Multiferroicity on the triangular lattice

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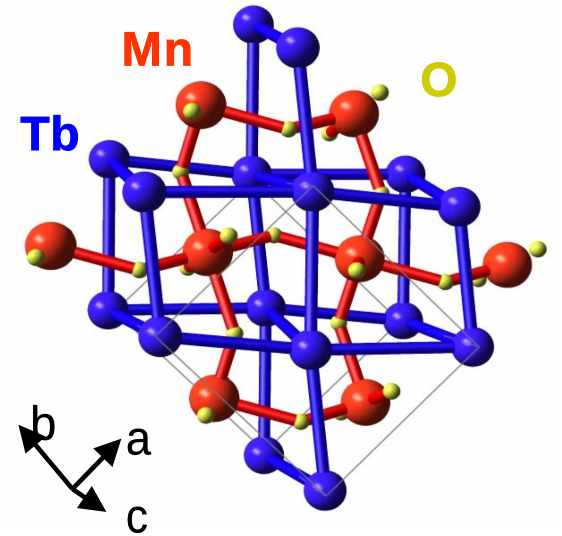
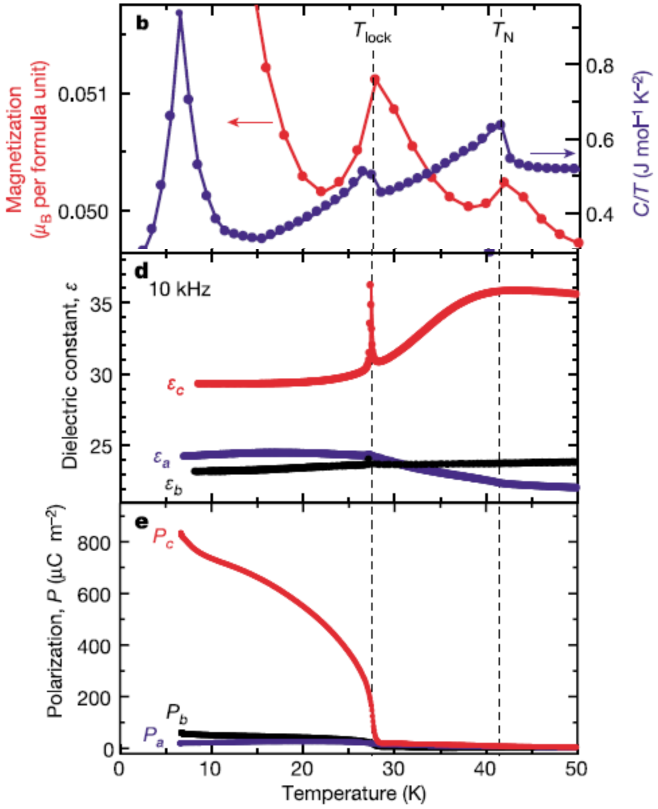
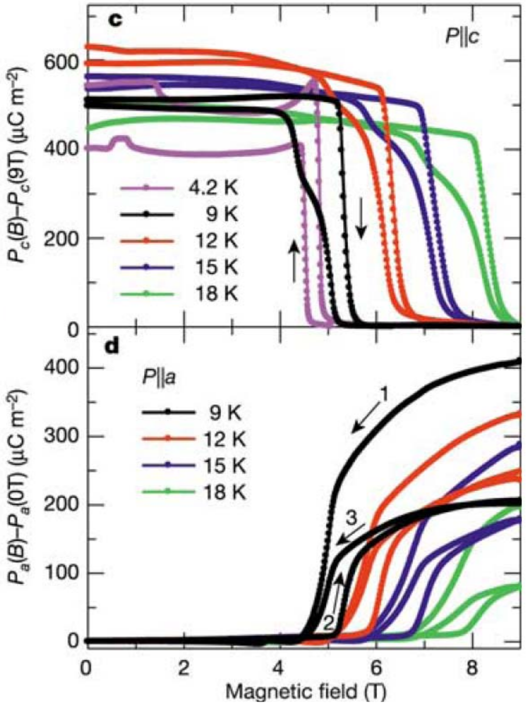
Ferroelectricity in $TbMnO_3$

