Neutron scattering studies of RMn_2O_5 multiferroics (R=Tb, Dy)

$TbMn_2O_5$: Phys Rev B, 140301(R), 2013

C. Doubrovsky S. Petit, V. Balédent, M. B. Lepetit, M. Greenblatt, B. Wanklyn, and P. Foury-Leylekian

$DyMn_2O_5$: Scientific Report 7:14506 | DOI:10.1038/s41598-017-15150-w

S. Chattopadhyay, S. Petit, E. Ressouche, S. Raymond, V. Balédent, G. Yahia, W. Peng, J. Robert, M.-B. Lepetit, M. Greenblatt & P. Foury-Leylekian

Long term collaboration bewteen LPS - LLB - CEA/MDN & ILL-Néel

Multiferroics

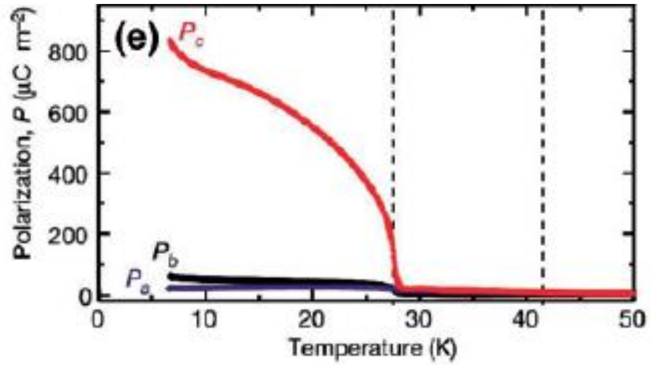
« Definition »

Two coupled order parameters in the ground state :

- Magnetization
- Electric polarization

- Frustrated magnets with complex magnetic structures
- Peculiar excitations « electromagnons », basically electro-active spin waves

$TbMnO_3$



$T_c \sim T_N$

Kimura et al, Nature 426 (2003)

Reviews :
Cheong and Mostovoy, Nature materials, 6 (2007)
Wang et al, Advances in Physics (2009)

Applications

Take advantage of the coupling between ferroelectricity and magnetism

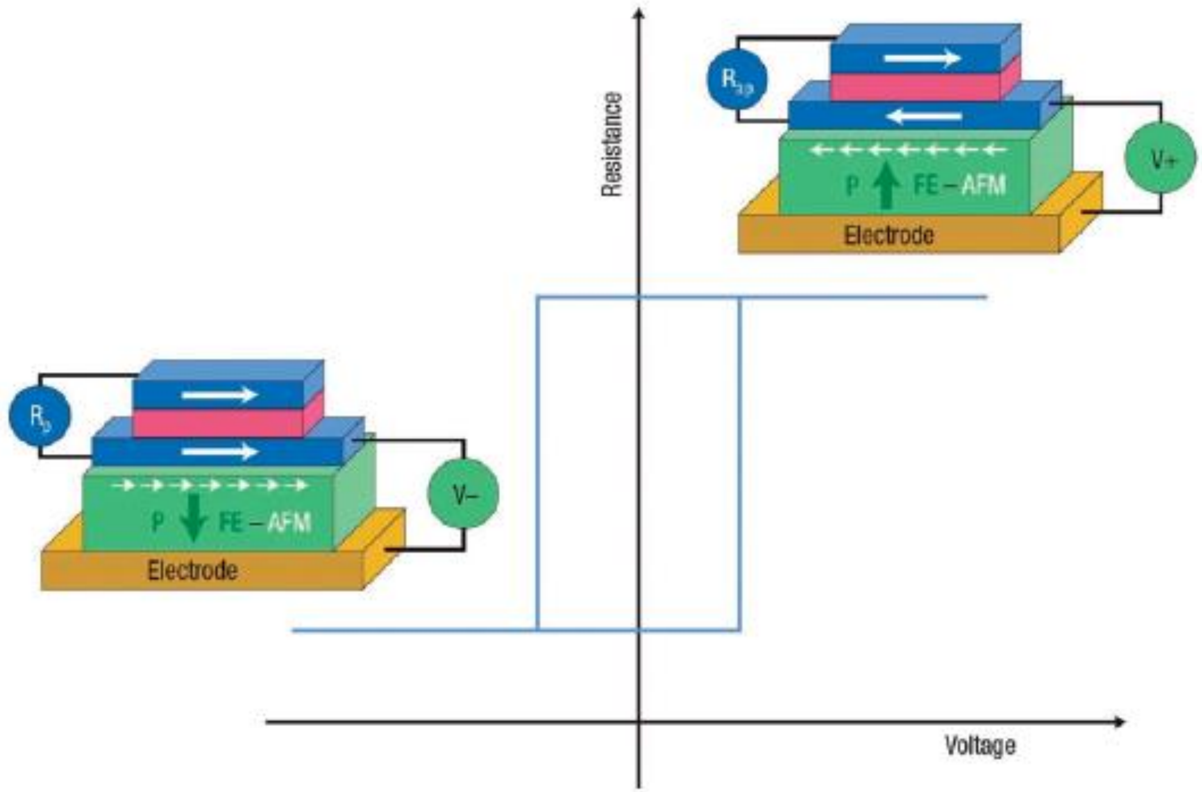


Figure 38. A possible multiferroic random-access memory element using antiferromagnetic multiferroic materials. (Reproduced with permission from [214]. Copyright © 2008 Macmillan Publishers Ltd/Nature Materials.)

Nature Mater. 7 (2008), p. 425

Outline

1) Introduction :

- Inverse DMI
- Symmetric exchange
- Archetypes compounds $TbMnO_3$ and YMn_2O_5

2) $TbMn_2O_5$

3) $DyMn_2O_5$

4) Conclusions

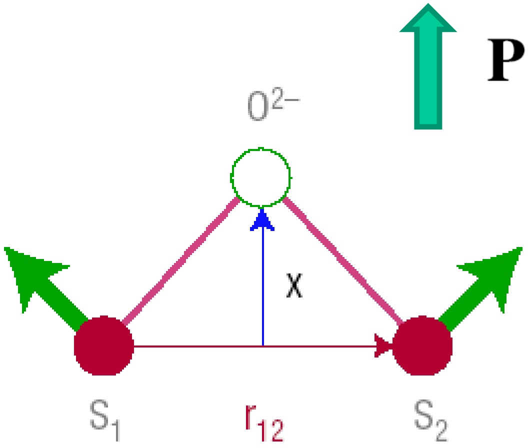
Magnetically induced ferroelectricity

« Inverse Dzyaloshinskii-Moriya model »

$$\mathbf{D}_{n,n+1} \cdot \mathbf{S}_n \times \mathbf{S}_{n+1}$$

$$\mathbf{D}_{n,n+1} \sim \mathbf{x} \times \mathbf{r}_{n,n+1}$$

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

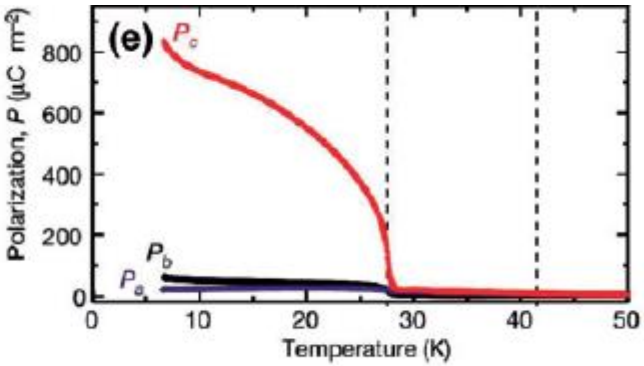


Ferroelectricity rises at $T_c \sim T_N$

Katsura, PRL 95 057205, 98 027203
Sergienko PRB 73 094434

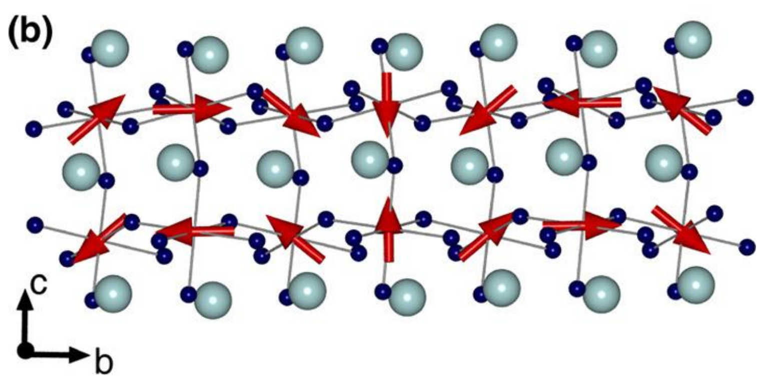
$TbMnO_3$

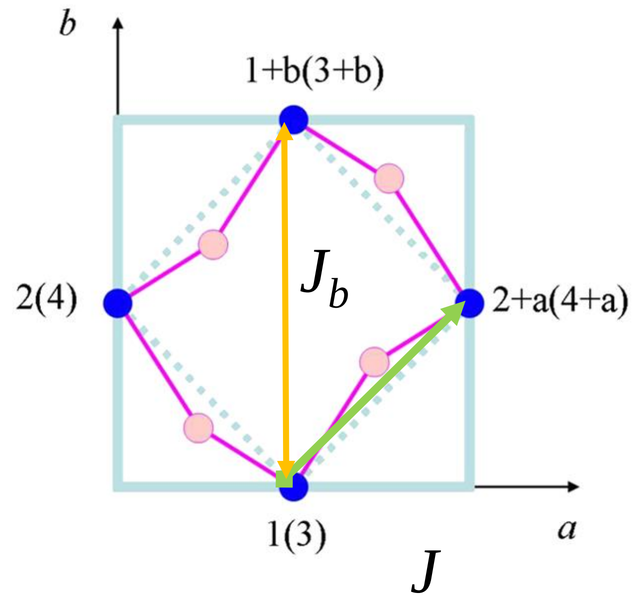
$TbMnO_3$



$T_c \sim T_N$

Kimura et al, Nature 426 (2003)

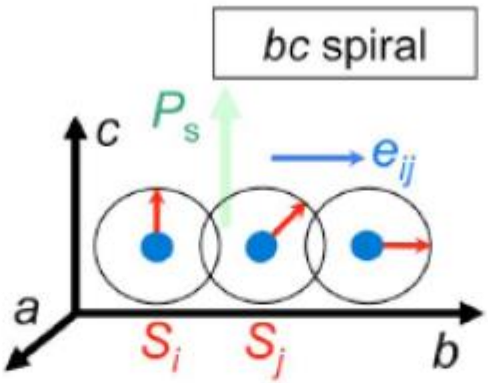


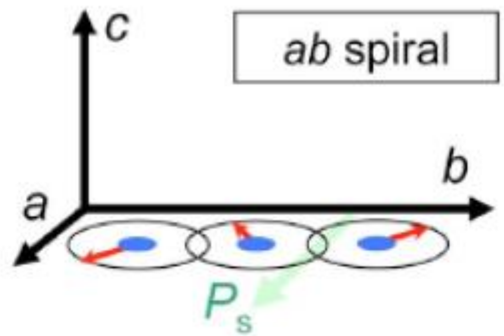


$$\cos\frac{Q}{2} = \frac{|J|}{2J_b}$$

$TbMnO_3$

Inverse DMI explains the flop of P induced by the flop of the spiral under applied H

