Frustrated Magnets with Pyrochlores Structure.

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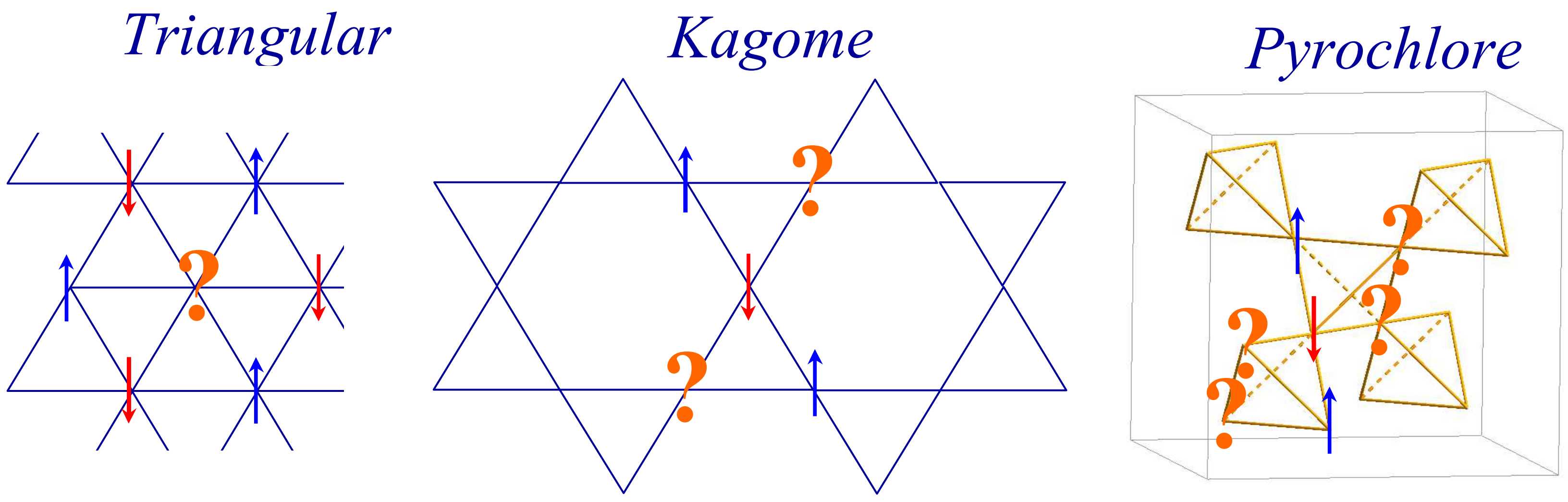
OUTLINE

- **Frustrated magnets**
- **Local anisotropy and Site Susceptibility Tensor**
- **Polarized neutron diffraction (PND)**
- ***Longitudinal* and *Transverse* susceptibilities in cubic pyrochlores**
- ***Ising* versus *XY* anisotropy in $R_2Ti_2O_7$ (R=Ho, Tb, Er and Yb) as seen by PND**