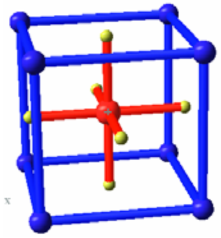


distorted perovskite structure
space group Pbnm
Mn^{3+} carries S=2





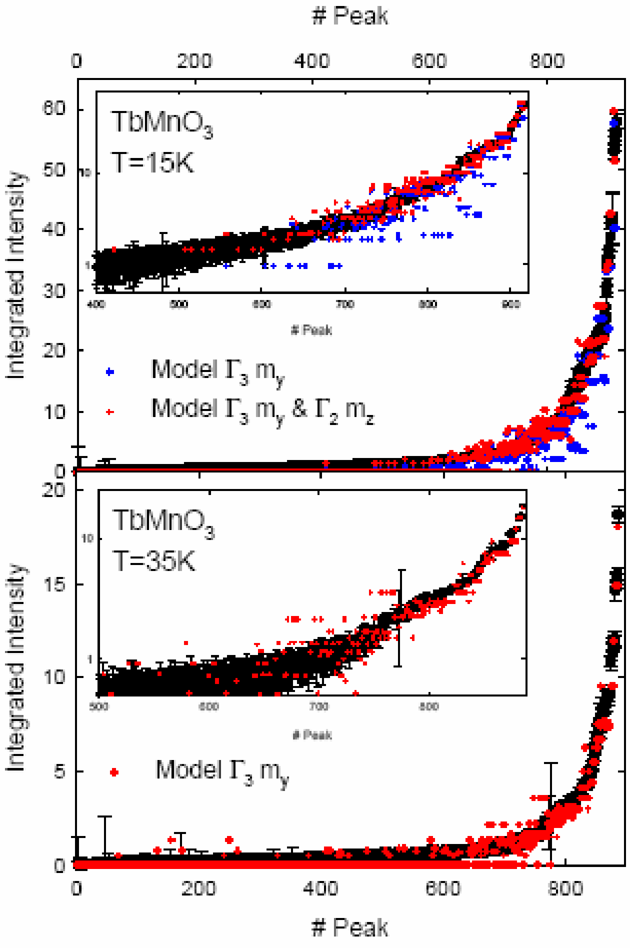
T. Kimura et al, Nature **426**, 55 (2003)