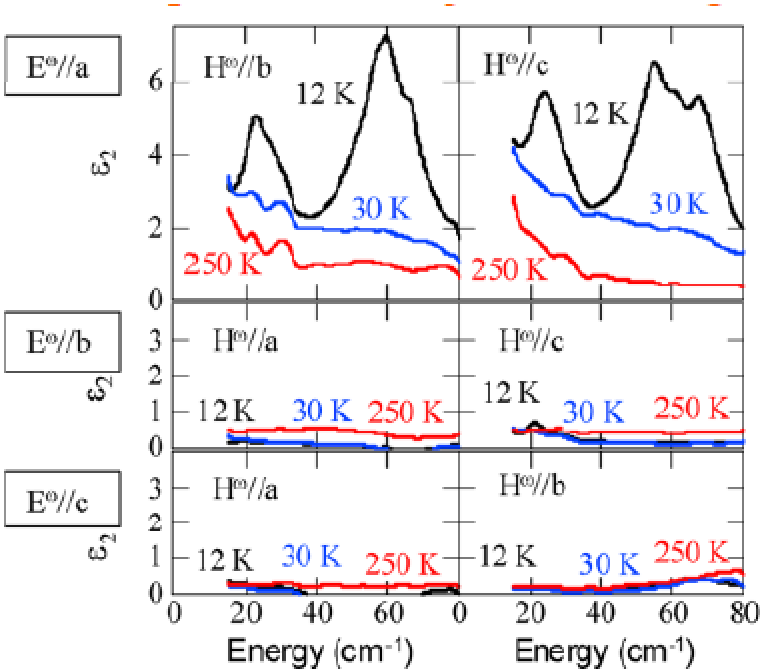




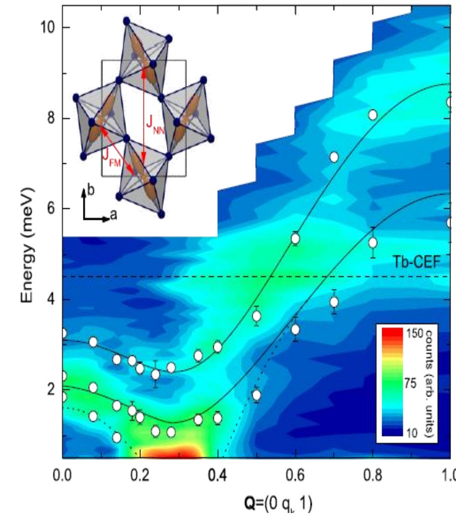
$TbMnO_3$

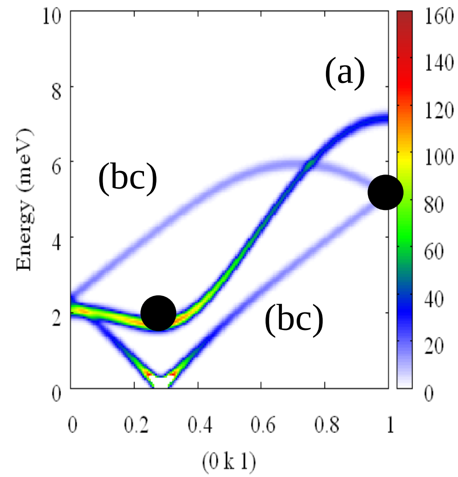
Electromagnons, electro-active spin waves :

Optical conductivity



Neutrons

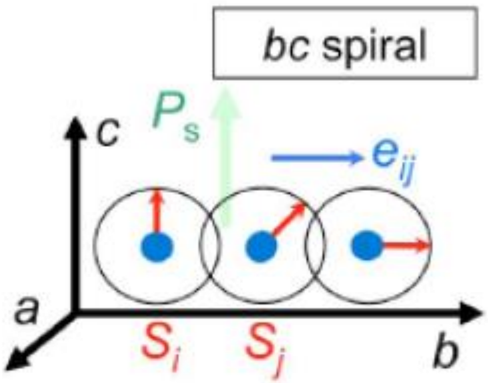




Pimenov, Nature Physics (2006
Takahashi, PRL 101, 187201 (2008),
Valdès, PRL 102, 047203 (2009)
Senff, PRL 98, 137206 (2007), J. Condens Matter 20 434212 (2008)

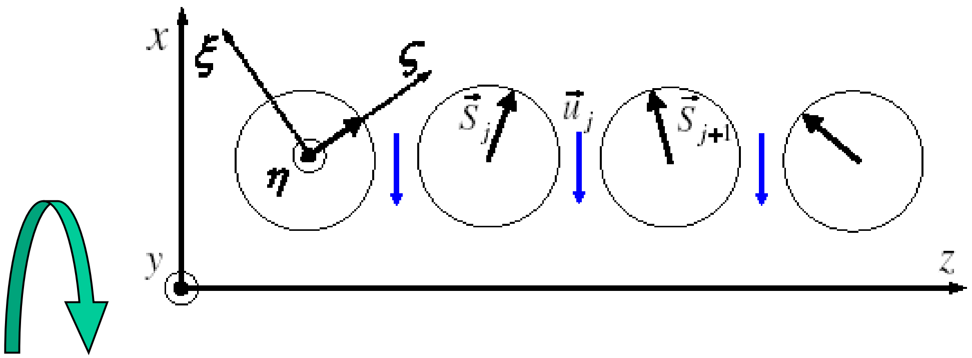
20 and 60 cm^{-1}
2.5 and 7.5 meV
E_w//a only

$TbMnO_3$



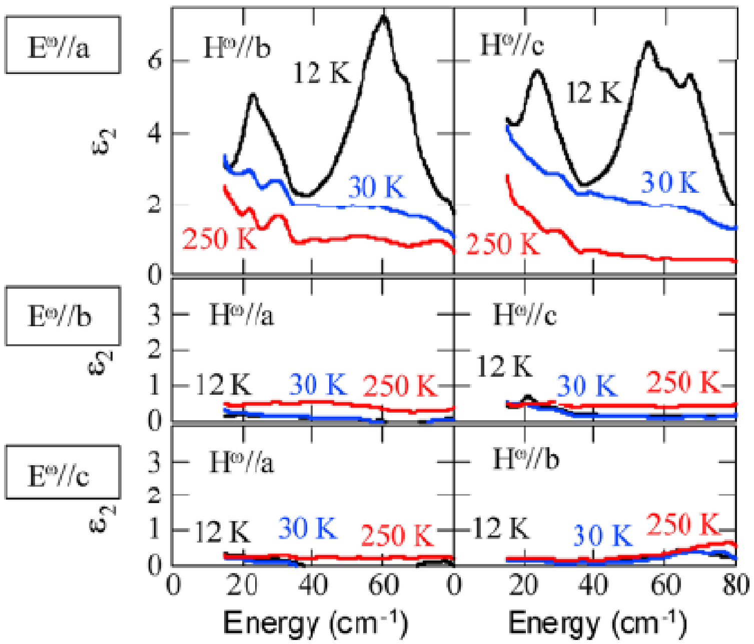
Inverse DMI electromagnon :

Excitation corresponding to a long wavelength rotation around the z axis of the spiral is coupled to P

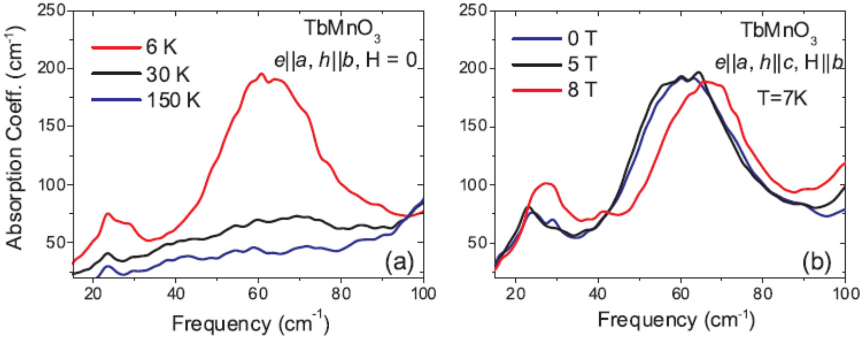


$TbMnO_3$

The selection rule Eω //a is independent of the orientation of the cycloid (should follow its flop under H)



Takahashi, PRL 101, 187201 (2008)

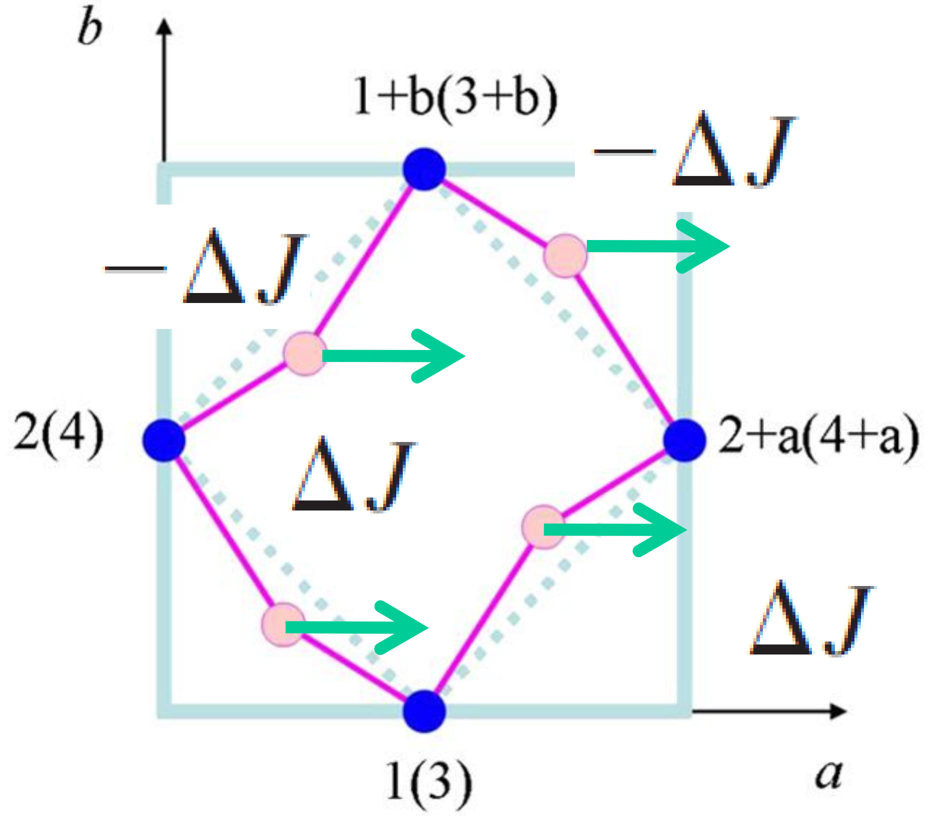


Valdès, PRL 102, 047203 2009

Inverse DMI does NOT explain the electromagnons …

$TbMnO_3$

Calls for another mechanism:
symmetric exchange modulated by a Q=0 deformation



Valdès, PRL 102, 047203 2009

Magnetically induced ferroelectricity

« Symmetric exchange model »

Exchange | Anisotropy

$$H = \frac{1}{2}\sum_{i,j} J_{ij}(\boldsymbol{P})(\boldsymbol{S}_i \cdot \boldsymbol{S}_j) - \frac{1}{2}\sum_{i\alpha} K_{i\alpha}(\boldsymbol{S}_i \cdot \hat{k}_{i\alpha})^2 - \sum_i \mu_i(\boldsymbol{S}_i \cdot \boldsymbol{H}) + V\left(\frac{\boldsymbol{P}^2}{2\chi_1^{(0)}} - \boldsymbol{P}\boldsymbol{E} - \frac{\chi_2 \boldsymbol{E}^2}{2}\right). \quad (2)$$