Geometrically frustrated magnets

Frustration: first neighbour interactions can not be satisfied simultaneously

High sensitivity to perturbations: pressure, magnetic field

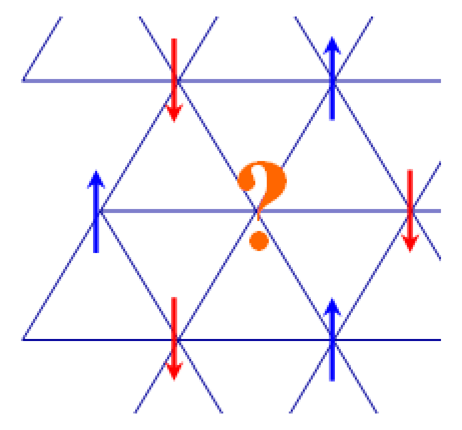


Frustration and ground state entropy increasing

A. P. Ramirez ***Nature*** *1999*
S. T. Bramwell ***Science*** *2001*

2D Geometrically frustrated magnets

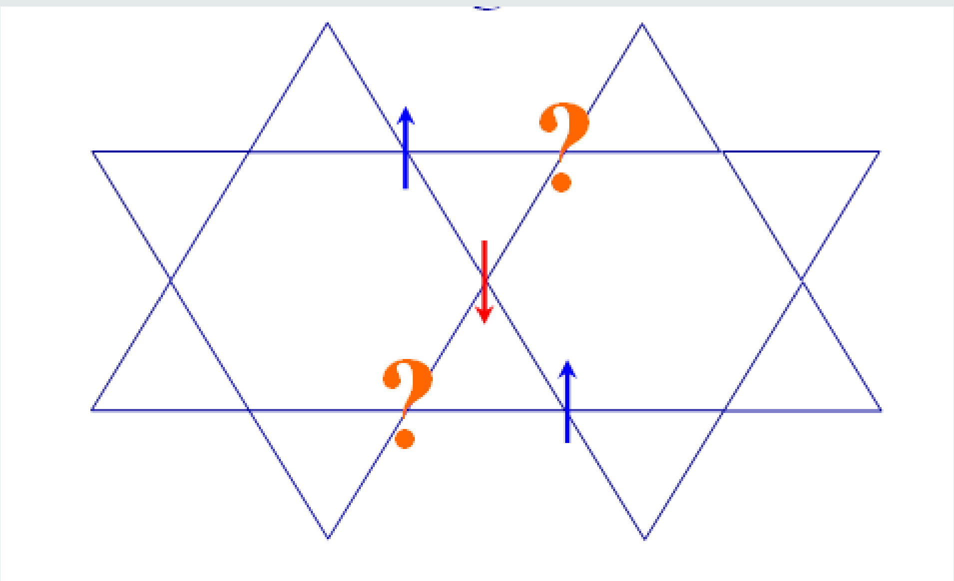
Triangular lattice



NNN-exchange can lift the SG degeneracy

2D Geometrically frustrated magnets

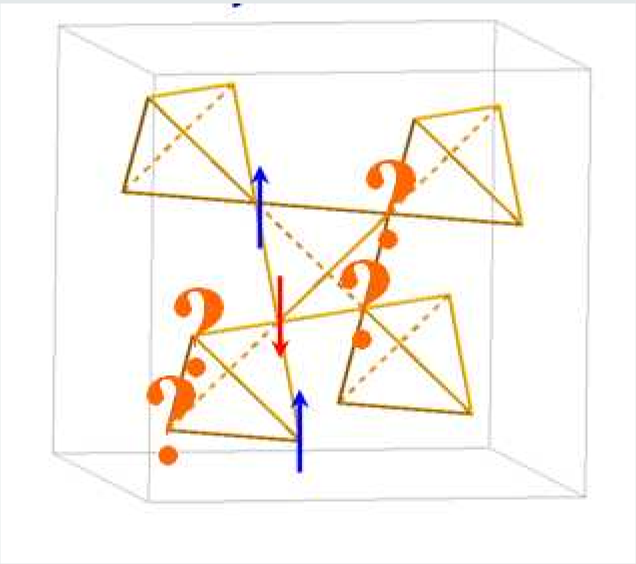
Kagome lattice



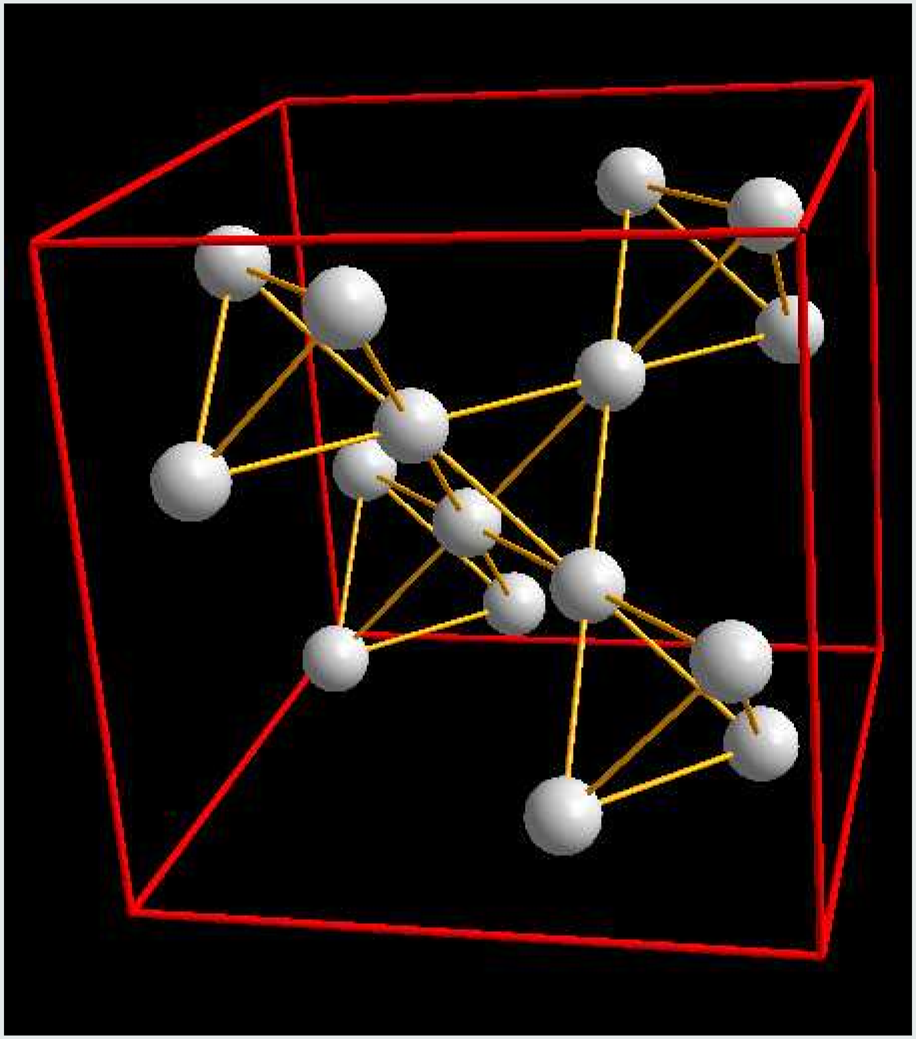
Less Next Nearest Neighbour more frustration

3D Geometrically frustrated magnets

Pyrochlore lattice



Symmetry of R Site in $R_2T_2O_7$



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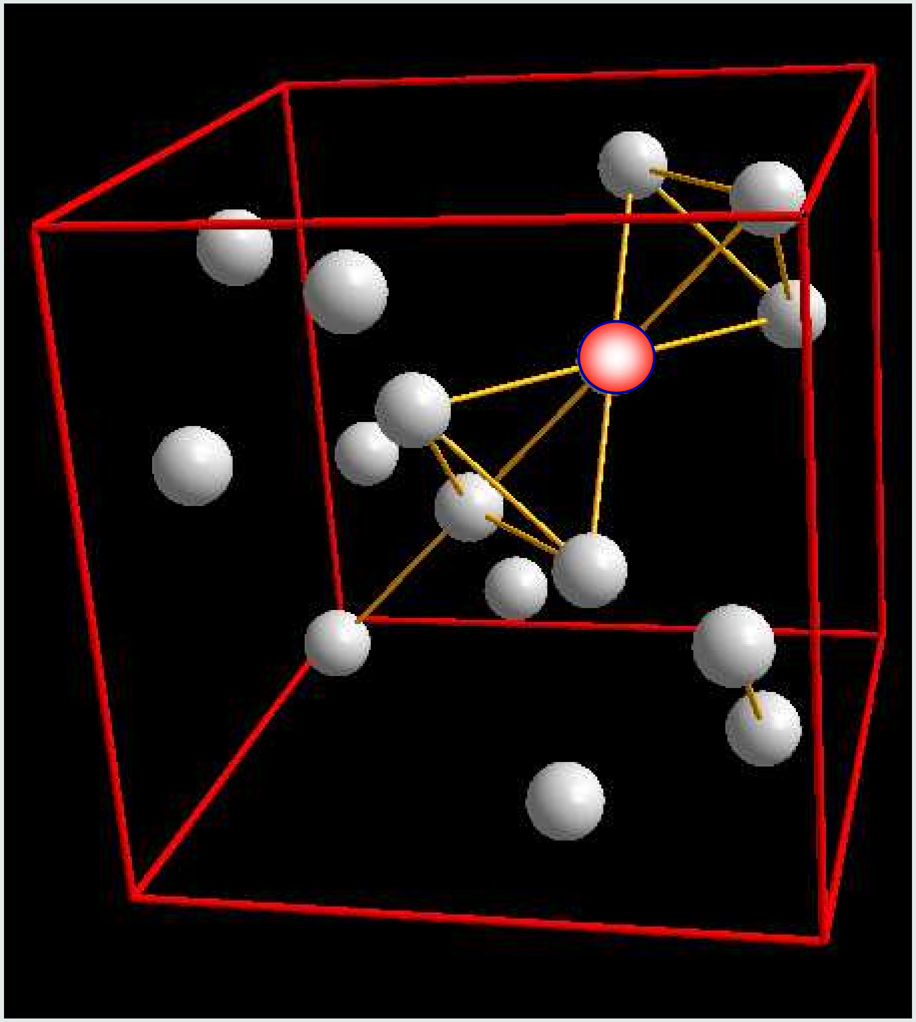

