

Title: Ferroelectricity out of magnetic frustration

Date: Apr 24, 2008 02:00 PM

URL: <http://pirsa.org/08040044>

Abstract: Responding electrically to magnetic stimuli and vice versa, multiferroics offer exciting possibilities for applications and challenge our understanding of coupled lattice and spin degrees of freedom in solids. I discuss how multiferroic properties can develop in frustrated magnets where competing interactions produce non-collinear spin order and symmetry breaking lattice distortions. Our experiments in TbMnO<sub>3</sub>, Ni<sub>3</sub>V<sub>2</sub>O<sub>8</sub>, and RbFe(MoO<sub>4</sub>)<sub>2</sub> show that when the low temperature magnetic order breaks spatial inversion symmetry it is accompanied by ferroelectricity [1-3]. Conversely, the application of an electric field favors one of the two inversion symmetry related antiferromagnetic domains. We infer that inversion symmetry breaking magnetic order acts as an effective electric field through magneto-elastic distortions that relieve frustration. We also present evidence for microscopic correspondence between the ferroelectric and the antiferromagnetic domain structure. The results presented are based on magnetic neutron diffraction, pyrocurrent measurements, and theoretical work by A. B. Harris [4]. [1] M. Kenzelmann, A. B. Harris, S. Jonas, C. Broholm, J. Schefer, S. B. Kim, C. L. Zhang, S.-W. Cheong, O. P. Vajk, and J. W. Lynn, Phys. Rev. Lett. 95, 087206 (2005). [2] G. Lawes, A. B. Harris, T. Kimura, N. Rogado, R. J. Cava, A. Aharony, O. Entin-Wohlman, T. Yildirim, M. Kenzelmann, C. Broholm, and A. P. Ramirez, Phys. Rev. Lett. 95, 087205 (2005). [3] M. Kenzelmann, G. Lawes, A.B. Harris, G. Gasparovic, C. Broholm, A.P. Ramirez, G.A. Jorge, M. Jaime, S. Park, Q. Huang, A.Ya. Shapiro, and L.A. Demianets, Phys. Rev. Lett. 98, 267205 (2007). [4] A. B. Harris, Phys. Rev. B 76 , 054447 (2007).



# Ferroelectricity out of Magnetic Frustration

Collin Broholm

Johns Hopkins University &

NIST Center for Neutron Research

# Outline

- Introduction
  - Magneto elastic effects frustrated magnets
- Inversion symmetry breaking magnetism
  - Kagome staircase magnetism  $\text{Ni}_3\text{V}_2\text{O}_8$
  - Triangular lattice AFM  $\text{RbFe}(\text{MoO}_4)_2$
  - Electric field control of spin chirality
- Conclusions and Outlook

# Magneto-striction

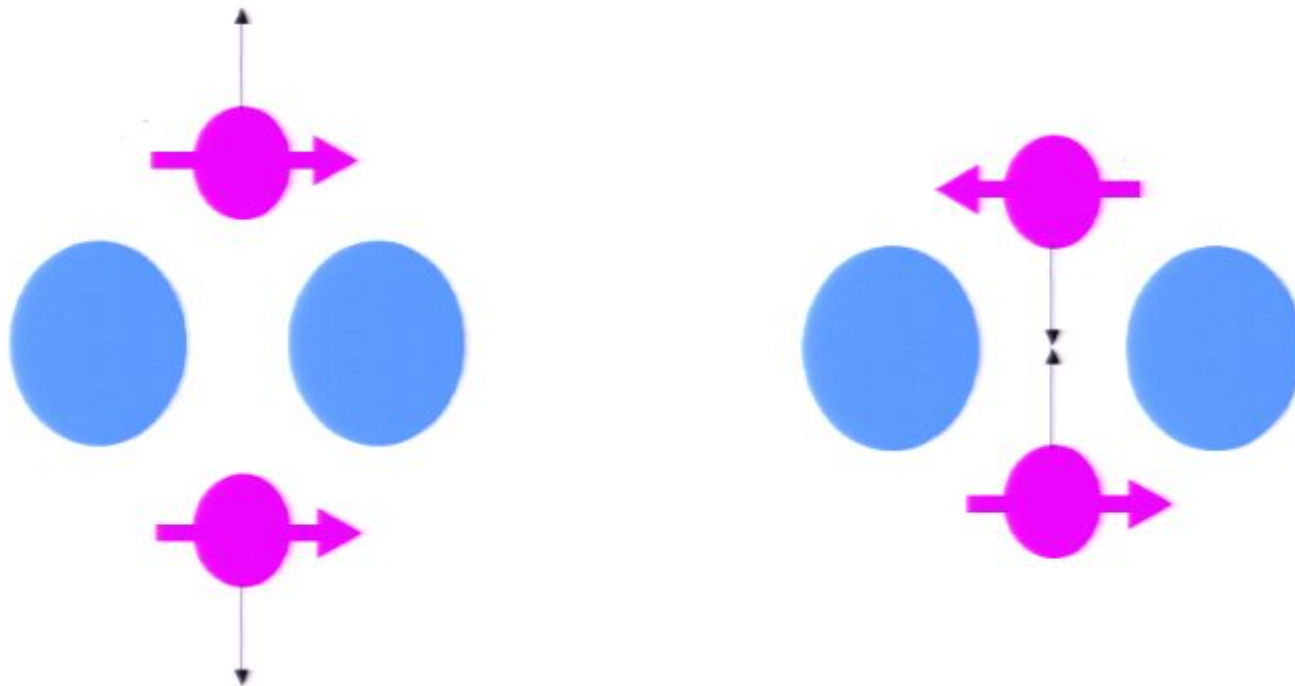
$$\mathbf{F}_{12} = -\nabla(J_{12}\langle\mathbf{S}_1 \cdot \mathbf{S}_2\rangle) = -\hat{\mathbf{r}}_{12} \frac{\partial J_{12}}{\partial r} \langle\mathbf{S}_1 \cdot \mathbf{S}_2\rangle$$



*Tchernyshyov et al. PRL (2001) and PRB (2002)*

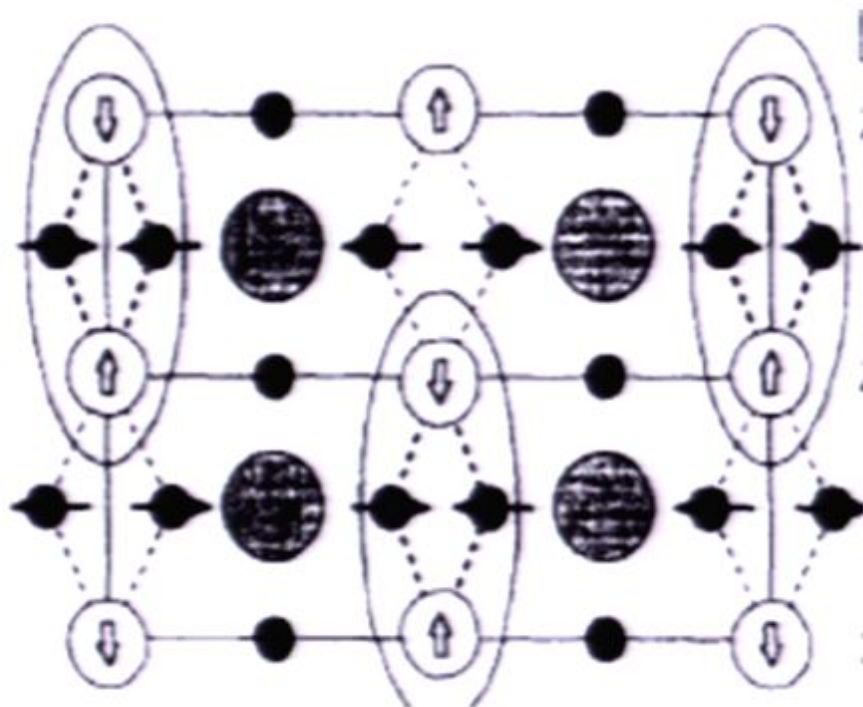
# Magneto-striction

$$\mathbf{F}_{12} = -\nabla(J_{12}\langle\mathbf{S}_1 \cdot \mathbf{S}_2\rangle) = -\hat{\mathbf{r}}_{12} \frac{\partial J_{12}}{\partial r} \langle\mathbf{S}_1 \cdot \mathbf{S}_2\rangle$$

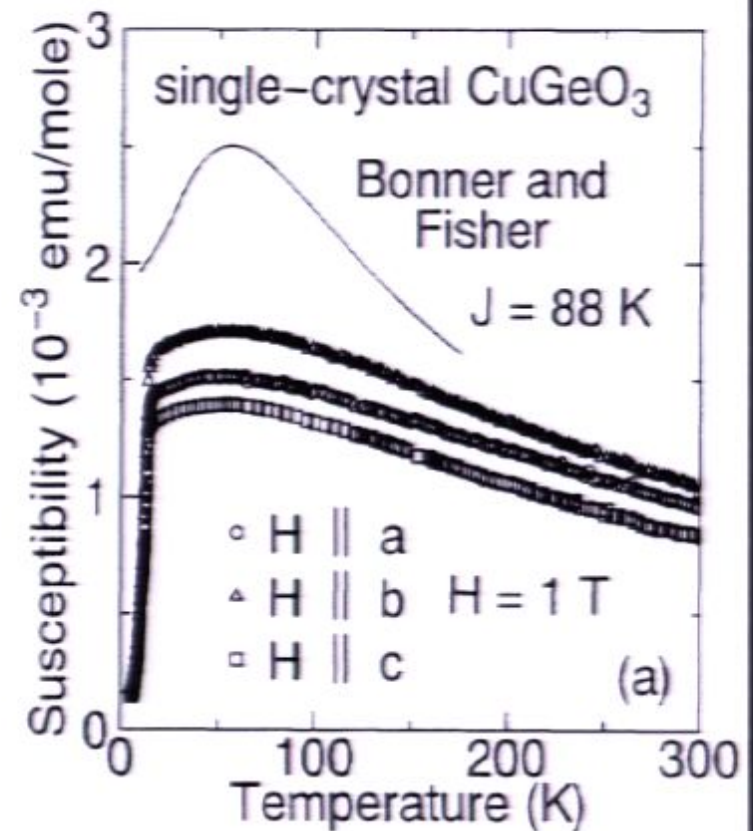


*Tchernyshyov et al. PRL (2001) and PRB (2002)*

# Quantum criticality and magneto-striction



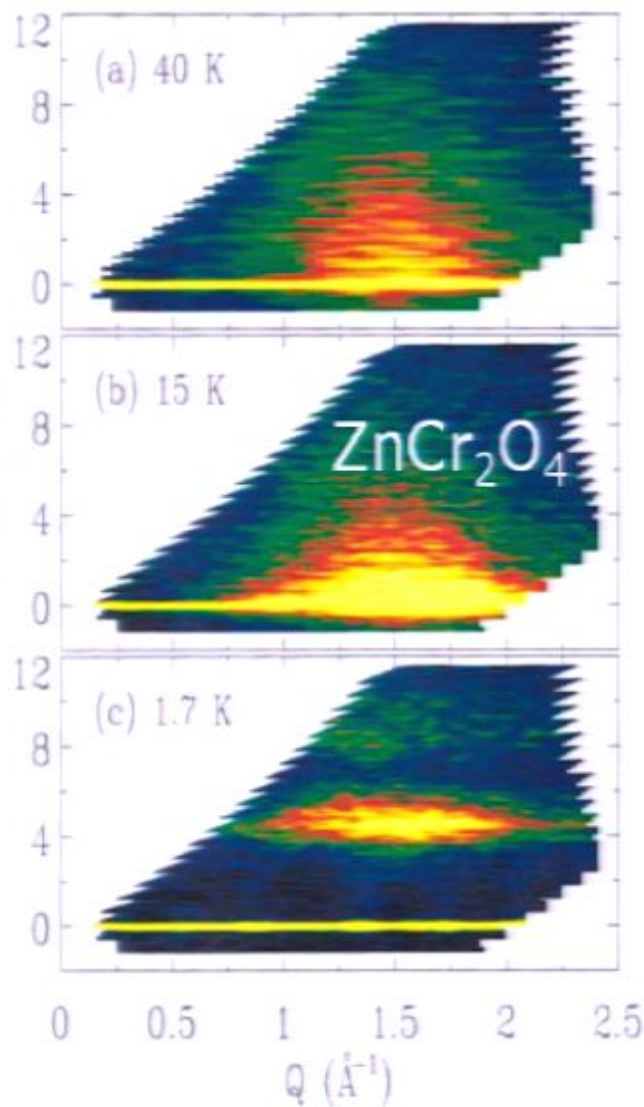
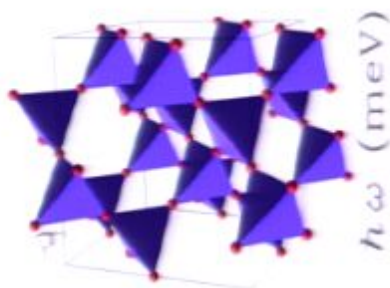
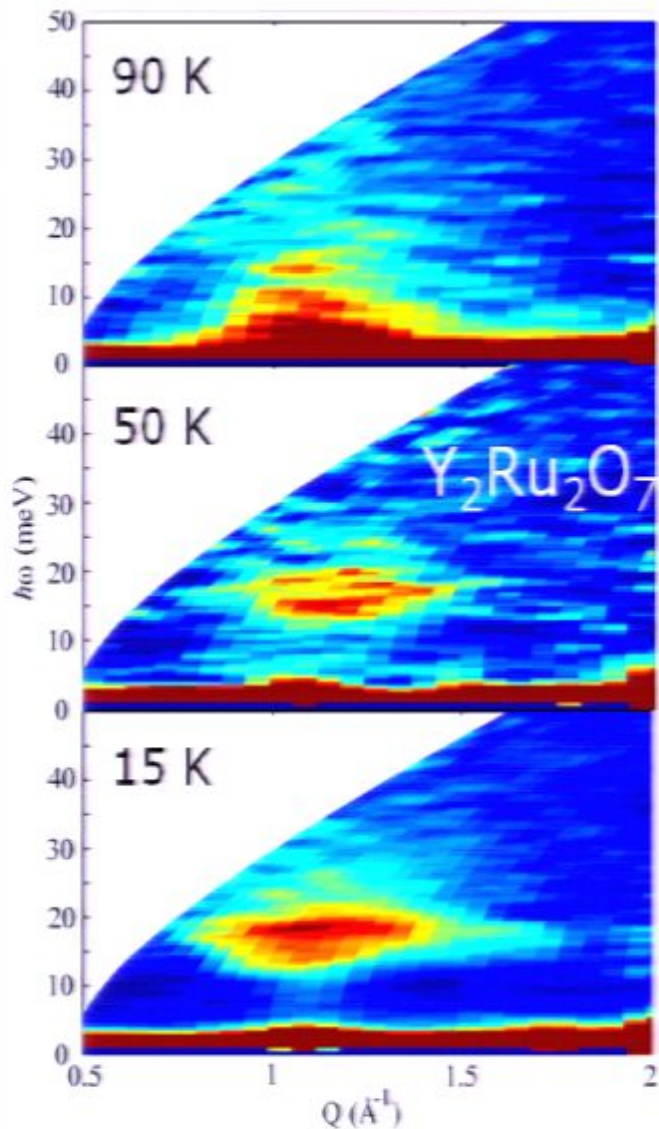
$$J_2 \approx 0.36 J_1$$



Hase et al (1993)

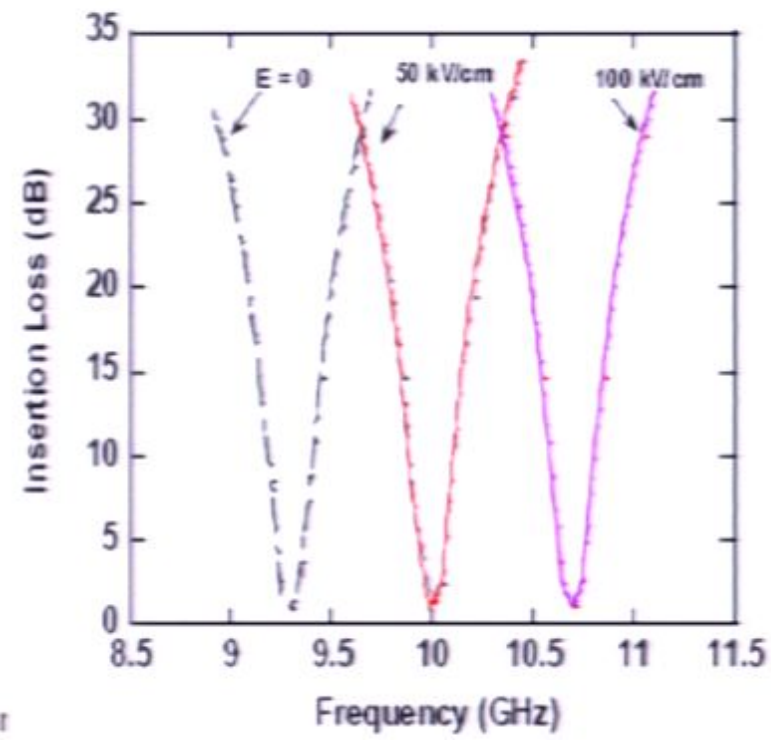
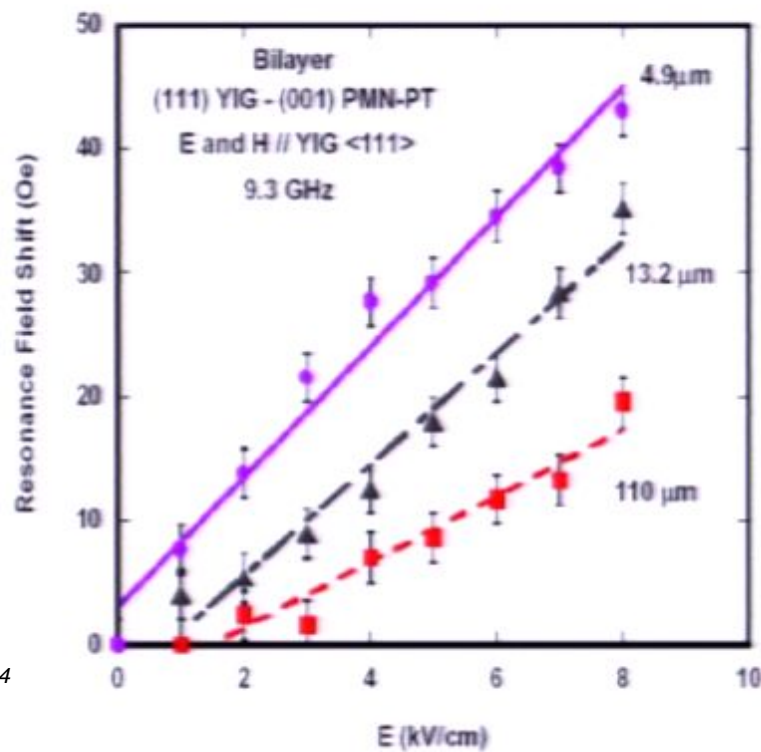
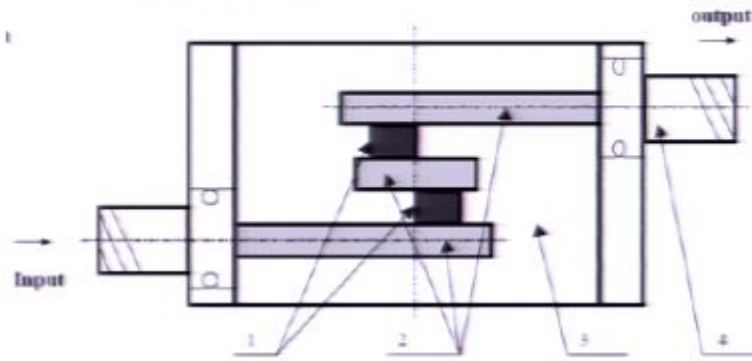
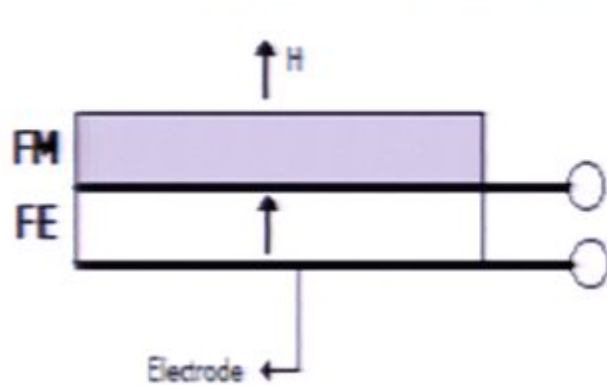


# Resonance upon ordering: pyrochlore & spinel



# Electric Field tuned RF filter

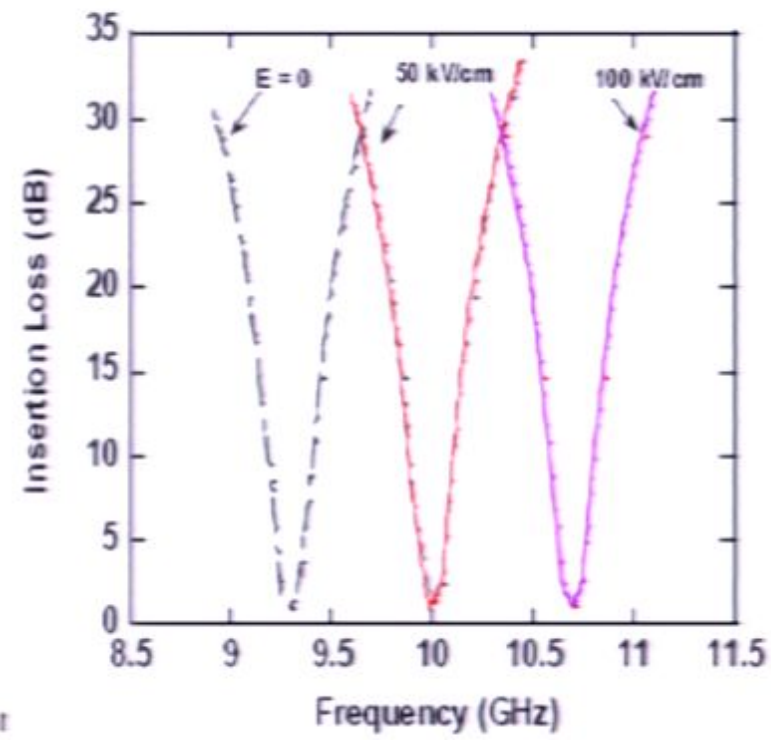
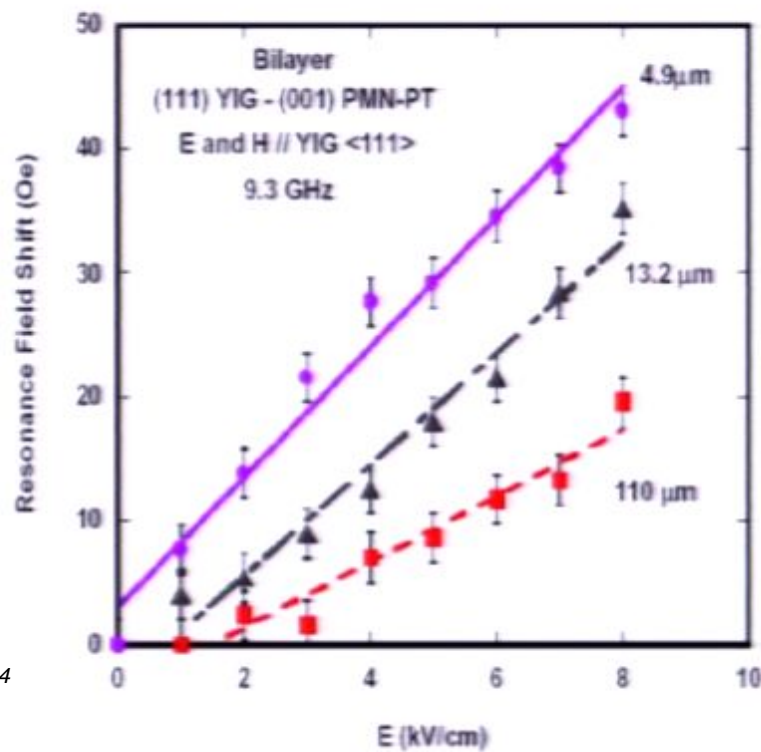
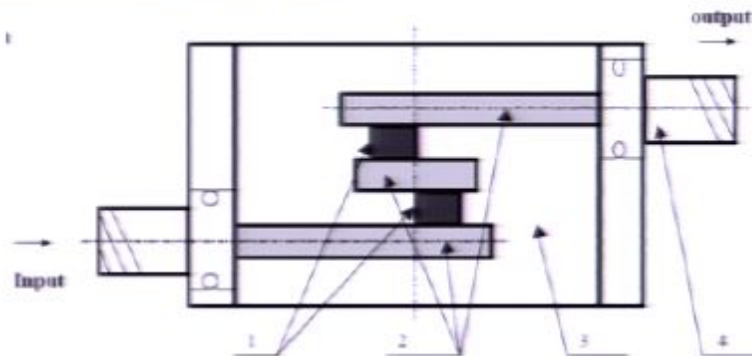
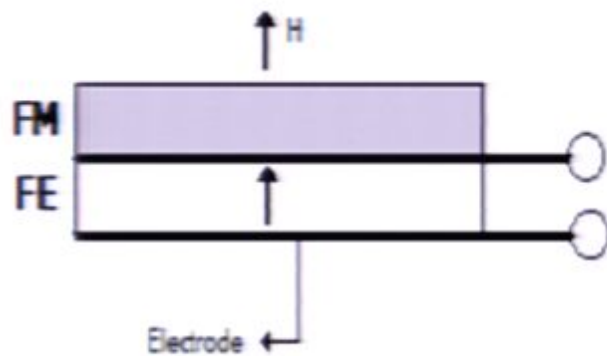
G. Srinivasan et al *Electronics Letters* (2005)





