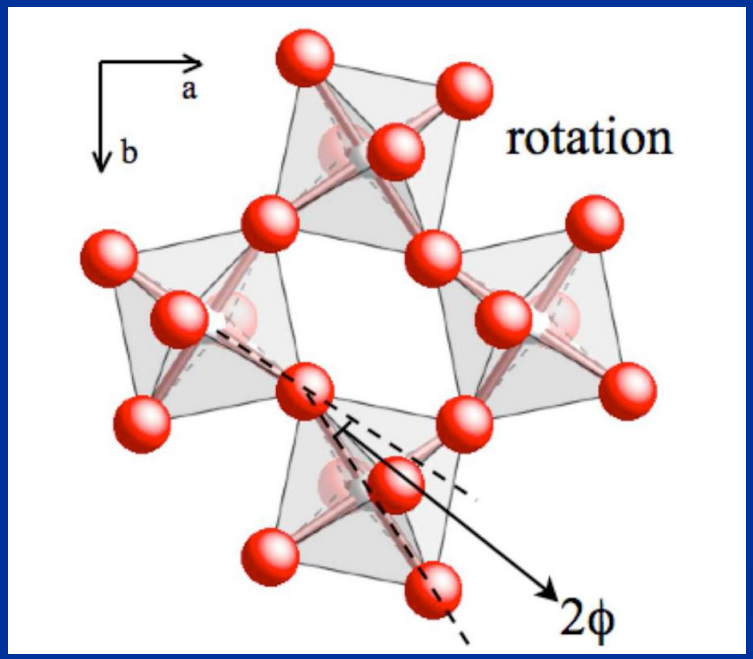
- Anomalies in resistivity at T_N and T_{MIT} for $x \geq 0.4$

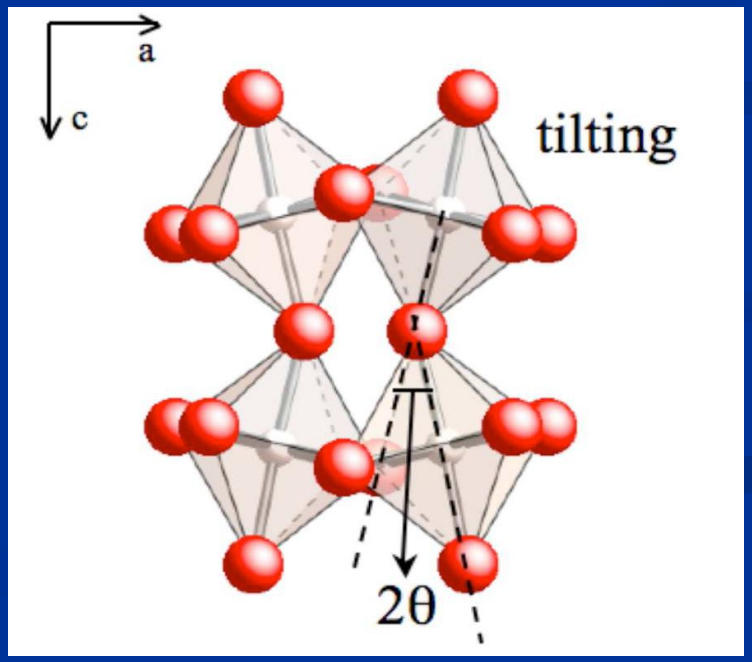
- Weak localization induced by magnetic scattering for $0.25 \leq x < 0.4$

Structural distortion in $(Sr_{1-x}Ca_x)_3Ru_2O_7$

- Ca substitution for Sr enhances structural distortion.

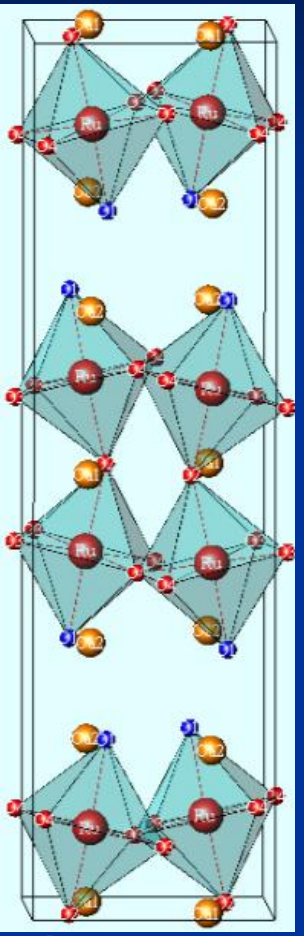
- The structural distortion occurs primarily via RuO_6 octahedral rotation and tilting





