Synchrotron X-ray Studies of Magnetic Order

ARIMA Taka-hisa

Inst. Multidisciplinary Res. Adv. Mat., Tohoku Univ.

& RIKEN SPring-8 Center

X-ray Studies of Magnetic Order: OVERVIEW

- •Resonance: Element-specific Investigation
- •LS Separation
- •Small Focus
- •High Q-resolution
- •No Killer Element

Complementary to Neutron Scattering

- •Magnetic Circular Dichroism in Ferromagnetic Matter
- •Magnetic Diffraction in Magnetic Matter
  - ✓Spectroscopy: Wave Function
  - ✓Polarization Analysis: LS Separation
  - ✓Circular Polarization: Spin Chirality
- •Superlattice Modulation in Frustrated Magnet
- •Nonreciprocal Directional Dichroism & Scattering:
  Magnetism at Non-centrosymmetric Sites

PEEM, Magnetic Compton, Auger, Moessbauer, …

1^{st} Topic

Magnetic Structure and 5d Orbital State in Sr_2IrO_4

Collaborators

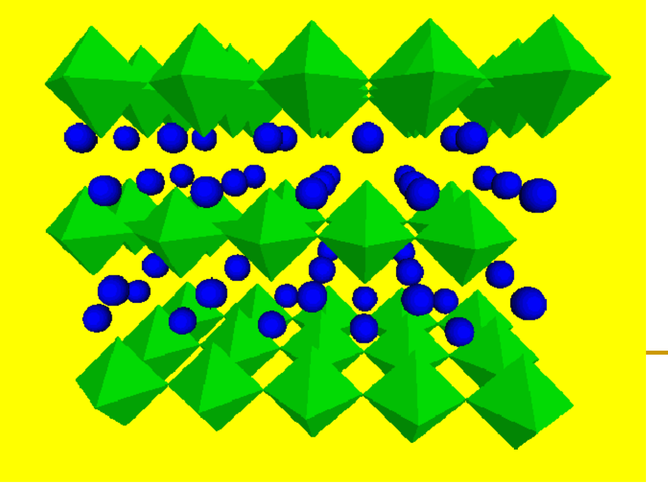
B. J. Kim, S. Fujiyama, J. Matsuno,

H. Takagi (Univ. Tokyo & RIKEN ASI)

T. Komesu, S. Sakai, T. Morita,

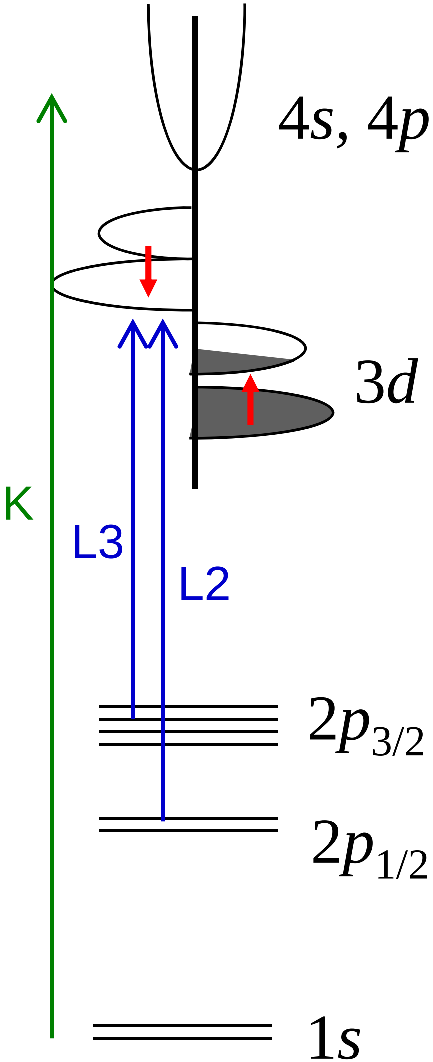
H. Ohsumi (RIKEN SP8)

S. Fujiyama

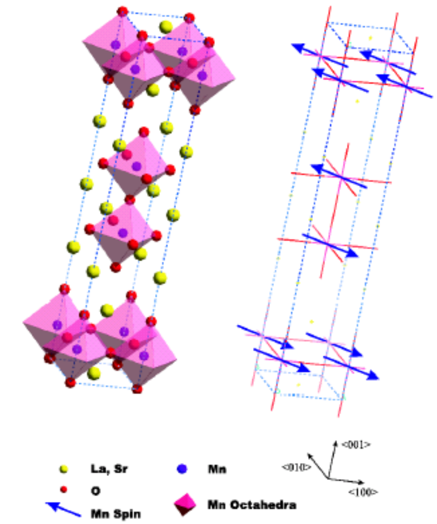


B. J. Kim *et al.*, submitted
S. Fujiyama *et al.*, unpublished

Resonant Magnetic X-ray Scattering

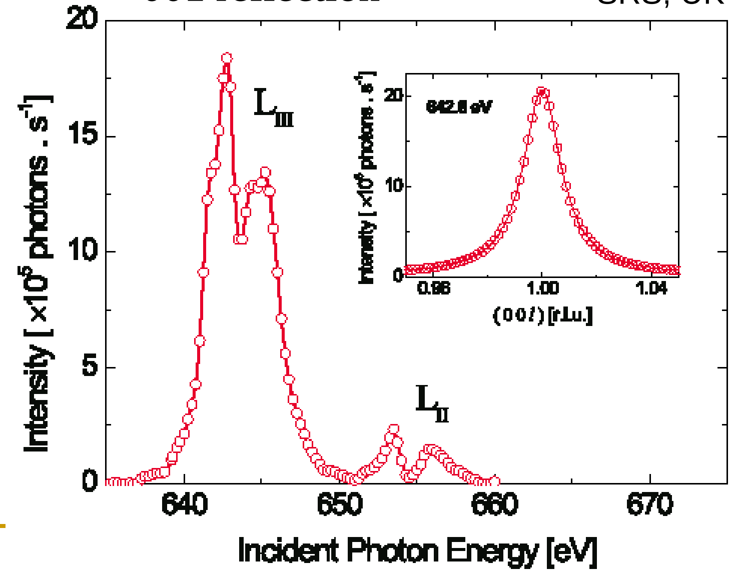


L-edge resonant x-ray scattering would provide useful information on magnetic *d* electrons.