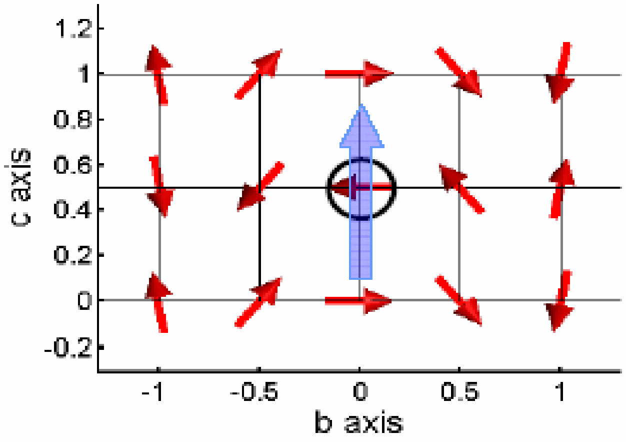


$BaTiO_3$

ferroelectric below 27K
direct coupling to
magnetic field observed

Magnetic structure of $TbMnO_3$



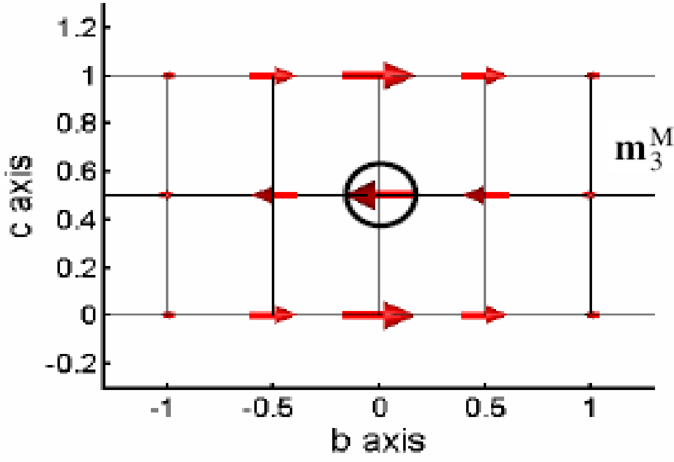


$$\mathbf{m}_3^{\mathrm{Mn}} = (0.0(5), 3.9(1), 0.0(7))\mu_B$$

$$\mathbf{m}_3^{\mathrm{Tb}} = (0, 0, 0(1))\mu_B$$

$$\mathbf{m}_2^{\mathrm{Mn}} = (0.0(1), 0.0(8), 2.8(1))\mu_B$$

$$\mathbf{m}_2^{\mathrm{Tb}} = (1.2(1), 0(1), 0)\mu_B$$



$$\mathbf{m}_3^{\mathrm{Mn}} = [0.0(8), 2.90(5), 0.0(5)]\mu_B$$

$$\mathbf{m}_3^{\mathrm{Tb}} = [0, 0, 0.0(4)]\mu_B$$

M. Kenzelmann et al, Phys. Rev. Lett. **95**, 087206 (2005)

spiral phase breaks inversion symmetry

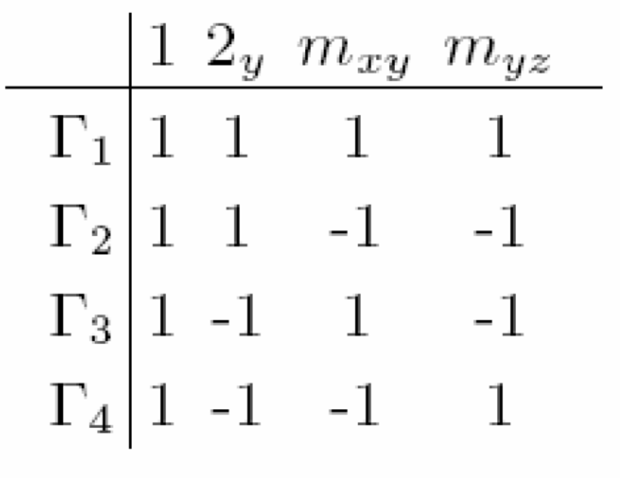
Polar axis from magnetic order

$$\mathcal{H} = \sum_{\alpha\beta\gamma,\mathbf{q}} a_{\alpha\beta\gamma}(\mathbf{q}) M_\alpha(\mathbf{q}) M_\beta(-\mathbf{q}) P_\gamma(0) \ .$$

Trilinear Coupling of M and P is allowed

→ **P** has to transform as M_α (q) M_β(-q)

Representational Analysis: M order parameters transform to one of the four irreducible representations in $TbMnO_3$: $\Gamma_2+\Gamma_3$

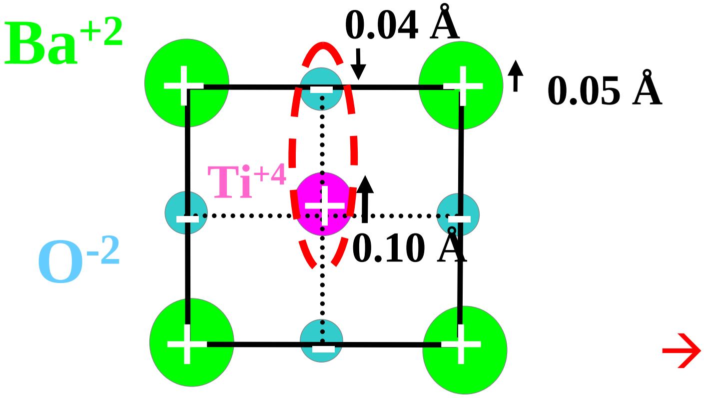


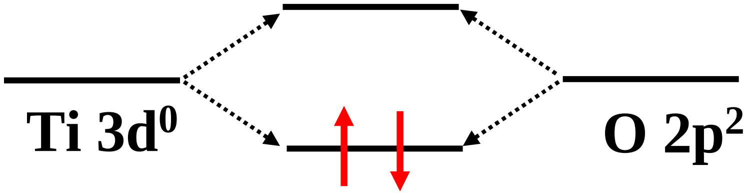
	1	2_y	m_{xy}	m_{yz}
Γ_1	1	1	1	1
Γ_2	1	1	-1	-1
Γ_3	1	-1	1	-1
Γ_4	1	-1	-1	1

→ Electric polarization is only allowed along the c-direction as observed (P has to be even under 1 & m_{yz} and odd under 2_y & m_{xy})