# Electric Field tuned RF filter

G. Srinivasan et al *Electronics Letters* (2005)



# Collaborators

## TbMnO<sub>3</sub>

M. Kenzelmann  
A. B. Harris  
S. Jonas  
C. Broholm  
J. Schefer  
S. B. Kim  
C. L. Zhang  
S.-W. Cheong  
O. P. Vajk  
J. W. Lynn  
T. Yildirim

## RbFe(MoO<sub>4</sub>)<sub>2</sub>

M. Kenzelmann  
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A.B. Harris  
G. Gasparovic  
C. Broholm  
A.P. Ramirez  
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A. Smirnov  
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R. J. Cava  
A. Aharony  
O. Entin-Wohlman  
T. Yildirim  
M. Kenzelmann  
C. Broholm  
A. P. Ramirez  
W. Chen

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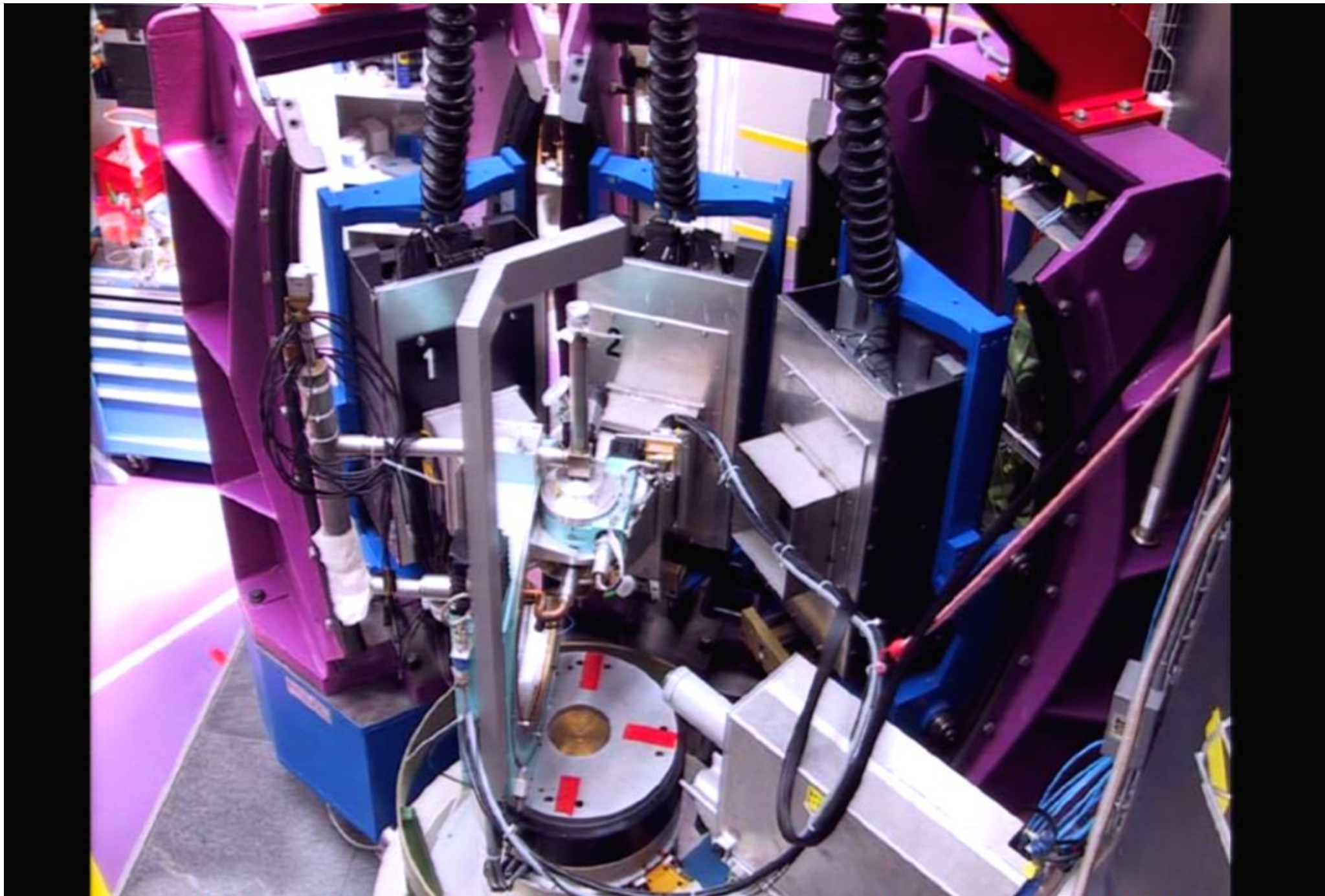
M. Kenzelmann

C. Broholm

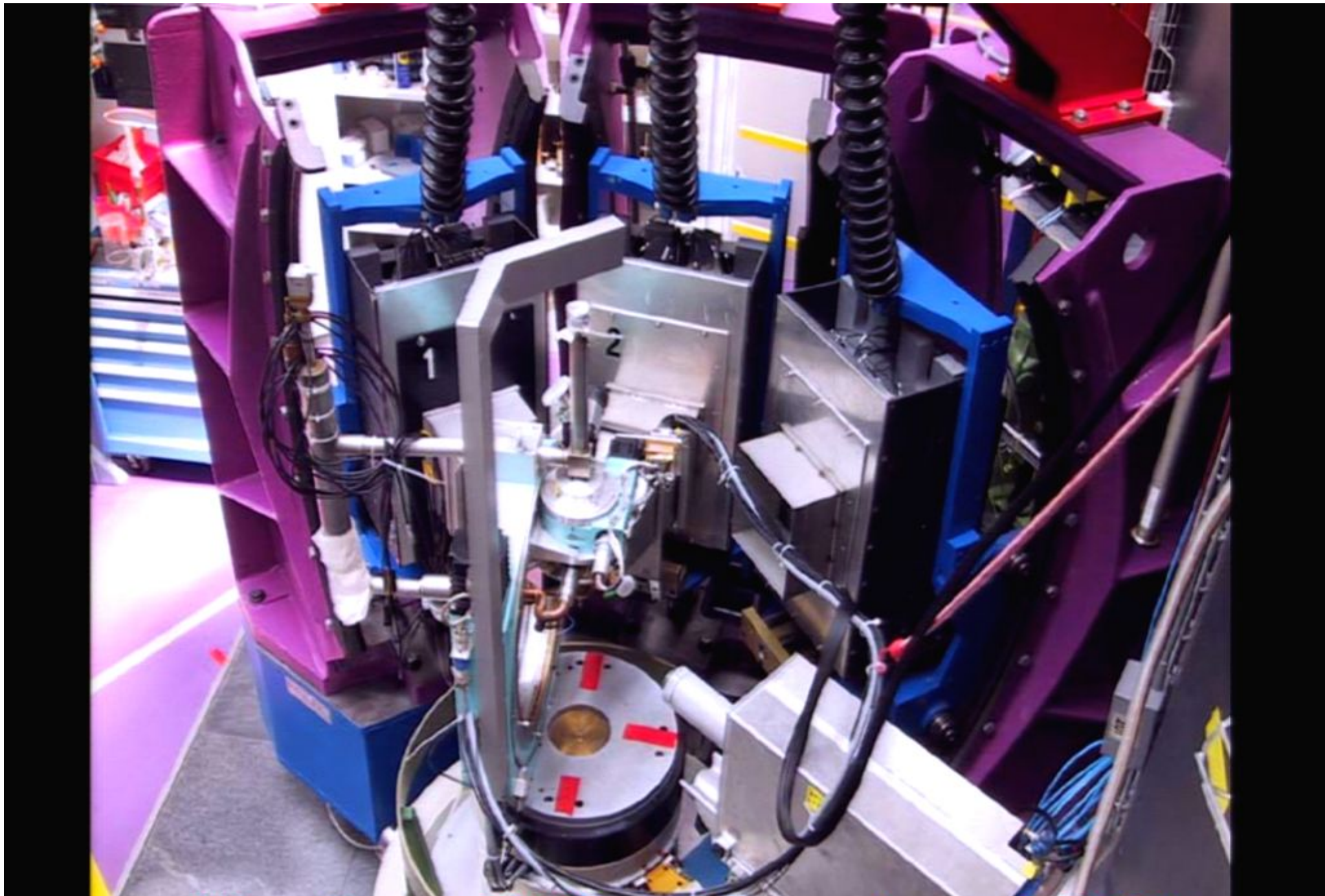
A. P. Ramirez

W. Chen









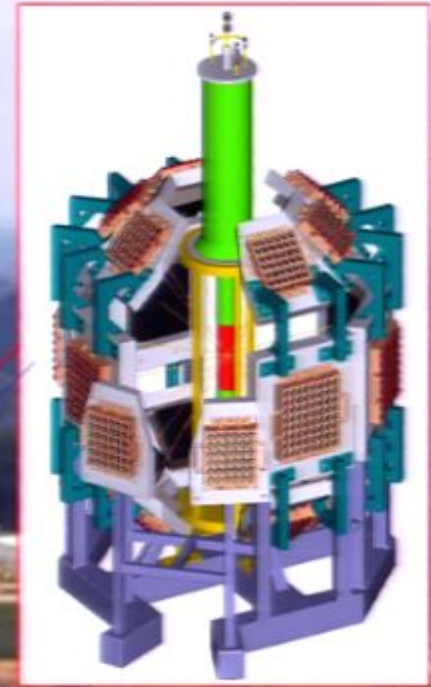


# Spallation Neutron Source at ORNL, TN

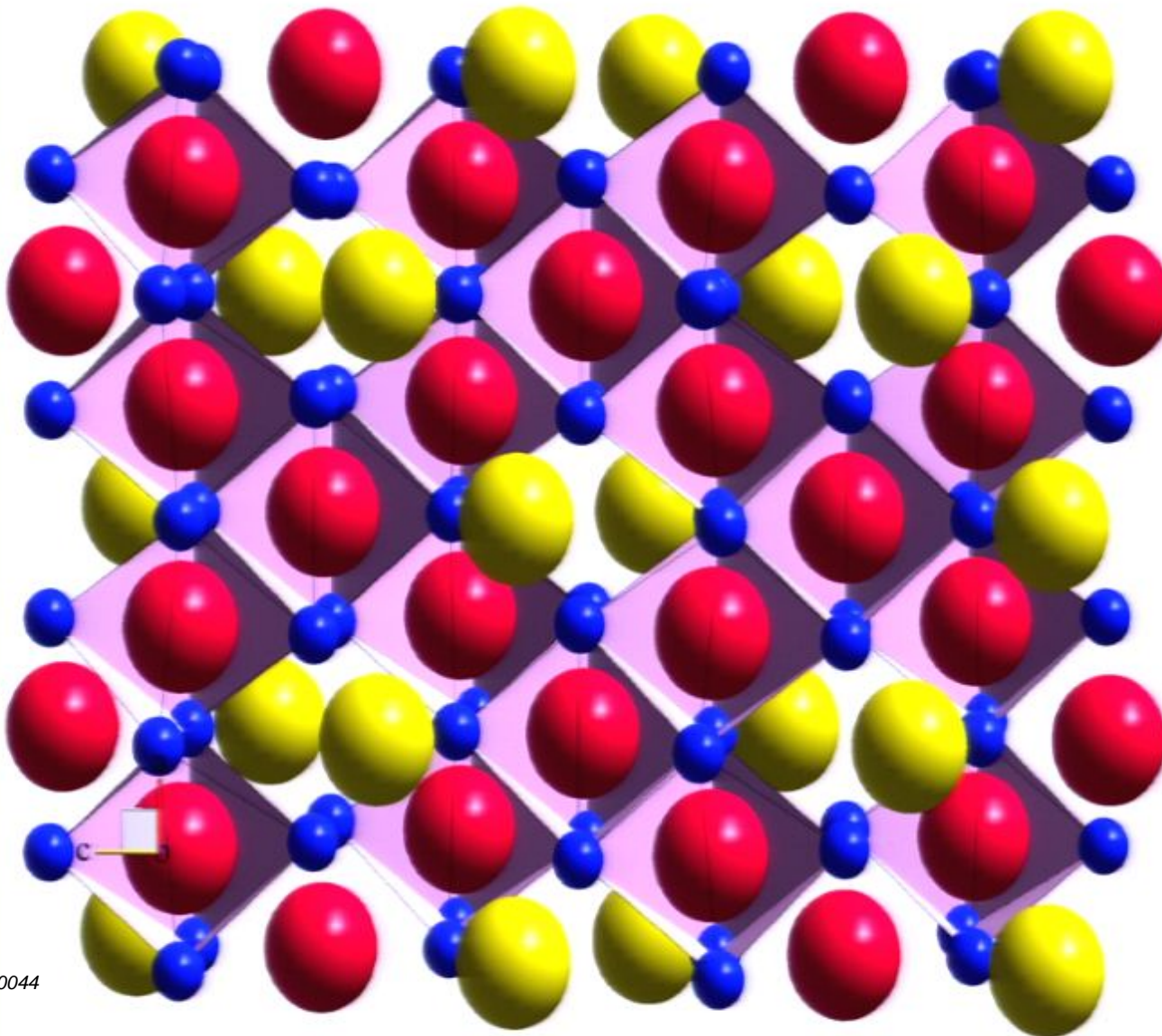




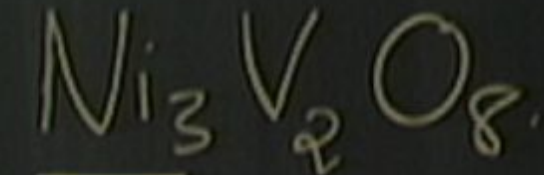
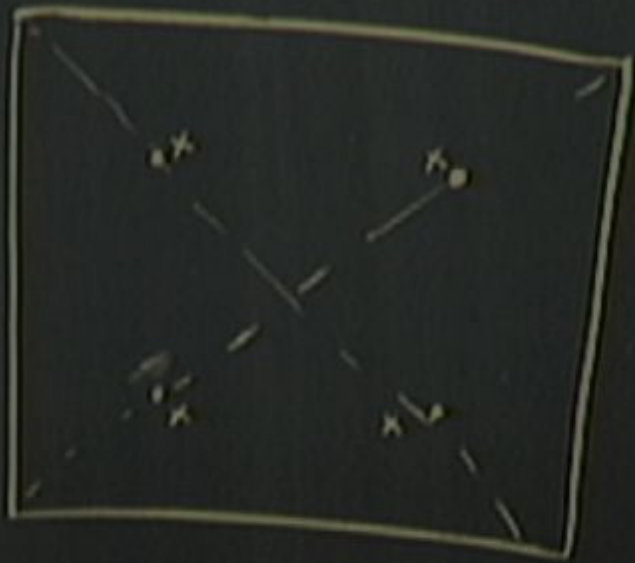
# Spallation Neutron Source at ORNL, TN



# A Kagome Staircase AFM

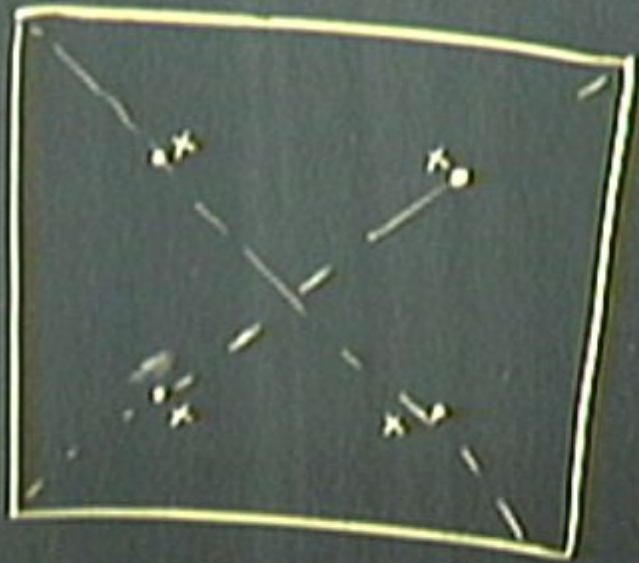






$$\underline{\underline{S=1}}$$

$$\Gamma(E) \sim E^{1+0.05}$$

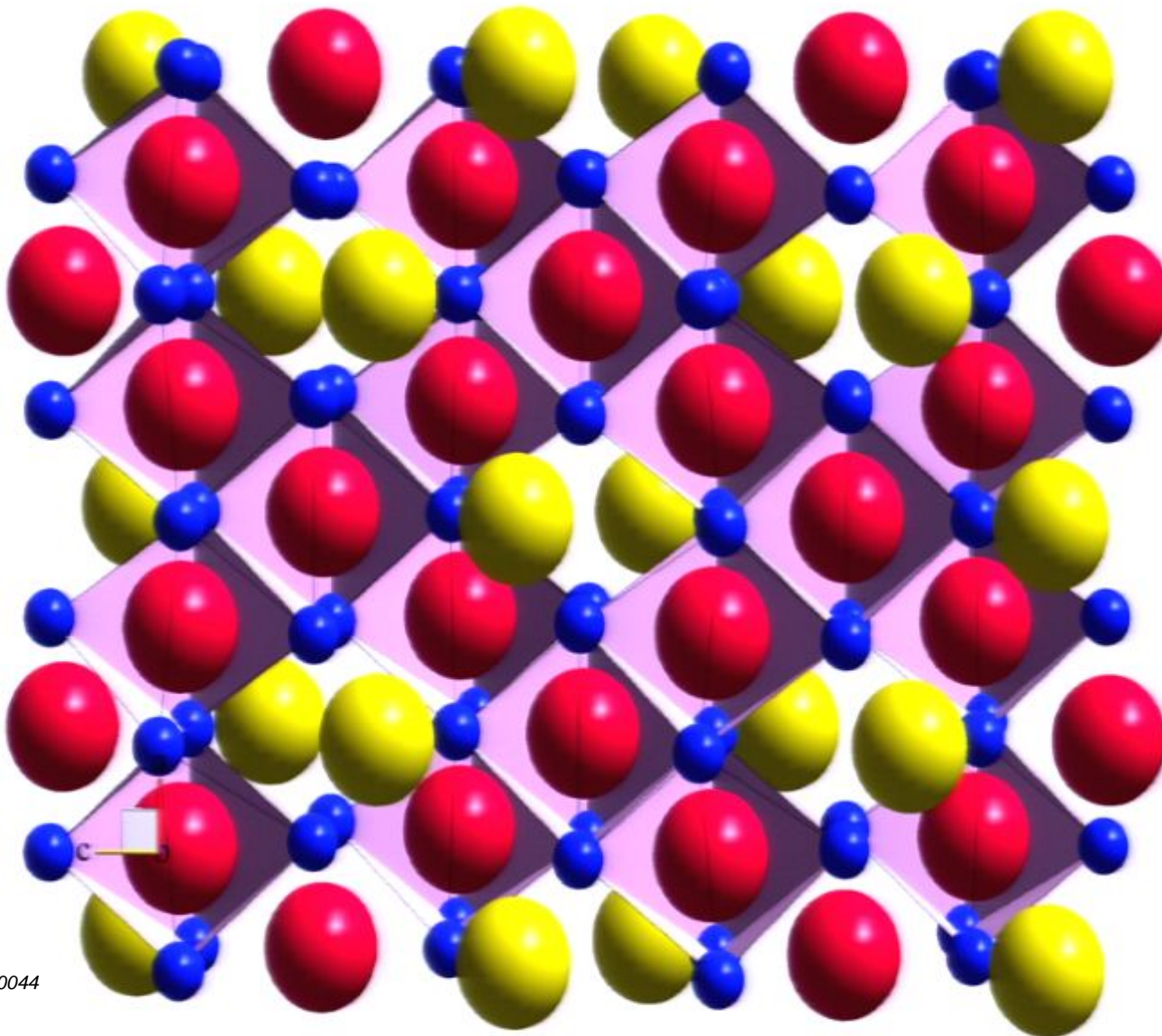


$$\underline{\underline{S=1}}$$

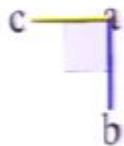
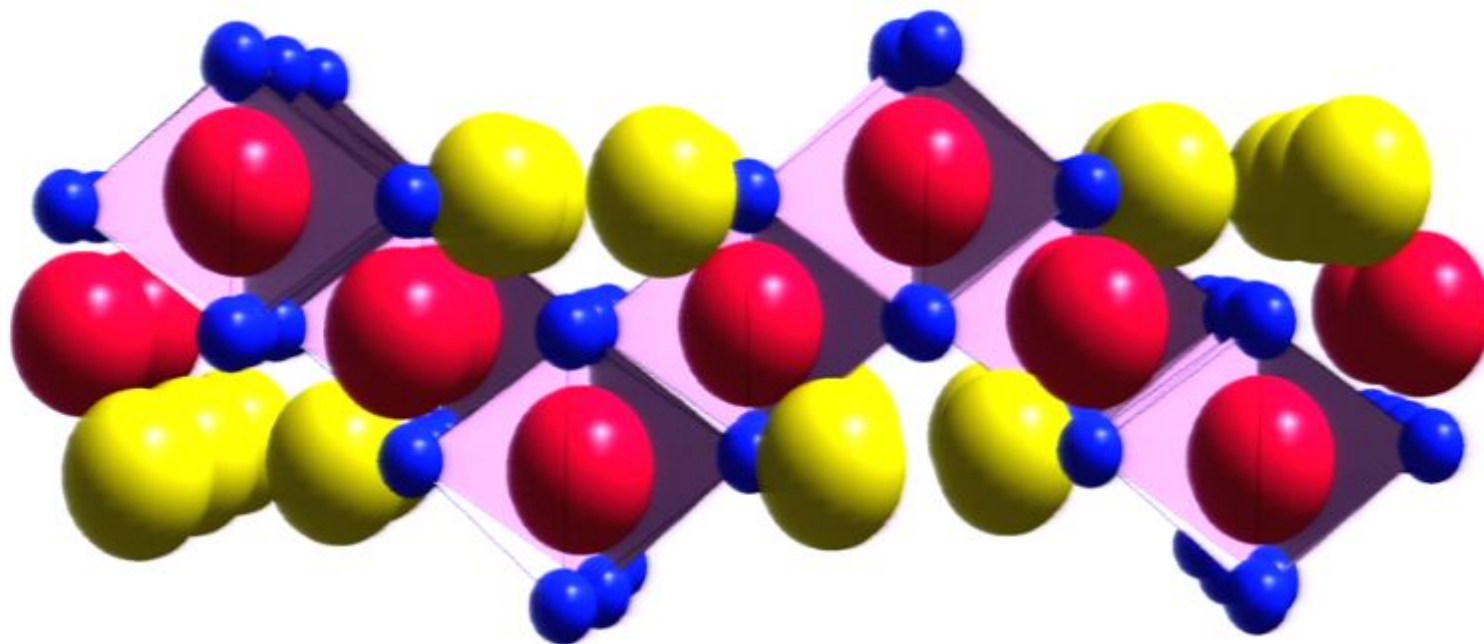
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# A Kagome Staircase AFM