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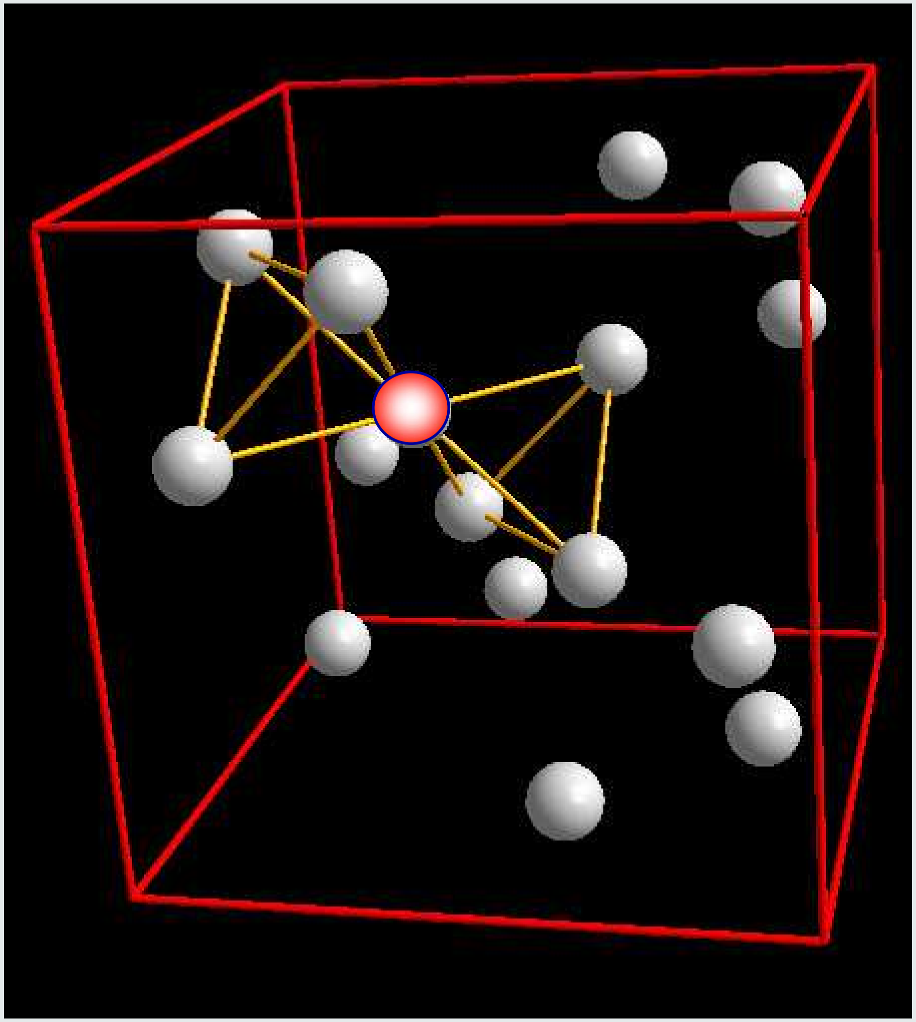

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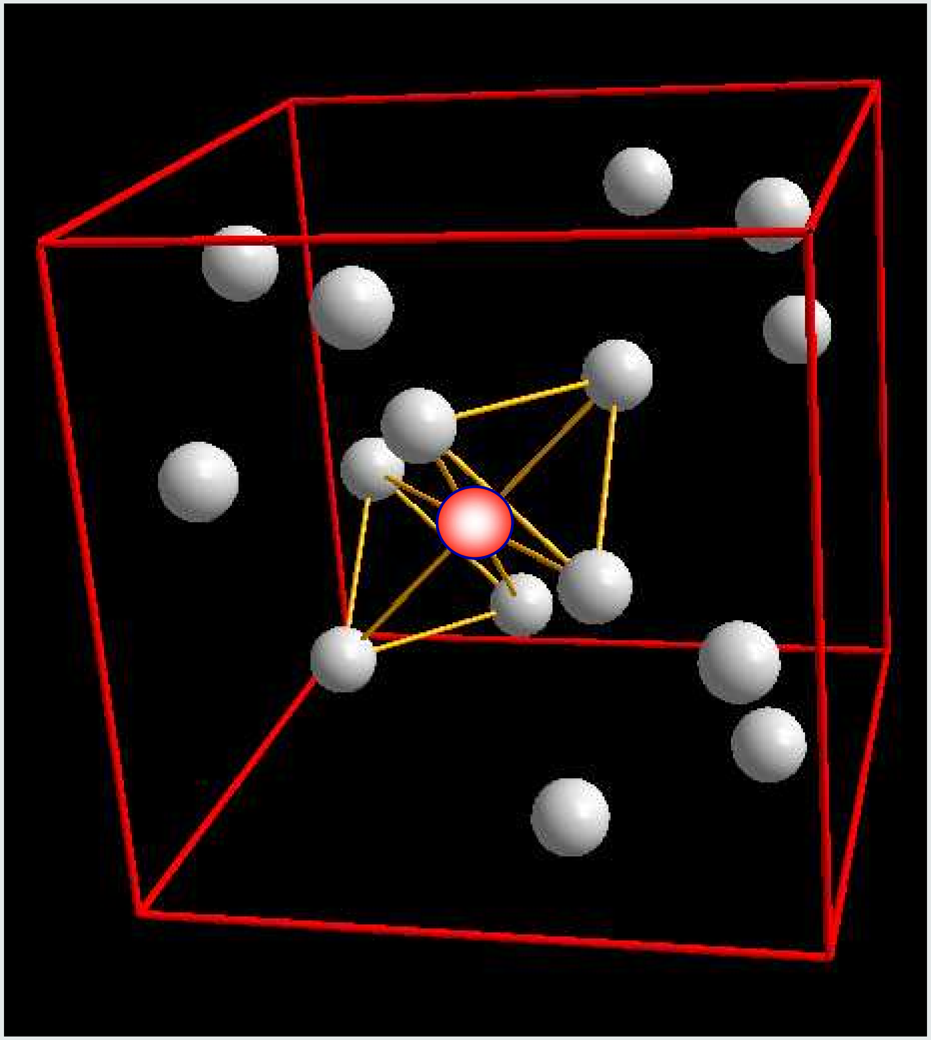