$(La,Sr)_3Mn_2O_7$ (x=0.475)
001 reflection

SRS, UK



S.B. Wilkins *et al.*, *Phys. Rev. Lett.* **90**, 187201 (2003)

Hard x-ray is a powerful tool for 5d system

	E_{2p} [eV]	λ [A]	E_{3d}	λ [A]
3*d* TM	450~900	14~28	-	-
4*d* TM	2.2k~3.4k	3.6~5.6	-	-
Lanthanides	5.7k~8.9k	1.3~2.2	450~1500	8~28
5*d* TM	9.6k~11.9k	1.0~1.3	-	-
Actinides	15.9k~	0.8~	3.2k~3.6k	3.4~3.8

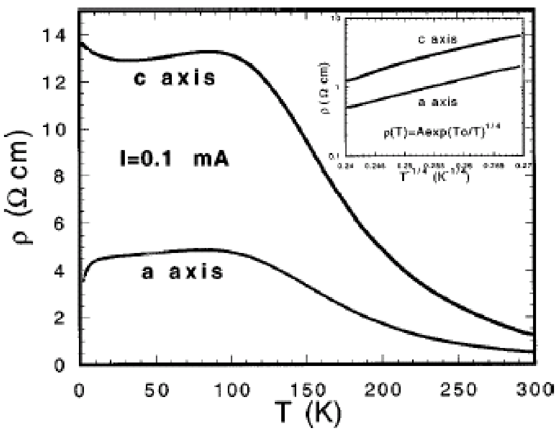
Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu
Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag
(Ln)	Hf	Ta	W	Re	Os	Ir	Pt	Au

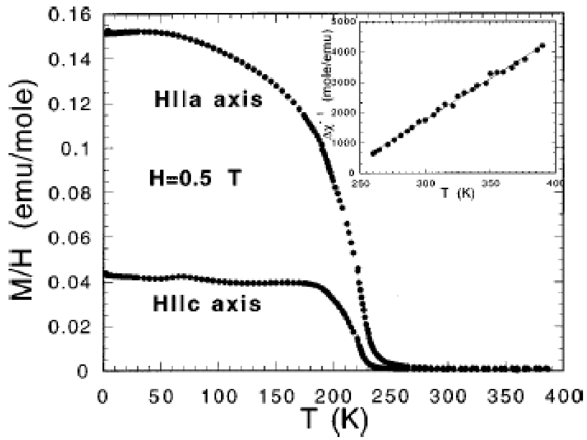
Strong LS
Strong Ligand Field
Wide Band

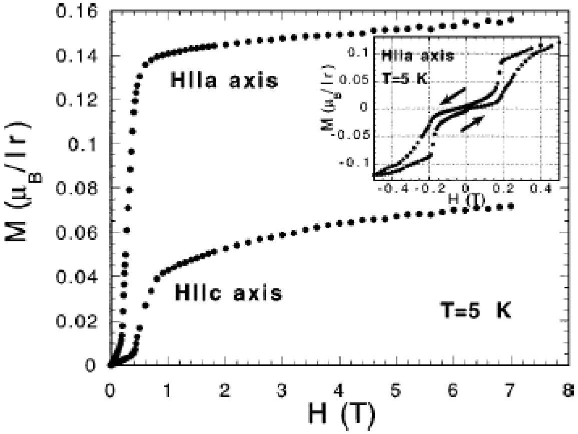
Sr_2IrO_4

Distorted K_2NiF_4-type Structure ($I4_1/acd$) $\sqrt{2}a \times \sqrt{2}a \times 2c$

Ir^{4+}: $5d^5$ Low-Spin







Semiconducting

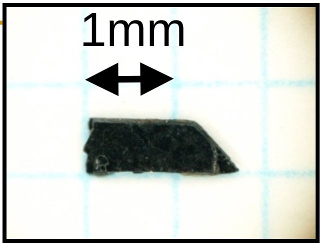
Magnetic Order below 230K

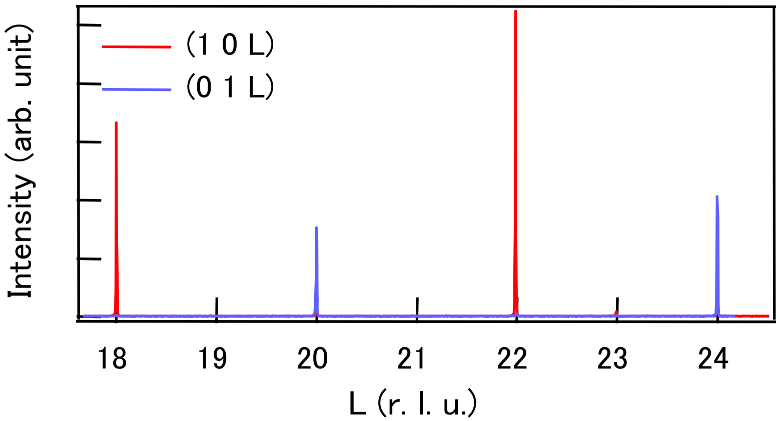
Meta-magnetism (~$0.15\mu_B$/Ir)

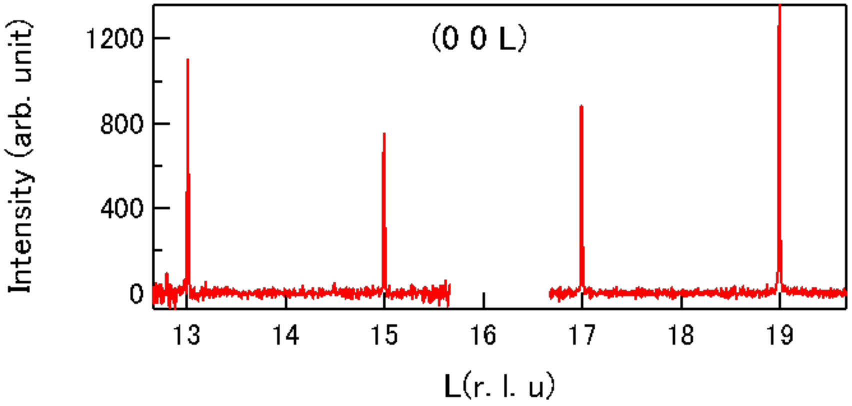
G. Cao *et al.*, *Phys. Rev. B* **57**, R11039 (1998)