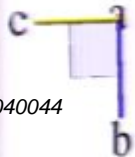
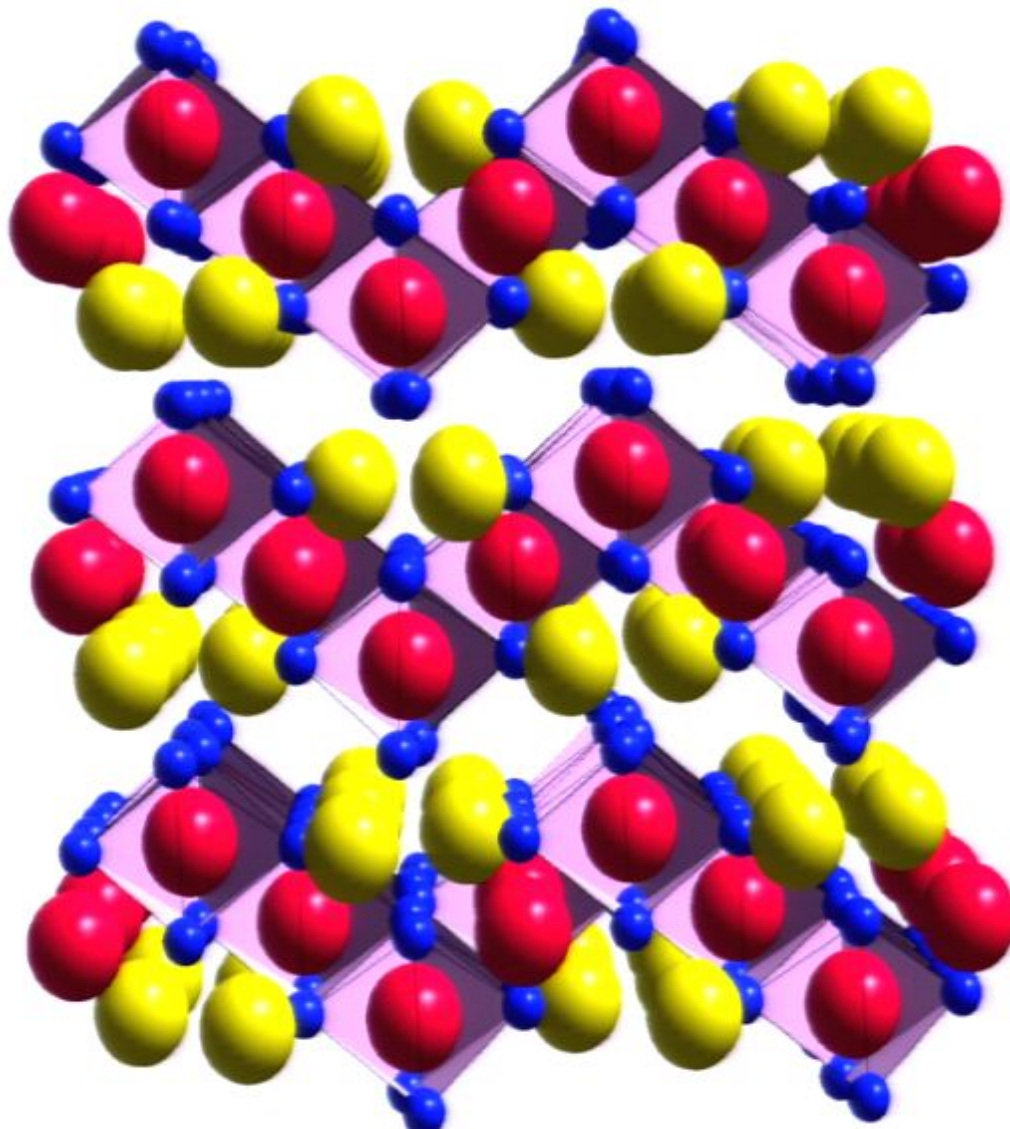


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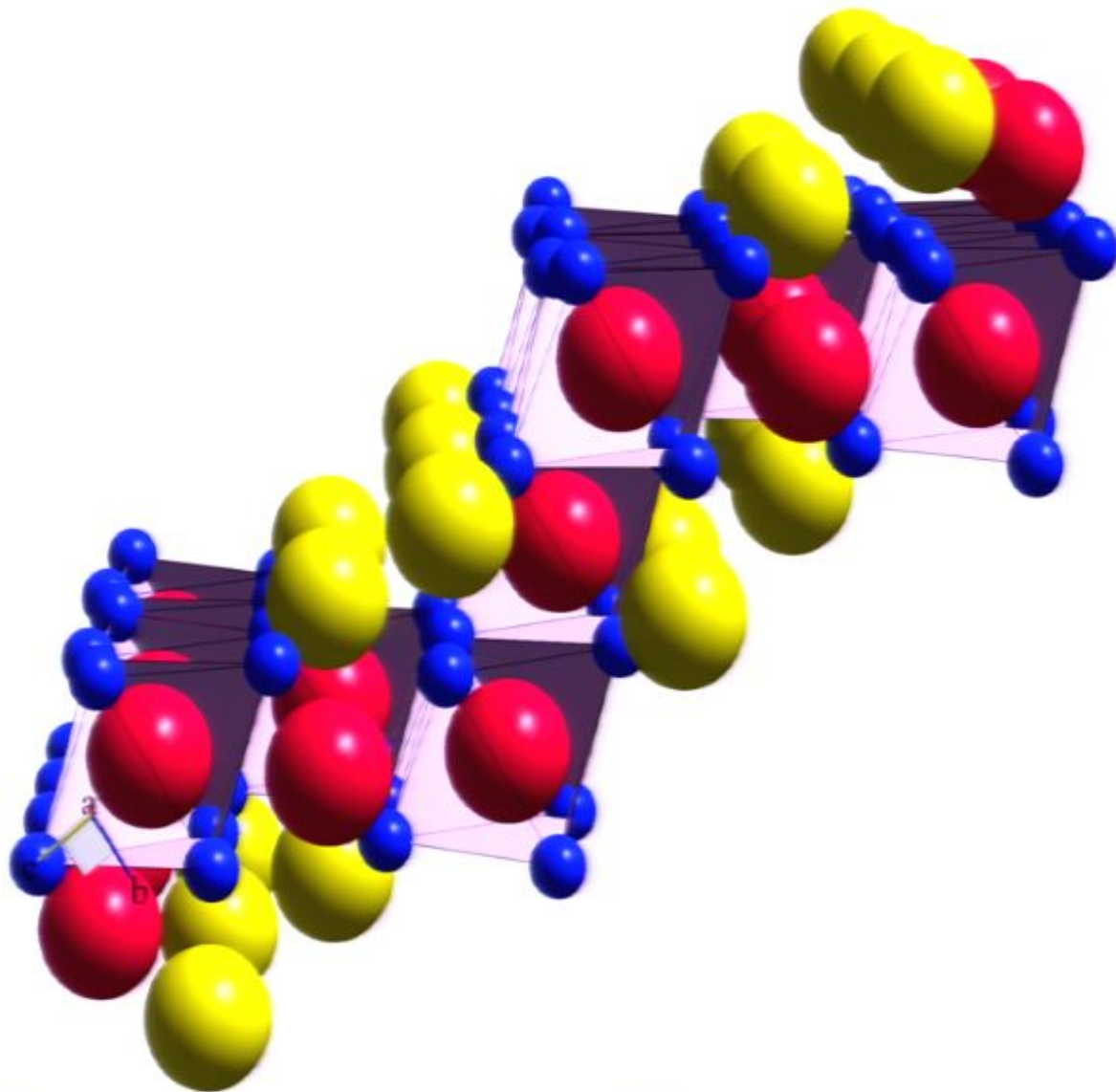




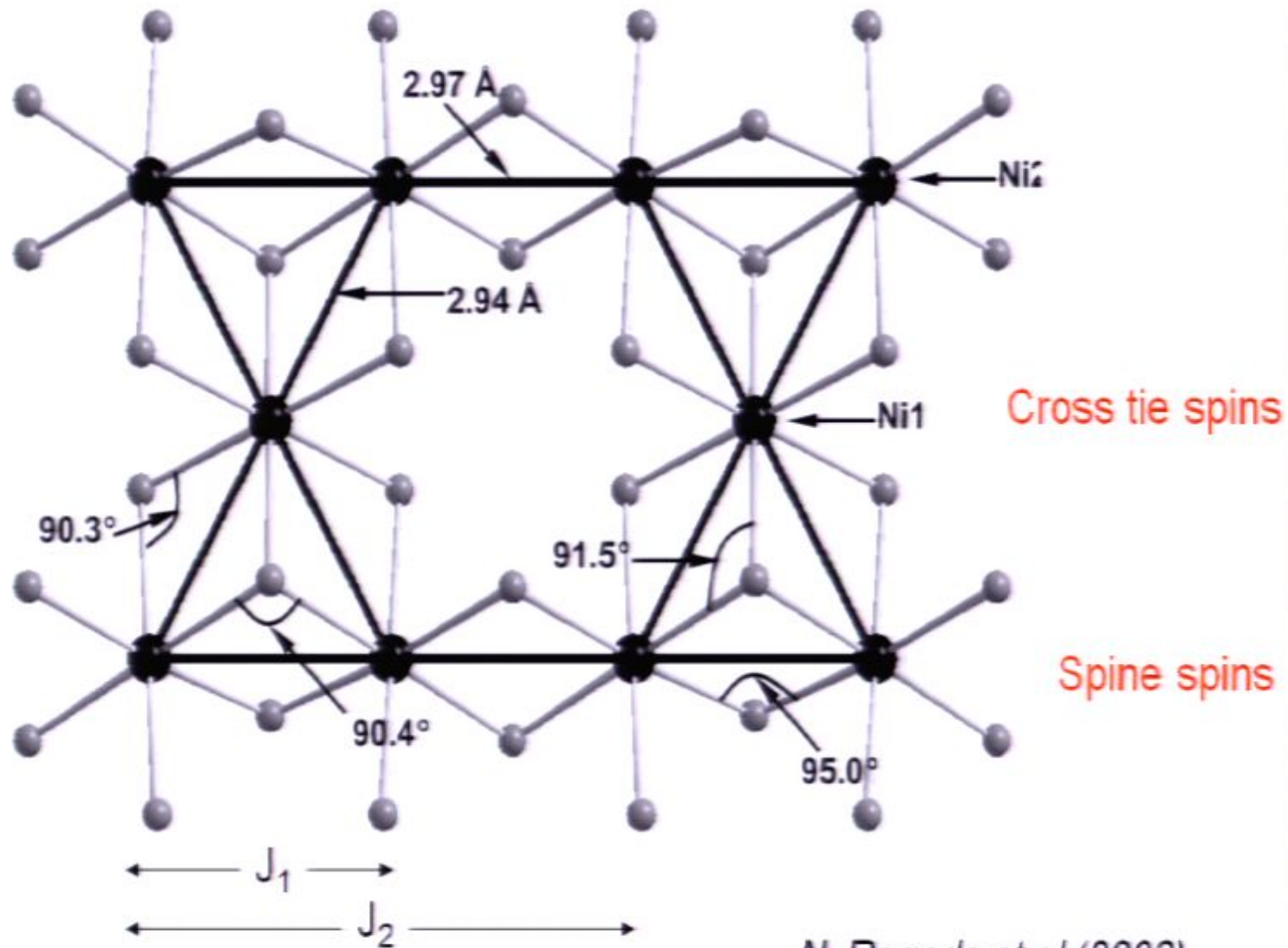
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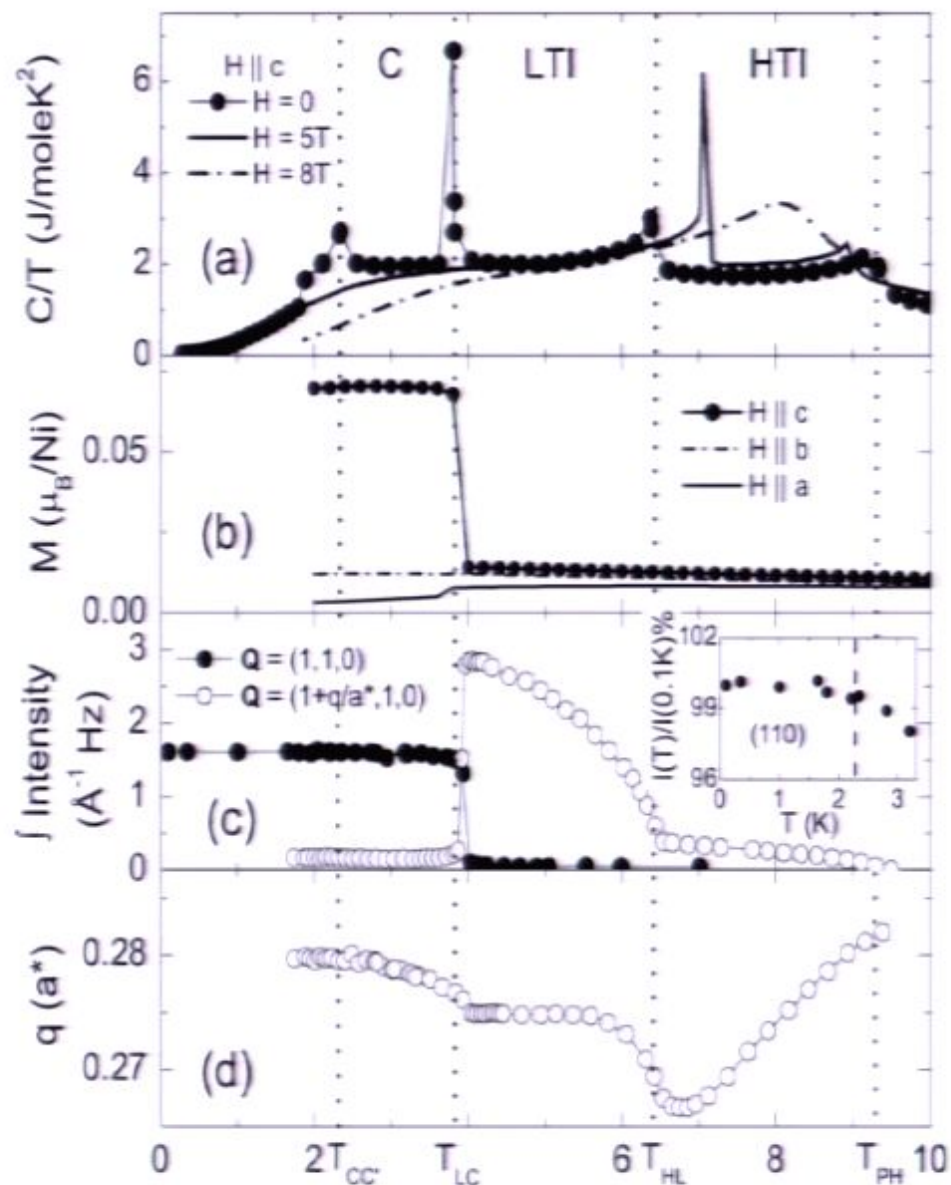
# Exchange Interactions



*N. Rogado et al (2003)*

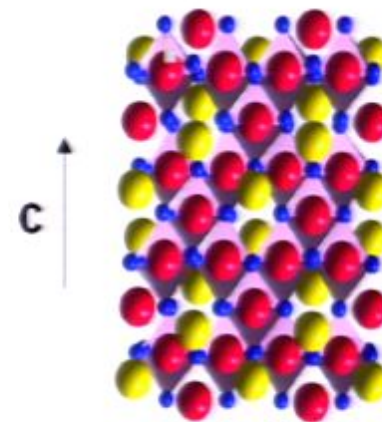
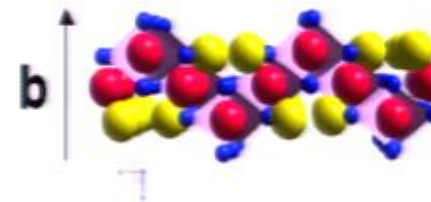
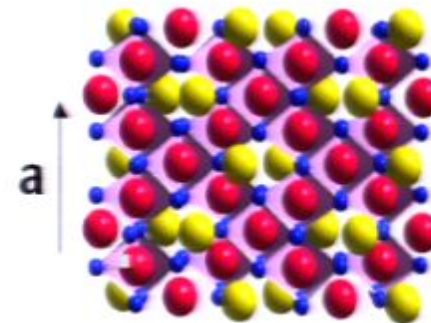
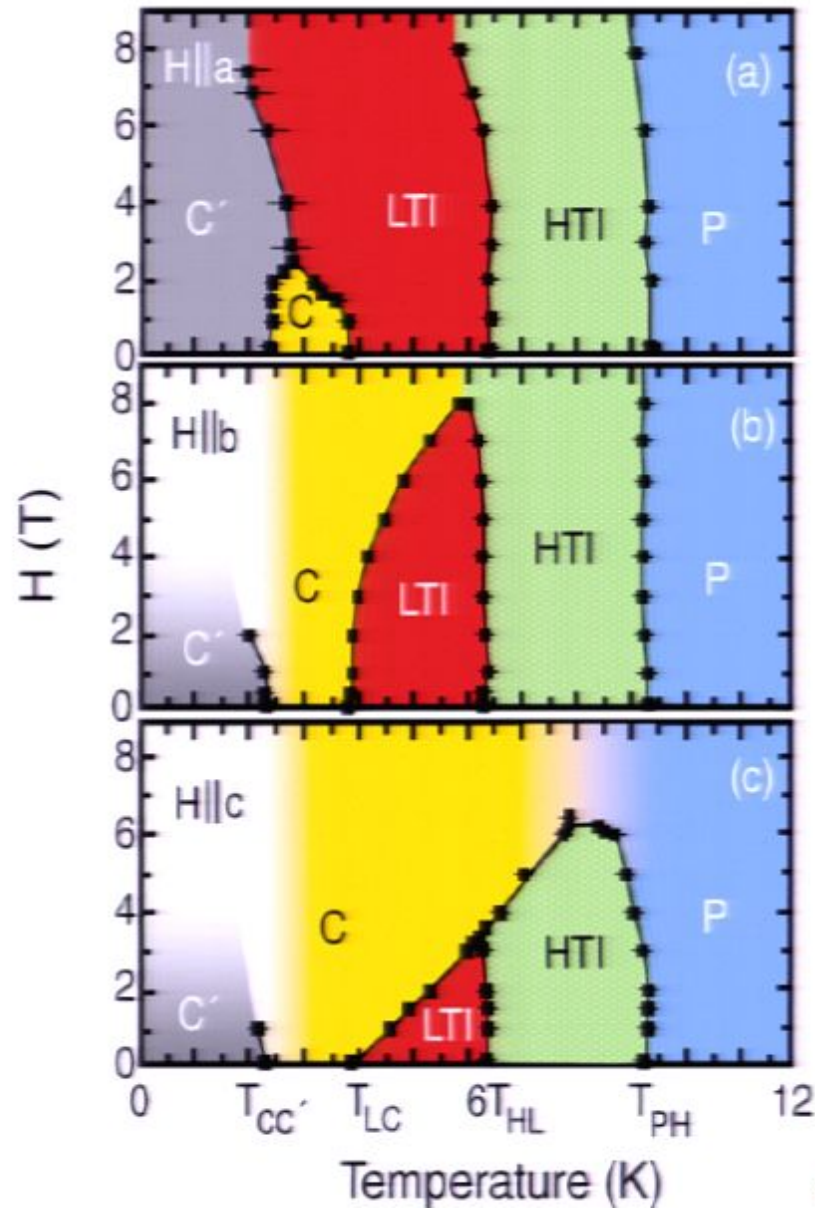