M. Kenzelmann et al, Phys. Rev. Lett. **95**, 087206 (2005)

Microscopic origin for ferroelctricity





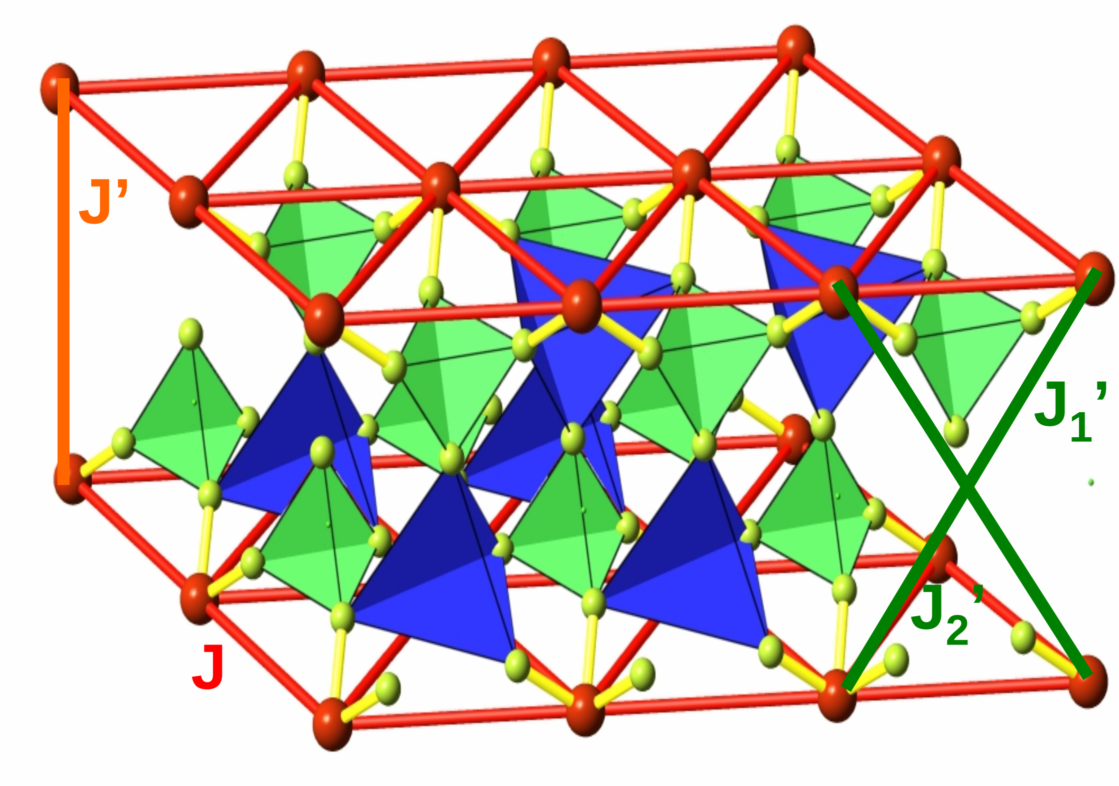
polarization from covalent bond of cation/anion

→Covalent bonds preclude coexistence between magnetism and ferroelectricity

1) orthorhombic $RMnO_3$
2) hexagonal $RMnO_3$
3) RMn_2O_5
4) $Ni_3V_2O_8$
5) $MnWO_4$
6) $LiCu_2O_2$
7) $CuFeO_2/CuFe_{1-x}Al_xO_2$
8) $RbFe(MoO_4)_2$
9) ……

→ New mechanism: ferroelectricity from long range magnetic order

$RbFe(MoO_4)_2$ Crystal Structure



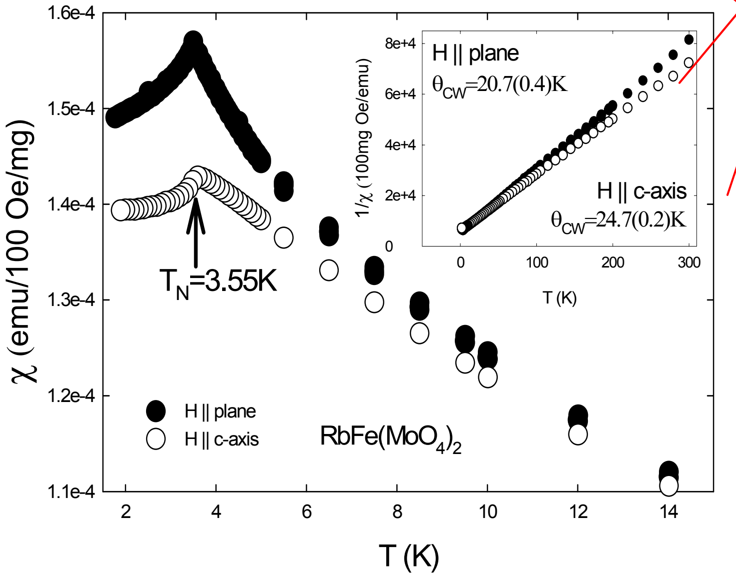
- 2D equilateral triangular lattice in trigonal crystal structure P-3
- Fe^{3+} carries "classical" S = 5/2
- spins interact via oxygen superexchange paths
- interplane frustration due to several nearest and next-nearest exchange paths