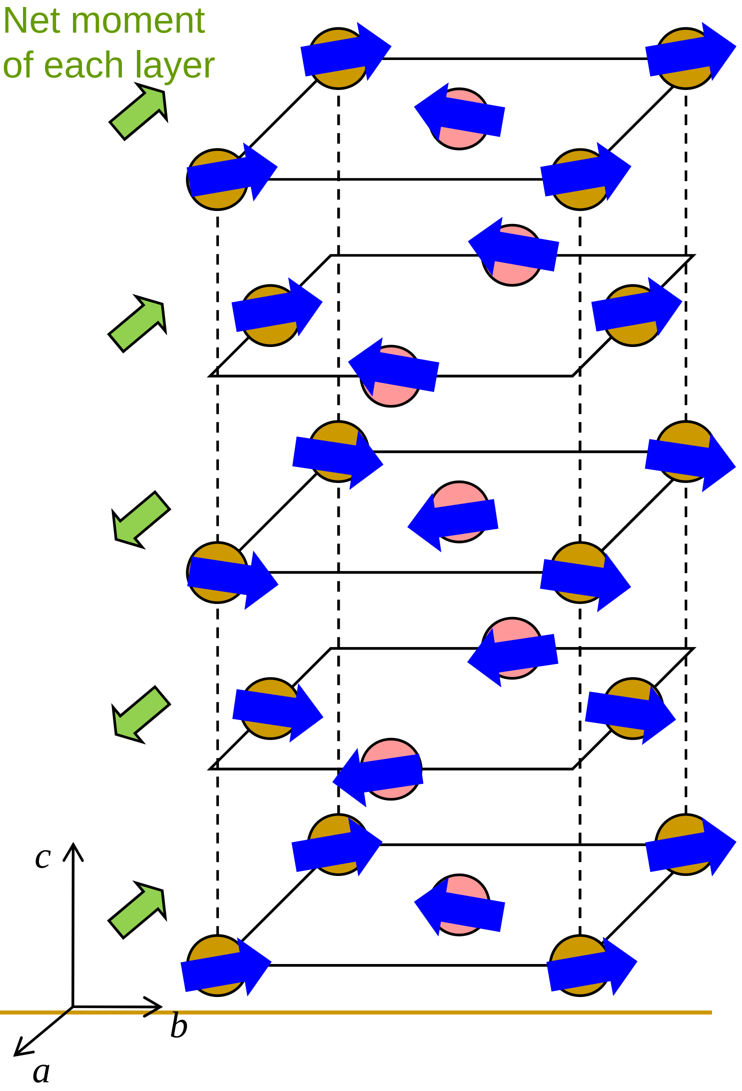
No Neutron Data

Resonant Magnetic Diffraction: Determination of Magnetic Structure





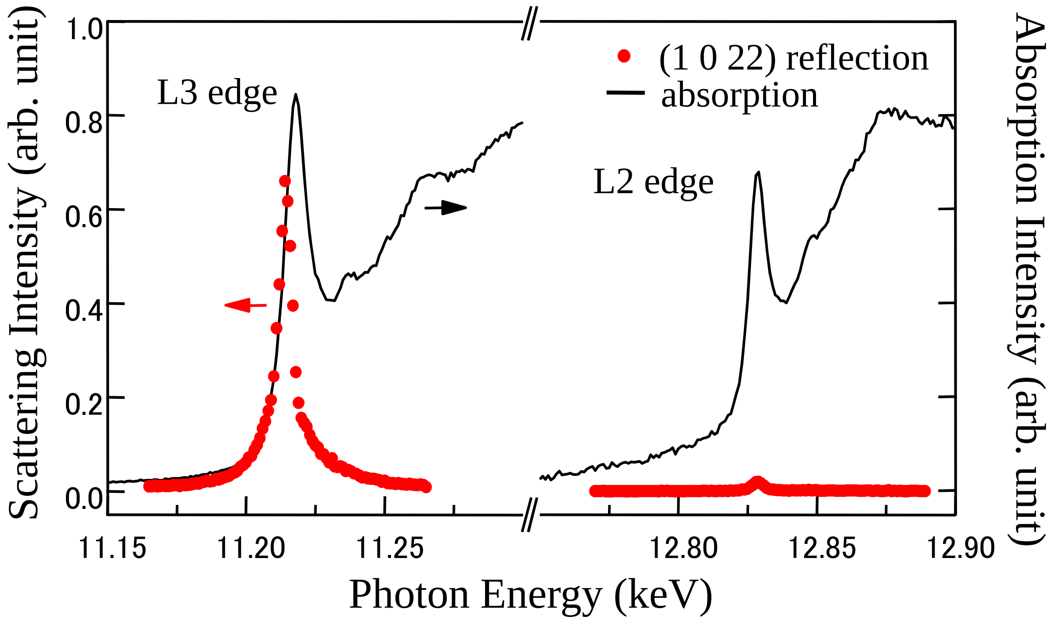


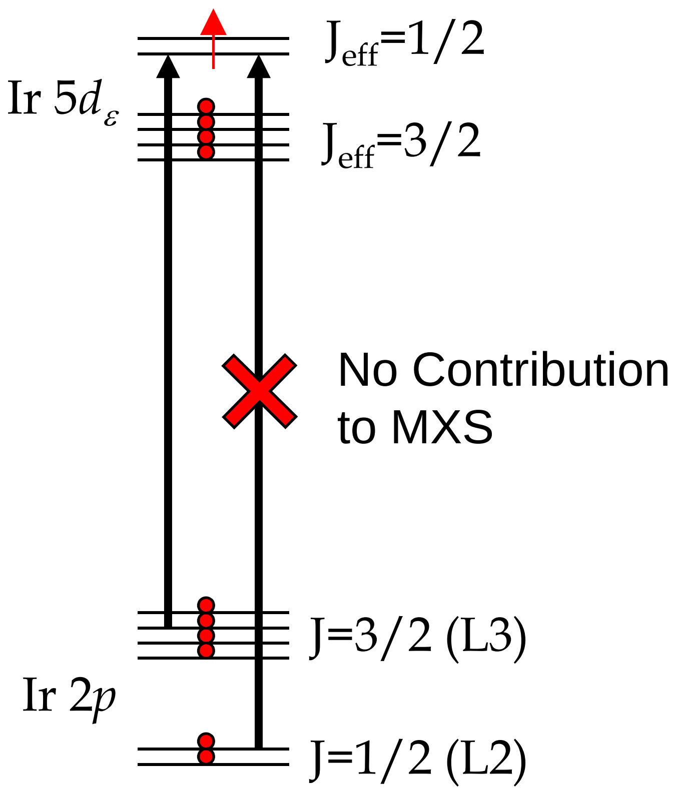


SPring-8 BL29XUL & BL19LXU

Resonant Spectrum: Wave Function

$\mathbf{J}_{\text{eff}} \equiv -\mathbf{L} + \mathbf{S}$



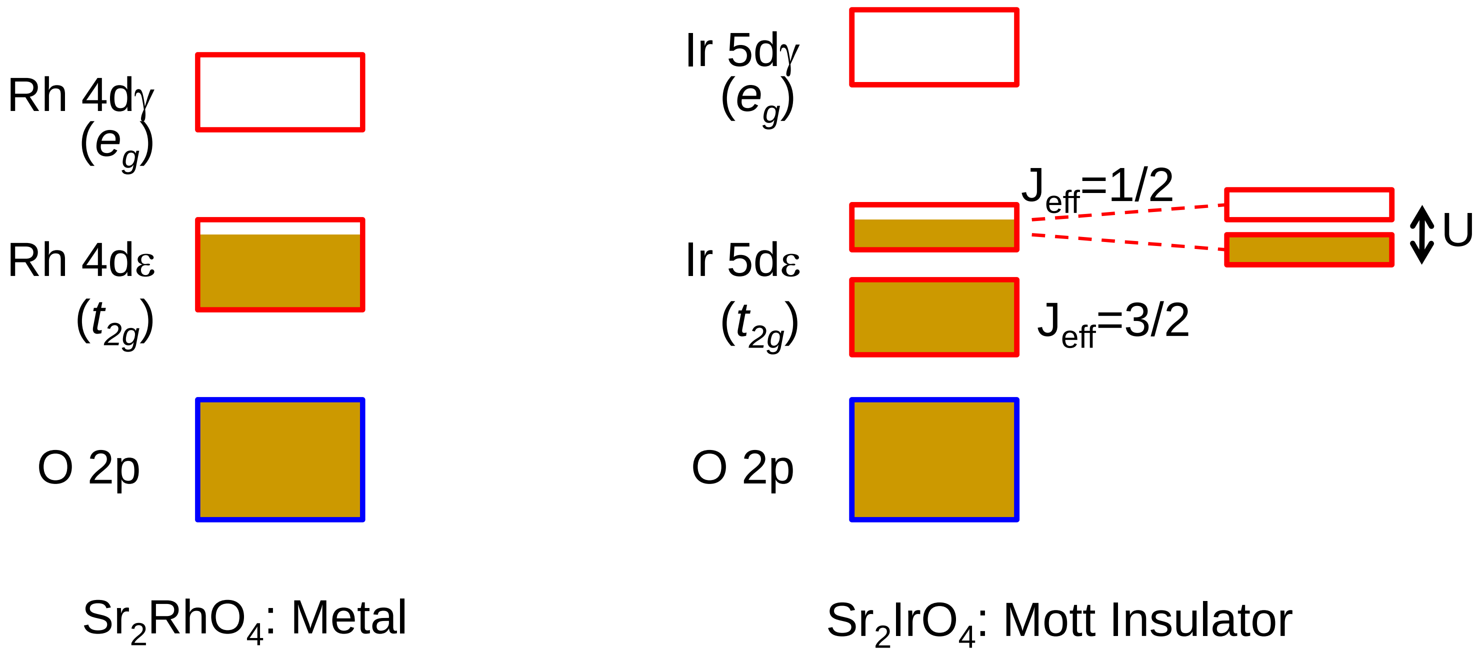


Spin-Orbital Coupled State!

Also see B. J. Kim *et al.*, Phys. Rev. Lett. **101**, 076402 (2008).