# Multiple Magnetic Phases





# Compiled Phase Diagram

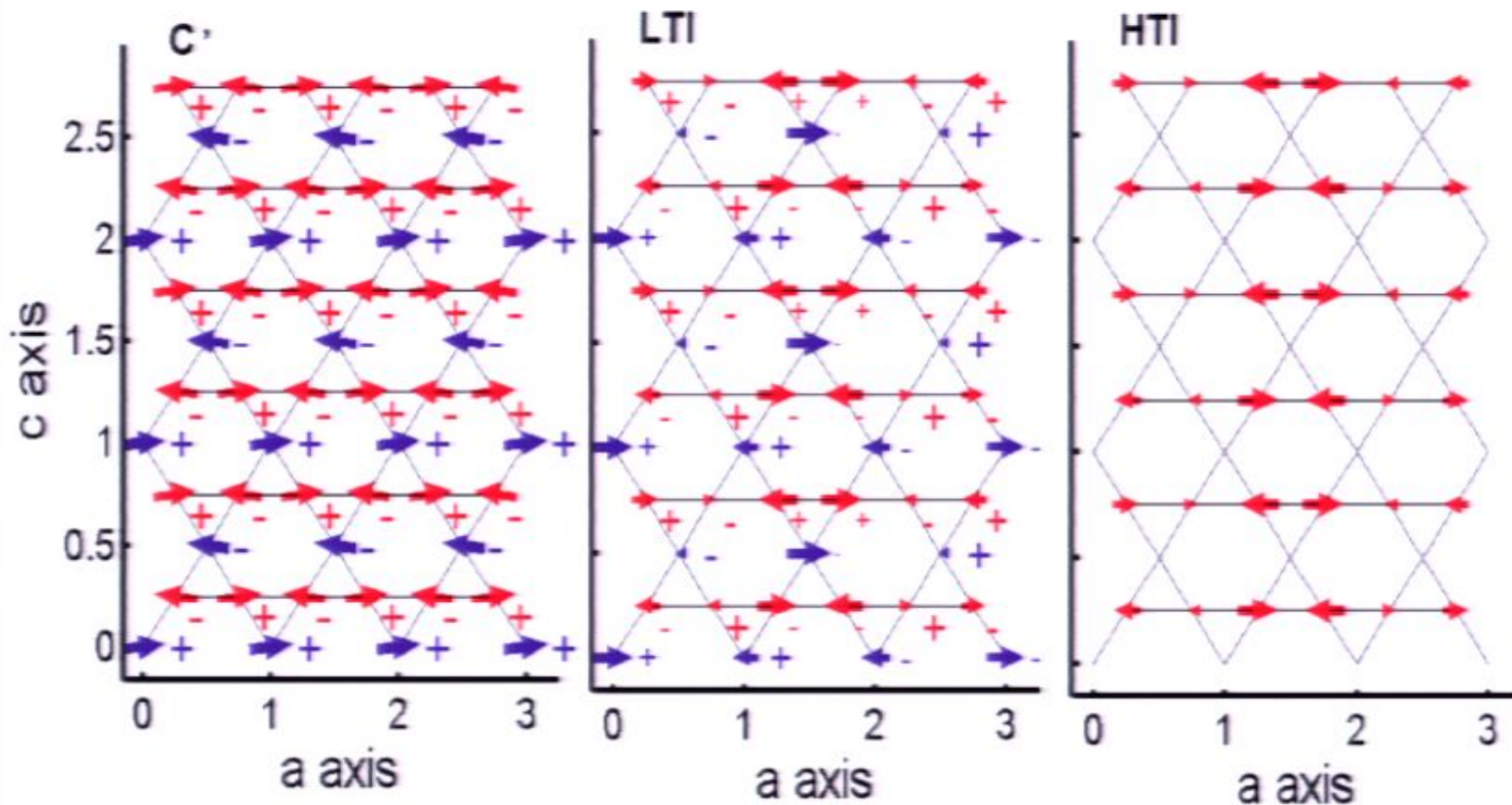


# Modulated magnetic phases

2.2 K < T < 4 K

4 K < T < 6.5 K

6.5 K < T < 9.2 K

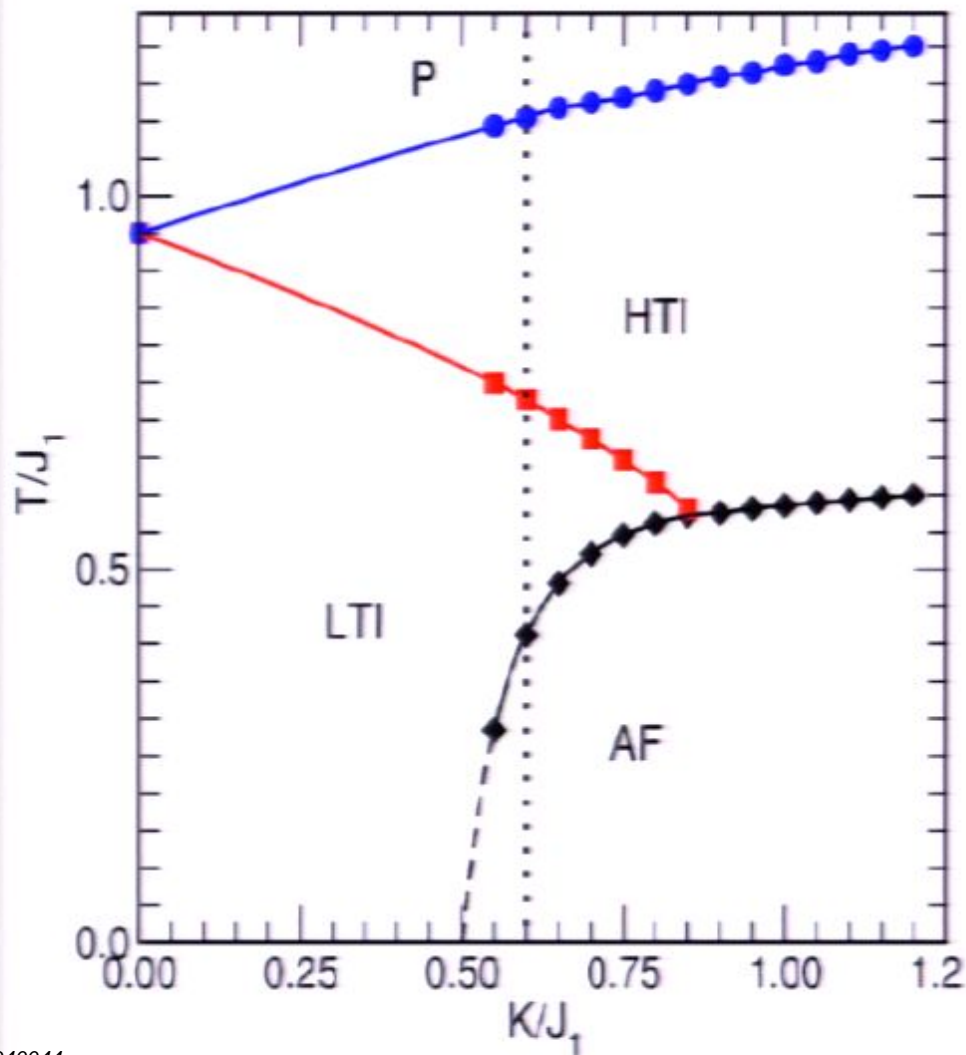


Commensurate  
Canted FM

Incommensurate  
Cycloidal

Incommensurate  
amplitude modulated

# Mean Field "ANNNI" Phase Diagram



$$\mathcal{H} = -K \sum_n S_a(n)^2 + J_1 \sum_n \mathbf{S}(n) \cdot \mathbf{S}(n+1) + J_2 \sum_n \mathbf{S}(n) \cdot \mathbf{S}(n+2).$$

$$\cos(\pi q_0 \alpha) = -J_{1\alpha} / (4J_{2\alpha})$$

$$J_1 / J_2 = 2.56$$

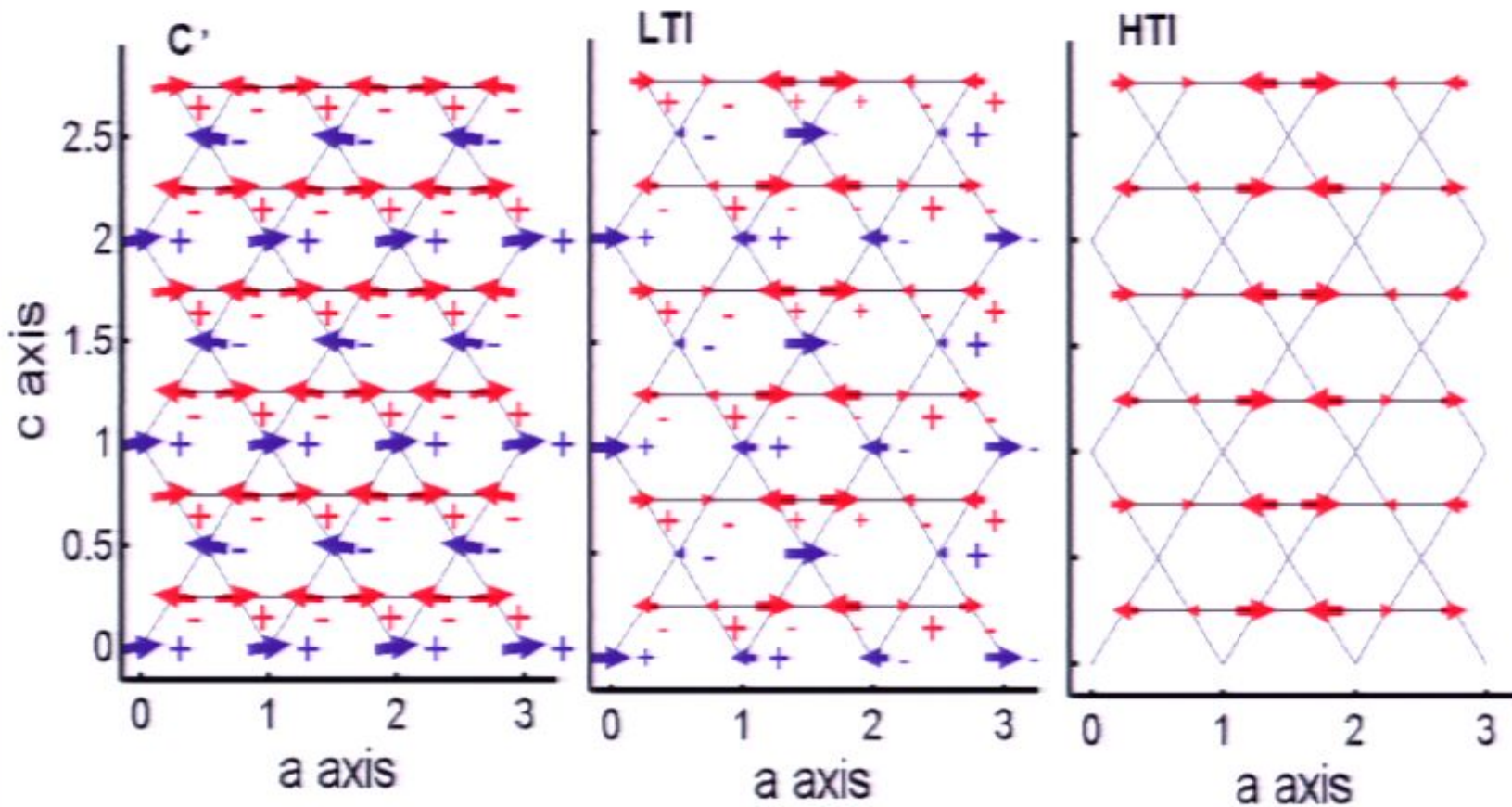


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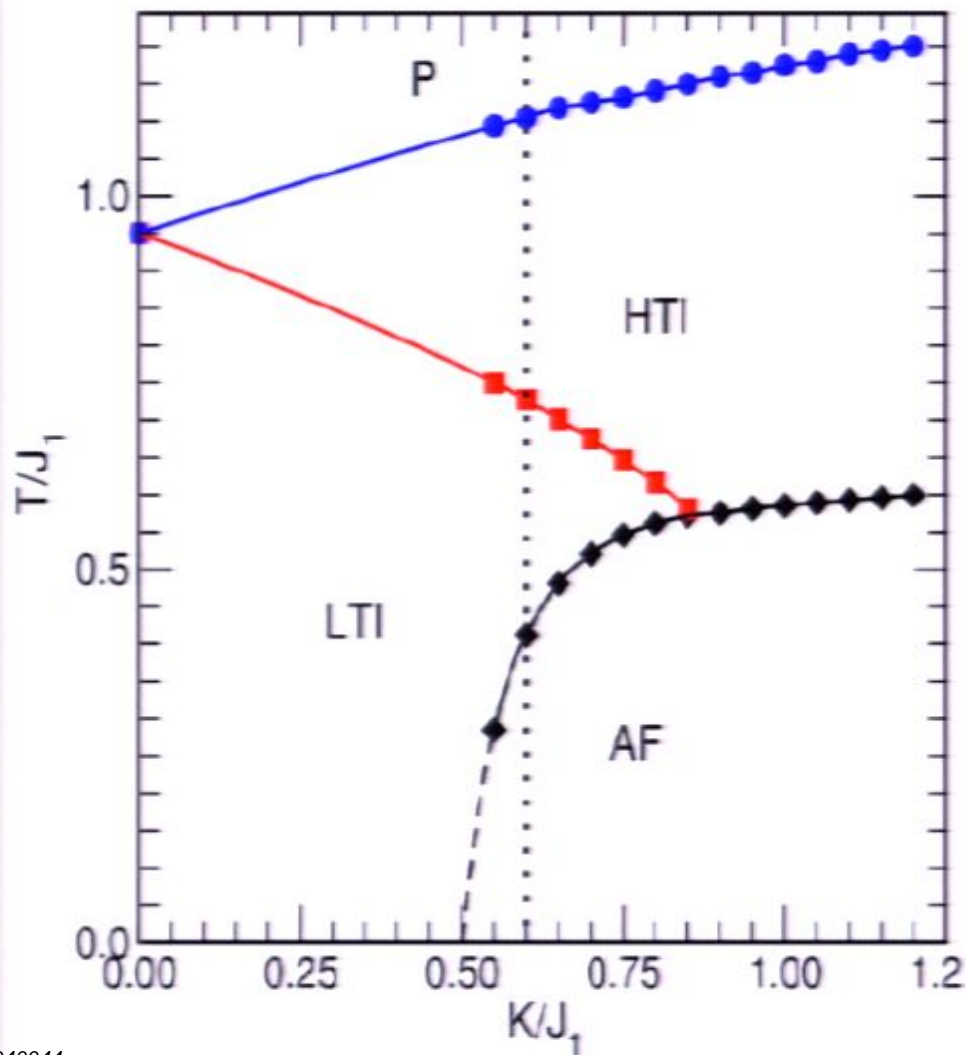


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# Mean Field "ANNNI" Phase Diagram

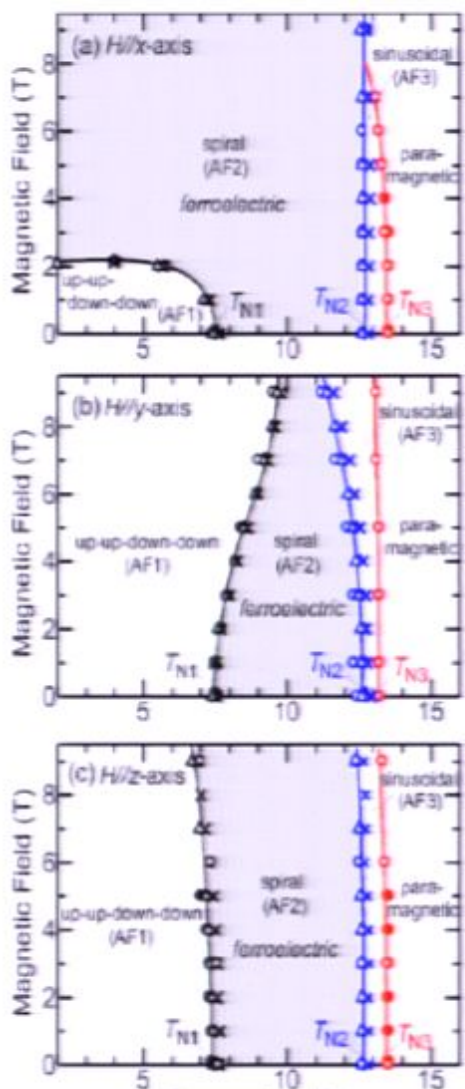


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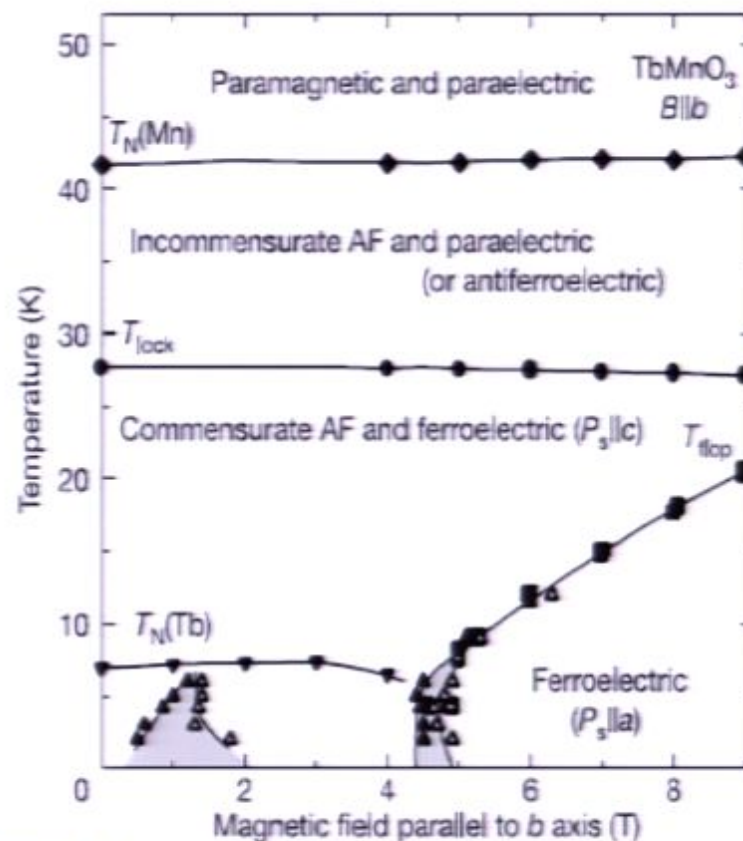
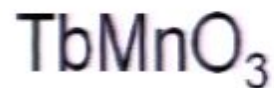
$$\cos(\pi q_0 \alpha) = -J_{1\alpha} / (4J_{2\alpha})$$

$$J_1 / J_2 = 2.56$$

# Related systems



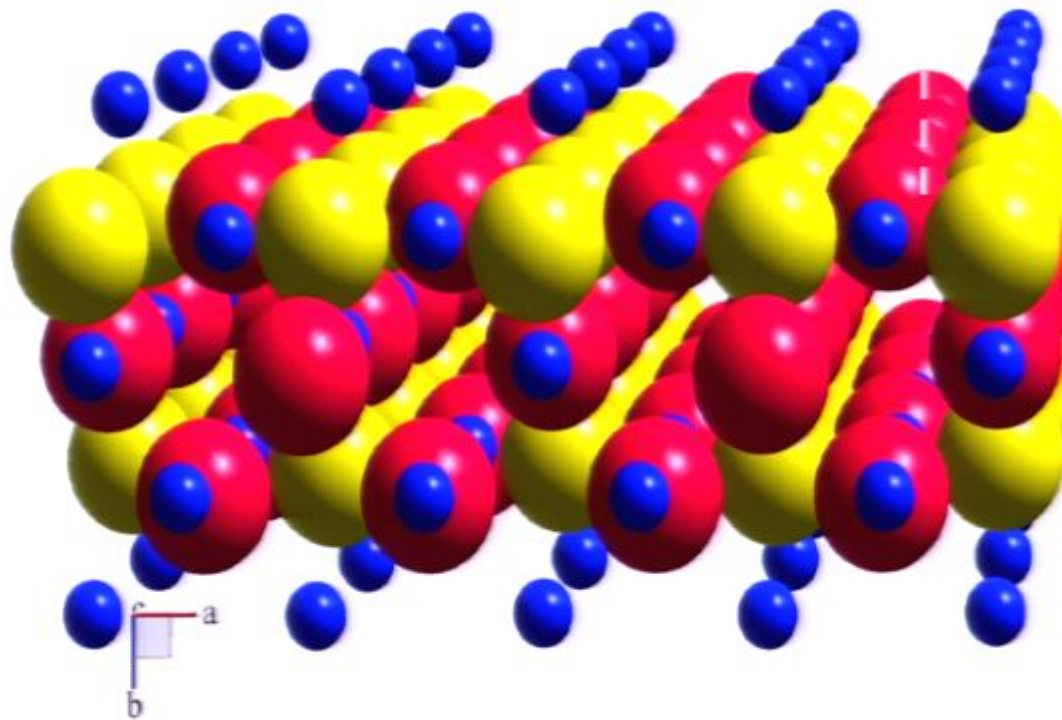
Arkenbout et al. PRB (2006)



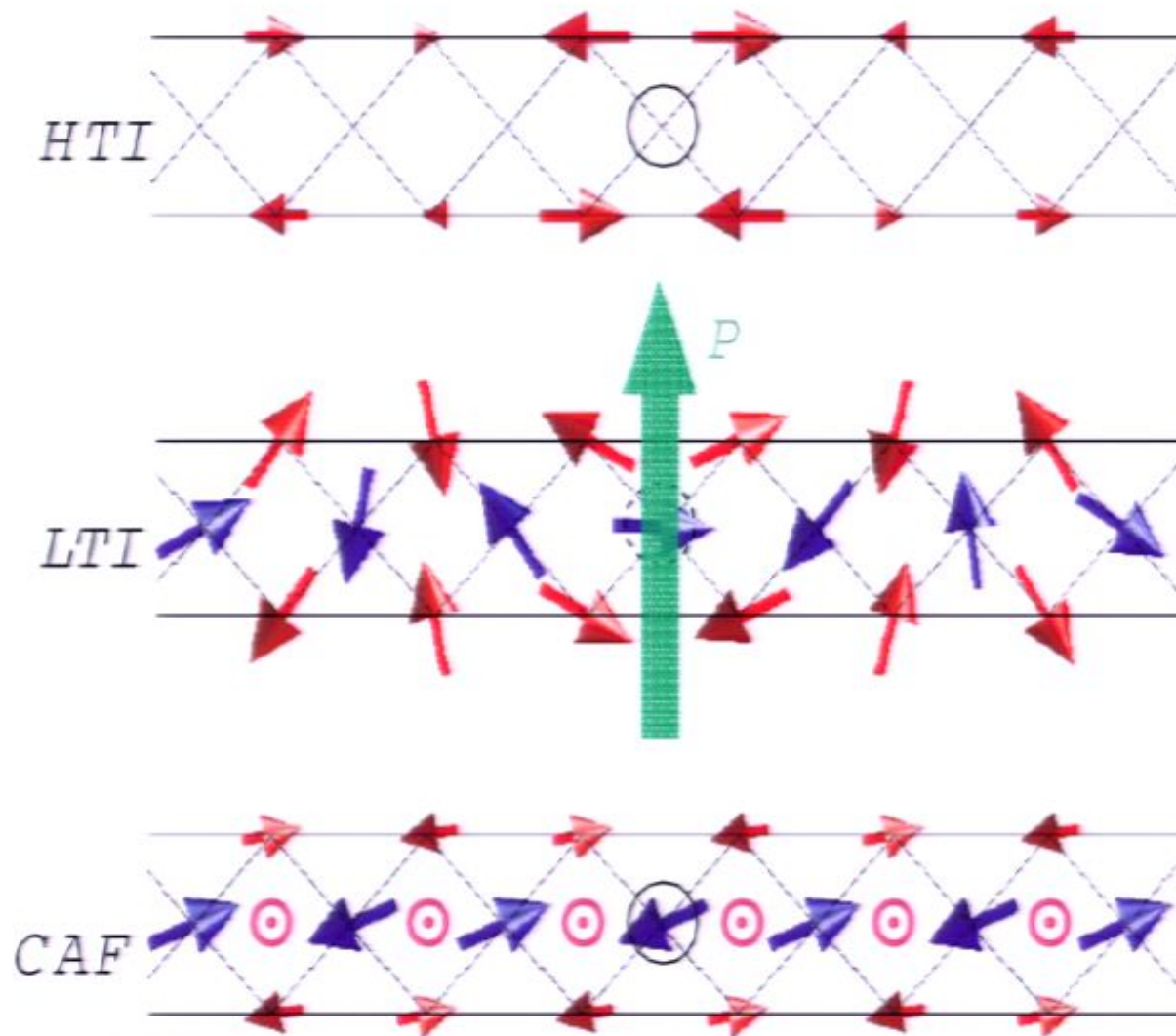
Kimura et al. Nature (2003)



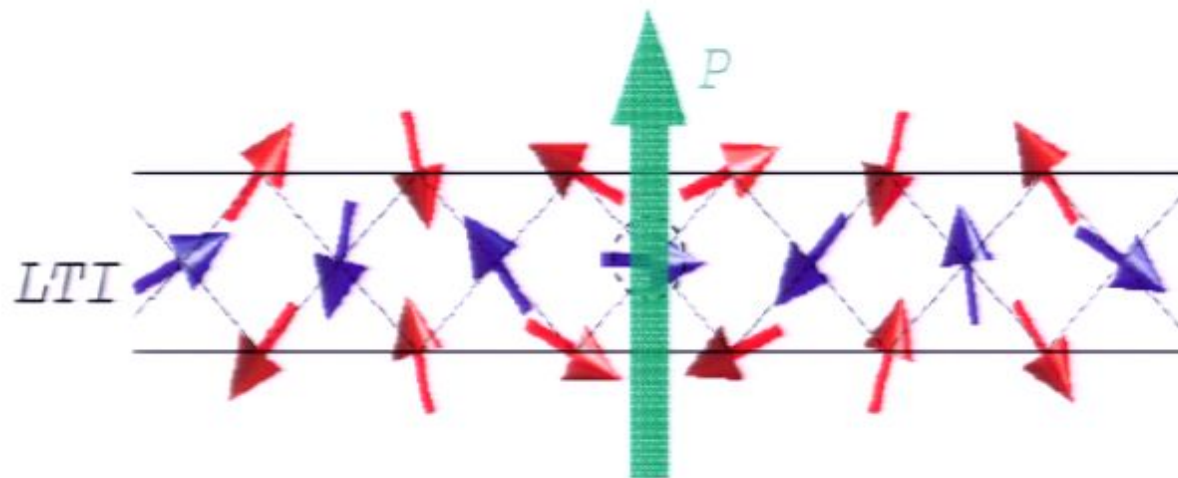
# Ferro-electricity on a kagome' staircase