Zeeman | Polarization

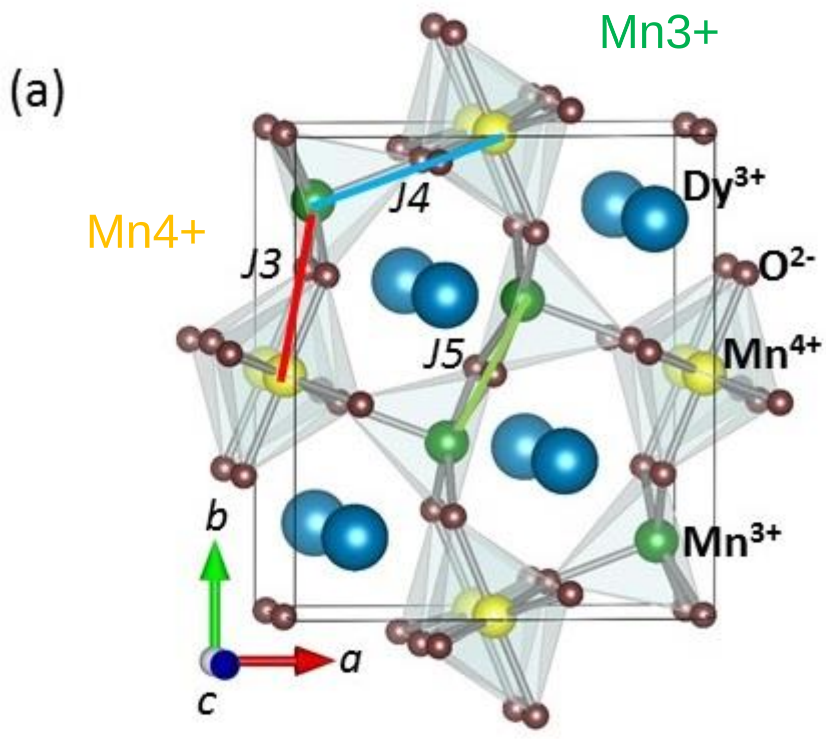
J vs P

$$J_{ij}(P_b) = J_{ij}(0) + J'_{ij}(0)P_b + \frac{1}{2}J''_{ij}(0)P_b^2 + \cdots .$$

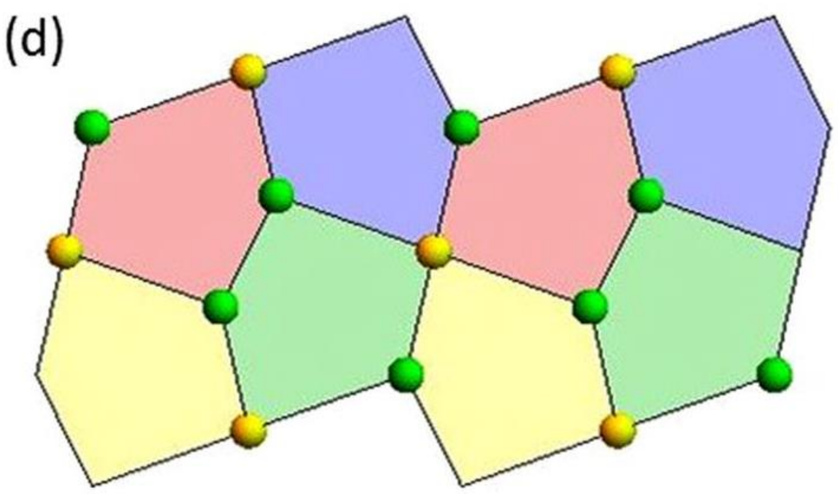
$$P_b \approx -\frac{\chi_1^{(0)}}{2V}\sum_{i,j} J'_{ij}(0)(\boldsymbol{S}_i \boldsymbol{S}_j),$$

From Mostovoy et al

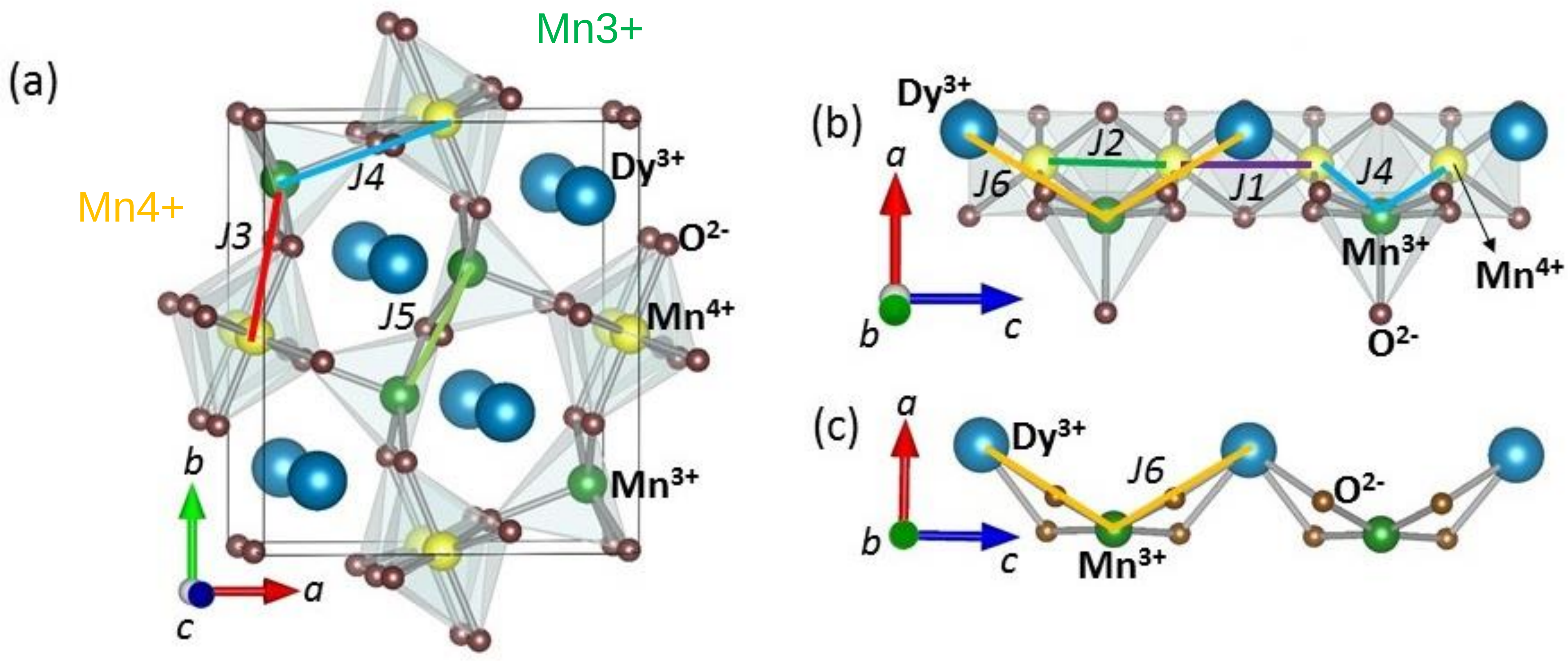
RMn_2O_5



Cairo pentagonal tiling

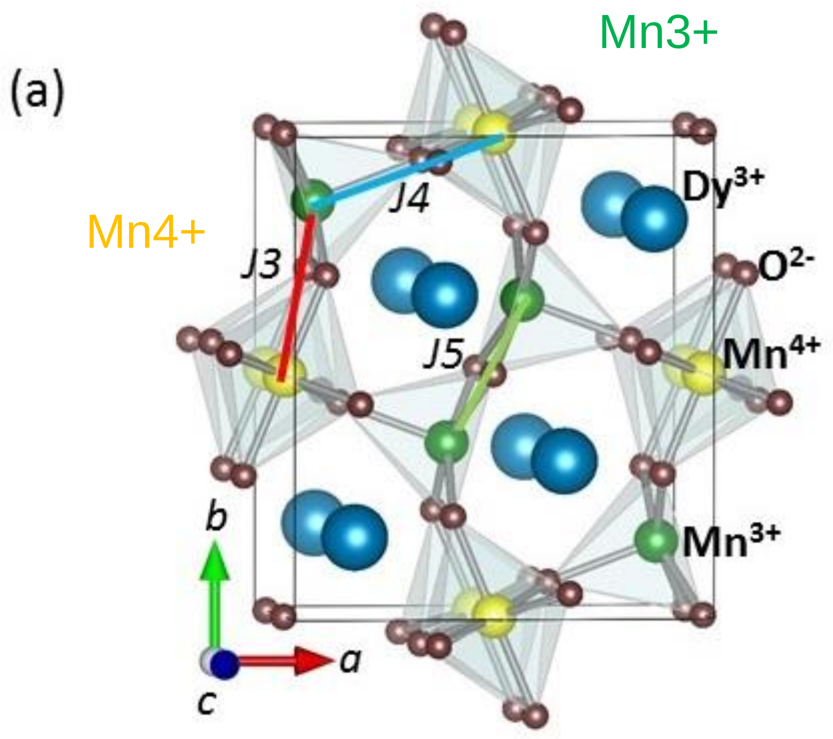


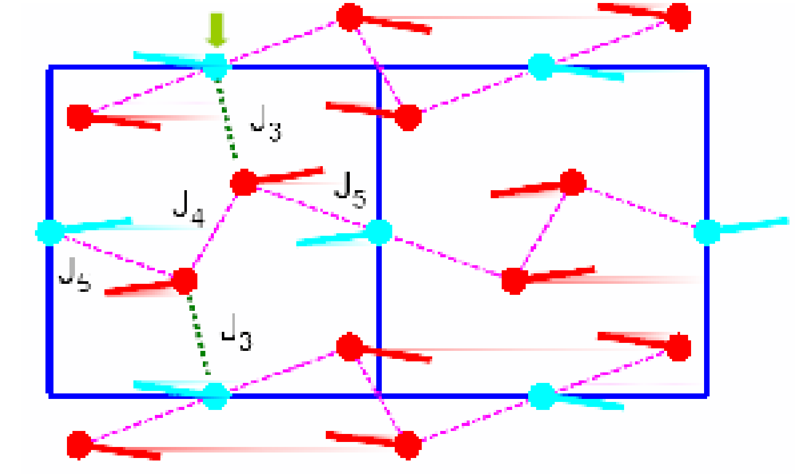
RMn_2O_5



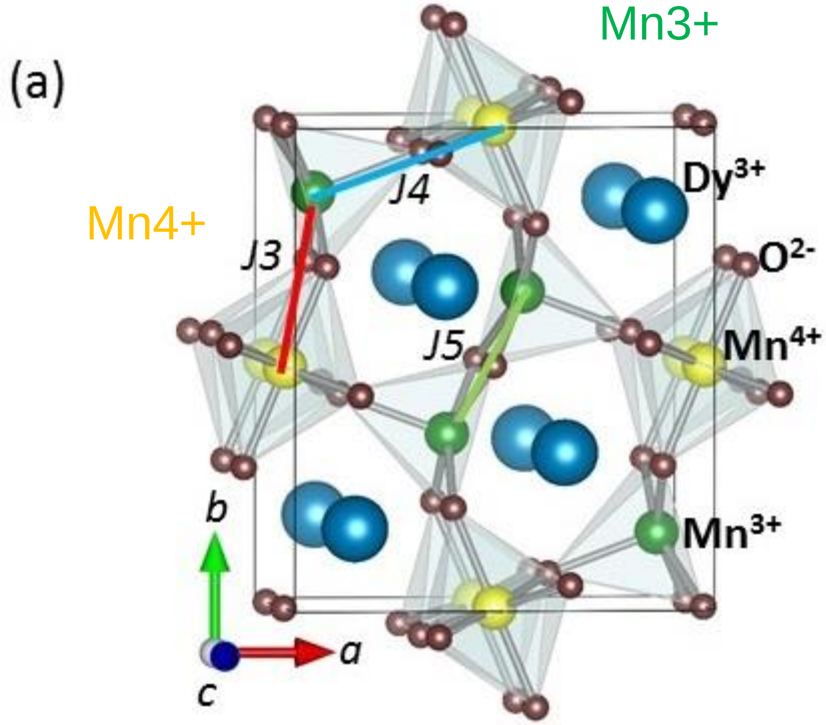
Mn3+
(a)
Mn4+
J4
J3
J5
Dy3+
O2-
Mn4+
Mn3+
b
a
c
(b)
Dy3+
J2
J6
J1
J4
a
Mn3+
Mn4+
O2-
b
c
(c)
a
Dy3+
J6
O2-
b
c
Mn3+