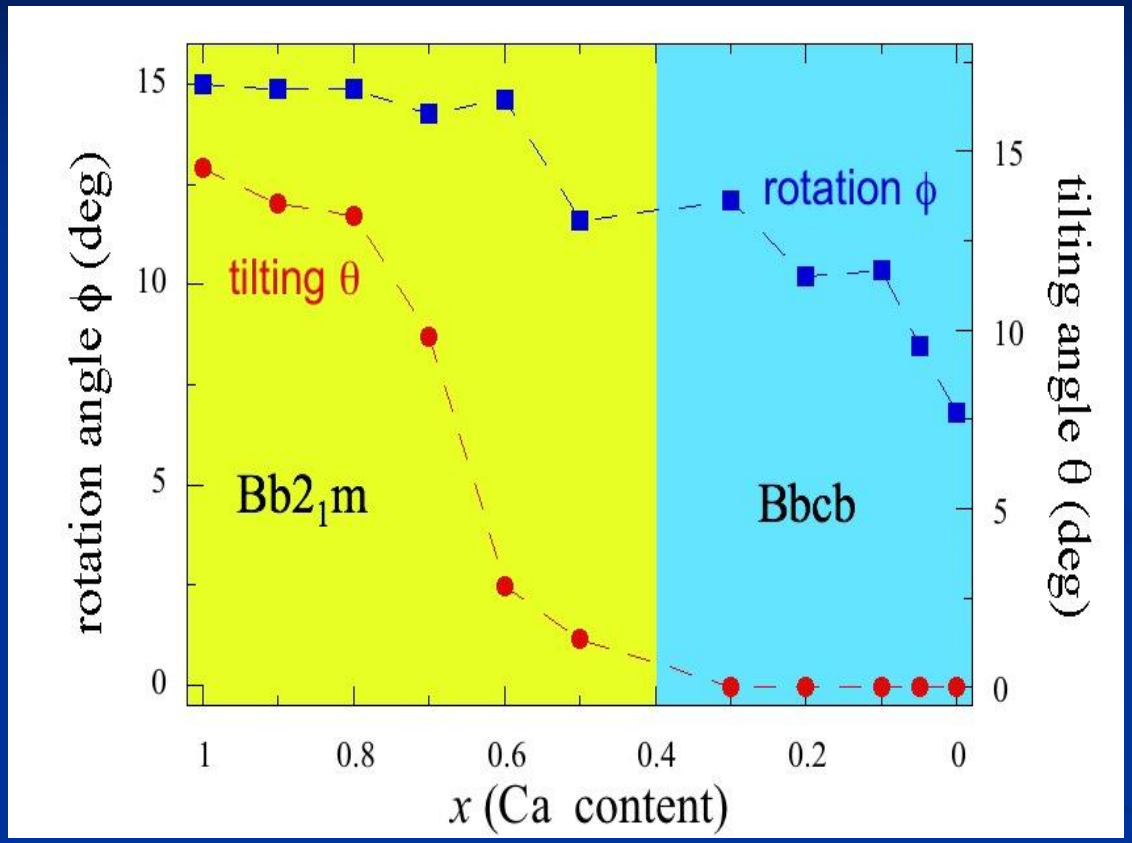
Structural distortion in $(Sr_{1-x}Ca_x)_3Ru_2O_7$

$Ca_3Ru_2O_7$



$Bb2_1m$

Yoshida *et al*. PRB 72, 054412

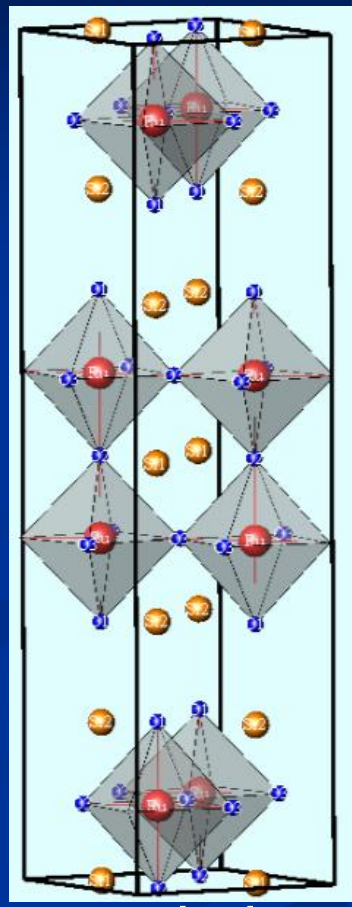


J. Peng, B. Qian and Z.Q.Mao *et al*. unpublished (Tulane)

Rotation: driving FM instability

Tilting: mediating the AFM coupling between layers.

$Sr_3Ru_2O_7$



Bbcb

Shaked and Jorgensen *et al.* J. Sol. Stat. Chem.154, 361-367

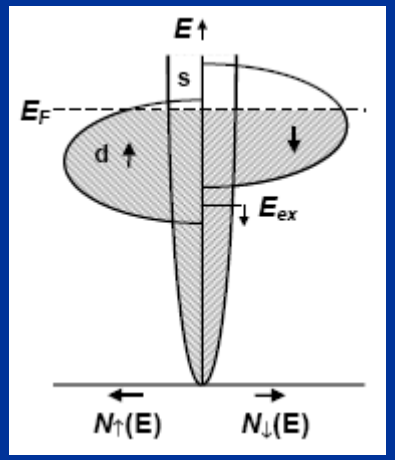
Conclusion

- Confirmed bulk-spin-valve as the origin of GMR in $Ca_3Ru_2O_7$.
- Established the phase diagram for $(Sr_{1-x}Ca_x)_3Ru_2O_7$ and discovered an unusual magnetic state in close proximity to 2D ferromagnetism.
- Demonstrated the strong interplay between charge, spin, and lattice degrees of freedom in double layered ruthenates.

Metamagnetism

- Empirically, a superlinear rise in magnetization at a specific field.

Itinerant metamagnetism:



- Driven by a Stoner instability
- Exchange interaction splits the DOS of spin-up and spin-down bands at the critical field
- Results in an imbalance of one spin species (IE, a spontaneous magnetic moment)

Stable if Stoner criterion satisfied:

$$I\,\mathrm{N}(E_{\mathrm{F}}) \geq 1$$