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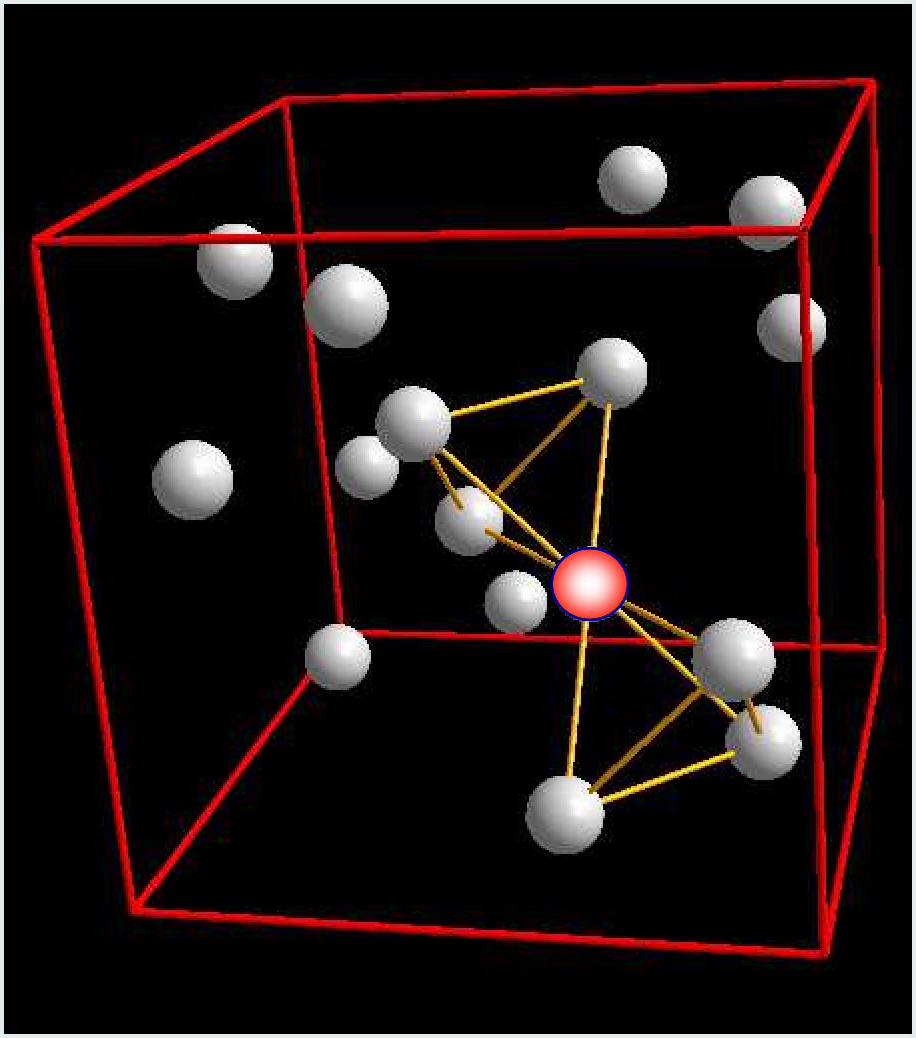