# Ferro-electricity on a kagome' staircase

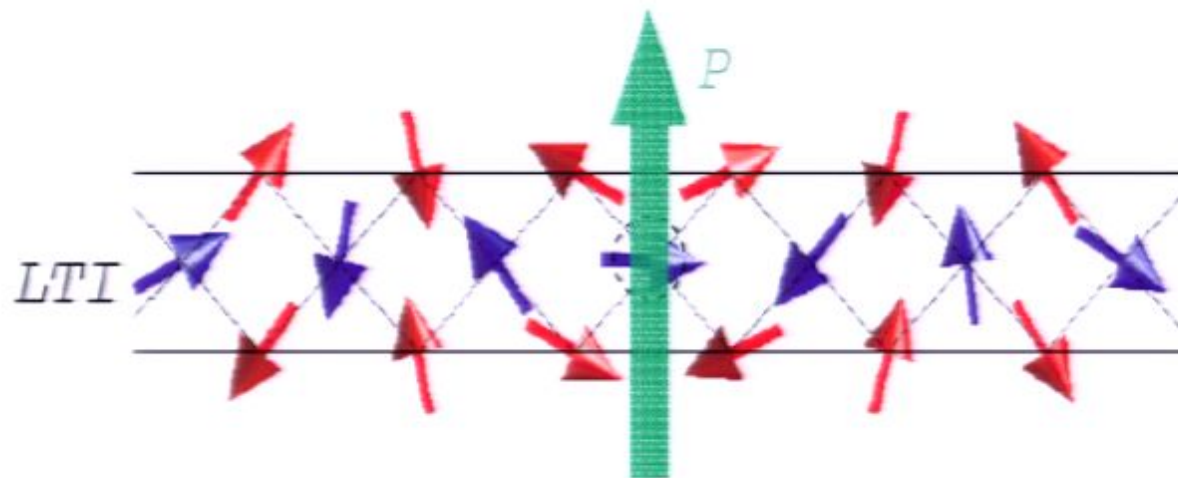


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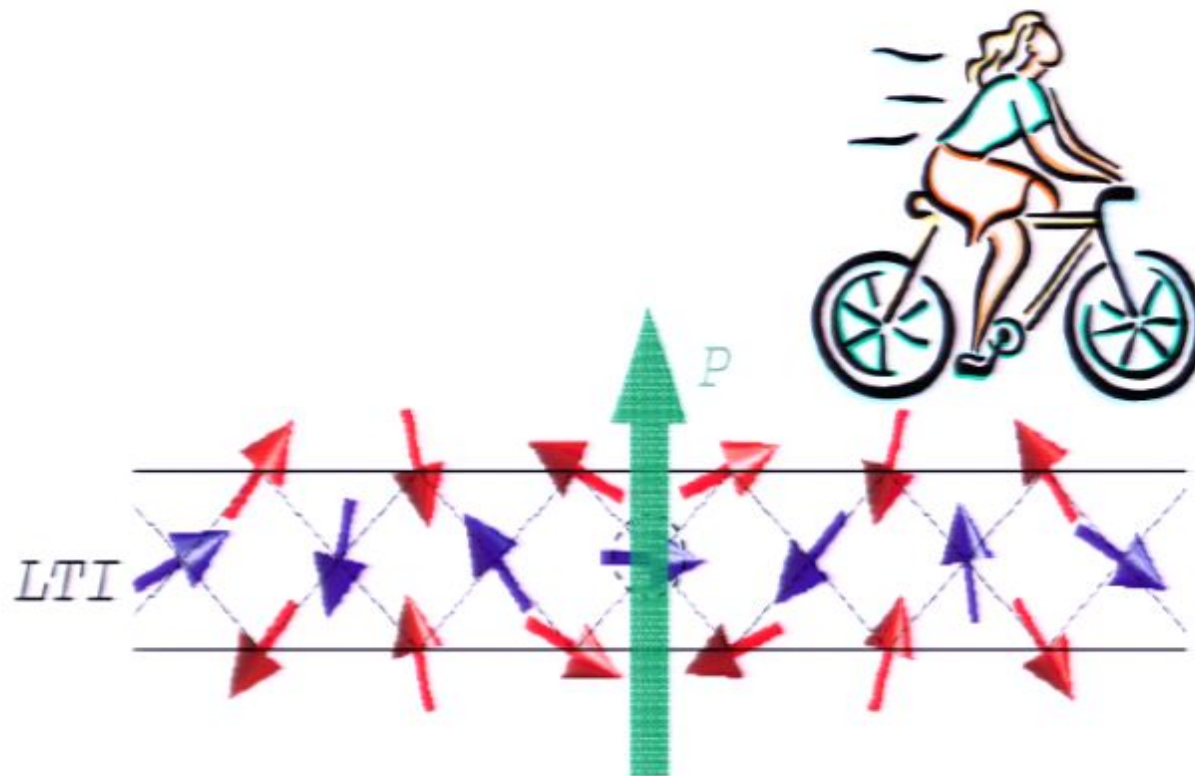




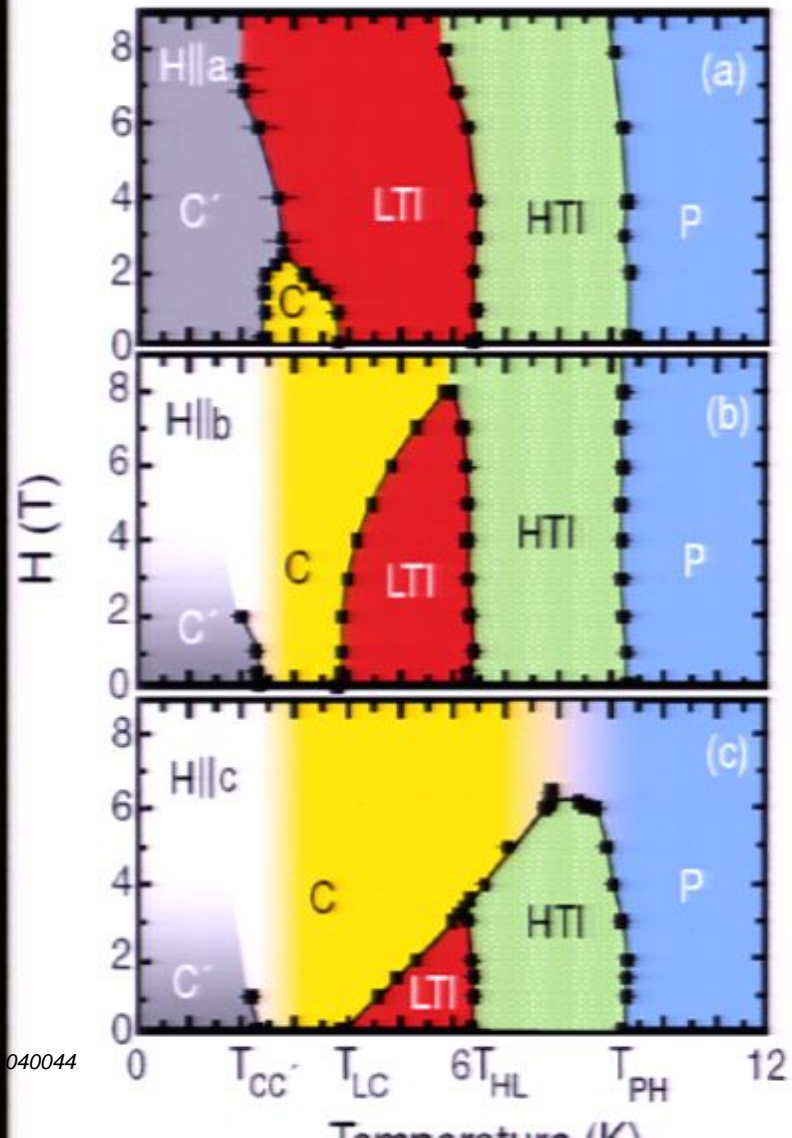
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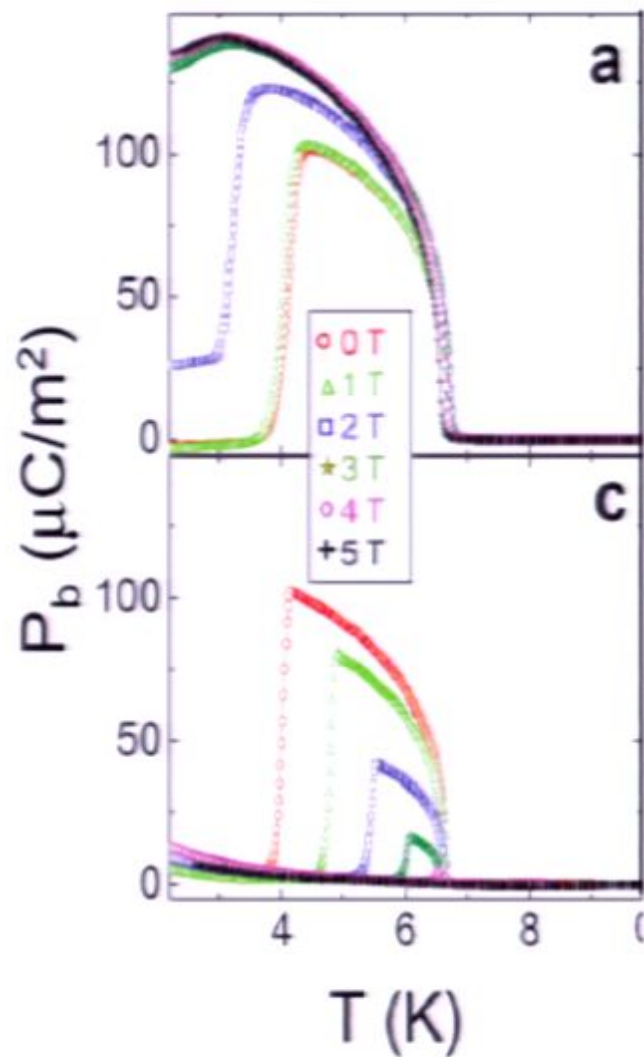
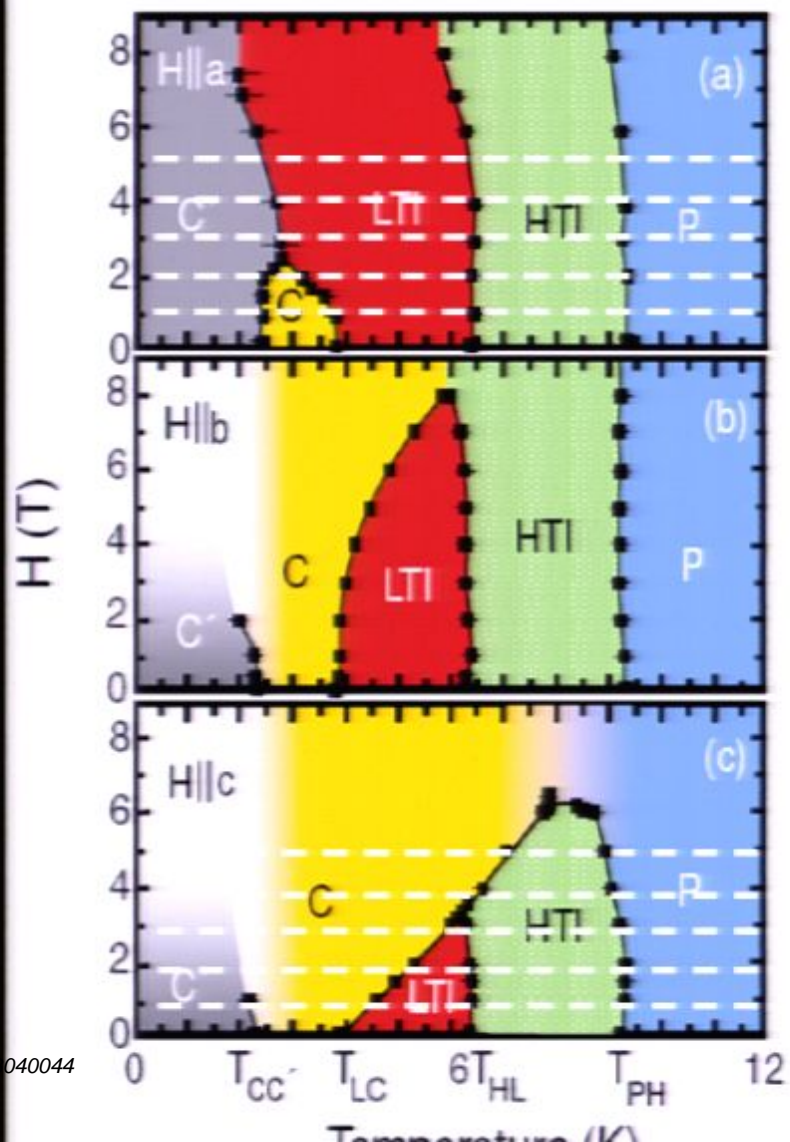


# Magnetism on a kagome' staircase

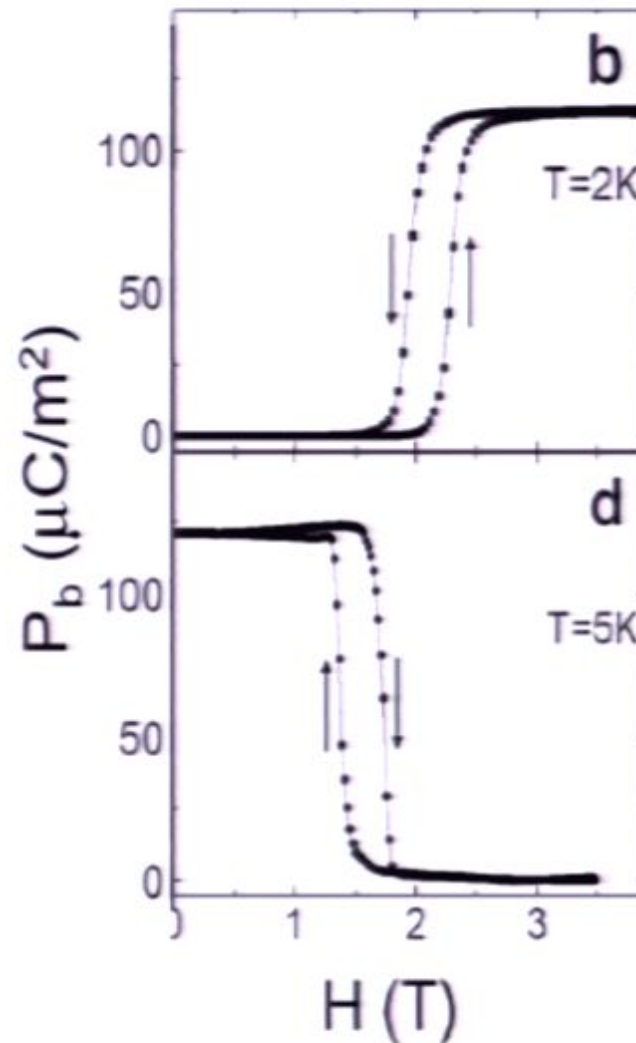
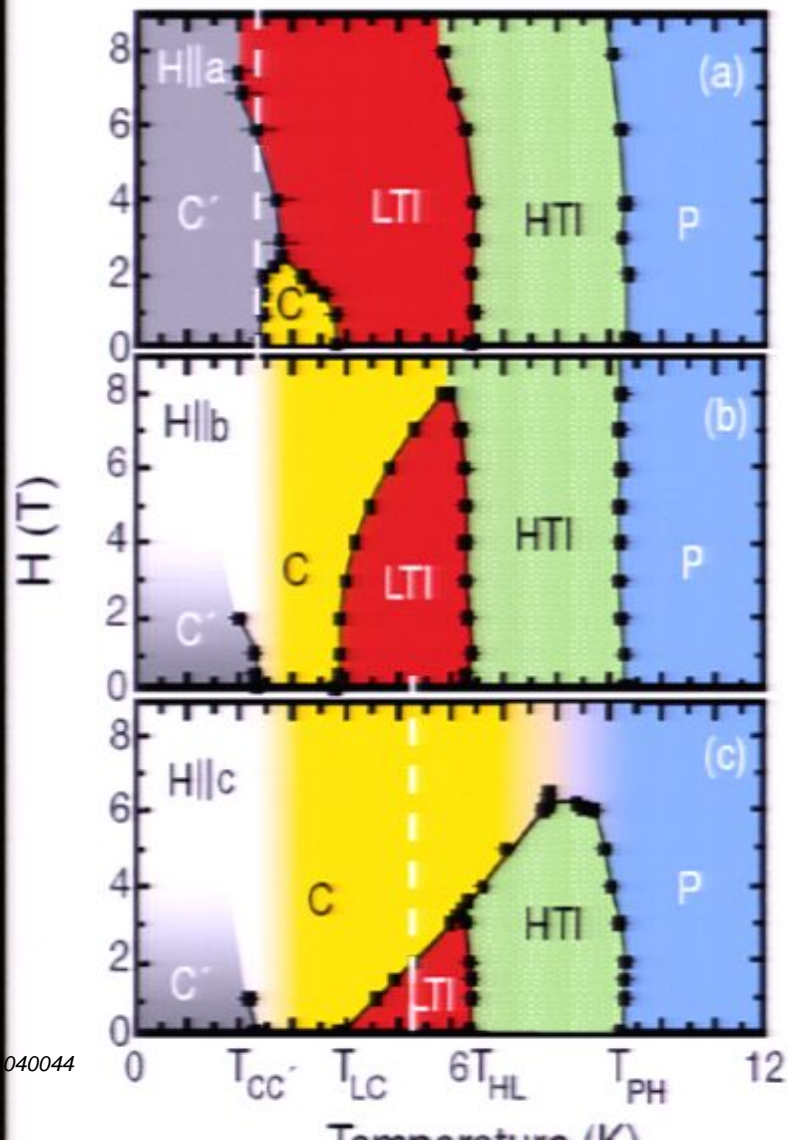




# Ferro-electricity on a kagome' staircase



# Ferro-electricity on a kagome' staircase



# Phenomenological observations

- Polarization direction is consistent with spin current model:

$$\mathbf{P} \propto \gamma \mathbf{e}_{ij} \times (\mathbf{S}_i \times \mathbf{S}_j)$$

- The magneto-electric phase is an incommensurate cycloid phase
- Magneto-electric phase develops from existing incommensurate order



# Landau Theory of Magneto-Electricity

A. B. Harris [arXiv:cond-mat/0610241](https://arxiv.org/abs/cond-mat/0610241)

General case:

$$V = \sum_{nm\gamma} c_{nm\gamma} \sigma_n(\mathbf{q}) \sigma_m(-\mathbf{q}) P_\gamma$$

IRREP	1	$2_X$	$\tilde{m}_y$	$\tilde{m}_z$
$\Gamma_1$	1	1	1	1
$\Gamma_2$	1	1	-1	-1
$\Gamma_3$	1	-1	1	-1
$\Gamma_4$	1	-1	-1	1

# Landau Theory of Magneto-Electricity

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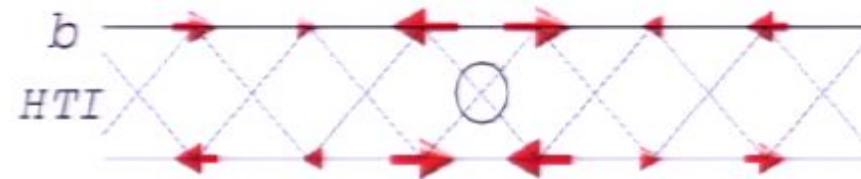
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$\Gamma_3$	1	-1	1	-1
$\Gamma_4$	1	-1	-1	1

Amplitude modulated ( $\Gamma_4$ )

$$V_{HTI} = \sum_\gamma c_{44\gamma} |\sigma_4(\mathbf{q})|^2 P_\gamma \equiv 0$$



# Landau Theory of Magneto-Electricity

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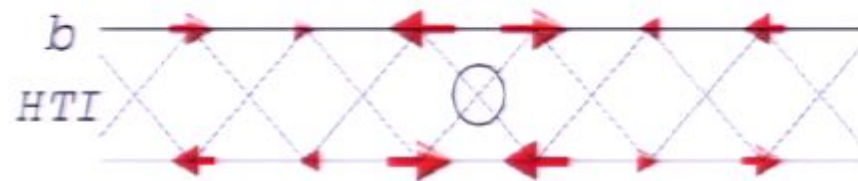
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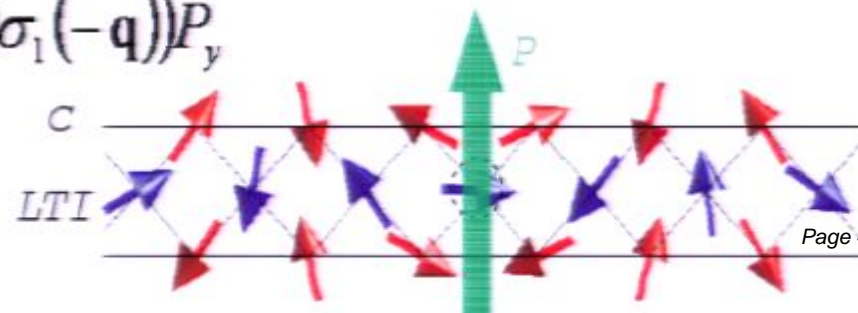
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$$V_{HTI} = \sum_{\gamma} c_{44\gamma} |\sigma_4(\mathbf{q})|^2 P_\gamma \equiv 0$$



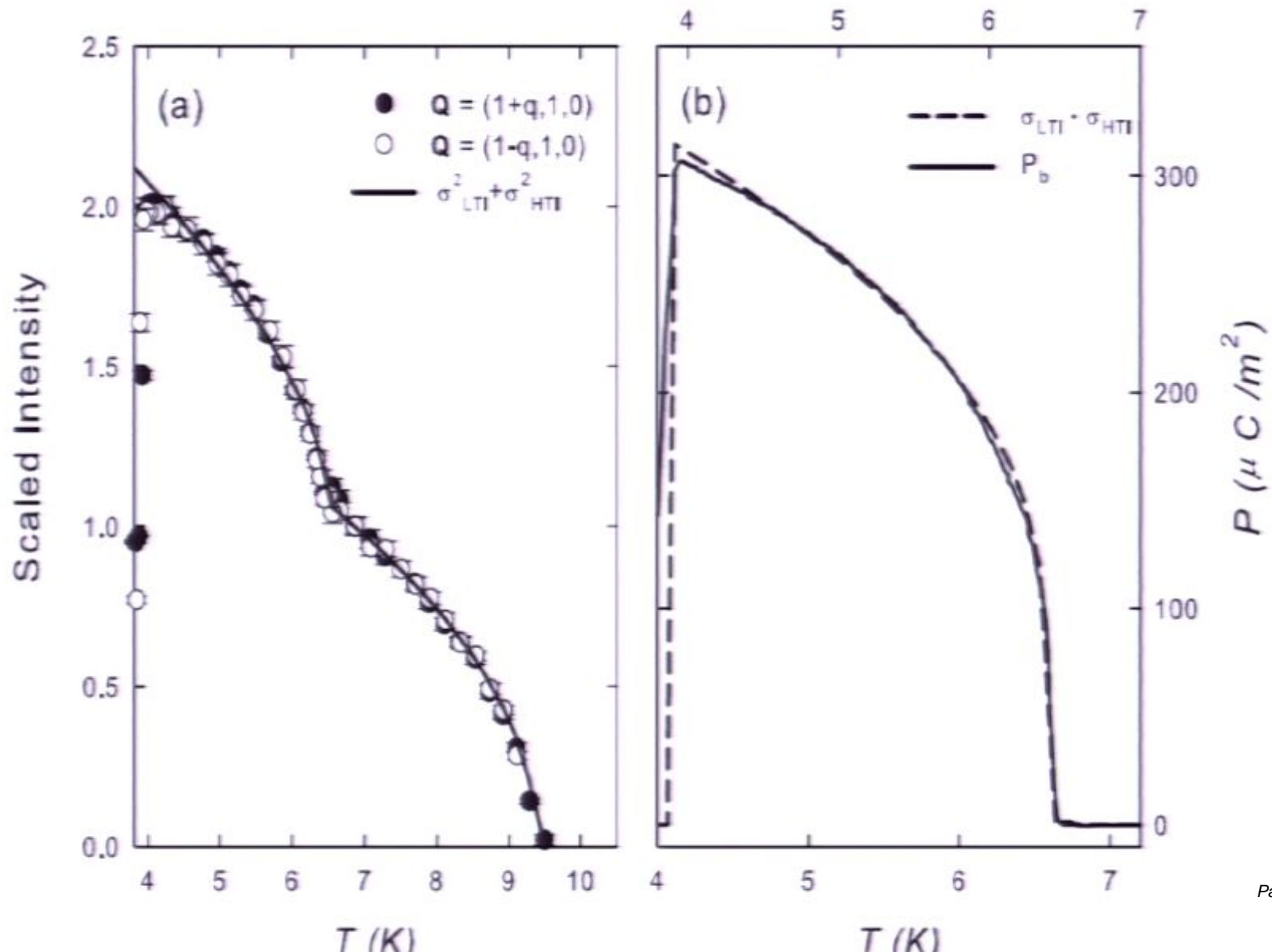
Cycloid state ( $\Gamma_4 + \Gamma_1$ )

$$V_{LTI} = c_{14y} (\sigma_1(\mathbf{q}) \sigma_4(-\mathbf{q}) + \sigma_4(\mathbf{q}) \sigma_1(-\mathbf{q})) P_y$$