RMn_2O_5

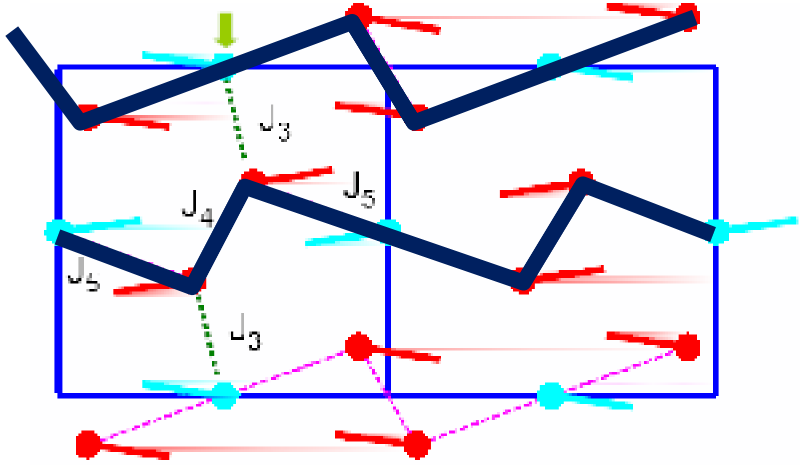




RMn_2O_5

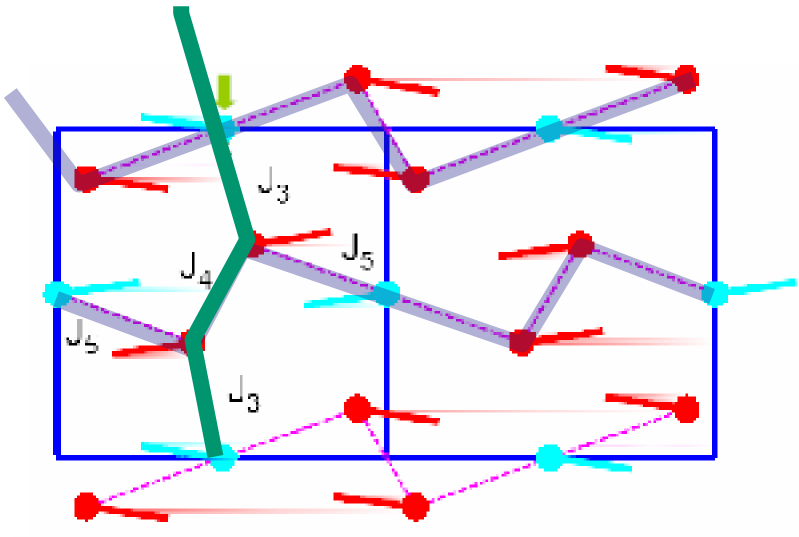


AF chains along a
Nearly commensurate structure

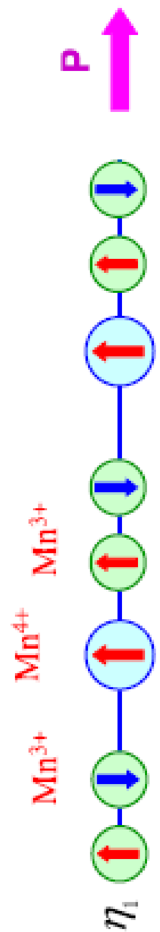


RMn_2O_5

Up Up Dw or Dw Dw Up chains along b



$$P_b \approx -\frac{\chi_1^{(0)}}{2V}\sum_{i,j} J'_{ij}(0)(S_i S_j),$$



P rises for CM magnetic structures

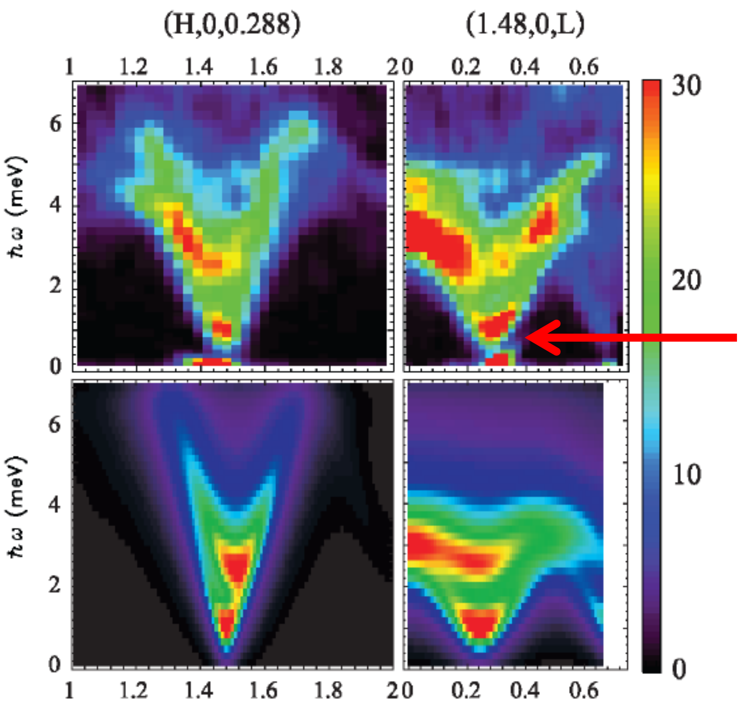
P is reduced or disappears for ICM structure

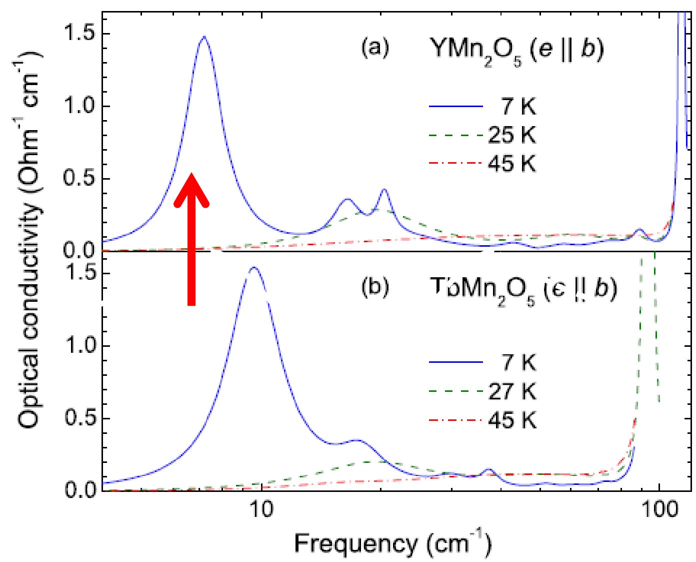
Conversely, no electromagnon for CM structure

.

The selection rule becomes Eω //b

YMn_2O_5





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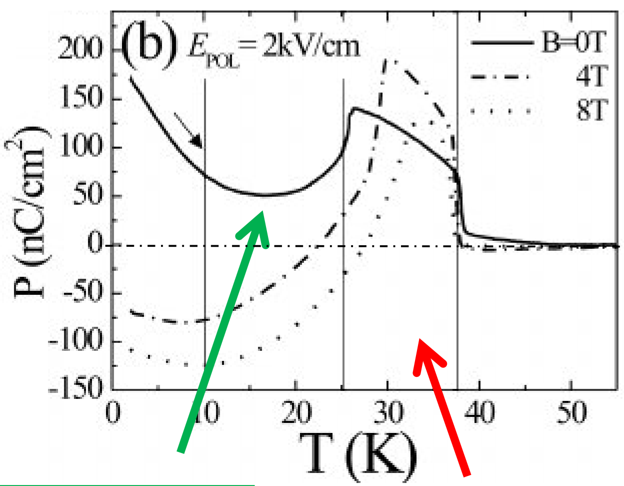
A. Pimenov, Nature Physics 2, 97 (2006)
R. ValdesAguilar, PRB 77, 092412 (2008)
N. Kida, PRB 78, 104414 (2008)
A. B. Sushkov, PRL 98, 027202 (2007)
B. Van Aken, Nature Materials 3, 164 (2004)
M. Cazayous, PRL 101, 037601 (2008)
Kim et al, PRL 107, 097401 (2011)
Blake et al, PRB 71 214402 (2005)

The electromagnon is the long wavelength perturbation of the ICM structure

$TbMn_2O_5$

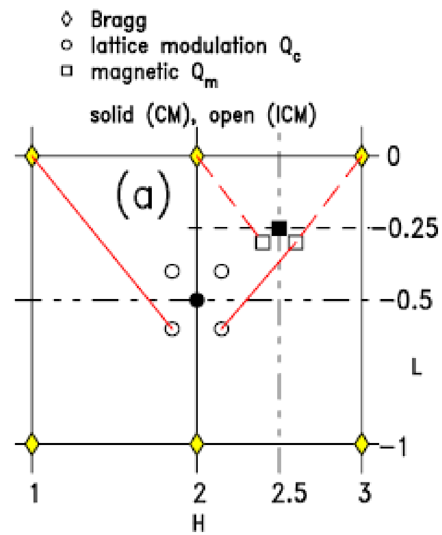
Polarization

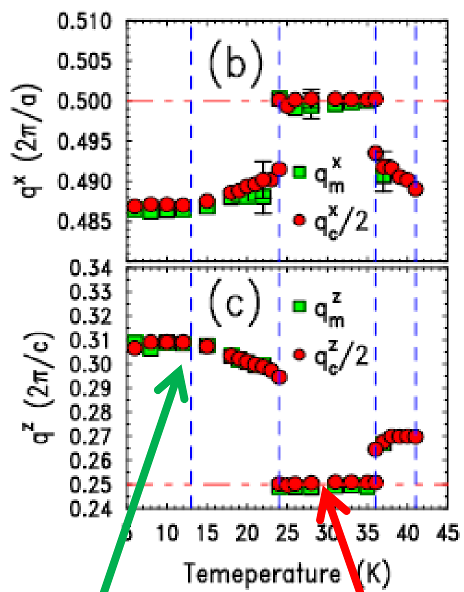


ICM (qx 0 qz)

CM (1/2 0 ¼)

Neutron diffraction





ICM (qx 0 qz)

CM (1/2 0 ¼)

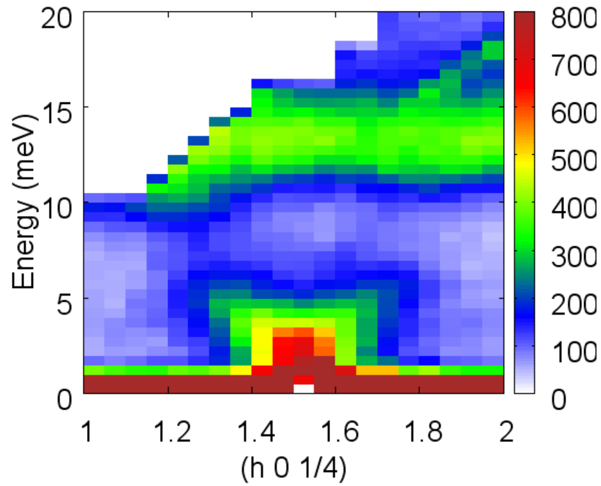
I. Kagomiya, et al, Ferroelectrics **280**, 131 (2002)
Inomata et al, J. Phys.: Condens. Matter **8**, 2673 (1996)
Kobayashi et al, J. Phys. Soc. Jpn. **73**, 1031 (2004), J. Phys. Soc. Jpn. **73**, 1593 (2004).
P.G. Radaelli and L.C. Chapon, J. Phys. Condens. Matter. 20, 434213 (2008)