Possible Impact of Strong LS Coupling in 5d system

Difference in Band Structure from 4d System



Possibly Good Arena for Spintronics or Magneto-optics

2nd Topic

Phase Transition & Spin-Lattice Coupling in Multiferroic $RMnO_3$

Collaborators

H. Sagayama, N. Abe, K. Taniguchi (IMRAM Tohoku Univ.)

T. Kimura, T. Goto, Y. Yamasaki, D. Okuyama,

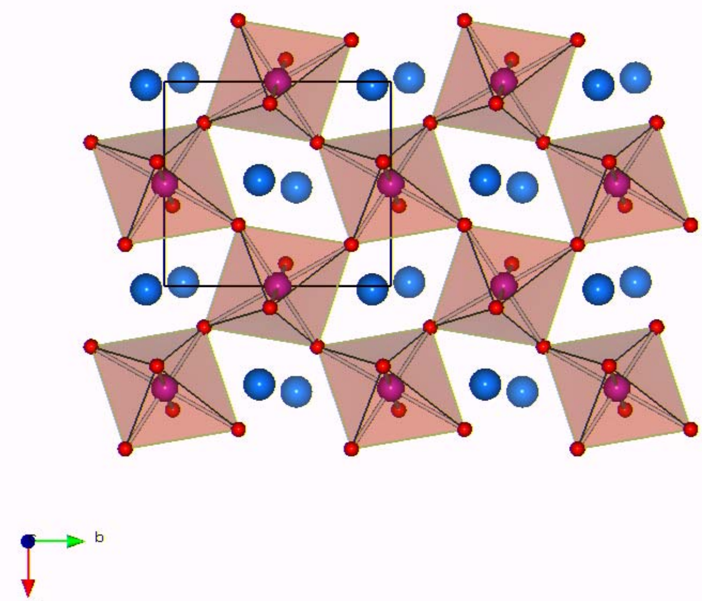
Y. Tokura (Univ. Tokyo & ERATO JST & RIKEN ASI)