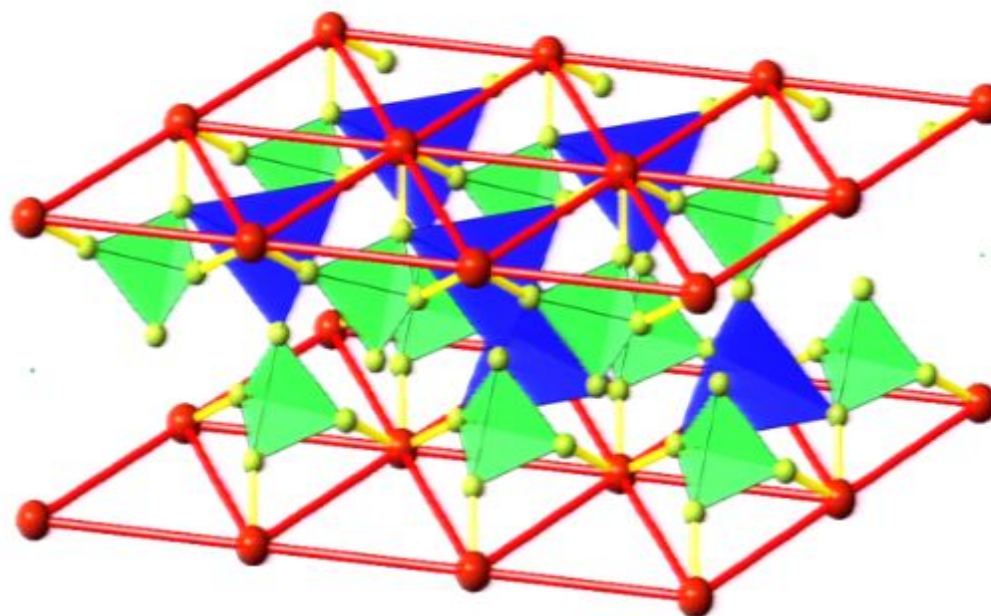




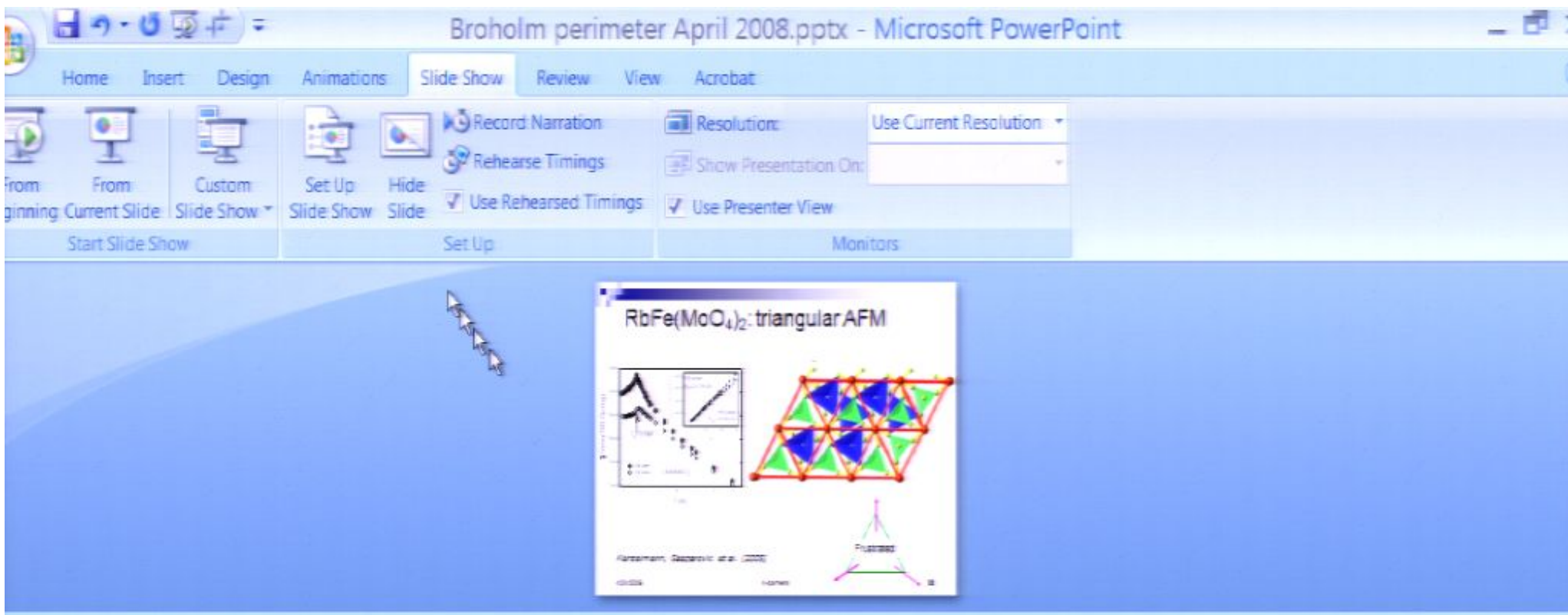
# Tri-linear coupling in $\text{Ni}_3\text{V}_2\text{O}_8$



# RbFe(MoO<sub>4</sub>)<sub>2</sub>: triangular AFM

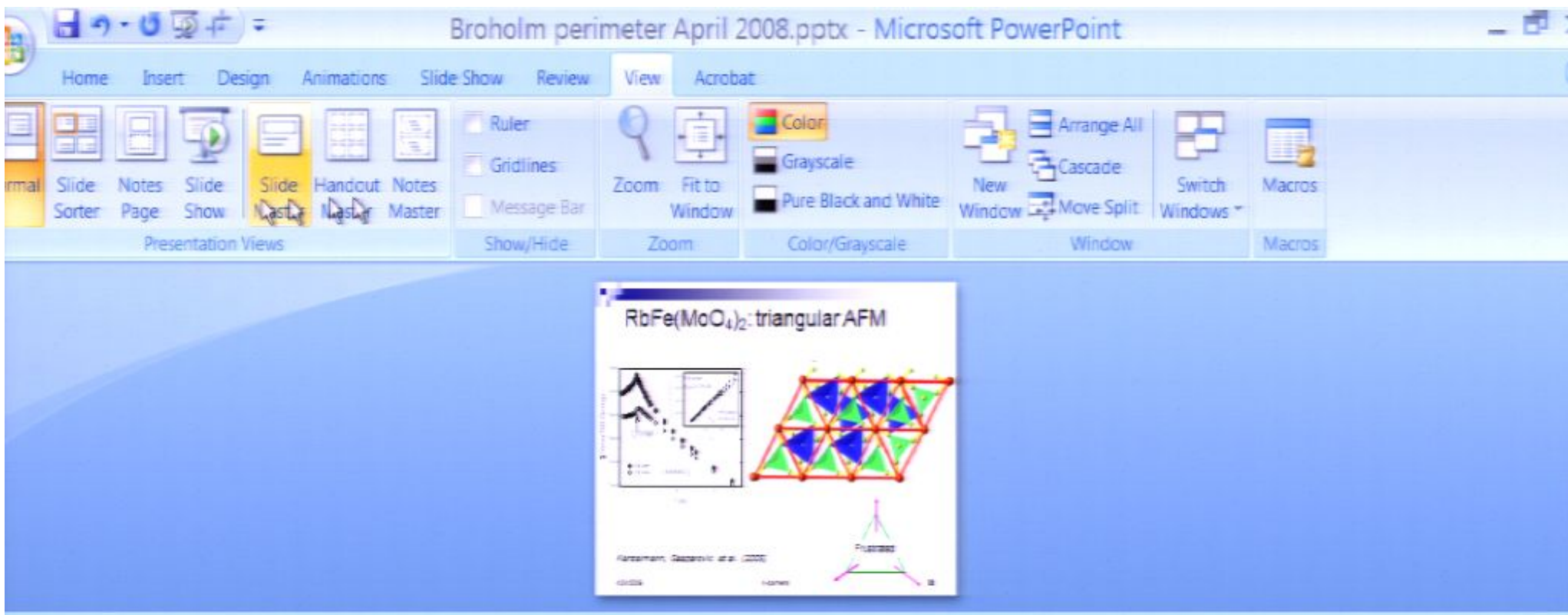


*Kenzelmann, Gasparovic et al. (2005)*



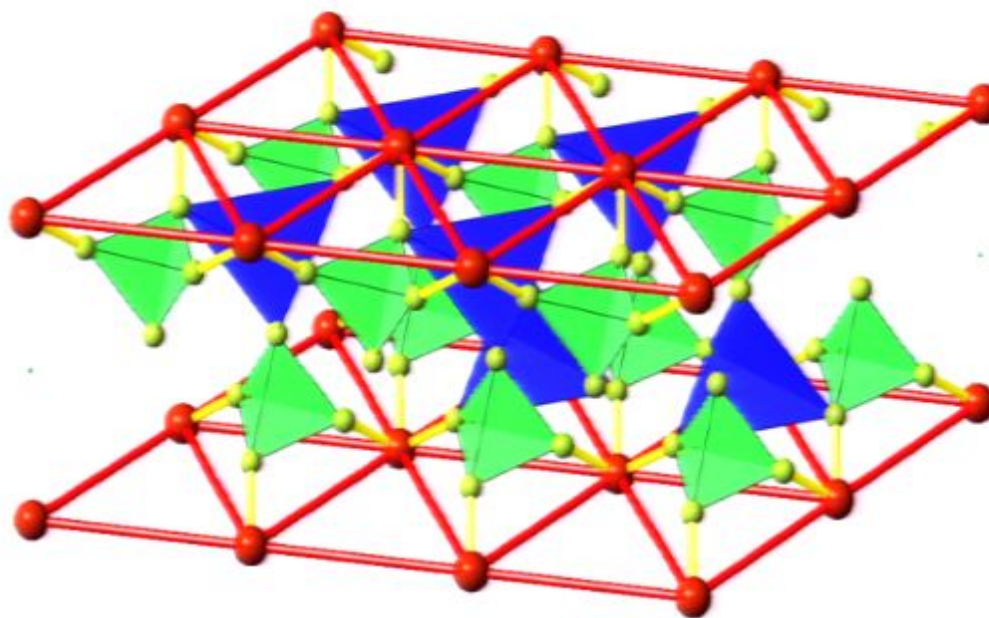
It is a stacked triangular lattice as shown here. Delightfully simple in there being just one magnetic Fe atom per unit cell. The susceptibility data show a magnetic phase transition at a temperature that is about an order of magnitude lower than the Curie-Weiss temperature. This indicates frustrated and quasi-two-dimensional magnetism and a potential for interesting lattice distortions and magneto-electricity.





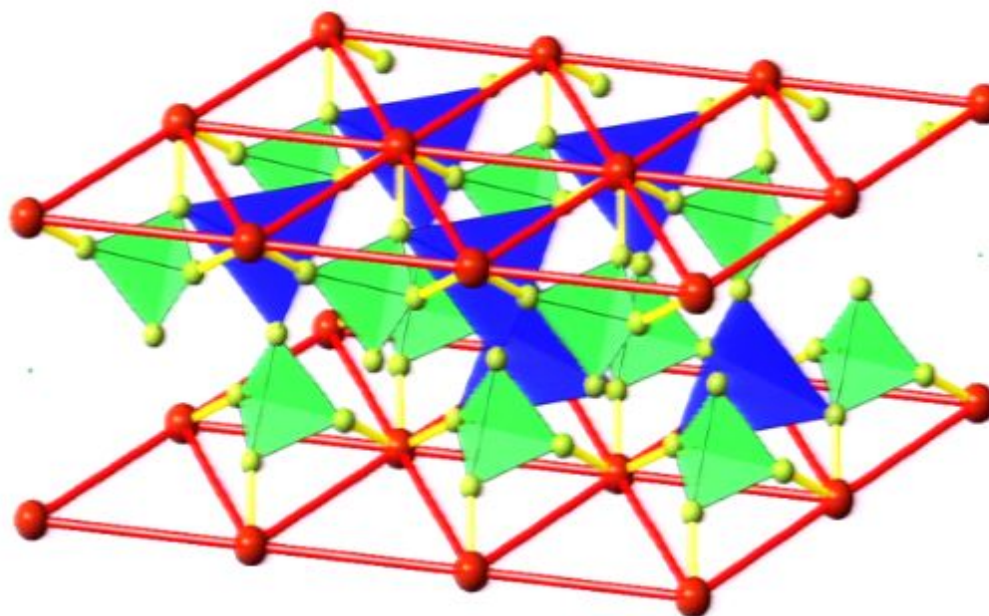
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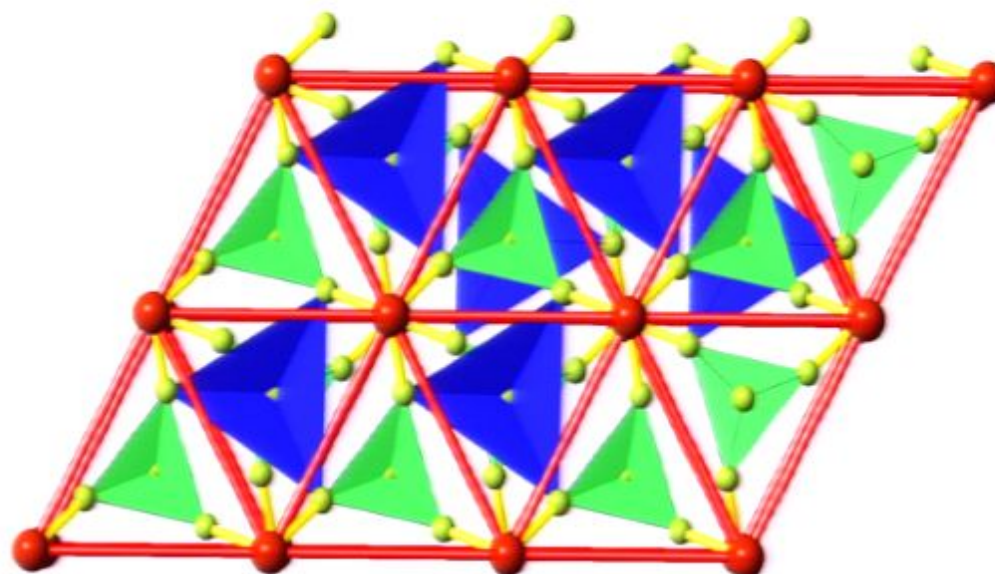
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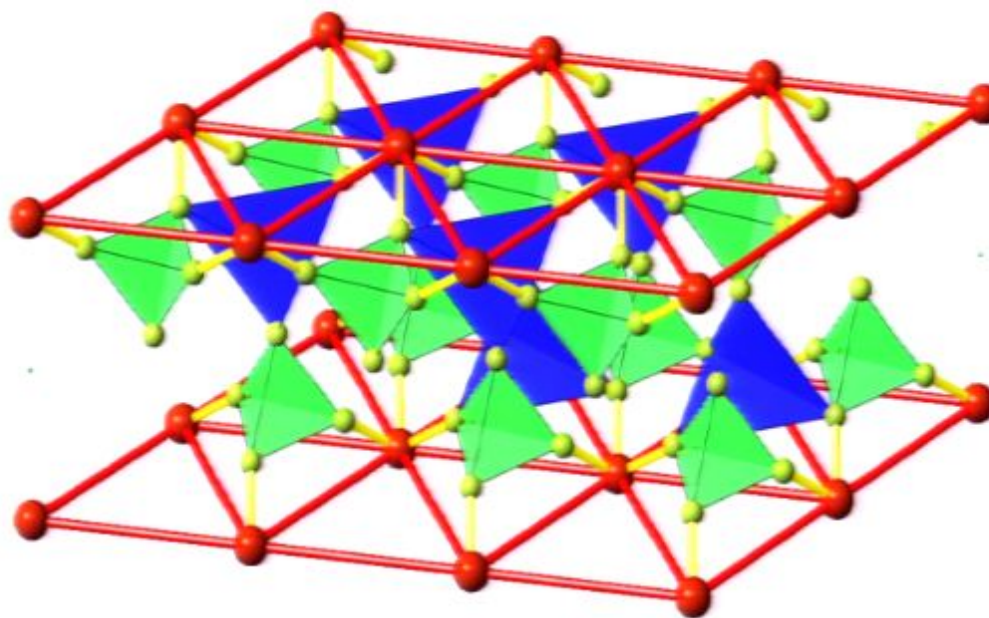


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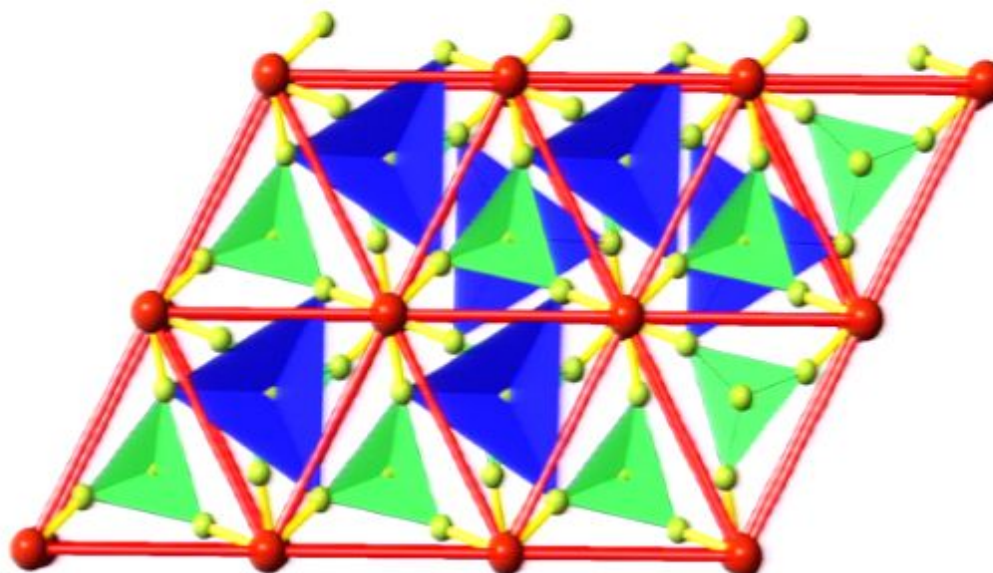
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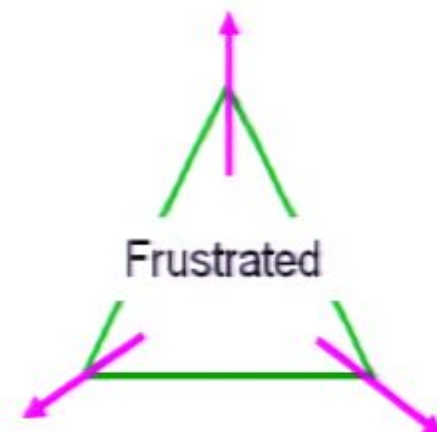
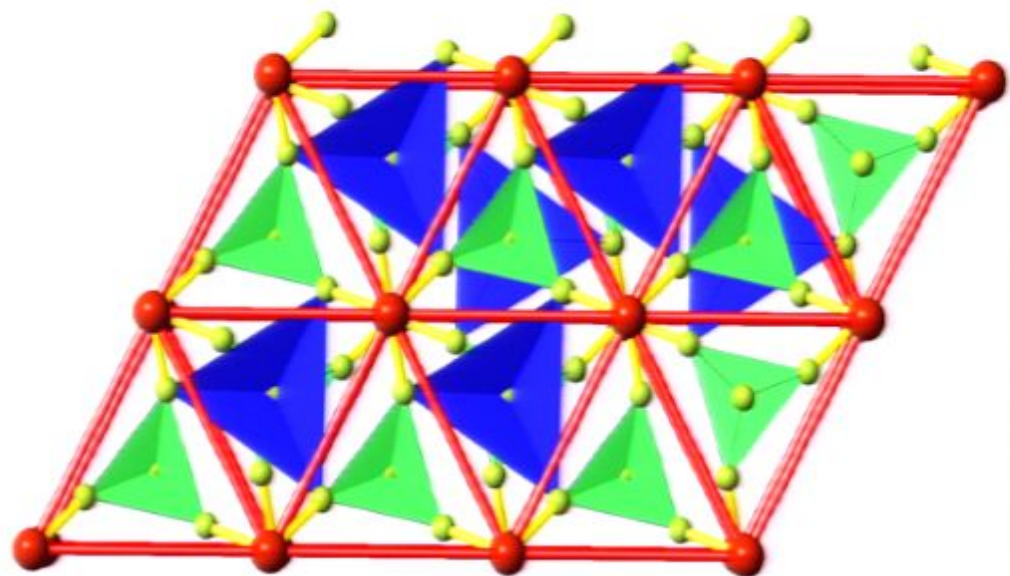
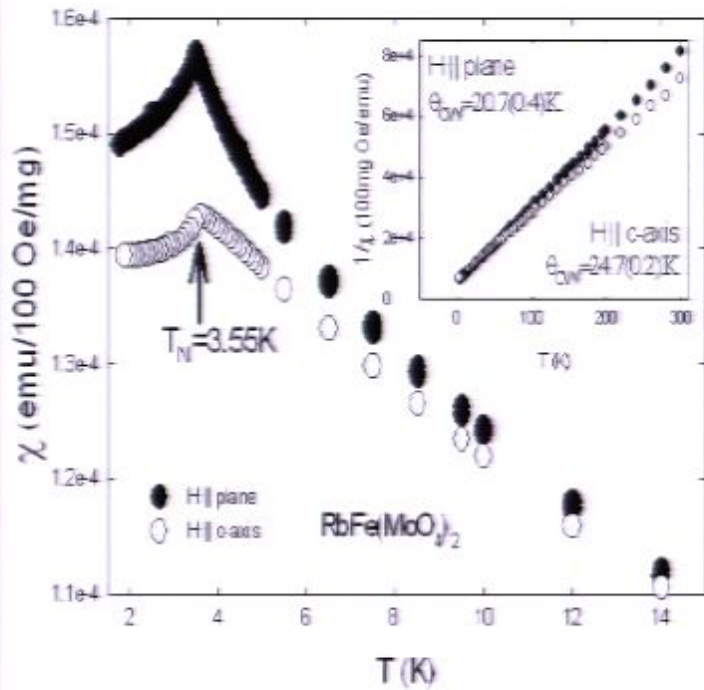
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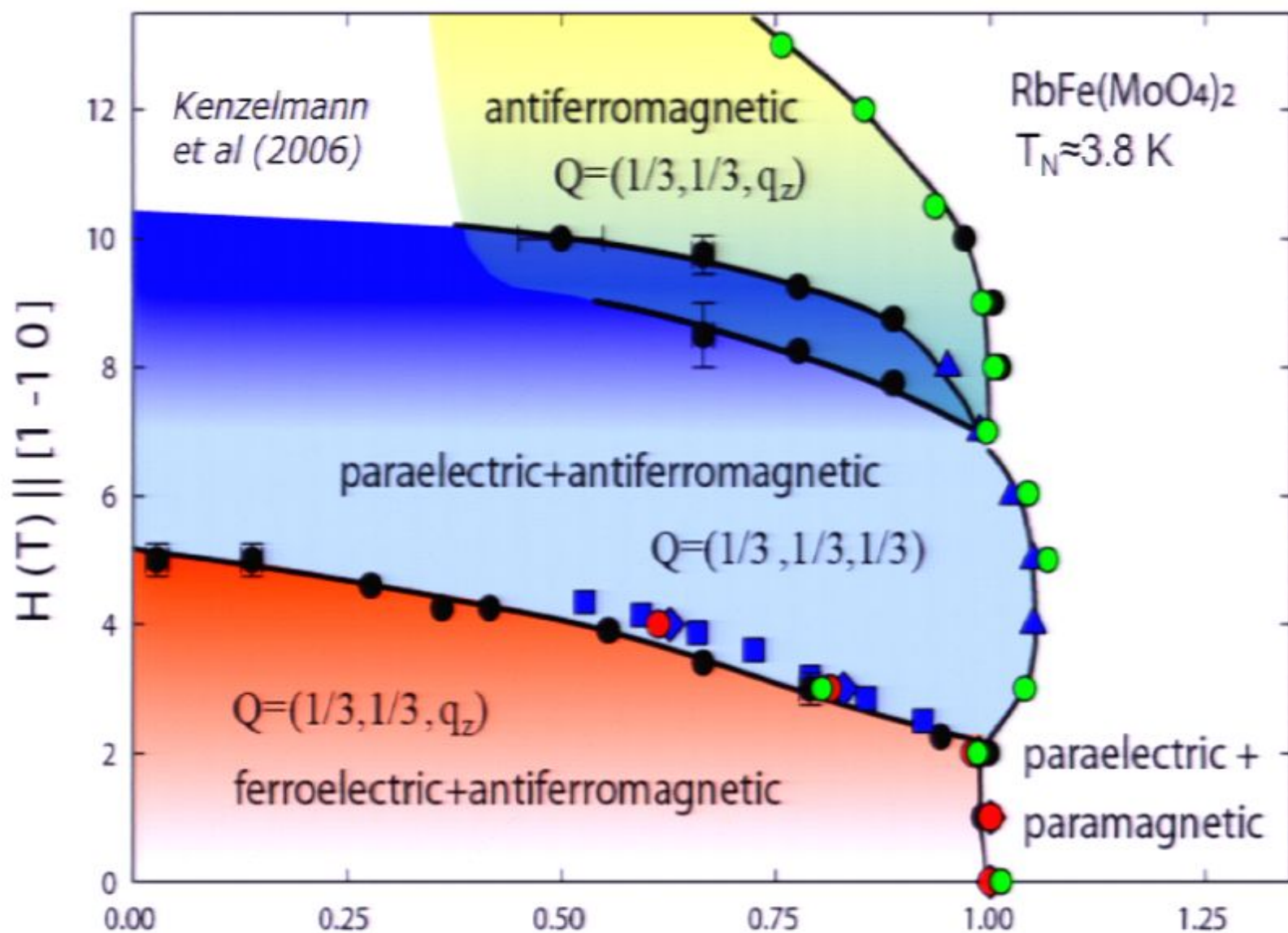