$TbMn_2O_5$

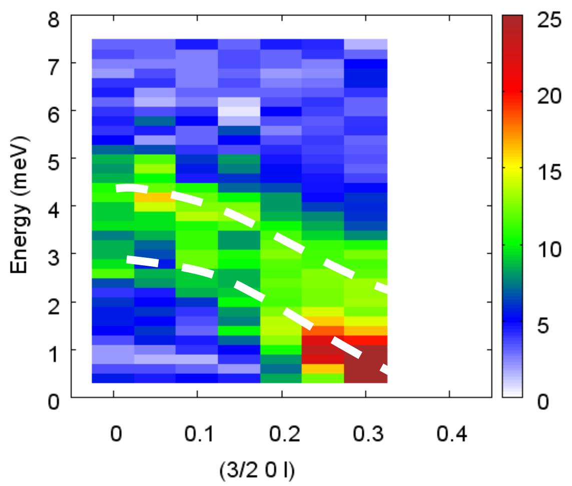
Unfortunately, it is not a 1 cm^3 sample …

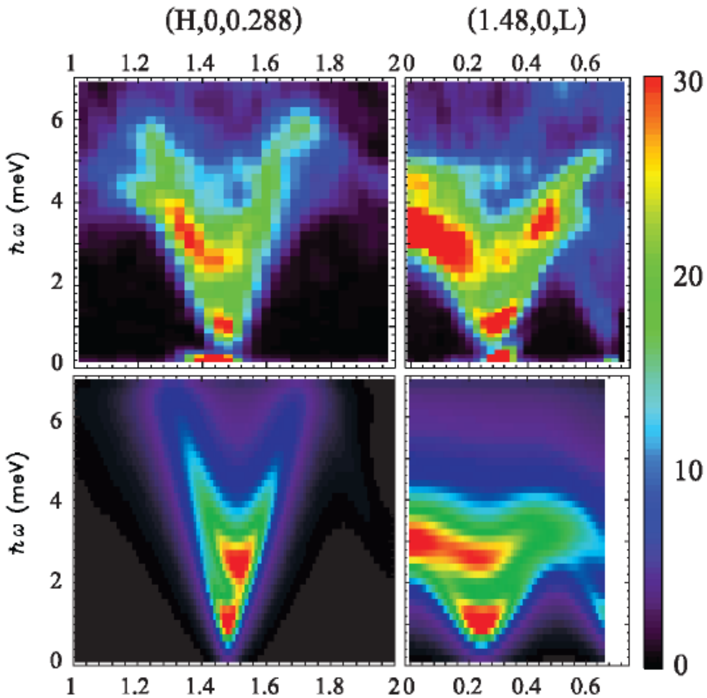
$TbMn_2O_5$

2T @ LLB



4F @ LLB

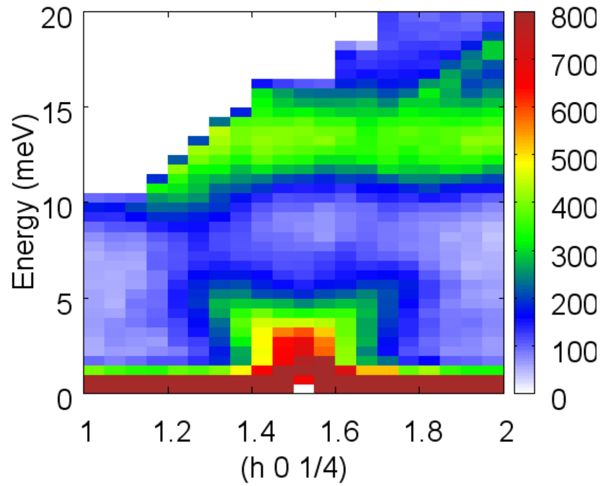




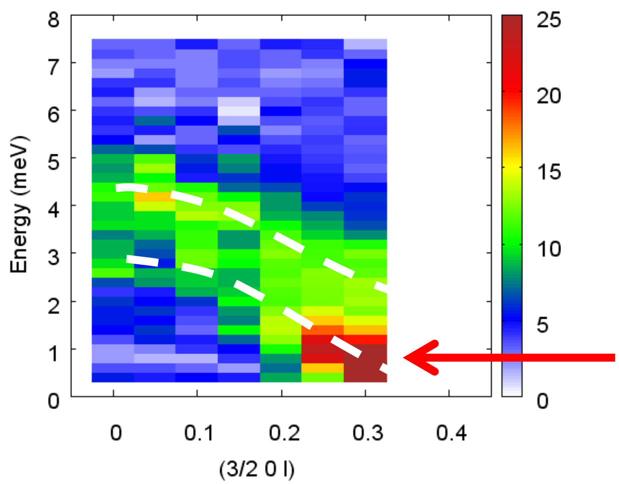
Kim et al, PRL 107, 097401 (2011)

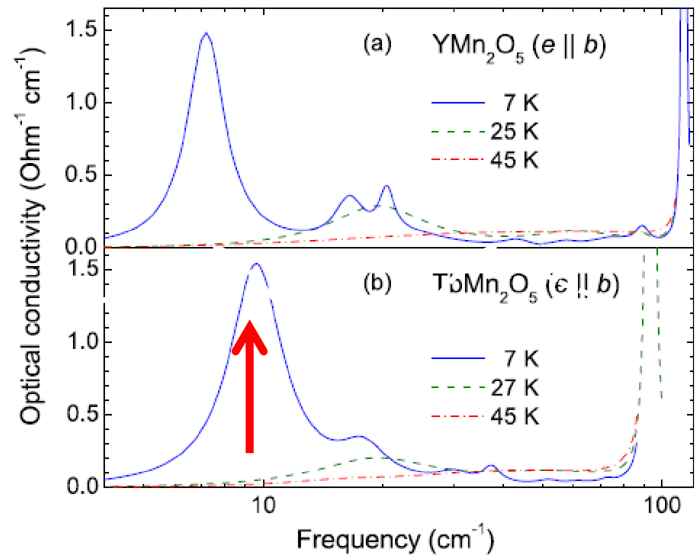
$TbMn_2O_5$

2T @ LLB



4F @ LLB





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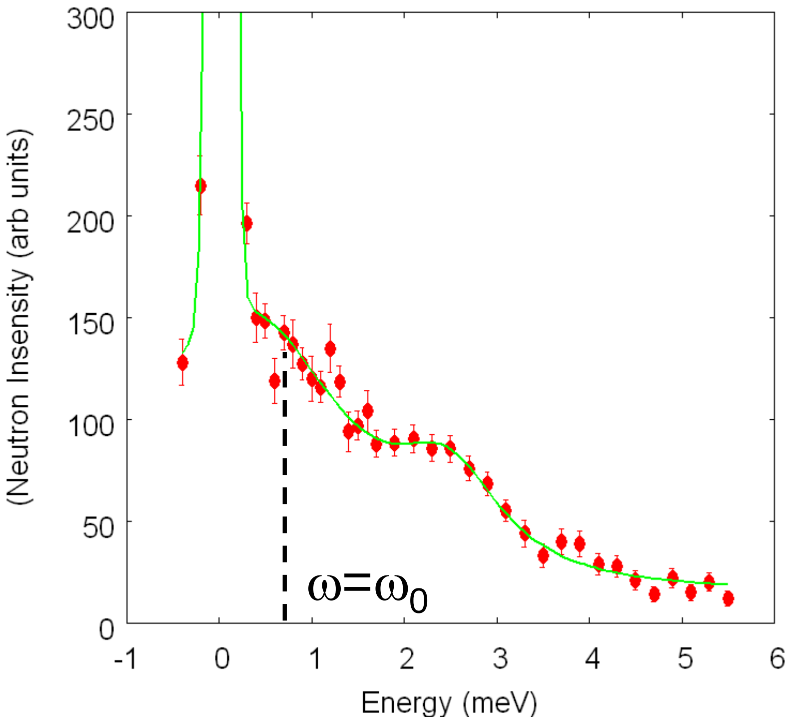
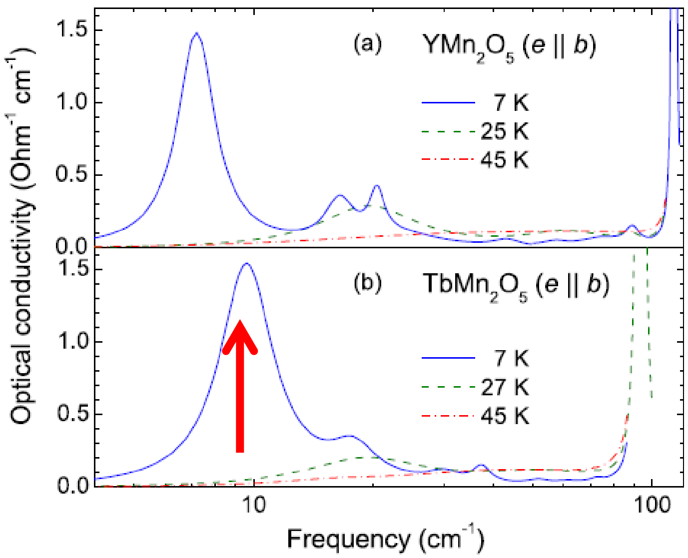
A. B. Sushkov, PRL 98, 027202 (2007)
B. Van Aken, Nature Materials 3, 164 (2004)
M. Cazayous, PRL 101, 037601 (2008)
Kim et al, PRL 107, 097401 (2011)
Blake et al, PRB 71 214402 (2005)

$TbMn_2O_5$

4F2 @ LLB

T=30K (CM)