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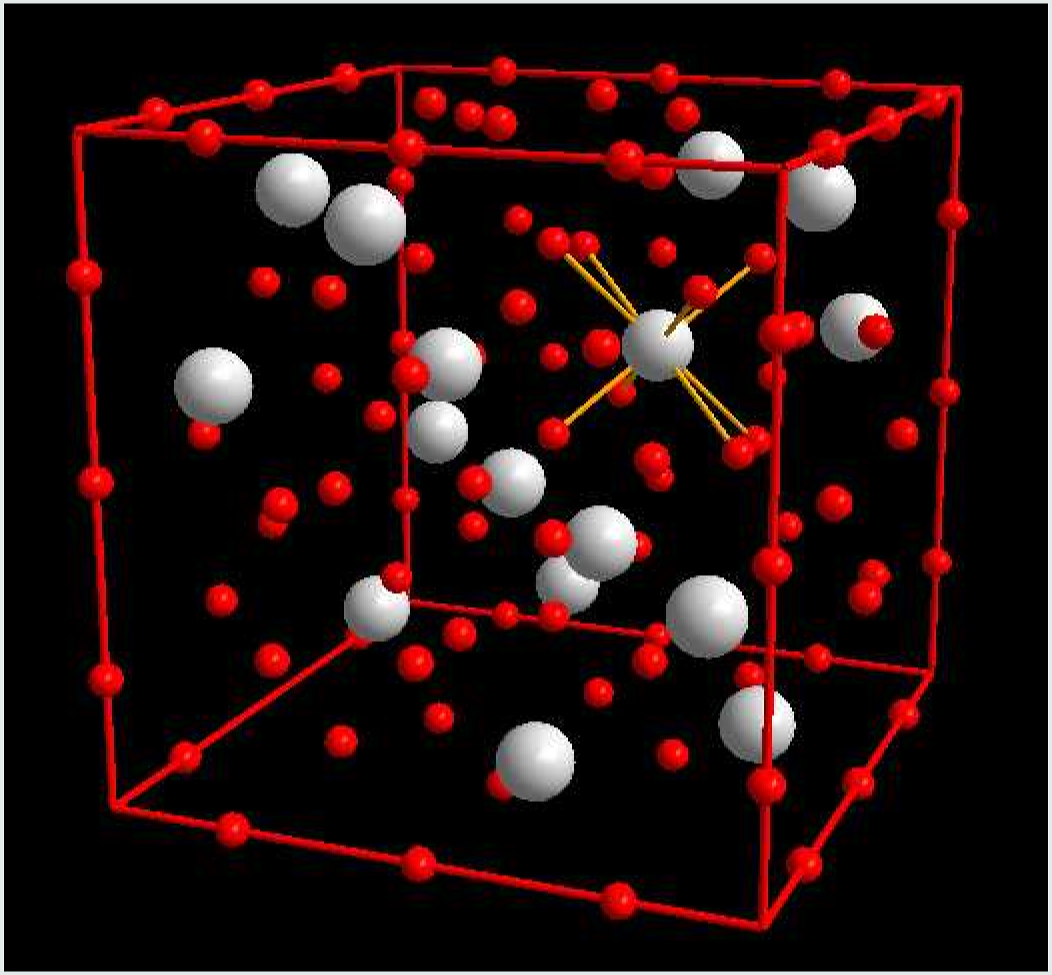
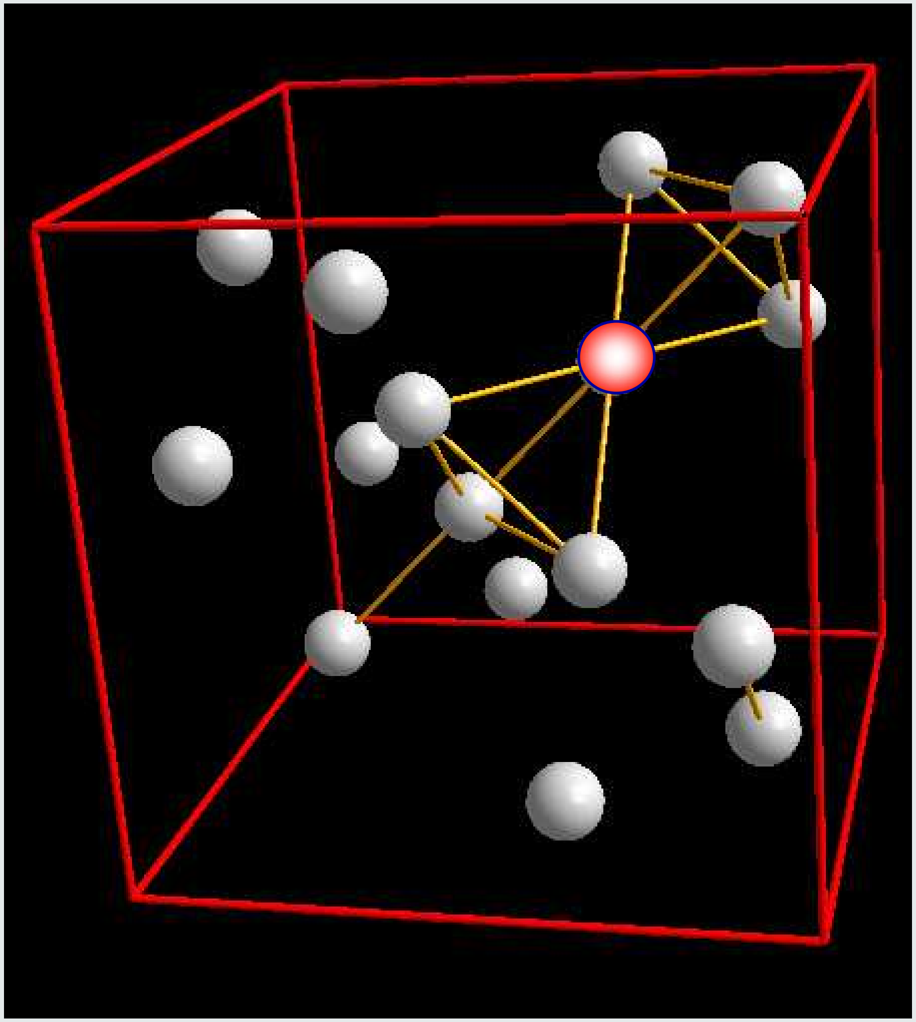