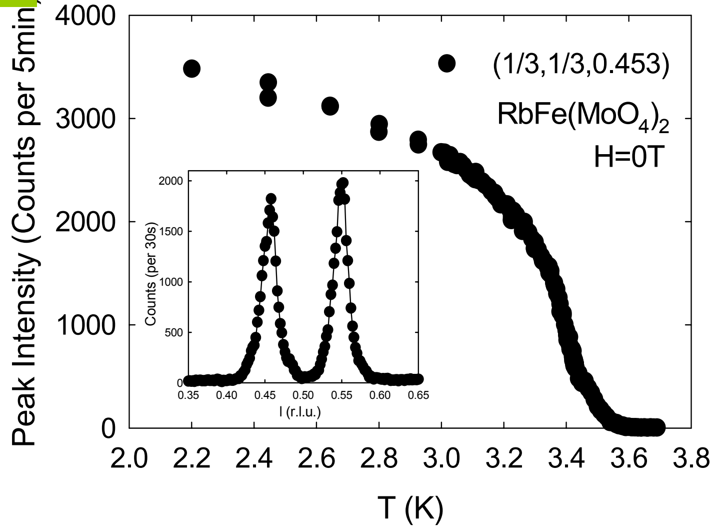
Antiferromagnetic order due interplane couplings

Magnetic susceptibility

J ~ 0.1meV

Neutron Diffraction

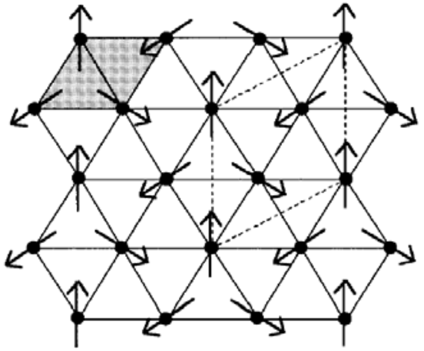




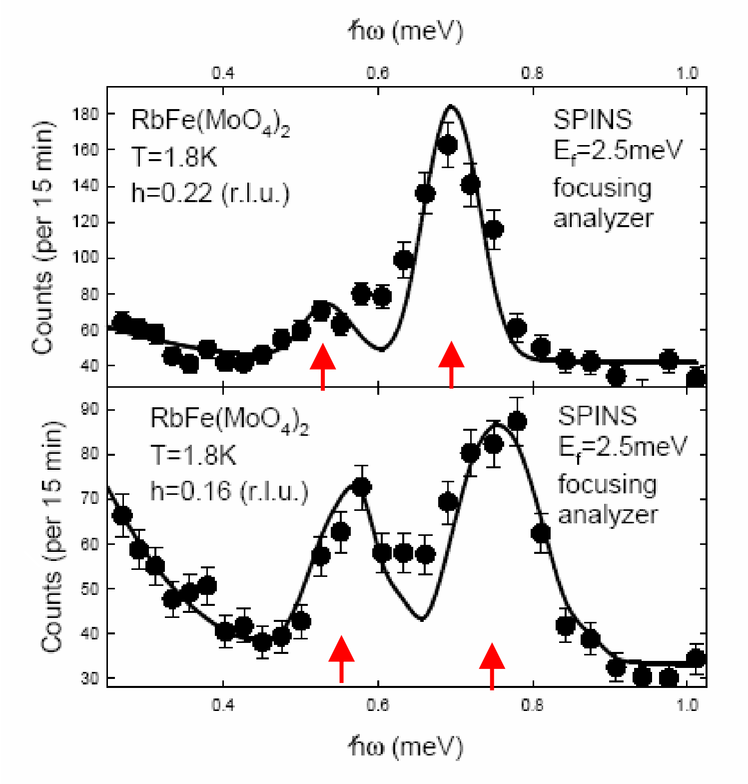
unpublished

Incommensurate AF order along c-axis due to diagonal interplane interactions observed with single-crystal and powder neutron diffraction

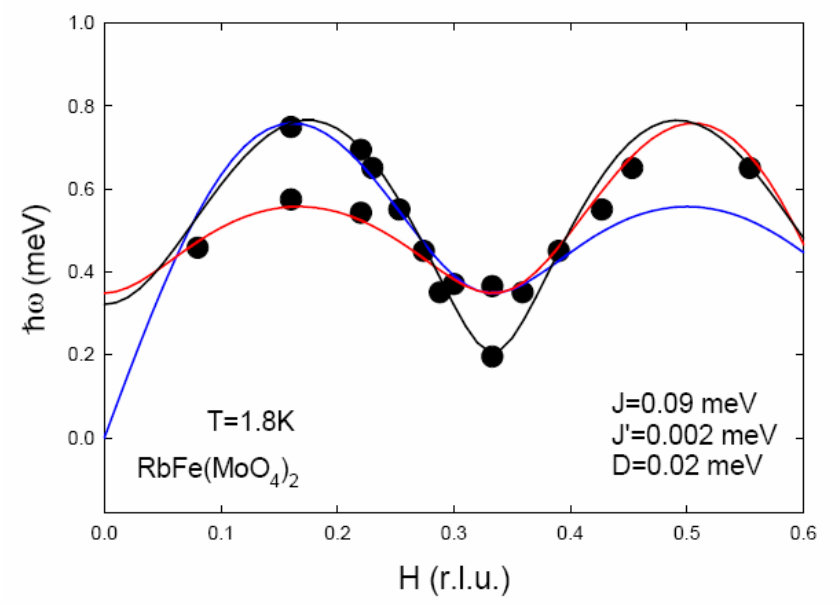
→ stacked 120^o structure as ordered ground state, ordered moment is $3.9\mu_B$ (about 80% of the available moment)