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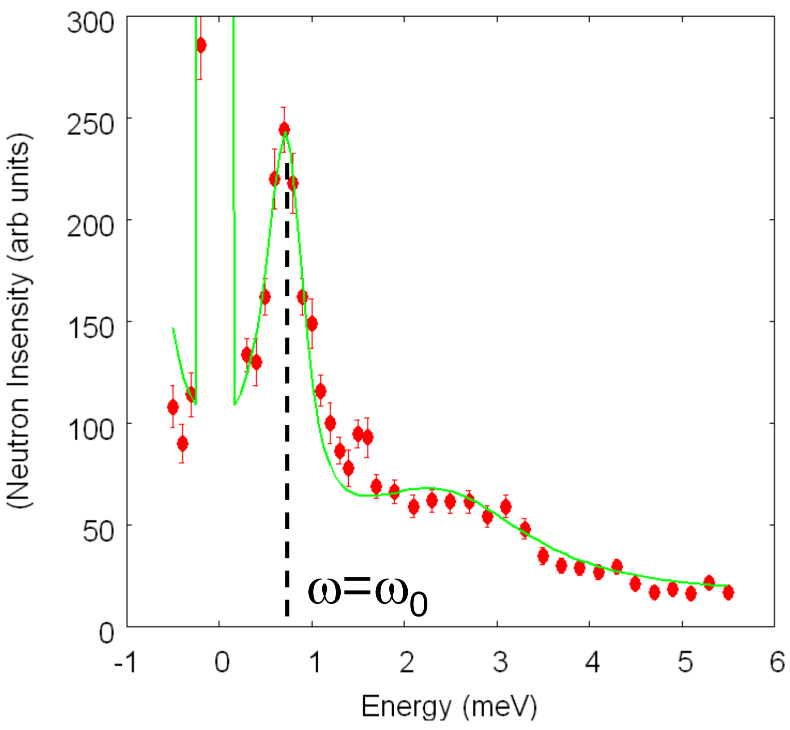
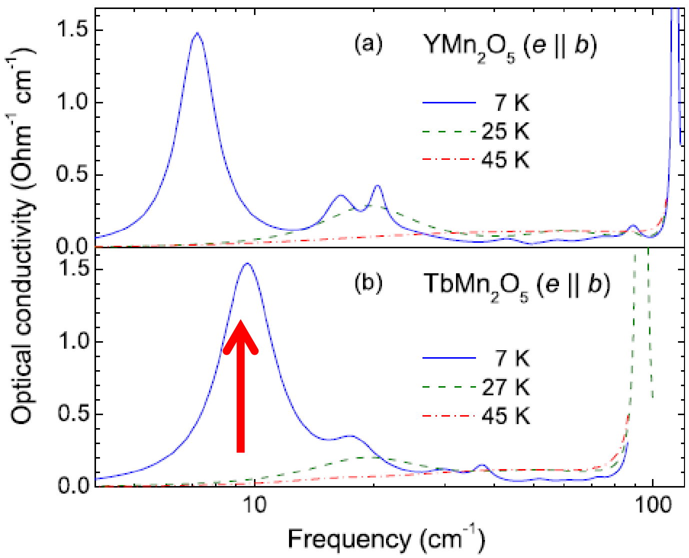
Kim et al, PRL 107, 097401 (2011)
Blake et al, PRB 71 214402 (2005)

$TbMn_2O_5$

4F2 @ LLB

T=17K (ICM2)





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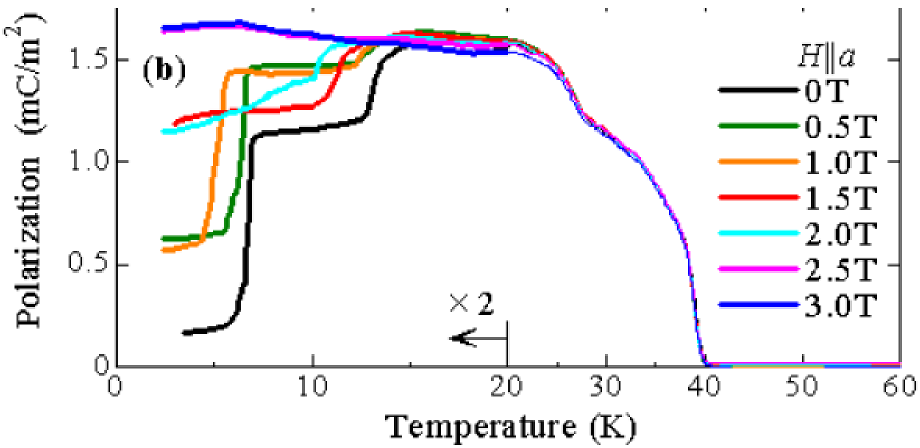
M. Cazayous, PRL 101, 037601 (2008)
Kim et al, PRL 107, 097401 (2011)
Blake et al, PRB 71 214402 (2005)

$DyMn_2O_5$

Polarization



Ferroelectric phases 8 < T < 39 K

Spiral along c

K = (1/2 0 ¼) at intermediate T

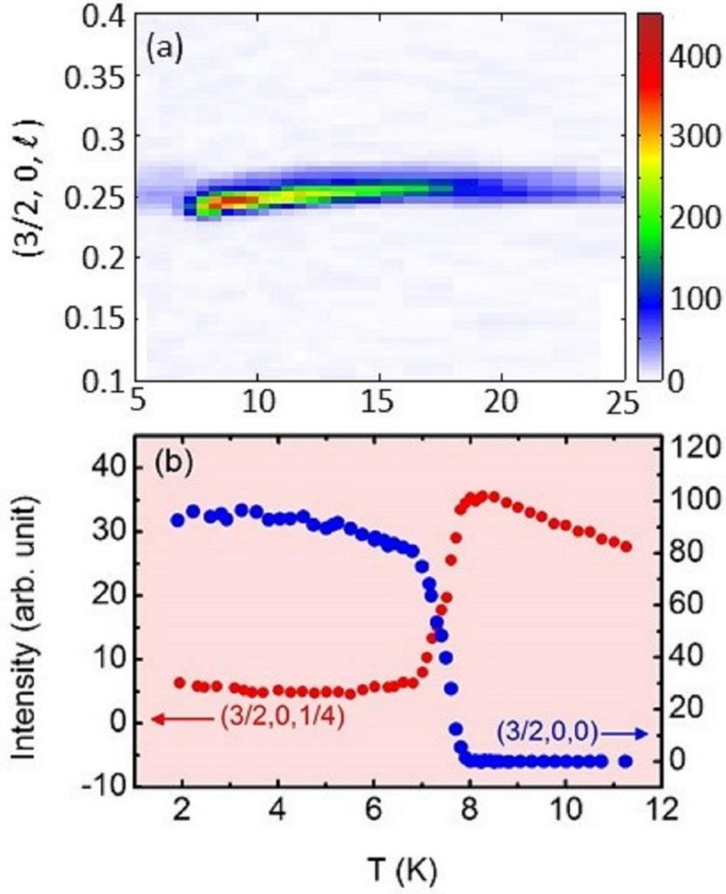
CM at low T

K = (1/2, 0 ,0)

No electromagnon (at H=0), so far

Neutron diffraction

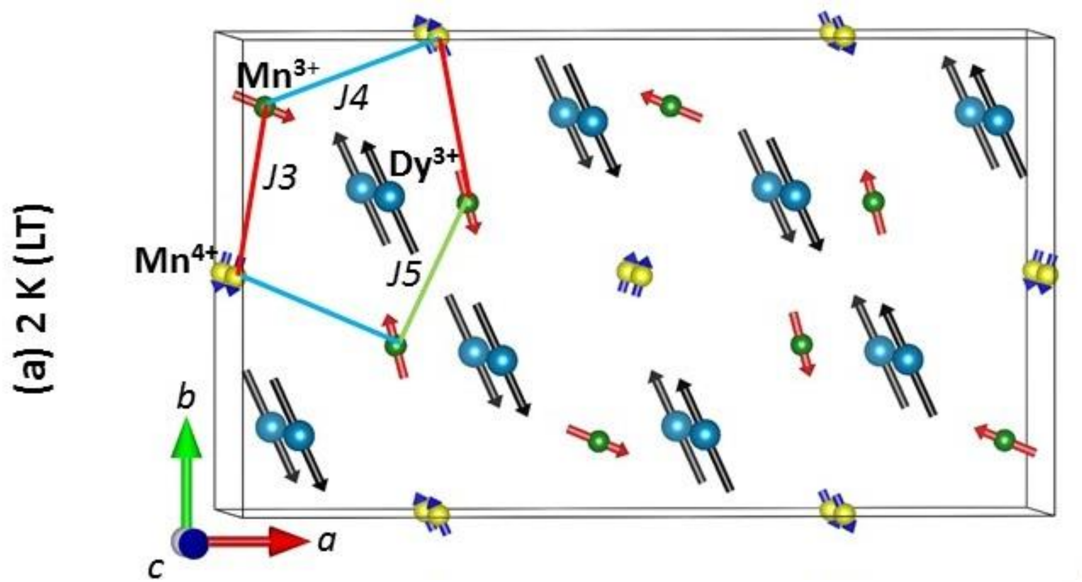
D23 CRG@ILL

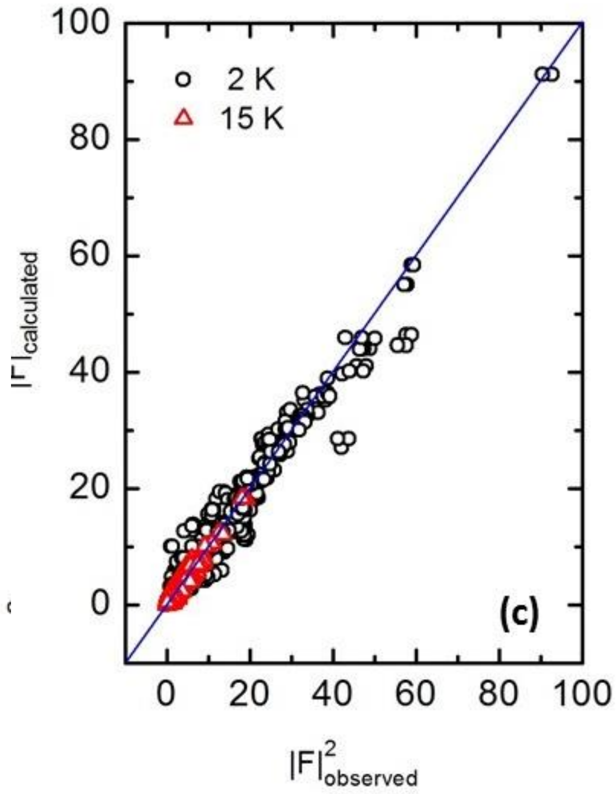


$DyMn_2O_5$

T < 8 K

D23 CRG@ILL



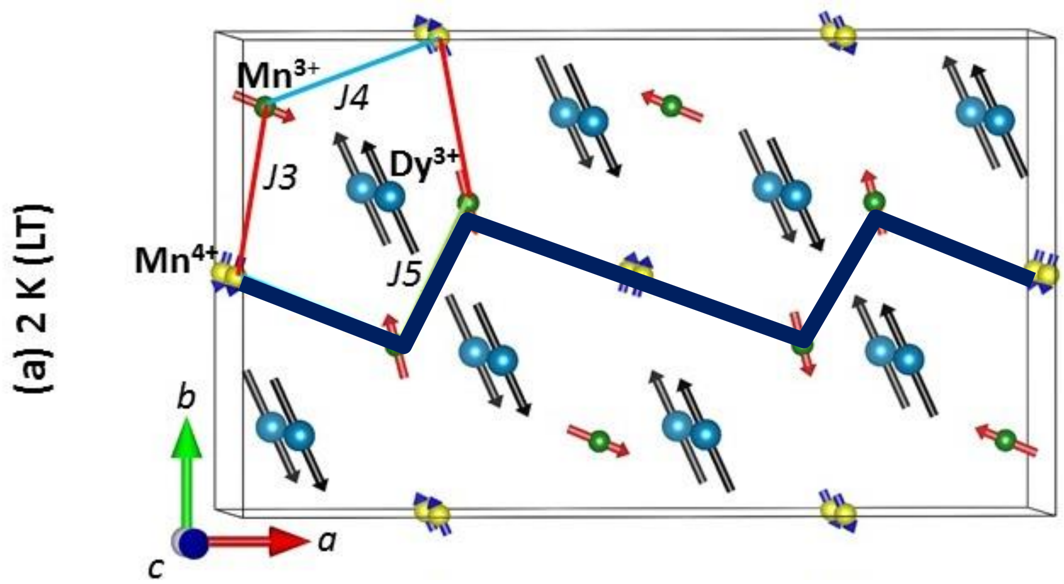


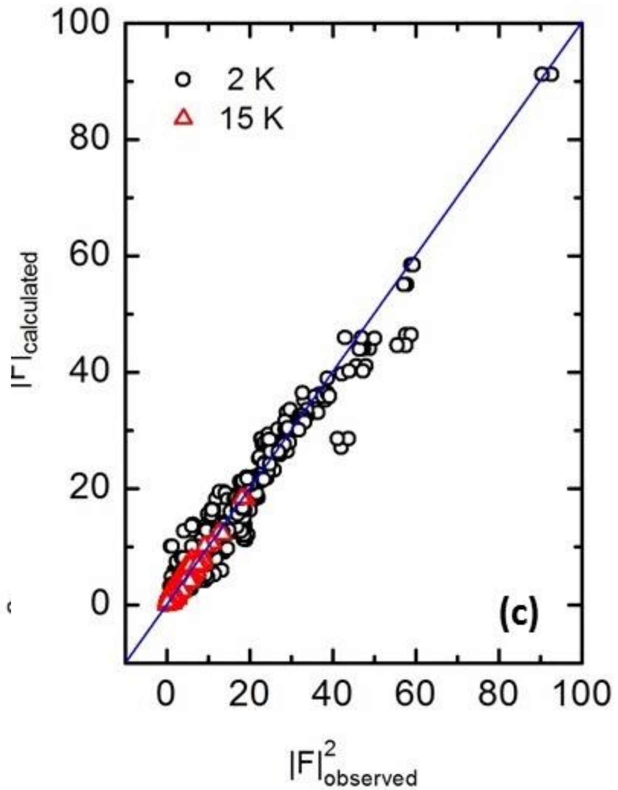
Solving the magnetic structure on a single crystal neutron diffractometer