2x Tb-O1 2.19 A

6 xTb-O2 2.51 A

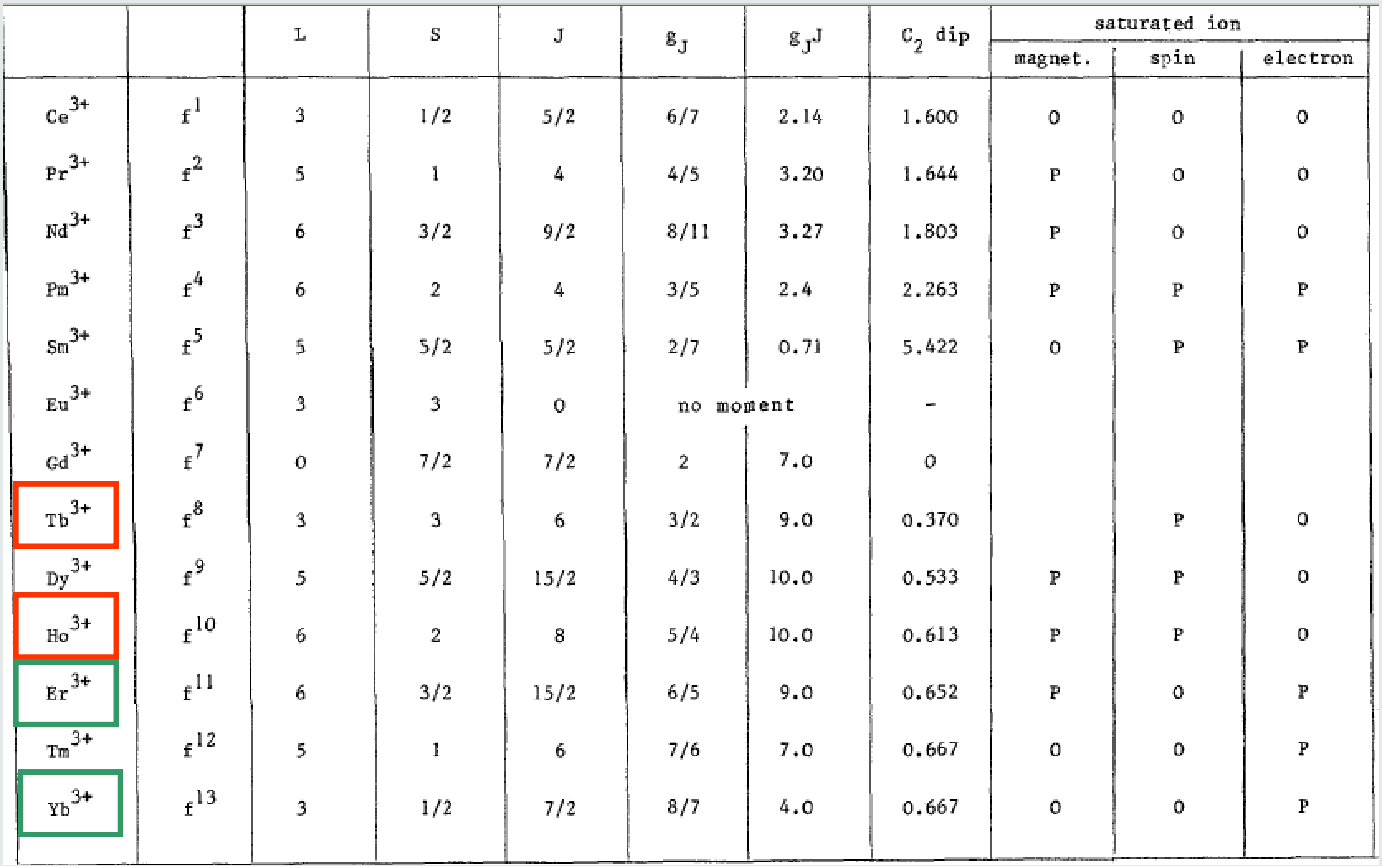
Symmetry of R Site in $R_2T_2O_7$



Tb-O1 2.19 A
Tb-O2 2.51 A

Orbital *L*, Spin *S* and Total *J* moments in 4f

Due to Strong *L-S* coupling *J* is a good quantum number



		L	S	J	g_J	$g_J J$	C_2 dip	saturated ion		
								magnet.	spin	electron
Ce^{3+}	f^1	3	1/2	5/2	6/7	2.14	1.600	0	0	0
Pr^{3+}	f^2	5	1	4	4/5	3.20	1.644	P	0	0
Nd^{3+}	f^3	6	3/2	9/2	8/11	3.27	1.803	P	0	0
Pm^{3+}	f^4	6	2	4	3/5	2.4	2.263	P	P	P
Sm^{3+}	f^5	5	5/2	5/2	2/7	0.71	5.422	0	P	P
Eu^{3+}	f^6	3	3	0	no moment		-			
Gd^{3+}	f^7	0	7/2	7/2	2	7.0	0			
Tb^{3+}	f^8	3	3	6	3/2	9.0	0.370		P	0
Dy^{3+}	f^9	5	5/2	15/2	4/3	10.0	0.533	P	P	0
Ho^{3+}	f^{10}	6	2	8	5/4	10.0	0.613	P	P	0
Er^{3+}	f^{11}	6	3/2	15/2	6/5	9.0	0.652	P	0	P
Tm^{3+}	f^{12}	5	1	6	7/6	7.0	0.667	0	0	P
Yb^{3+}	f^{13}	3	1/2	7/2	8/7	4.0	0.667	0	0	P

Pyrochlore compounds $R_2T_2O_7$

		R-R interaction	
		Ferro	Antiferro
R anisotropy	Ising	d **Dy$_2$Ti$_2$O$_7$** ge **Ho$_2$Ti$_2$O$_7$** e	**Not frustrated** **Long Range Order**
	Heisenberg	**Not frustrated** **Long Range Order**	**Tb$_2$Ti$_2$O$_7$** strated Short Range Order Spin Liquid

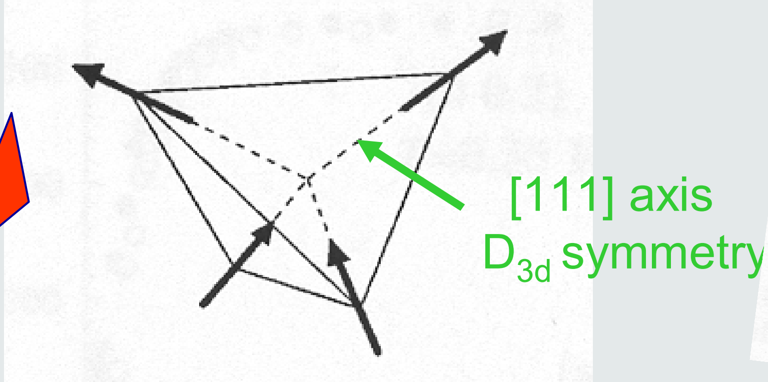
Ising ferromagnet (exchange + dipole): the pyrochlore « spin ice », a frustrated system

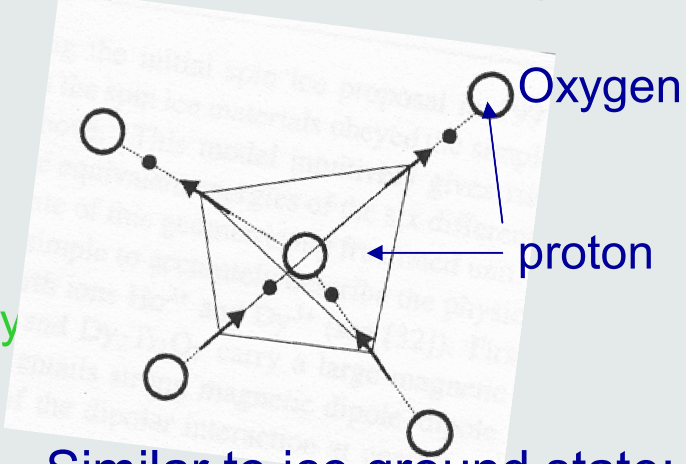
M.Harris, Nature **399** (1999) 311 & A.P.Ramirez *et al*, Nature **399** (1999) 333

Ising spins, constrained
to lie along (111) directions
(« in » or « out »)

⇔

Ice protons, constrained
to be on the (111) ligands
(close or near to O)