Inelastic neutron scattering from spin waves



In-plane spin wave dispersion

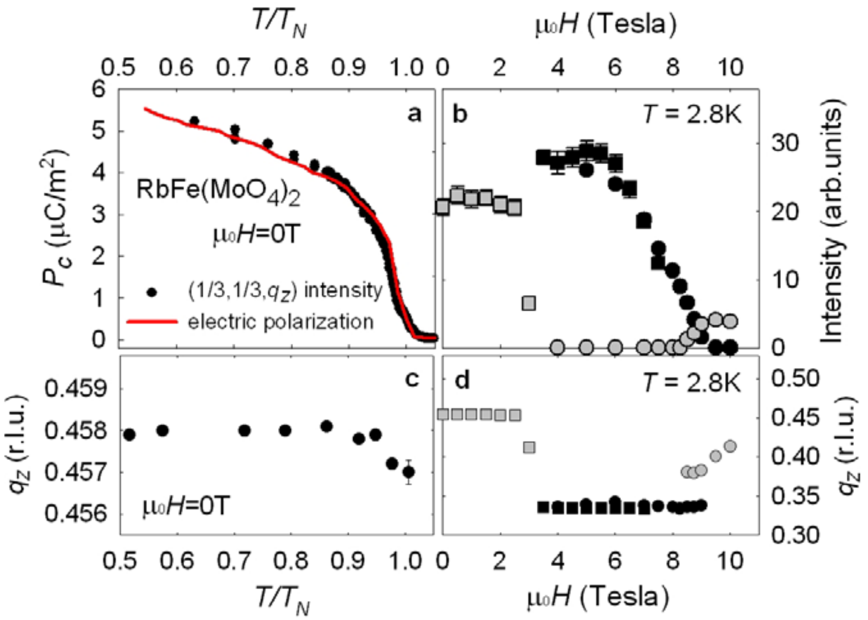


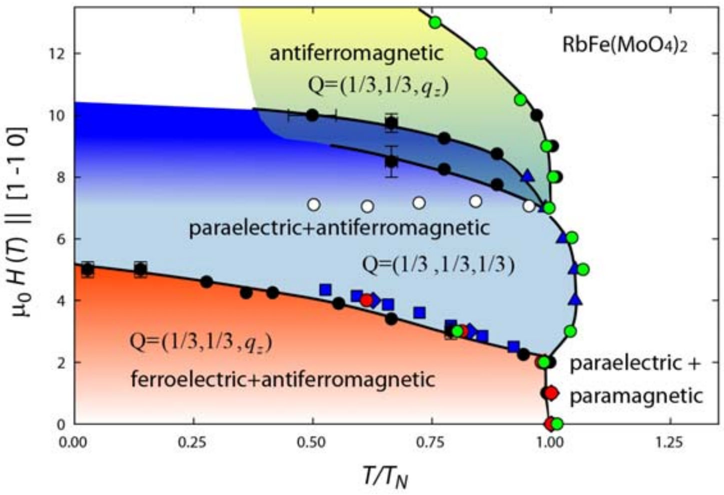
M. Kenzelmann et al, unpublished

Well-defined spin waves are observed in the ordered phase of RbFe(MoO$_4$)$_2$

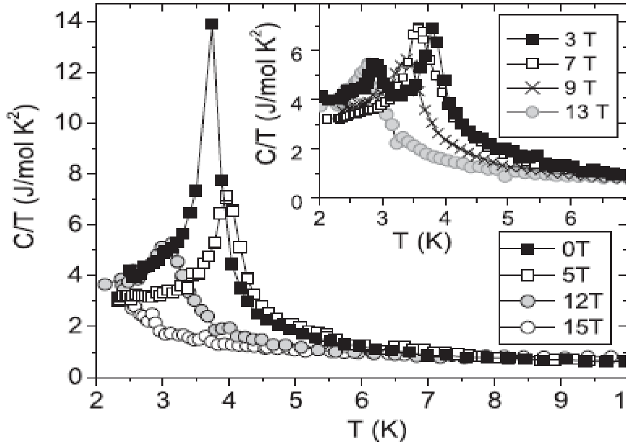
→ **In-plane spin anisotropy makes RbFe(MoO$_4$)$_2$ a XY-like 2D triangular lattice**