$DyMn_2O_5$

T < 8 K

D23 CRG@ILL



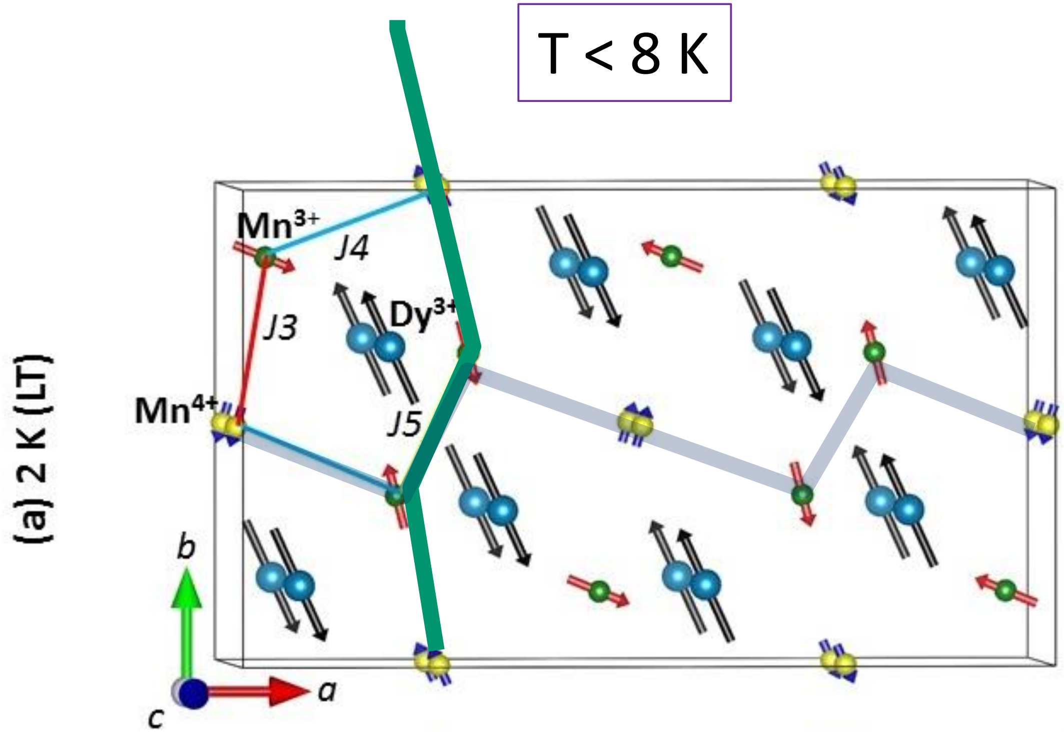


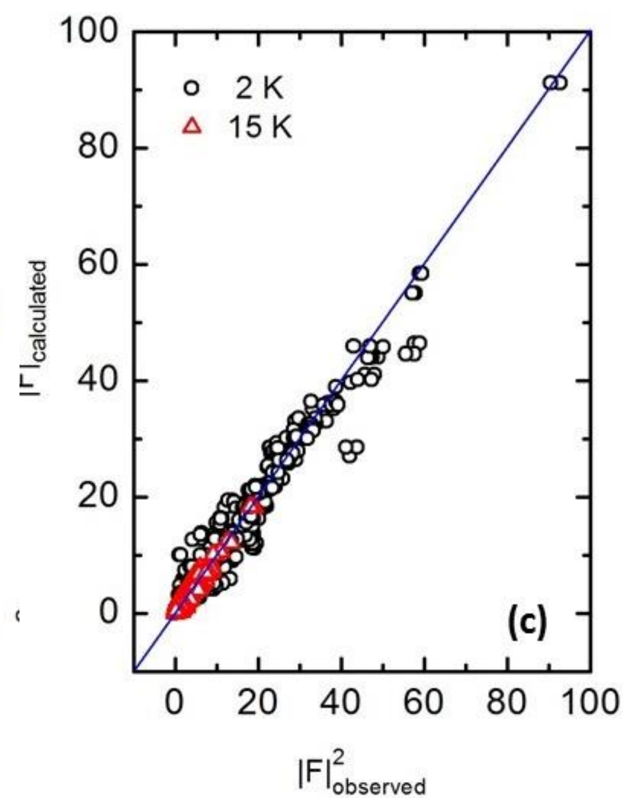
AF chains along a
Up-Up-Dw chains along b