T. Inami, K. Ishii, Y. Murakami (JAEA)

Y. Wakabayashi, H. Sawa (KEK-PF)

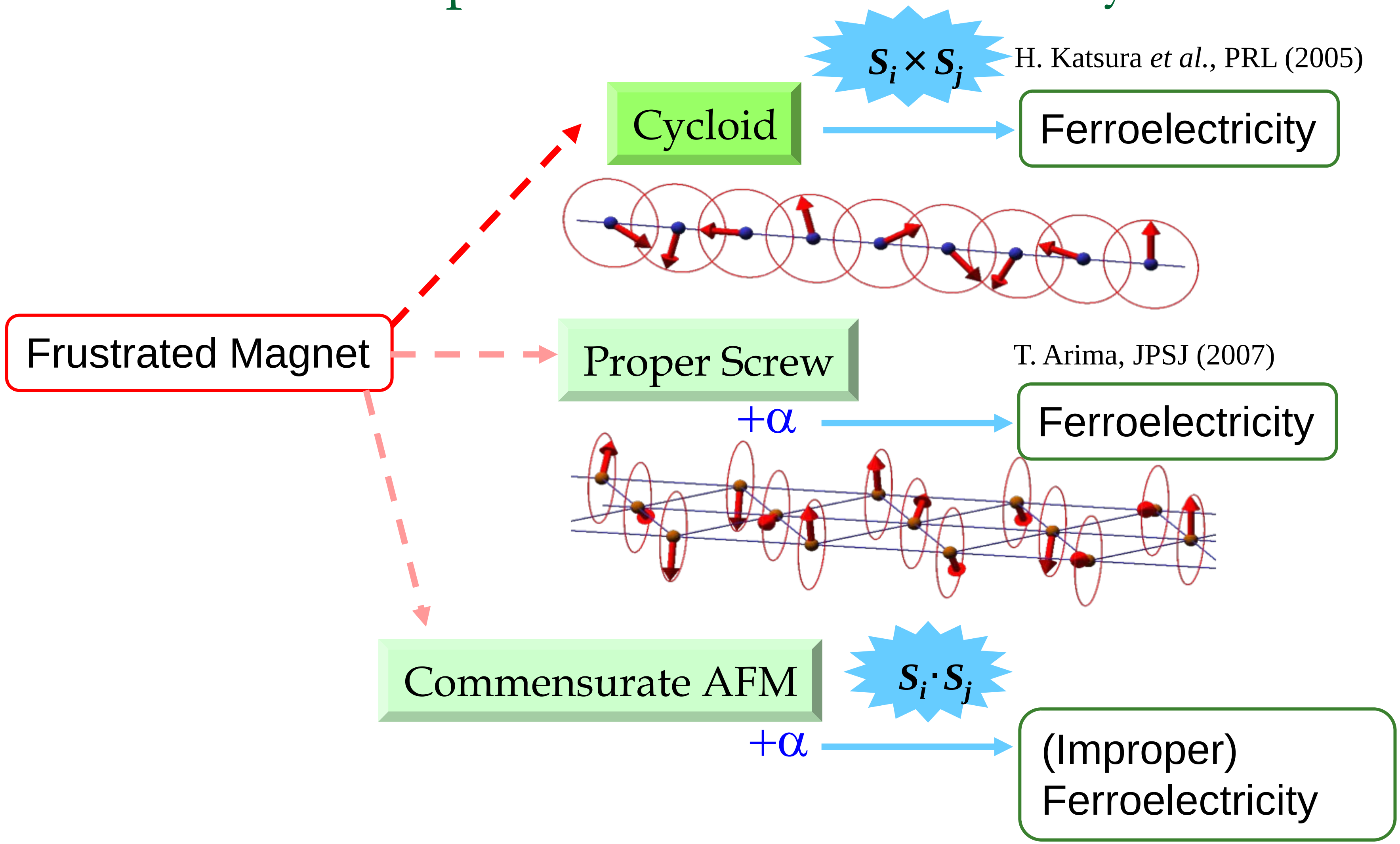
H. Ohsumi, S. Sakai (RIKEN SP8)

H. Sagayama



T. Kimura *et al.*, Phys. Rev. B**68**, 060403(R) (2003).
T. Kimura *et al.*, Nature **426**, 55 (2003).
T. Arima *et al.*, Phys. Rev. B**72**, 100102R (2005).
T. Arima *et al.*, J. Phys. Soc. Jpn. **76**, 023602 (2007).

Mechanisms of Spin-driven Ferroelectricity



Such an (anti)ferromagnetic ferroelectric is often classified into 'Multiferroic'.

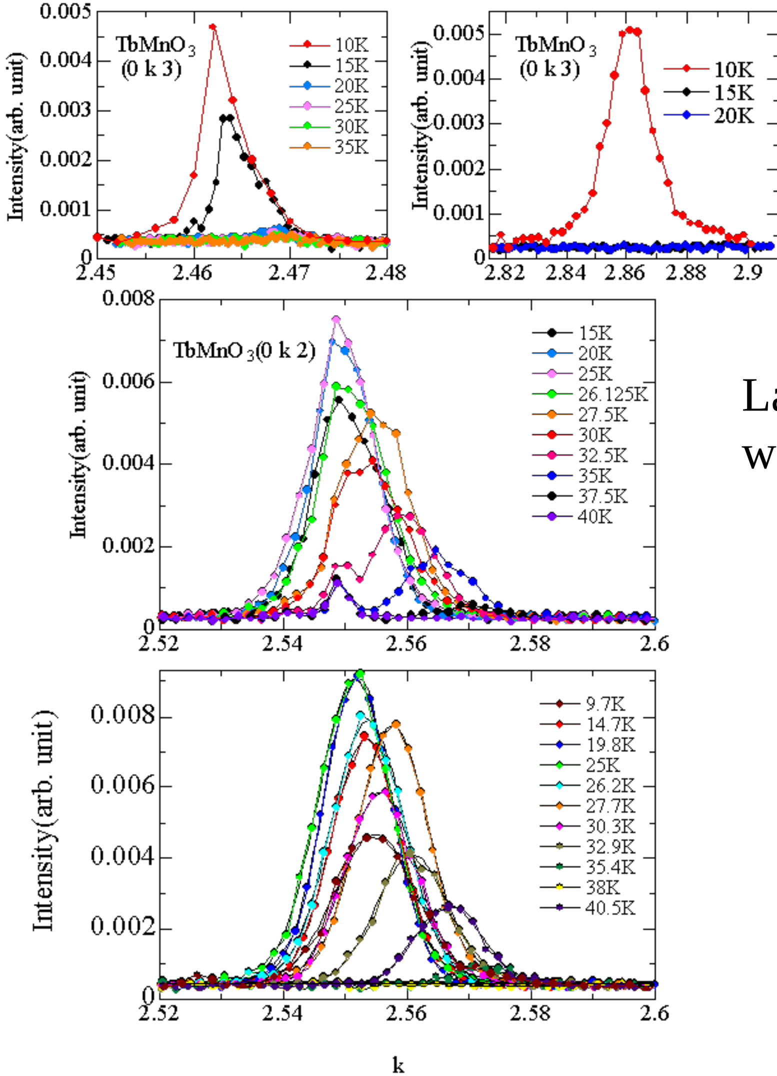
Role of Diffraction Studies in Multiferroics

Giant magnetoelectric effect in a frustrated system is nothing but a transition between nearly-degenerate different magnetic phases, some of which accompany ferroelectricity.