Ferroelectricity in $RbFe(MoO_4)_2$





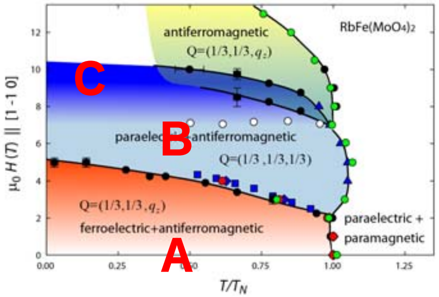
M. Kenzelmann et al, Phys. Rev. Lett. **98,** 267205 (2007)



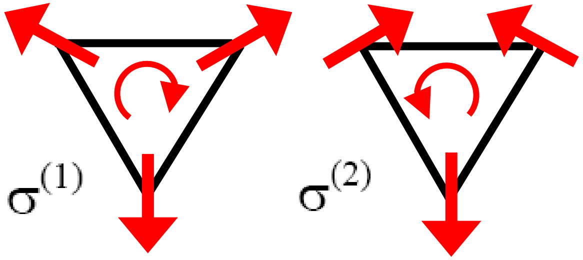
G.A. Jorge et al, Physica B **354**, 297 (2004).

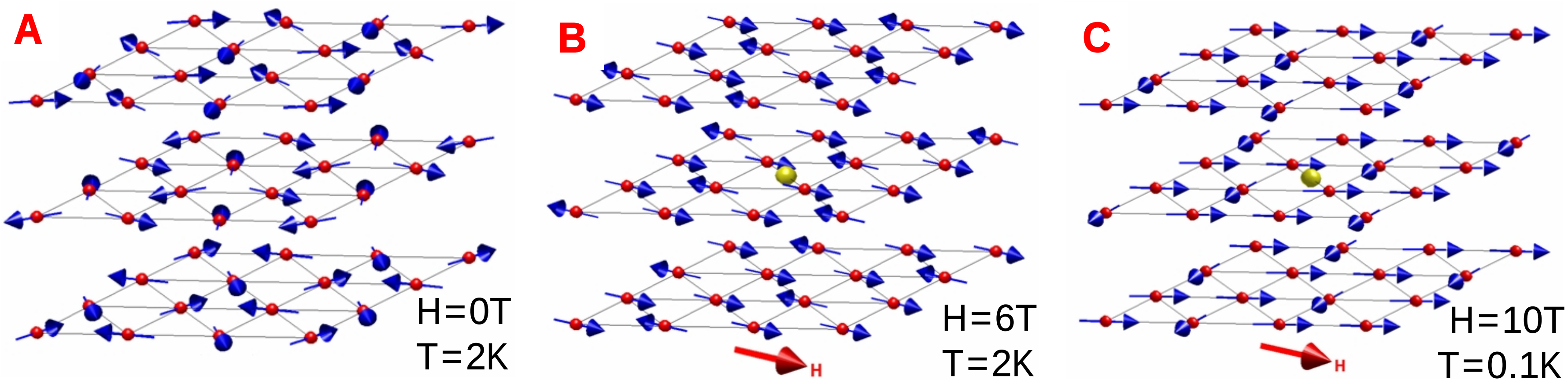
- direct transition from a disordered to a multiferroic state
- magnetic field suppressed ferroelectric polarization
- ferroelectric polarization perpendicular to spiral plane
- temperature dependence of $P_c \sim M^2$

Magnetic inversion symmetry breaking on the triangular lattice

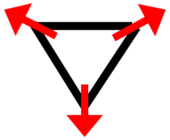


two chiral order paramaters

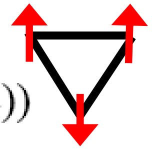




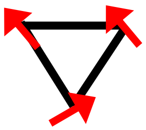
$\sigma^{(1)}$=3.9(5)
$\sigma^{(2)}$=0



$\sigma^{(1)} = (1.09(4) + \mathrm{i}\ 1.85)$
$\sigma^{(2)} = (1.09(4) + \mathrm{i}\ 1.94(4))$



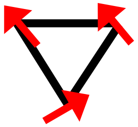
$\sigma^{(1)} = (2.16(3) + \mathrm{i}\,0.197)$
$\sigma^{(2)} = (-1.251 - \mathrm{i}\ 1.724)$



No inversion center for the “120º structure”, but inversion center for commensurate phase

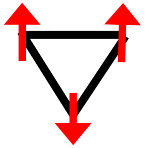
Generation of a polar axis on the triangular lattice

$$\sigma^{(1)} = (2.16(3) + i0.197)$$
$$\sigma^{(2)} = (-1.251 - i\,1.724)$$



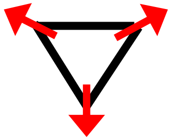
$$|\sigma^{(1)}|^2 = 2.17(4)$$
$$|\sigma^{(2)}|^2 = 2.13$$