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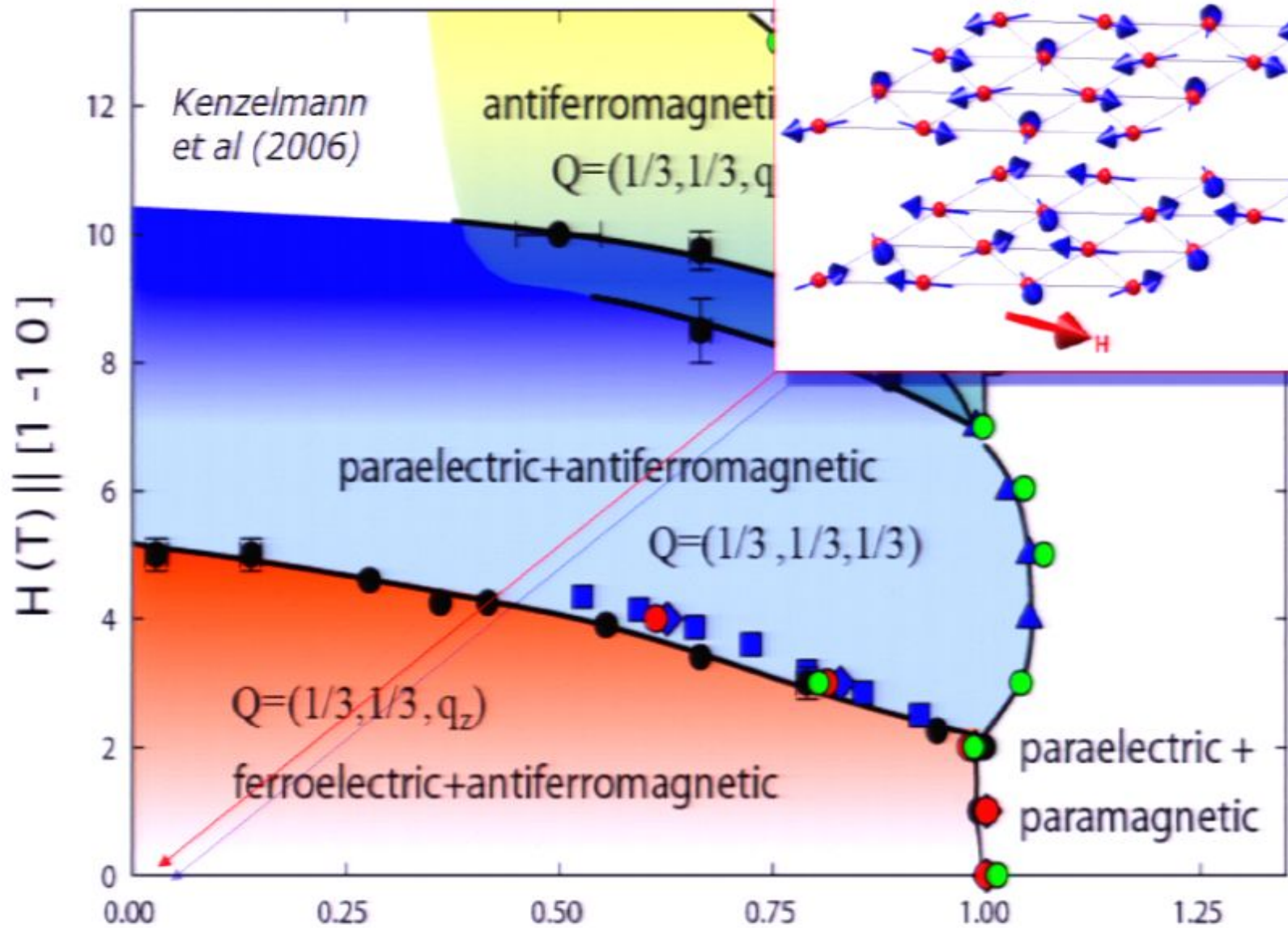


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# Field Dependent Long Range Order

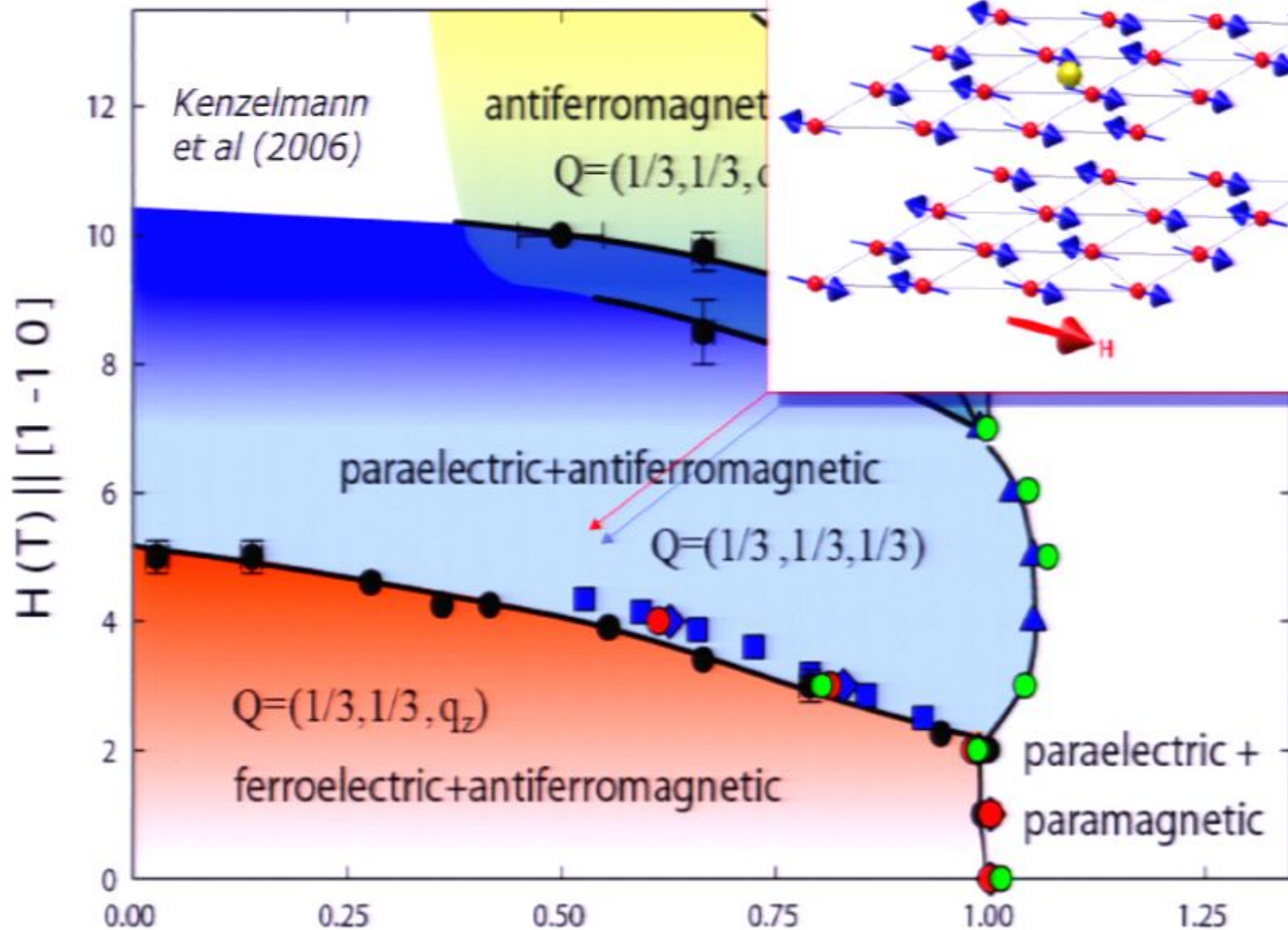


# Field Dependent Long

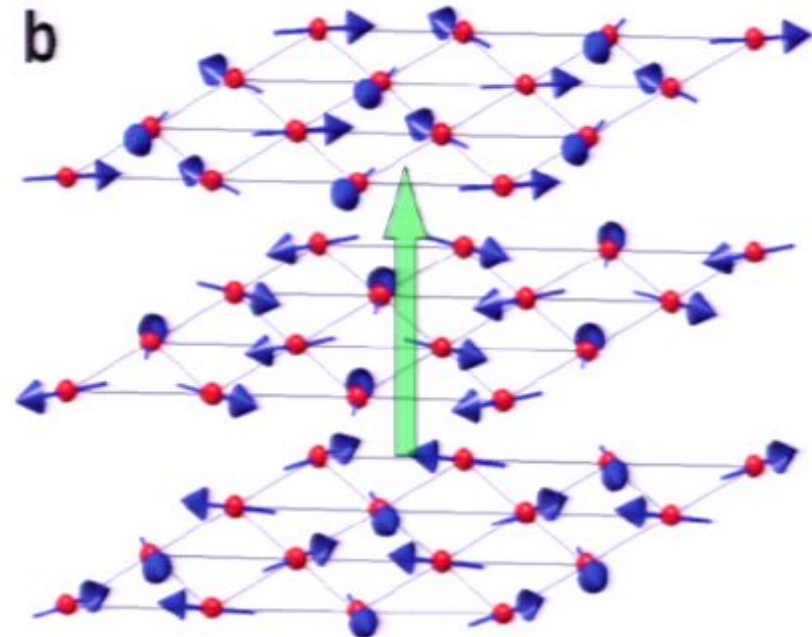
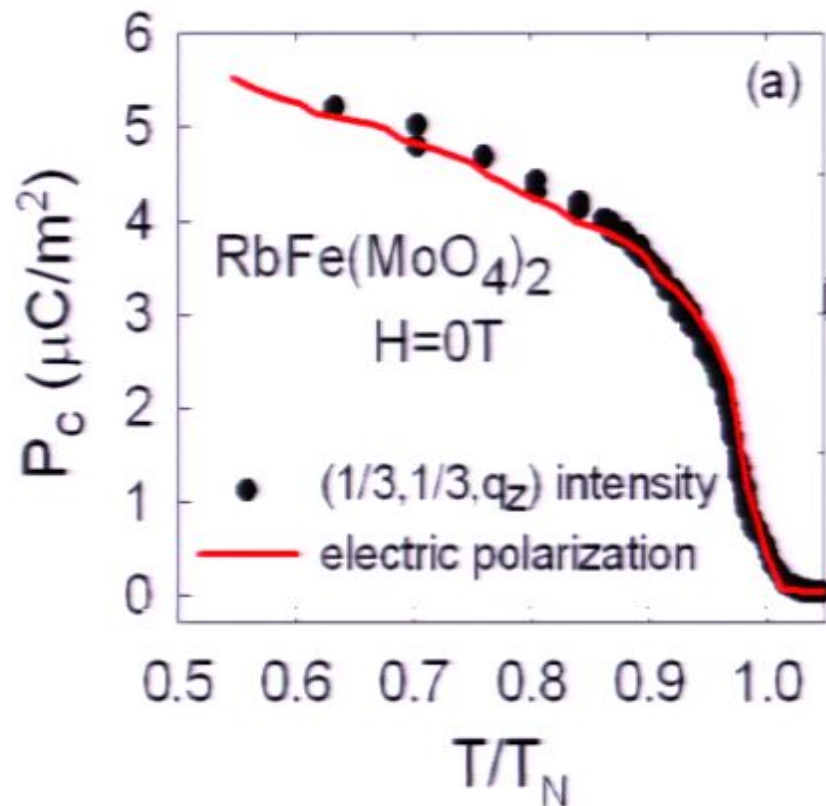




# Field Dependent Long



# RbFe(MoO<sub>4</sub>)<sub>2</sub>: Breaking the mold



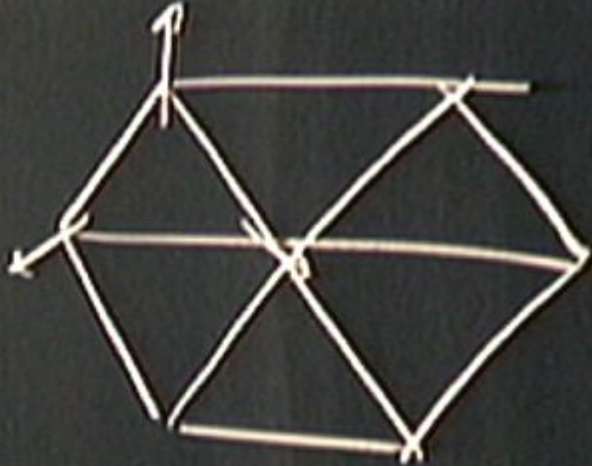
- Direct transition to magneto-electric state
- Polarization perpendicular to spin plane  $\mathbf{P} \neq \gamma \mathbf{e}_{ij} \times (\mathbf{S}_i \times \mathbf{S}_j)$
- Incommensurability irrelevant

# Landau theory for $\text{RbFe}(\text{MoO}_4)_2$

Spin structure expressed in terms of complex order parameters  $\sigma^{(i)}$

$$\mathbf{S}(\mathbf{r}) = \left[ \sigma^{(1)}(\mathbf{X}_1 + q_z \hat{\mathbf{z}})(\hat{\mathbf{x}} - i\hat{\mathbf{y}}) + \sigma^{(2)}(\mathbf{X}_1 + q_z \hat{\mathbf{z}})(\hat{\mathbf{x}} + i\hat{\mathbf{y}}) \right] \exp(i(\mathbf{X}_1 + q_z \hat{\mathbf{z}})) + c.c.$$





$$\nu_3 \nu_2 \nu_1$$

$$= 1$$

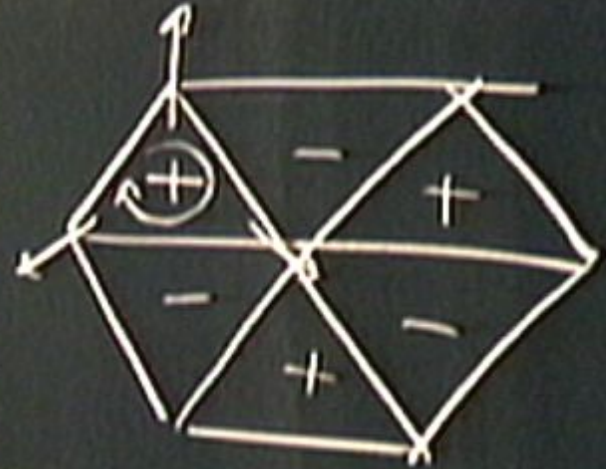
$$\Gamma(E) \sim E^{1+0.05}$$





$\underline{\underline{S=1}}$

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Effect of spatial inversion operator on order parameters:

$$\mathcal{I}\mathbf{S}(\mathbf{r}) = \mathbf{S}(-\mathbf{r}) \Rightarrow \mathcal{I}\sigma^{(1)}(\mathbf{q}_m) = \sigma^{(2)}(-\mathbf{q}_m) = \left( \sigma^{(2)}(\mathbf{q}_m) \right)^*$$



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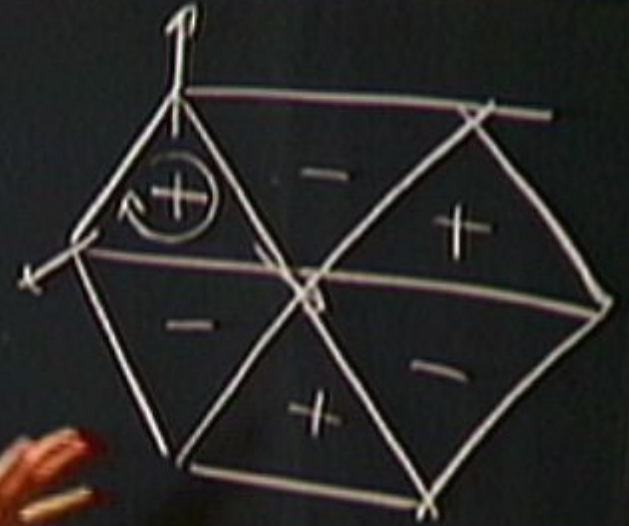
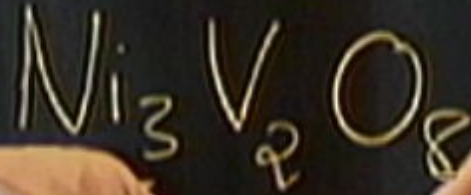
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Free energy at quadratic order:

$$F = \frac{1}{2} \alpha (T - T_c) \left( \left| \sigma^{(1)}(\mathbf{q}_m) \right|^2 + \left| \sigma^{(2)}(\mathbf{q}_m) \right|^2 \right) + \mathcal{O}(\sigma^4) \quad \text{Magnetic free energy}$$

$$+ \frac{1}{2} \chi^{-1} |P|^2 + \mathbf{E} \cdot \mathbf{P} \quad \text{Dielectric free energy}$$

$$+ K \left( \left| \sigma^{(1)}(\mathbf{q}_m) \right|^2 - \left| \sigma^{(2)}(\mathbf{q}_m) \right|^2 \right) P_z \quad \begin{array}{l} \text{Symmetry allowed} \\ \text{Tri-linear interaction} \end{array}$$



$$E = 1 + 0.05$$



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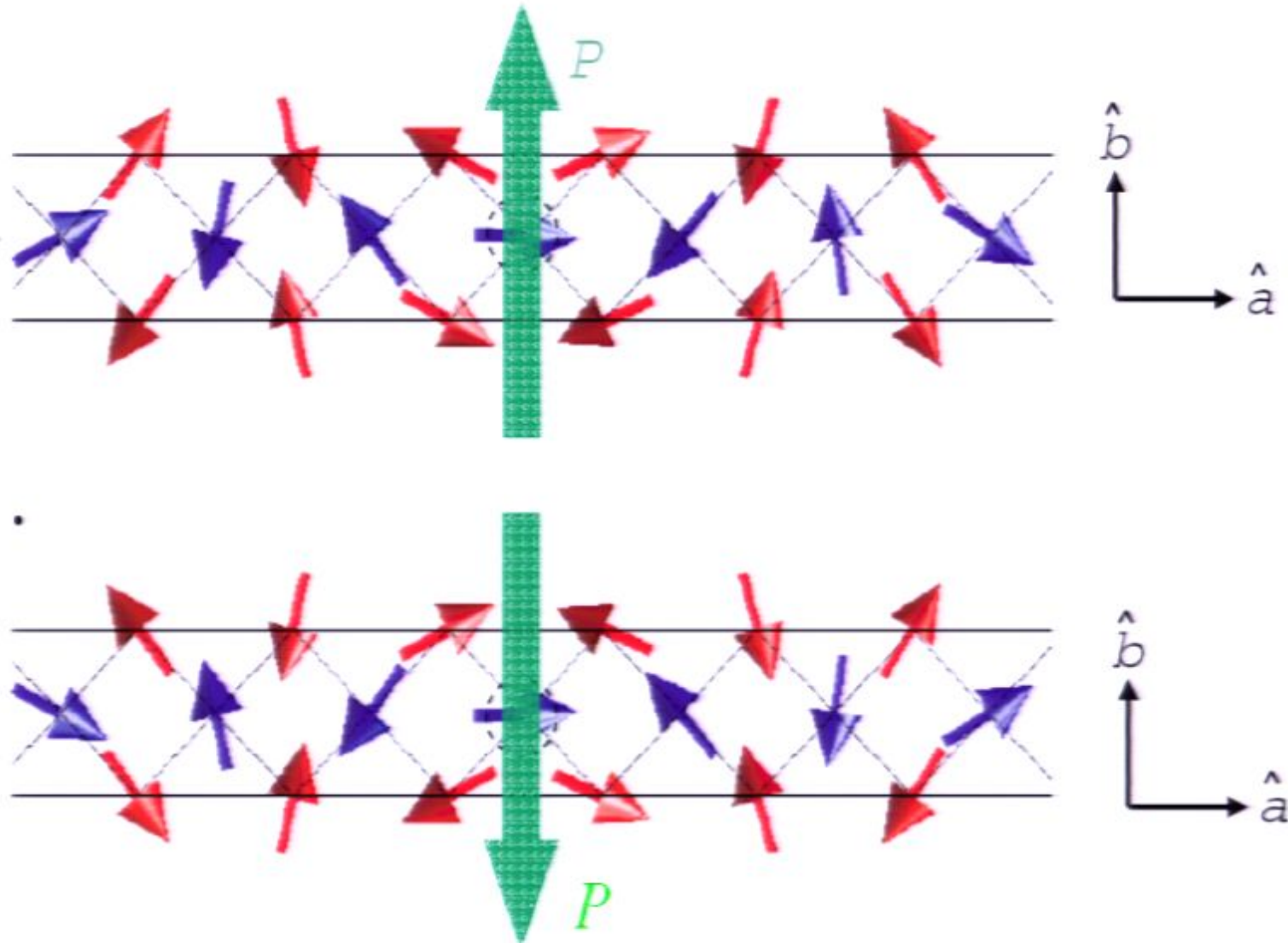
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Dielectric free energy