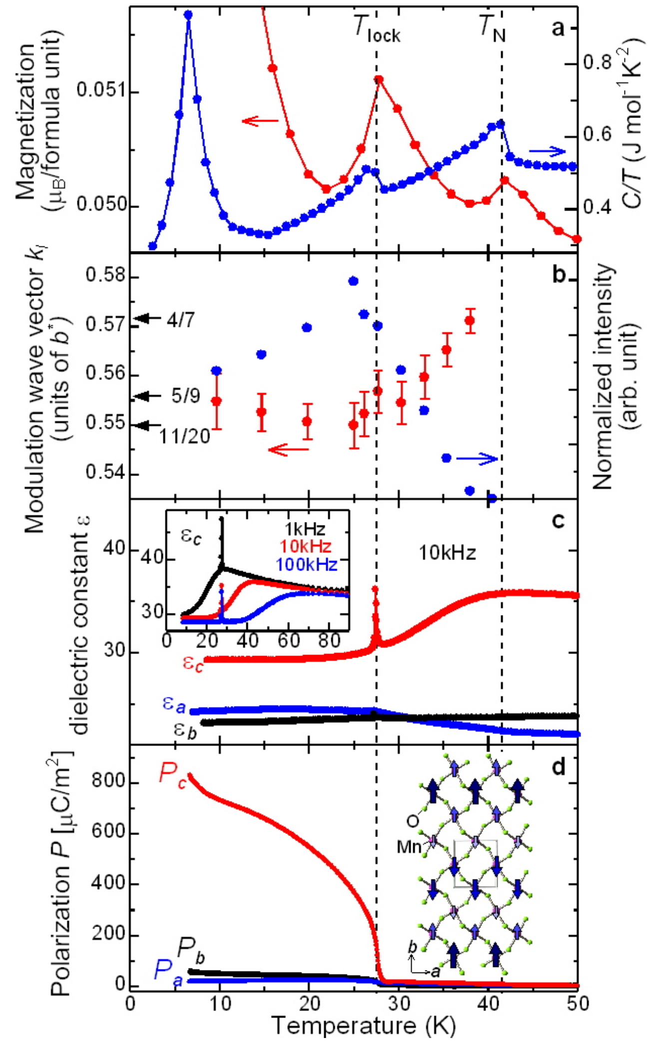
Diffraction Study →

- Magnetic Structure
- Phase Transition
- Spin-Lattice Coupling

First Synchrotron X-ray Investigation on $TbMnO_3$



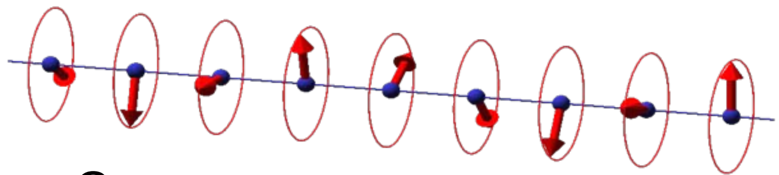
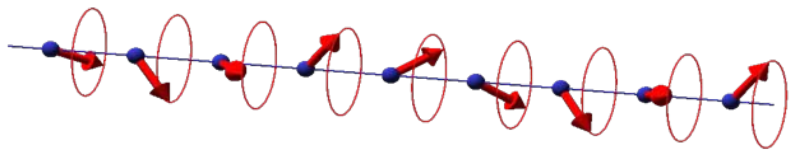
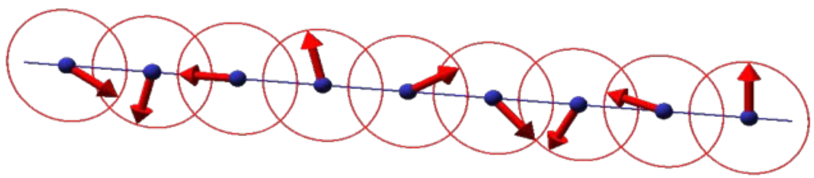
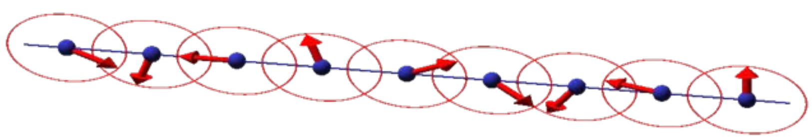
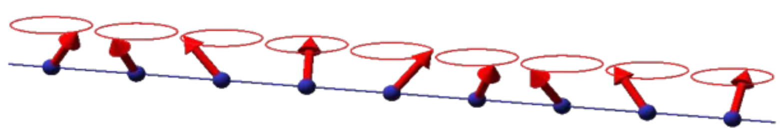
Lattice Modulation
with $2q_m$



BL-4C KEK-PF in 2002

T. Kimura *et al.*, Phys. Rev. B**68**, 060403(R) (2003).
T. Kimura *et al.*, Nature **426**, 55 (2003).

Lattice Modulation in Spiral Magnets

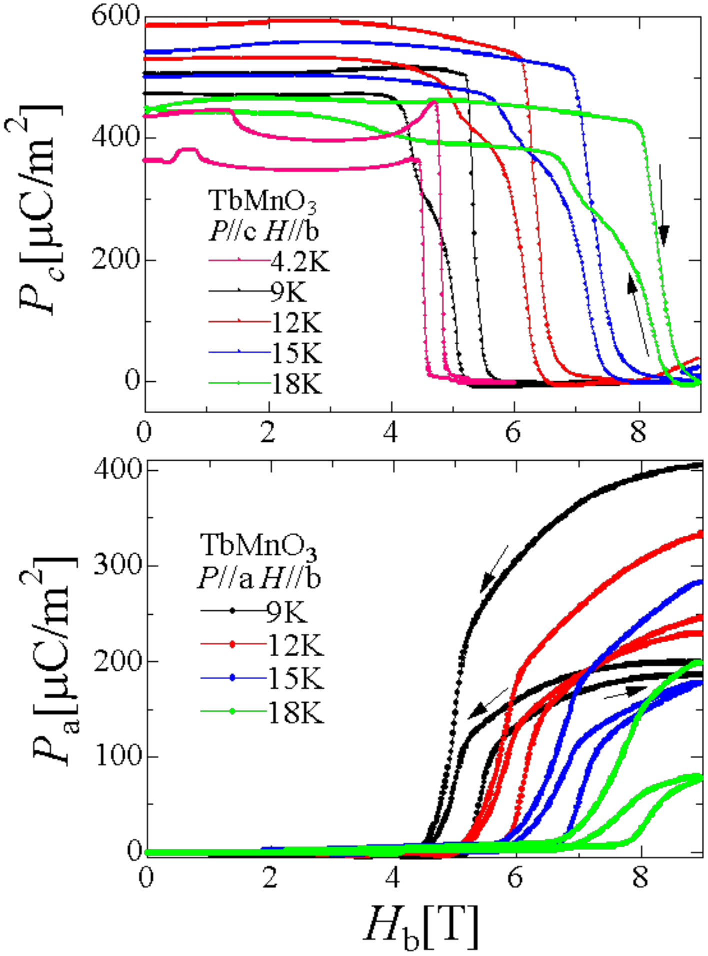
		$\vec{Q}_s = 0$	$\vec{Q}_s = \vec{Q}_m$	$\vec{Q}_s = 2\vec{Q}_m$
(a) Proper Screw		—	—	—
(b) Screw Cone		—	○	—
(c) Cycloid		○	—	—
(d) Elliptical Cycloid		○	—	○
(e) Cycloid Cone		○	○	—