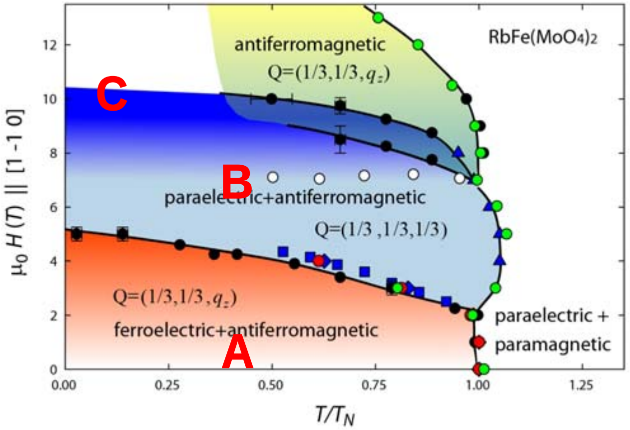
$$\sigma^{(1)} = (1.09(4) + i\,1.85)$$
$$\sigma^{(2)} = (1.09(4) + i\,1.94(4))$$



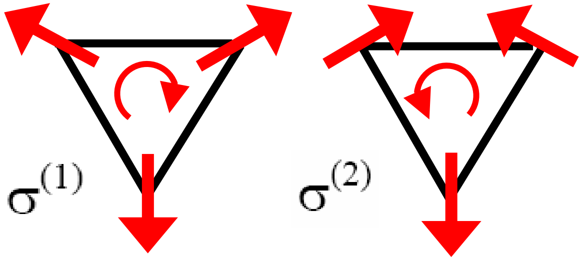
$$|\sigma^{(1)}|^2 = 2.15(3)$$
$$|\sigma^{(2)}|^2 = 2.11(9)$$

$$\sigma^{(1)} = 3.9(5)$$
$$\sigma^{(2)} = 0$$





two chiral order paramaters



Brooks Harris' Landau theory:

$$F = \frac{1}{2}\chi^{-1}P^2 + K\left[\left|\sigma^{(1)}(q_z)\right|^2 - \left|\sigma^{(2)}(q_z)\right|^2\right]P_c$$

$\mathbf{P} \,||\, \mathbf{e}_3 \times \mathbf{Q}$ not valid

Ferroelectricity is allowed for unequal population of the chiral order paramater $\sigma^{(1)}$ and $\sigma^{(2)}$ is **perpendicular** to the spiral plane (in contradiction to most theories on magneto-electric coupling)

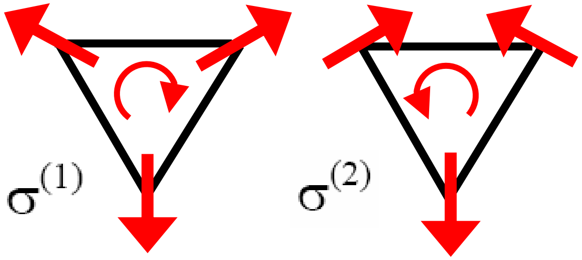
Chirality vs helicity

$$F = \frac{1}{2}\chi^{-1}P^2 + K\left[\left|\sigma^{(1)}(q_z)\right|^2 - \left|\sigma^{(2)}(q_z)\right|^2\right]P_c$$

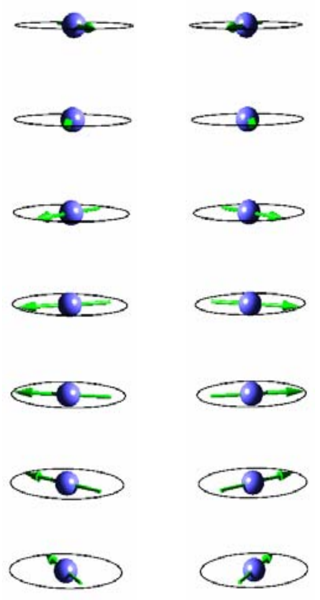
Trigonal stacked triangular antiferromagnets
with a 120° magnetic order
should show magneto-electric order

Magnetic chirality determines the direction of the ferroelectric polarization, not magnetic helicity

two chiral order paramaters



opposite helicity



Conclusions

- **Novel multiferroic material**
- **Direct transition from disordered to ferroelectric/antiferromagnetic phase**
- **Trilinear coupling** puts stringent symmetry rules on any **microscopic mechanism**
- None of the mechanisms proposed for the manganite multiferroics directly apply