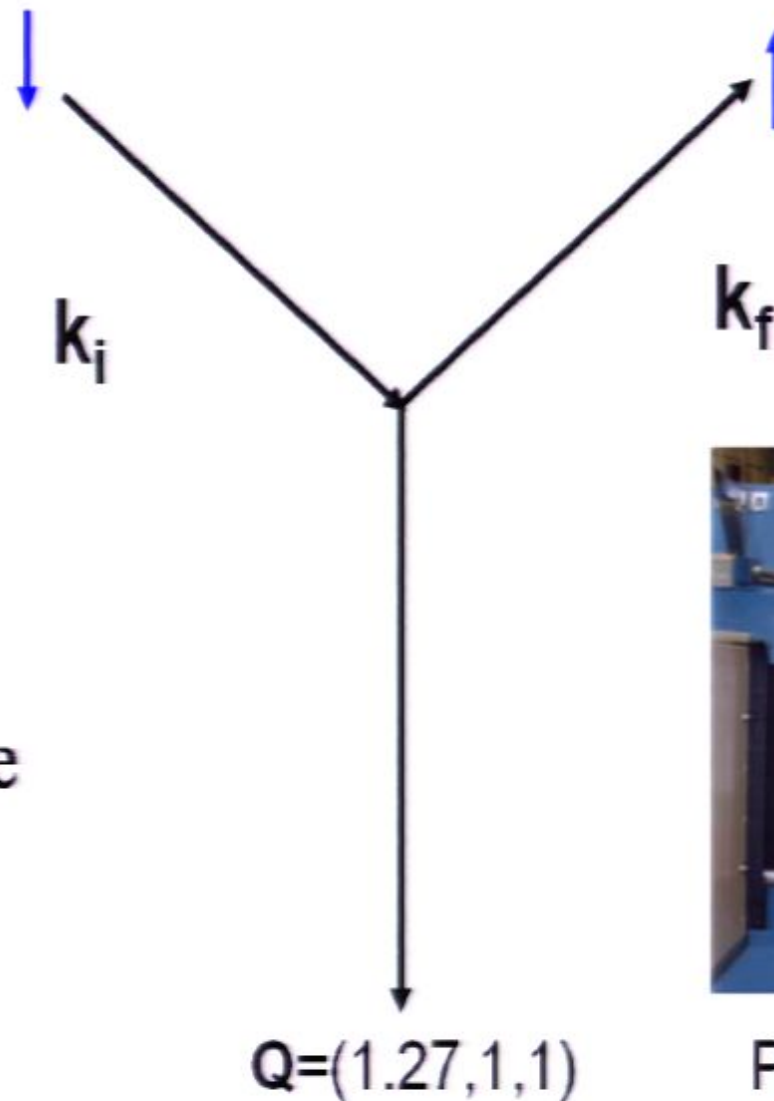
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# Hypothesis: Chiral FE domains



# Detecting chiral domains in $\text{Ni}_3\text{V}_2\text{O}_8$



No effect if:

$$\mathbf{S}_n \perp \mathbf{P}_e$$

$\mathbf{Q} \parallel$  spin plane

$$\mathbf{S}_n \perp \mathbf{q}_m$$

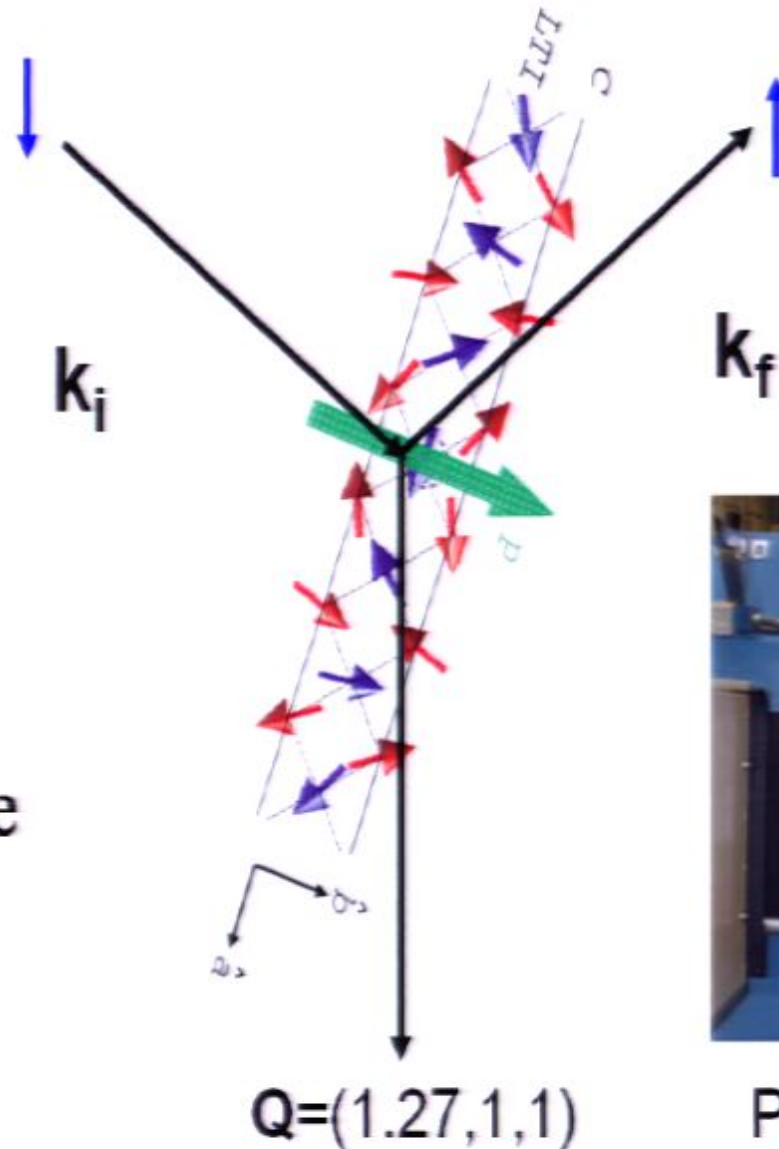


Polarized  $^3\text{He}$   $\rightarrow$

Polarized neutrons



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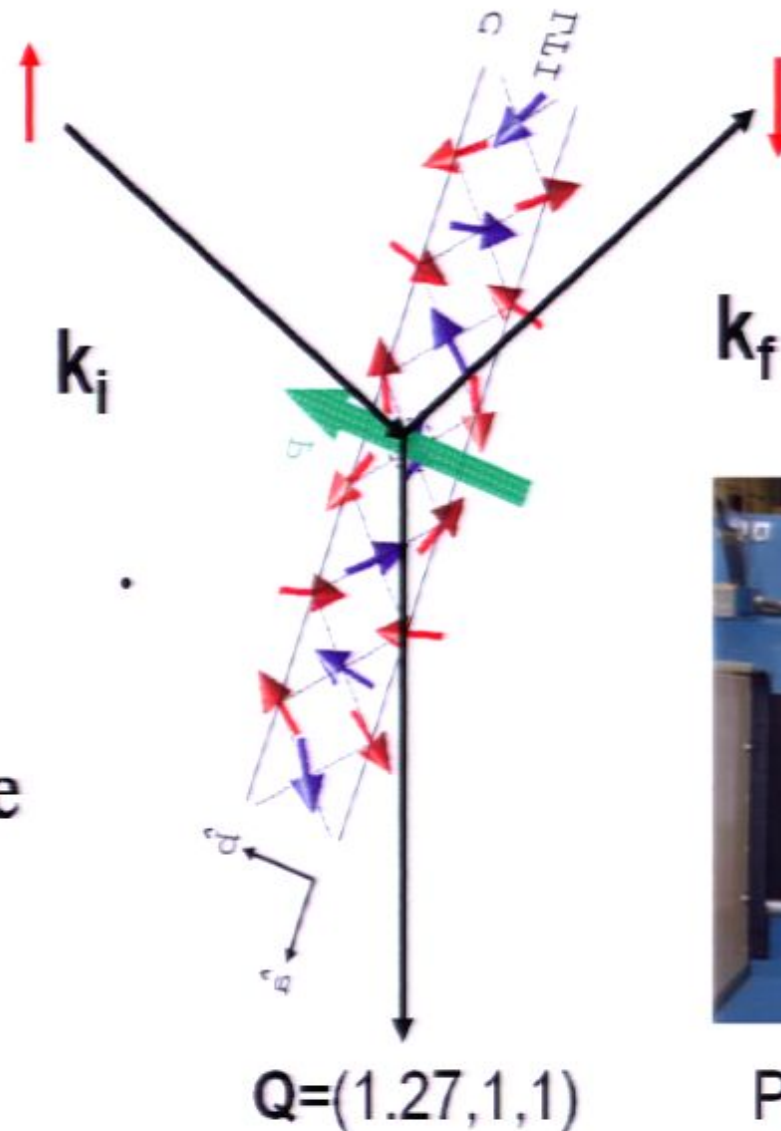
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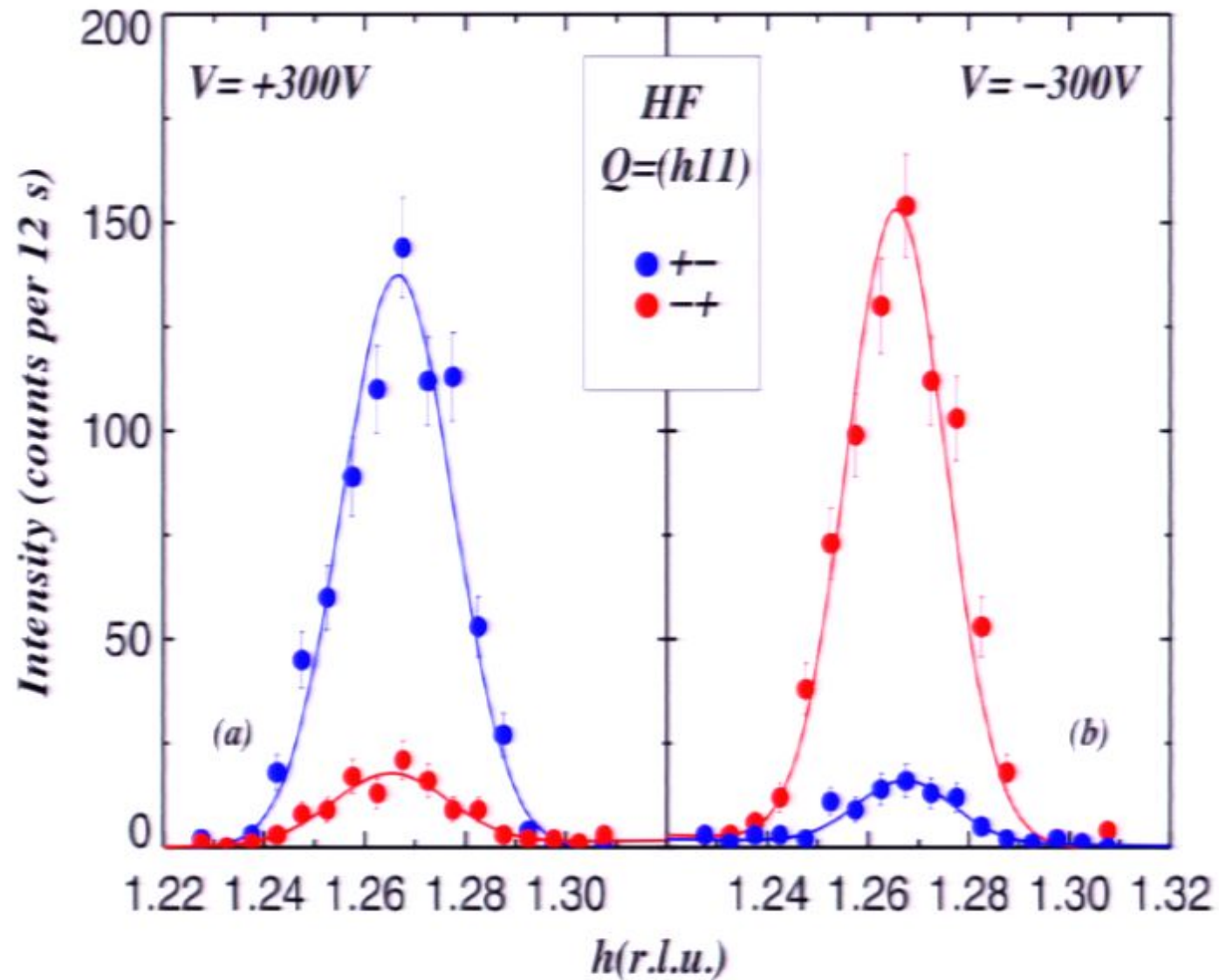
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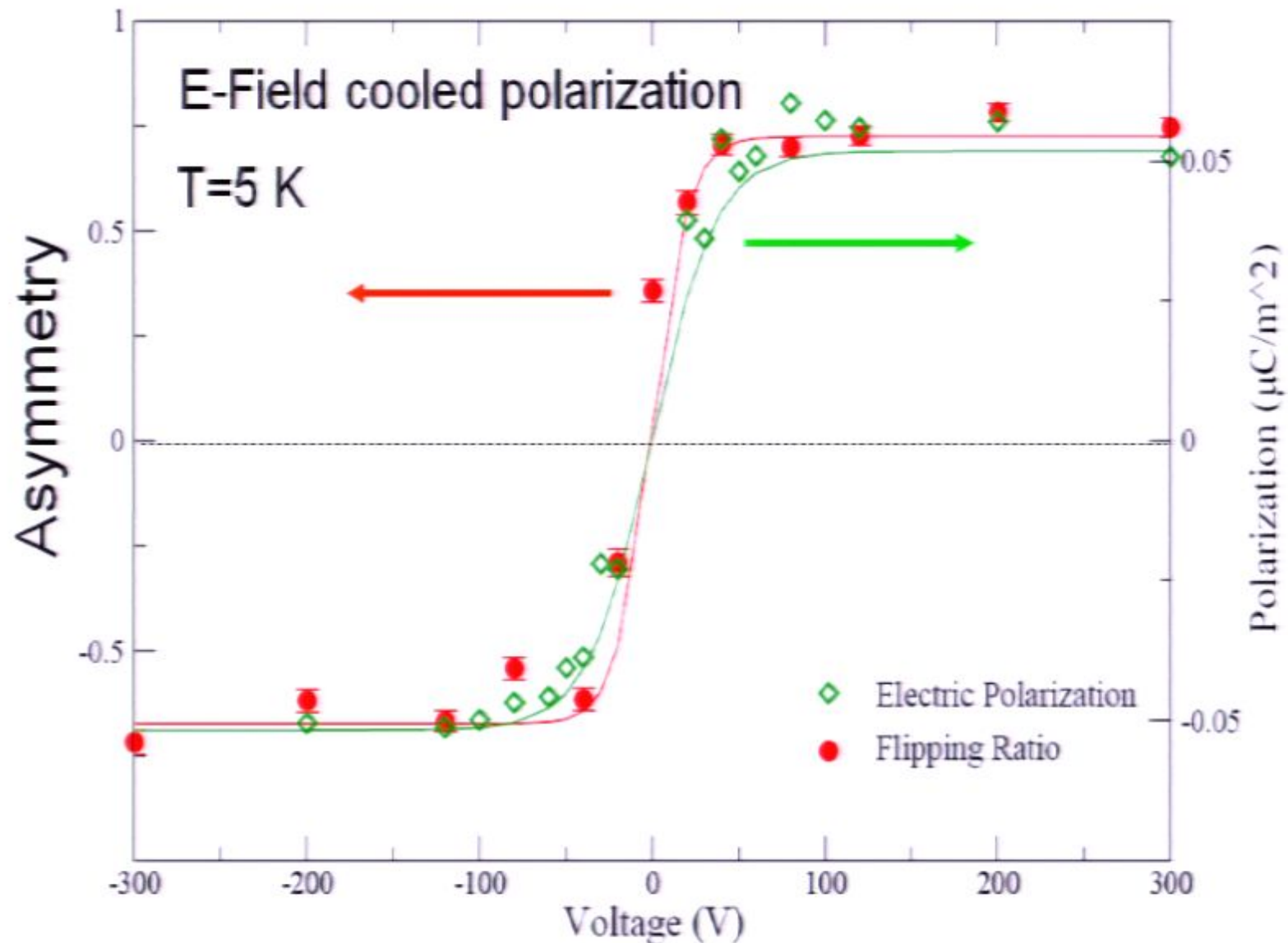
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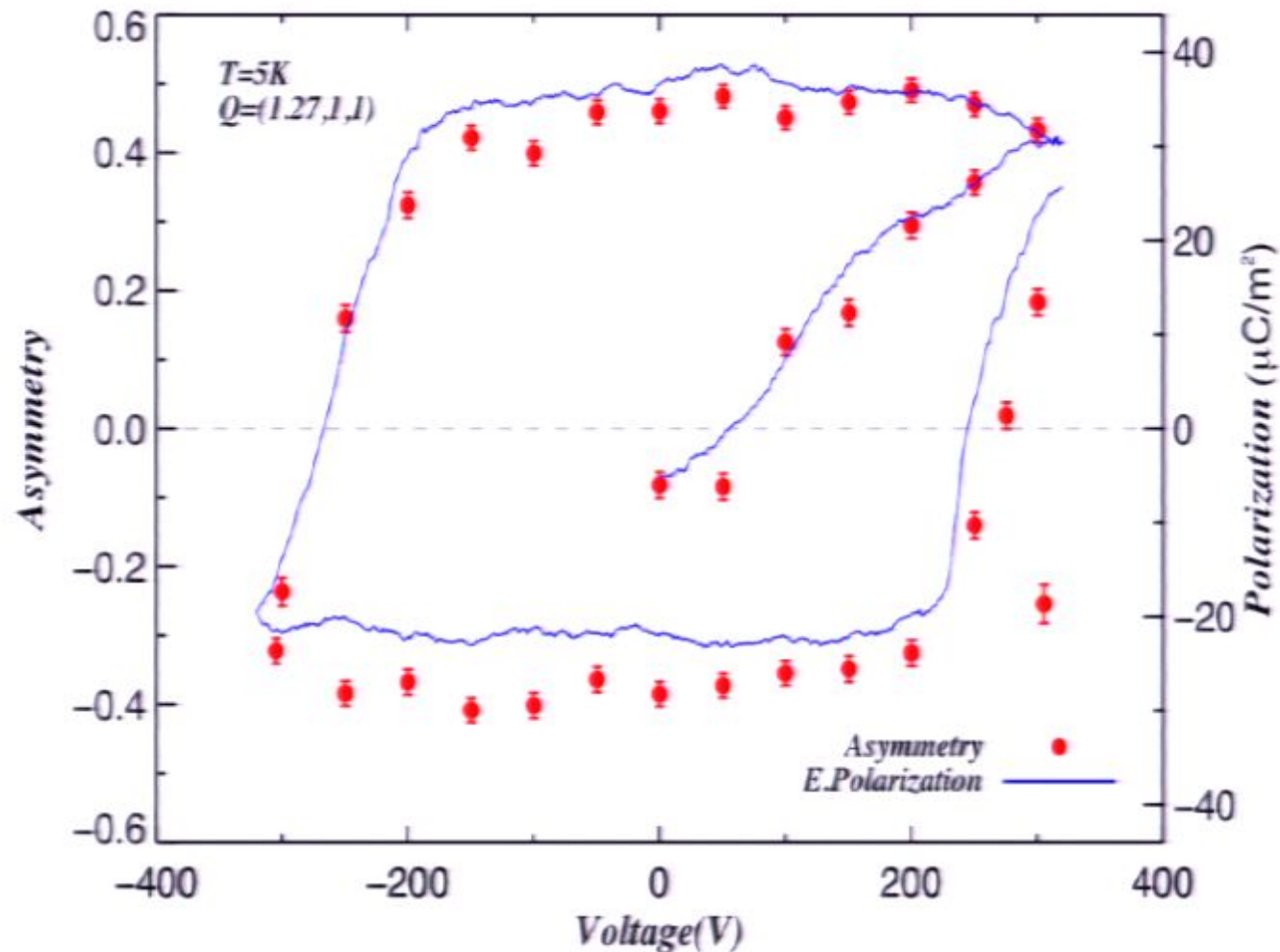




# Ferroelectric & Chiral domains



# Chirality hysteresis in FE state



# Conclusions

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  - Non-collinear inversion symmetry breaking structures
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2. Outline

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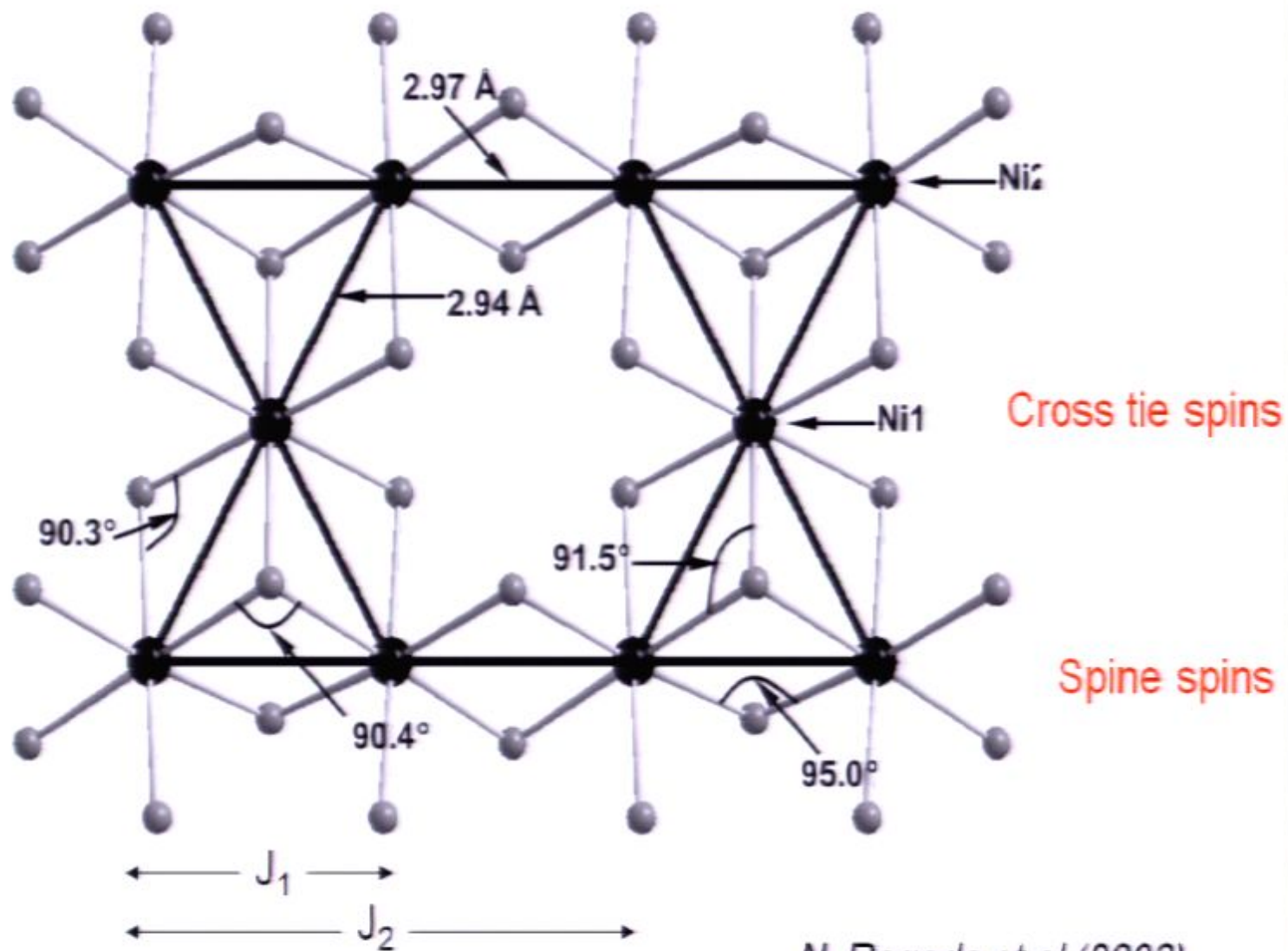
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# Exchange Interactions



*N. Rogado et al (2003)*

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