$DyMn_2O_5$

T < 8 K

D23 CRG@ILL





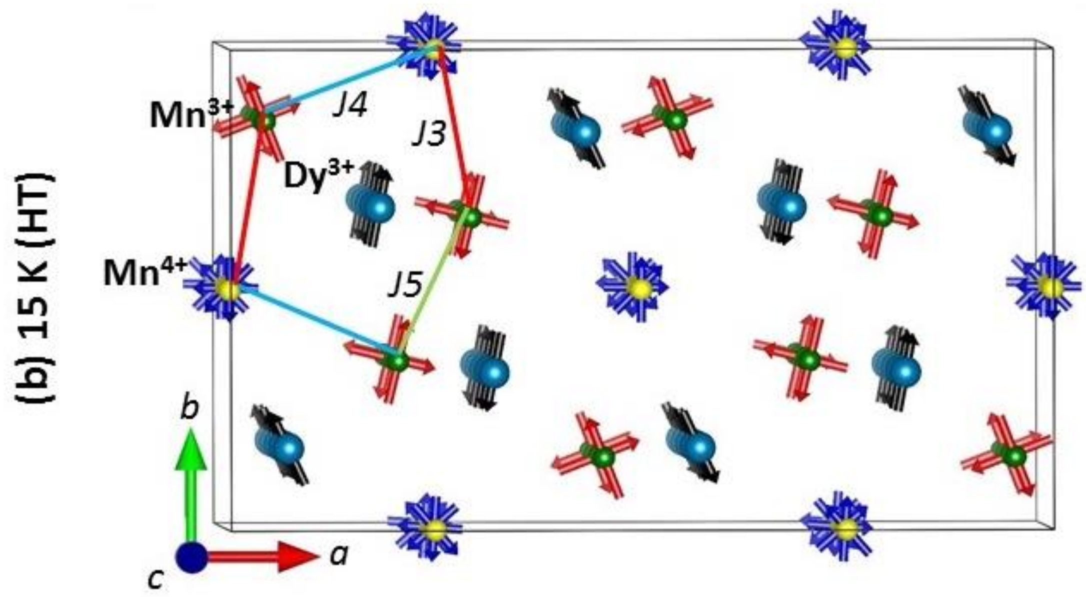
AF chains along a

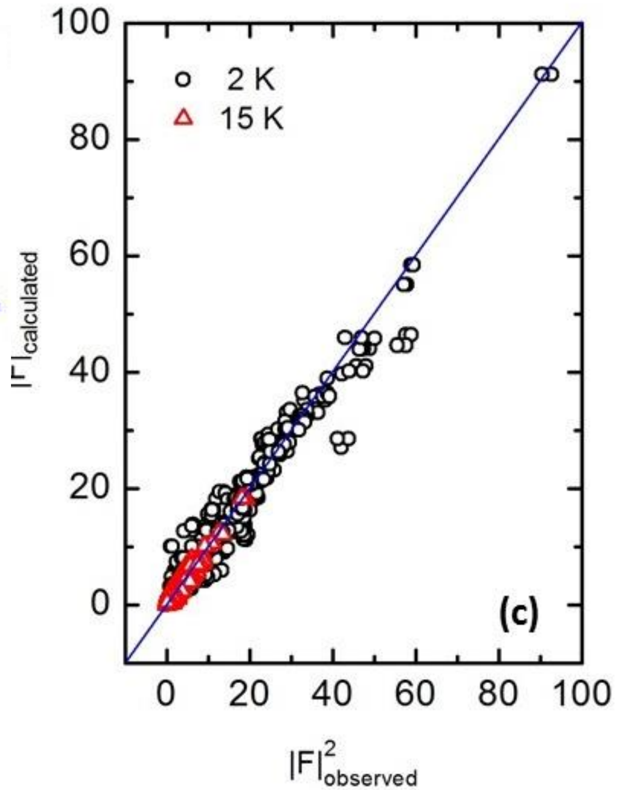
Up-Up-Dw chains along b

$DyMn_2O_5$

T > 8 K

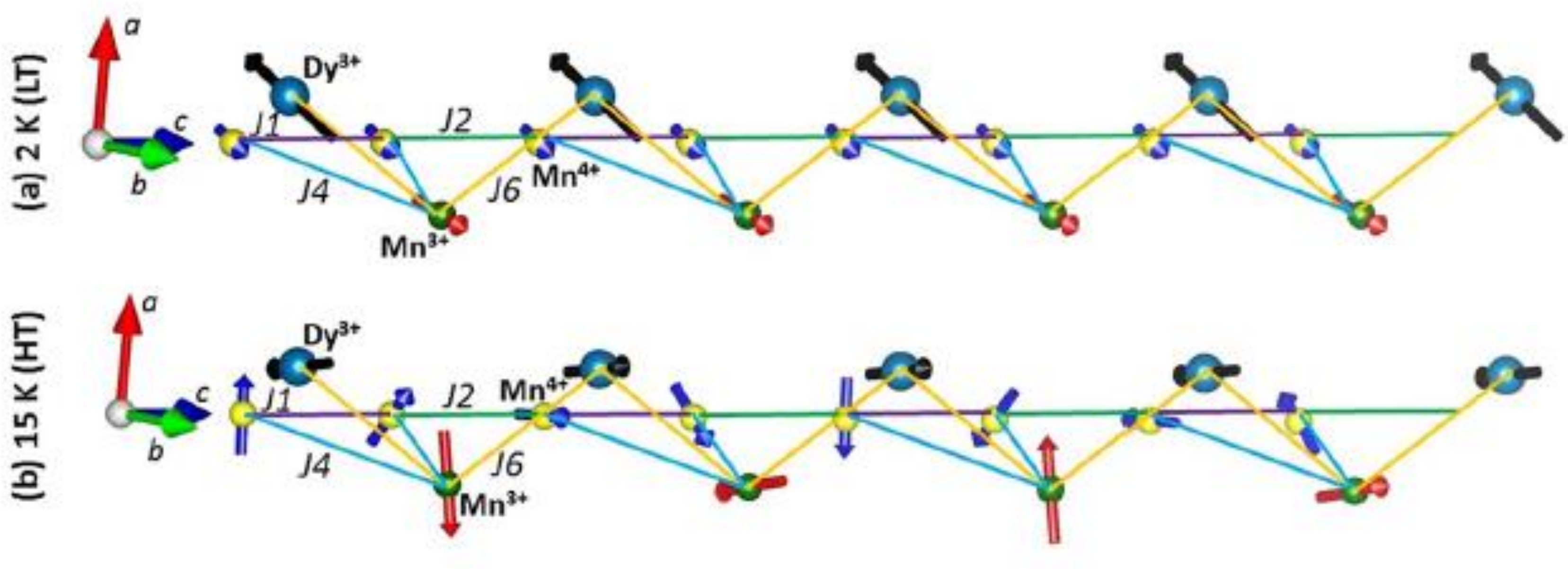
D23 CRG@ILL





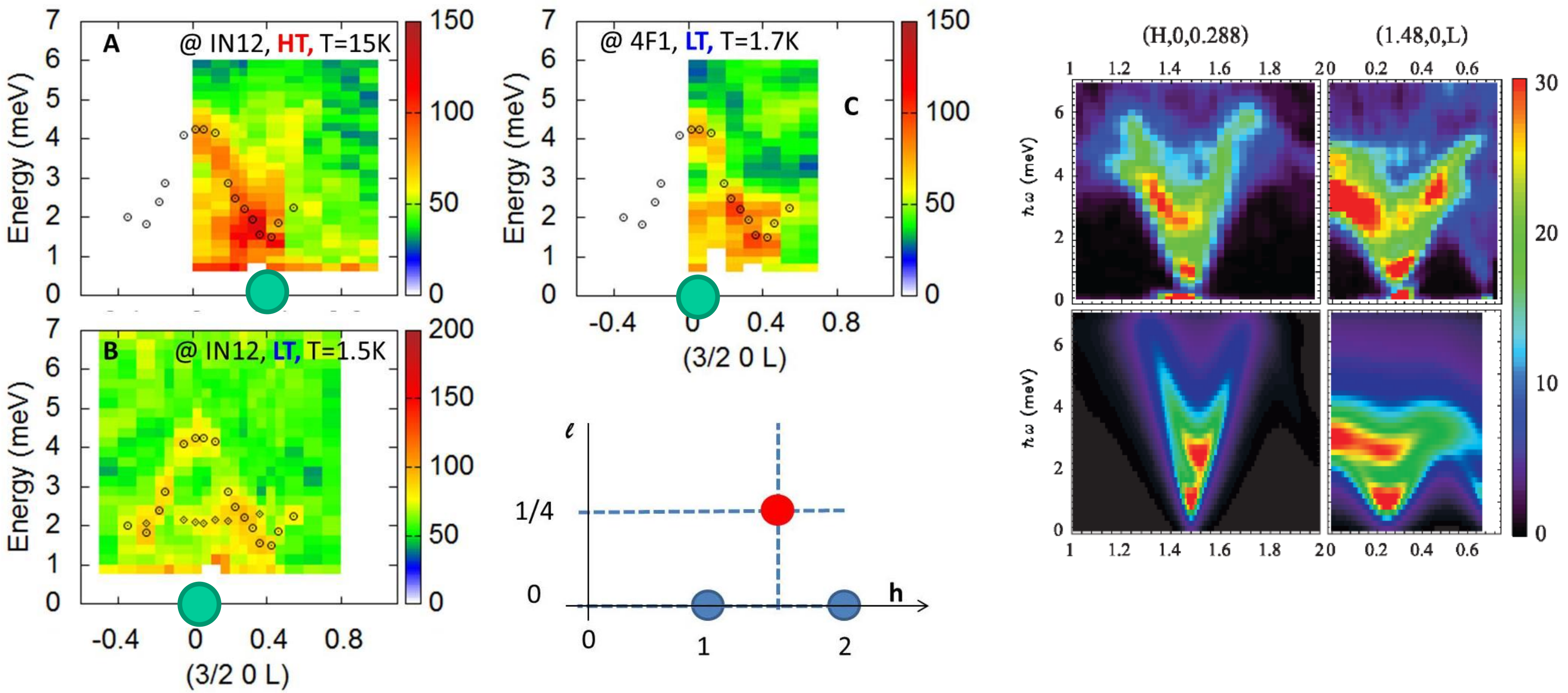
Mn4+ rotate by 45°
Mn3+ rotate by 90°
Dy is Up-Up-Dw-Dw

$DyMn_2O_5$



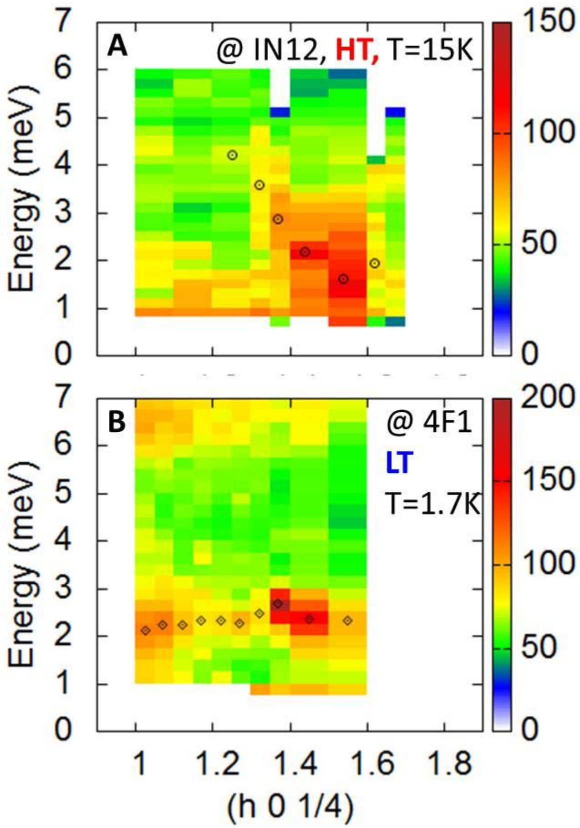
Suggests that Dy is likely characterized by a strong easy axis, since those spins keep more or less the same direction whatever T

$DyMn_2O_5$

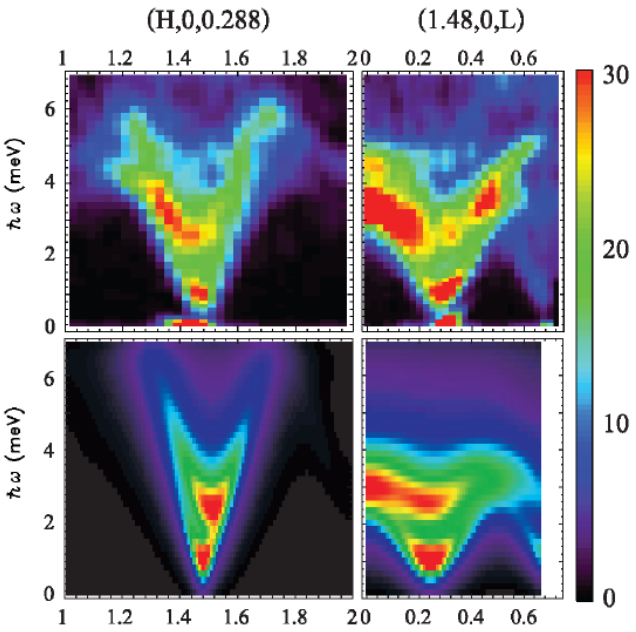


Little evolution of the dispersion despite the change of the magnetic structure
Suggests that the K=(1/2 0 0) LT phase is likely NOT stabilized by J1-J2-J3-J4-J5 exchange.

$DyMn_2O_5$



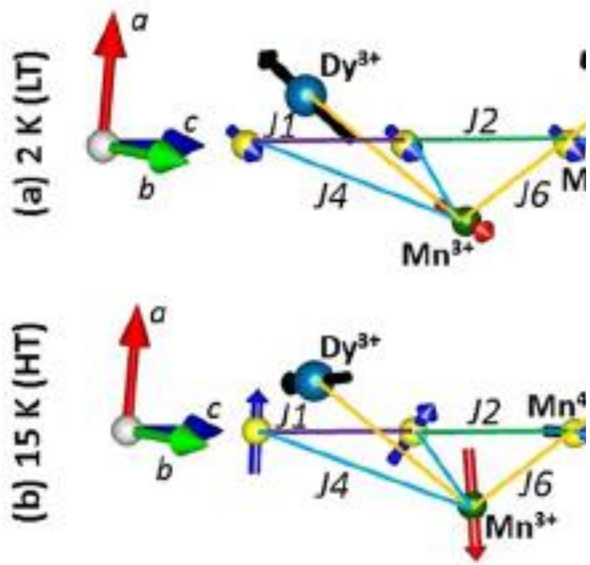
A
@ IN12, HT, T=15K
B
@ 4F1
LT
T=1.7K
Energy (meV)
(h 0 1/4)

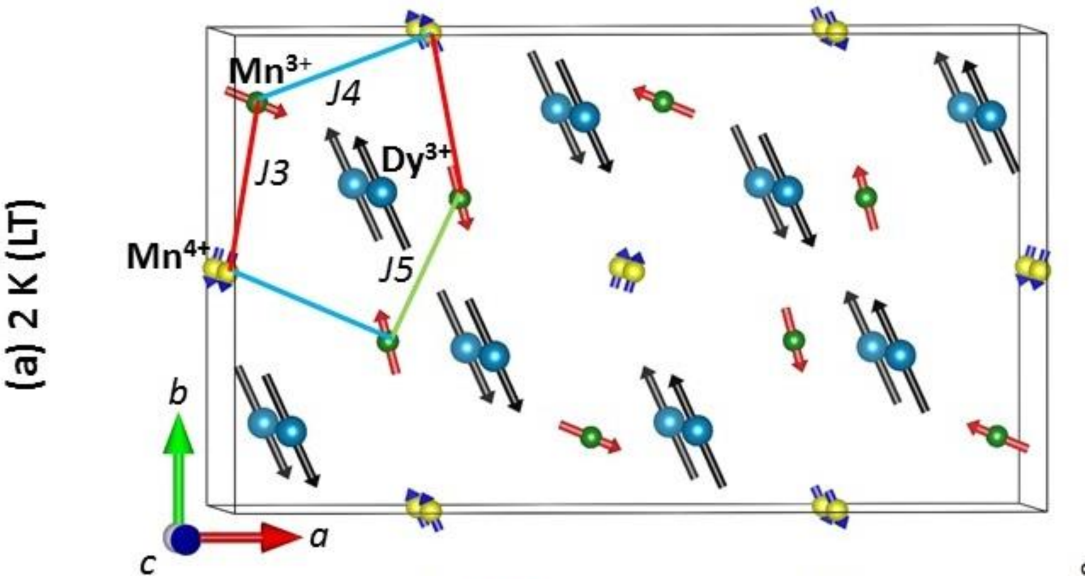


(H,0,0.288)
(1.48,0,L)
ħω (meV)

$DyMn_2O_5$

- Introduce an effective J6 coupling bewteen 3d and 4f spins





- Assume very large easy axis anisotropy for Dy
- J6 may overcome J1-J2-J3 at low T

Conclusions

- ❖ $TbMn_2O_5$ follows theoretical precictions. Resembles much YMn_2O_5

- ❖ $DyMn_2O_5$ is different ! Back to the IDM model ?

  - Similar SW dispersion in the ICM phase, yet no electromagnon

  - Our work suggests that this is likely due to the 3d-4f coupling which remains to be incorporated in the theory

Thank you for your attention