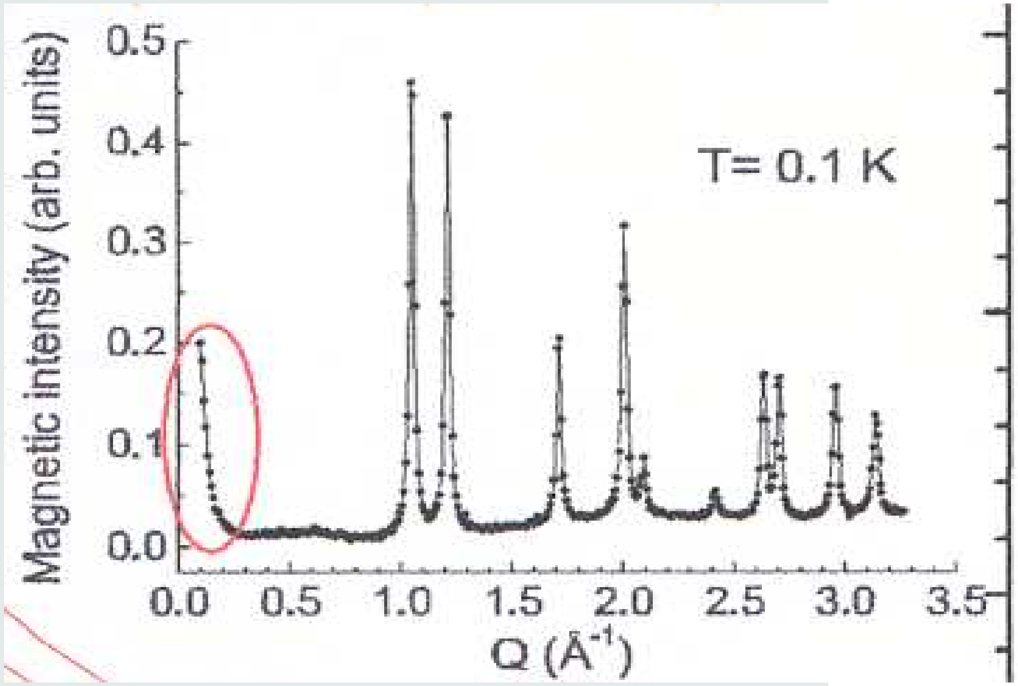
If ferromagnetic interactions,
the ground configuration is
« two in – two out » spins

⇒

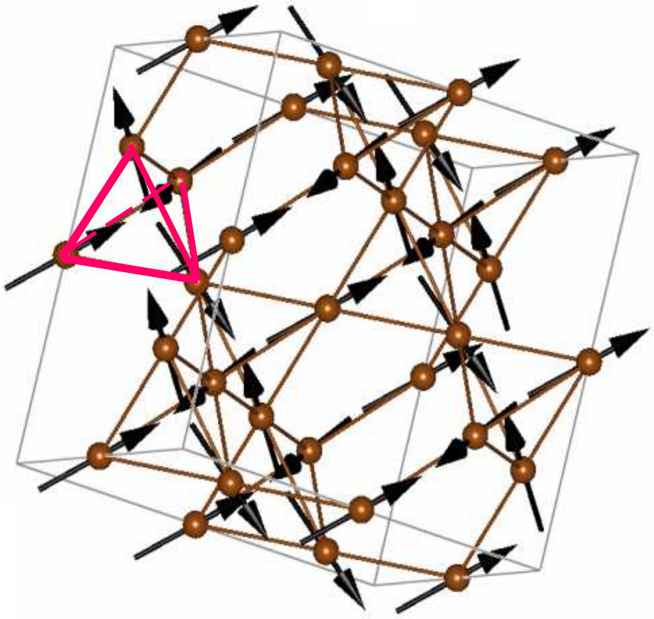
Similar to ice ground state:
« two close – two far » protons
with zero point entropy

« Spin ice » (e.g. $Dy_2Ti_2O_7$, with Ising ion Dy^{3+}):
large GS degeneracy and no LRO

$Tb_2Sn_2O_7$ (quasi-Ising F) - T_C = 0.87K
Magnetic diffractio



I.Mirebeau *et al*, PRL **94** (2005) 2464

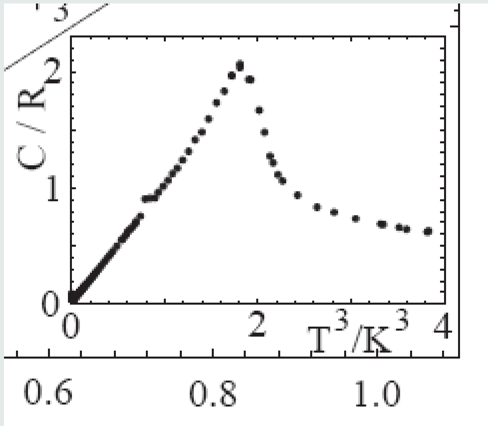


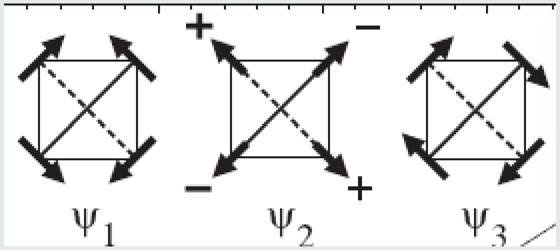
$L_c \cong 20nm$

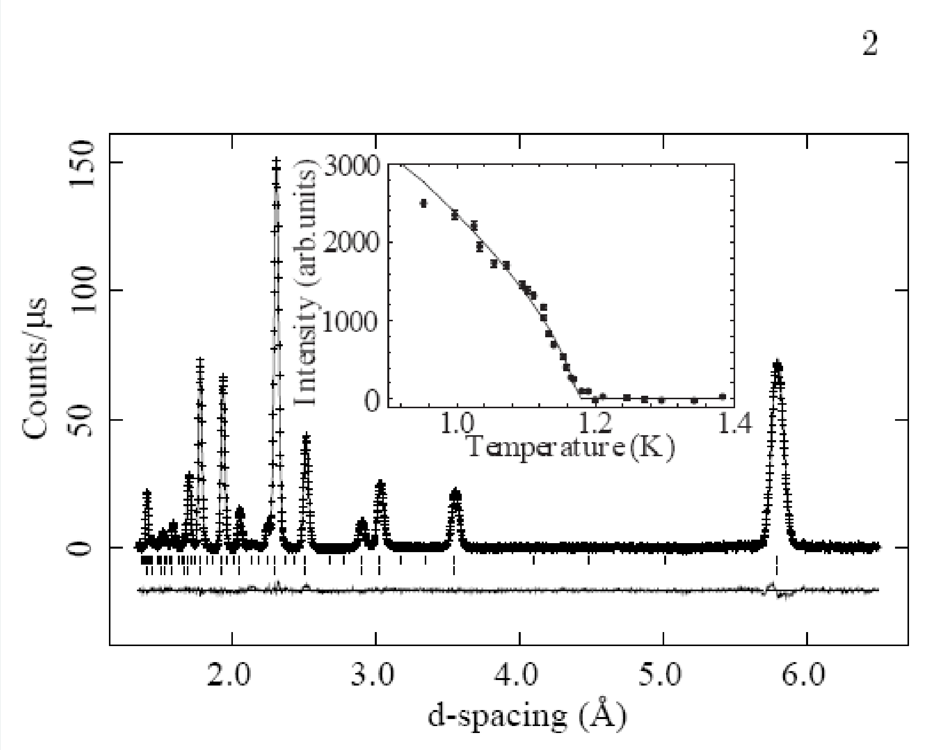
- presence of q=0 scattering below 0.9K ⇒ Ferro. correlations
- « ordered spin-ice »: quasi-« two in – two out » with LRO
- saturated Tb^{3+} moment: m(Tb)= 5.9(1)μ_B