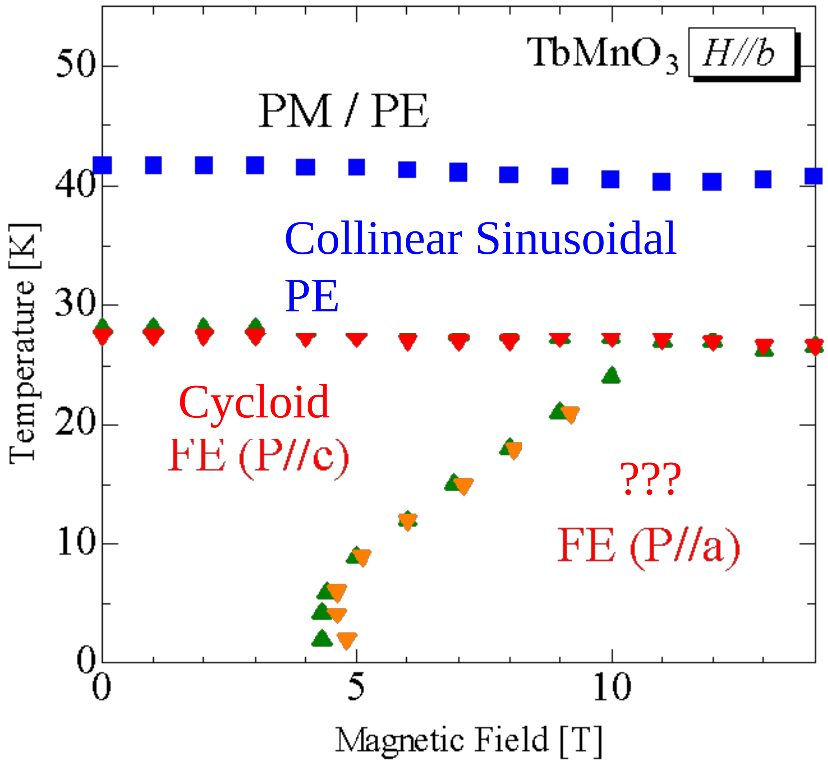
$\vec{Q}_s = 0$ ⇩ Ferroelectricity

$\vec{Q}_s = \vec{Q}_m$, $\vec{Q}_s = 2\vec{Q}_m$ ︸ Superlattice

T. Arima *et al.*, J. Phys. Soc. Jpn. **76**, 023602 (2007).

$\boldsymbol{H}$-induced $\boldsymbol{P}$ rotation ("flop") in $TbMnO_3$

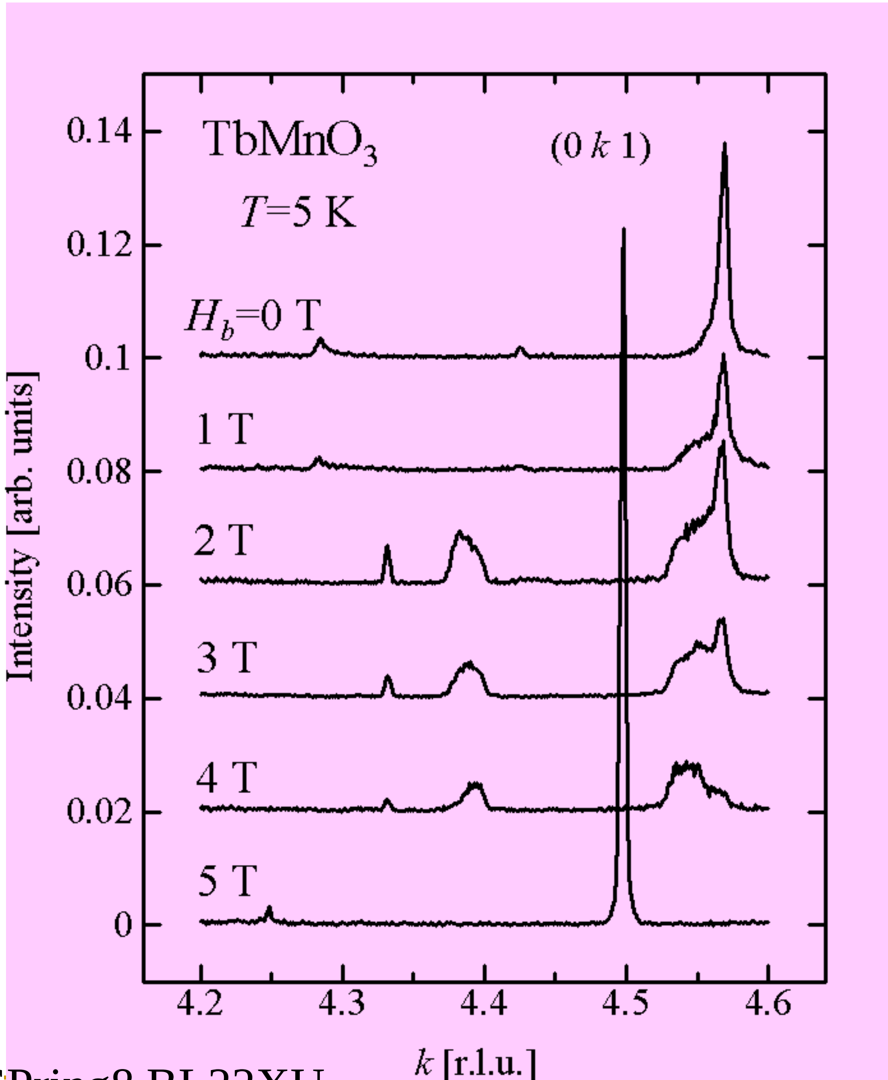


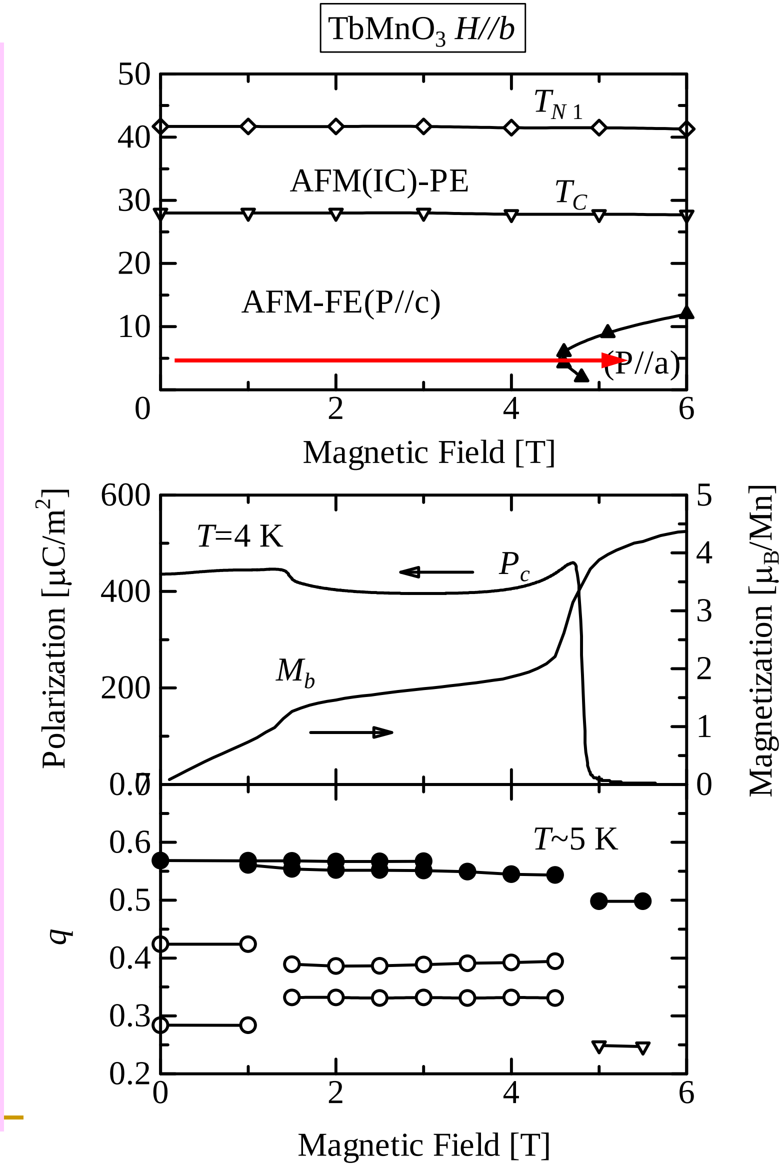


T. Kimura *et al.*, Nature **426**, 55 (2003)

What sort of magnetic phase appears in the $P \parallel a$ phase?

Incommensurate-Commensurate Transition upon ***P***-Flop

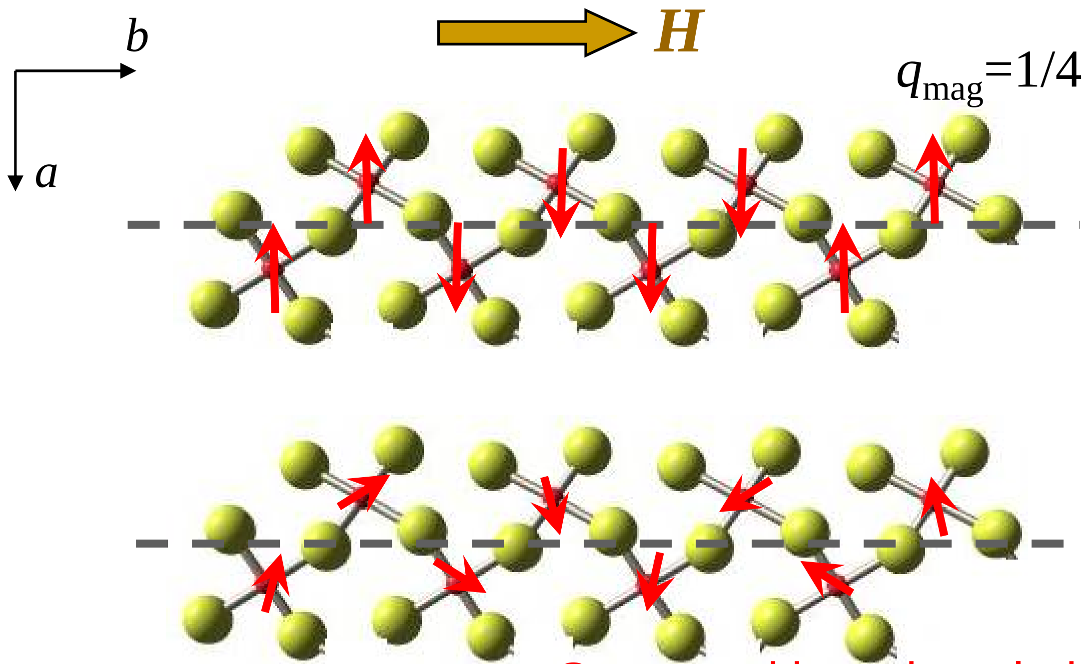




SPring8 BL22XU

T. Arima *et al.*, Phys. Rev. B**72** 100102R (2005).

Two Models of Magnetic Structure in *P* || *a* Phase



Commensurate AFM with Q=1/4

Cycloid with moments in the *ab* plane

Supported by spin-polarized neutron diffraction

Y. Yamasaki *et al.*, PRL **101**, 097204 (2008)

Detection of spin chirality is crucial.

1. Spin-polarized Neutron
2. Circularly-polarized X-ray

M. Blume, Phys. Rev. **130**, 1670 (1963). M. Blume & D. Gibbs, Phys. Rev. B**37**, 1779 (1988).

Summary

- Magnetic structures below and above Neel temperature as well as the L/S ratio and wave function of Ir ion in Sr_2IrO_4 have been determined by x-ray magnetic scattering.
- Magnetic wave vector and spin-lattice coupling in multiferroics have been investigated through superlattice reflections.
- Spin chirality can be investigated by circularly polarized x-ray.

In regard to resonant magnetic scattering,

- Resonant magnetic scattering for 4d systems with photons of 2.2~3.5 keV would provide much useful information.

Thank you for your Attention!!