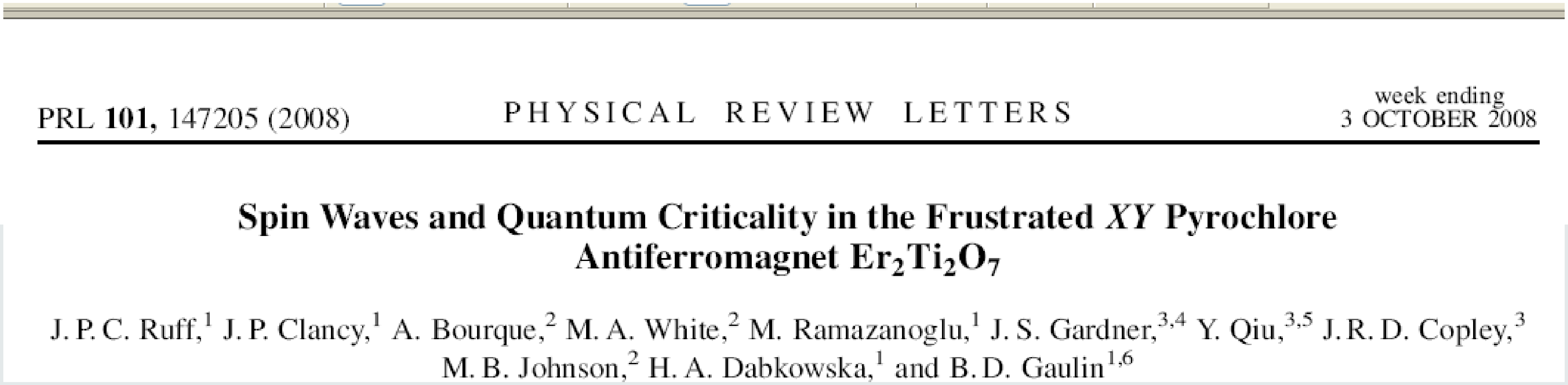
$Er_2Ti_2O_7$: Evidence of Quantum Order by Disorder in a Frustrated Antiferromagnet

J. D. M. Champion[1,2,3], M. J. Harris[2], P. C. W. Holdsworth[3], A. S. Wills[4], G. Balakrishnan[5],





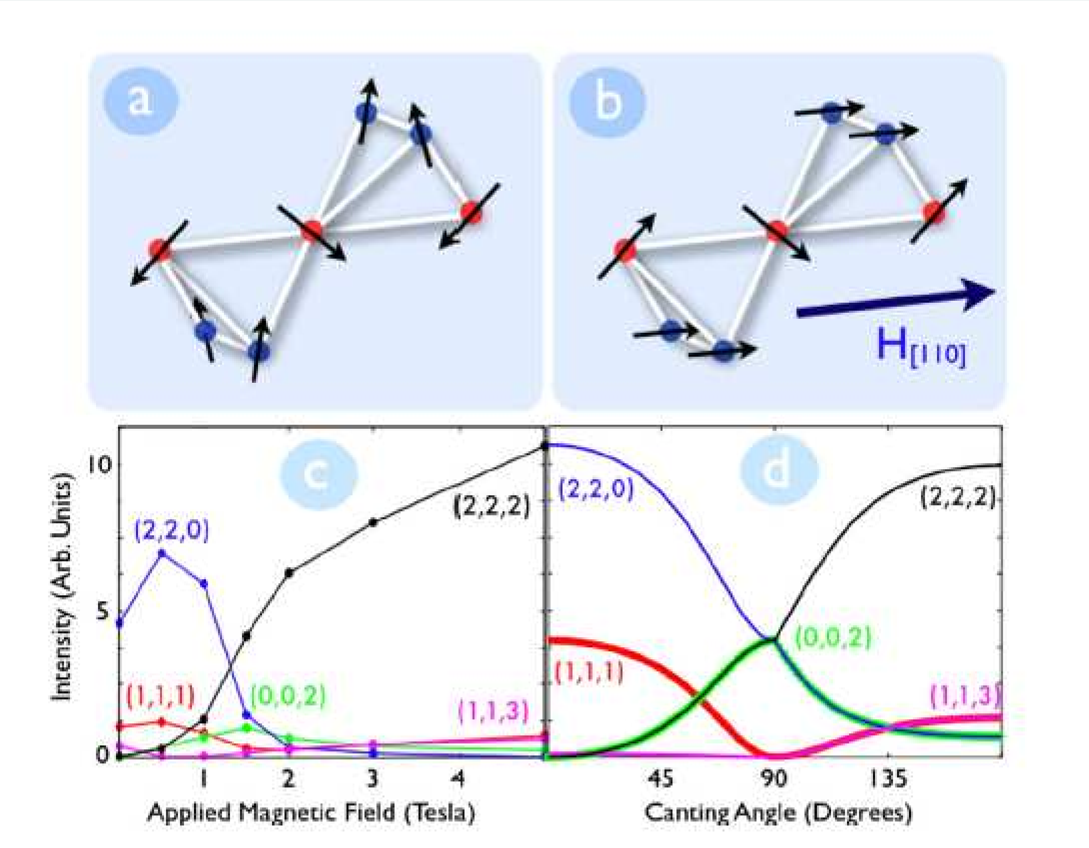




PRL **101,** 147205 (2008) PHYSICAL REVIEW LETTERS week ending 3 OCTOBER 2008

Spin Waves and Quantum Criticality in the Frustrated *XY* Pyrochlore Antiferromagnet $Er_2Ti_2O_7$

J. P. C. Ruff,[1] J. P. Clancy,[1] A. Bourque,[2] M. A. White,[2] M. Ramazanoglu,[1] J. S. Gardner,[3,4] Y. Qiu,[3,5] J. R. D. Copley,[3]
M. B. Johnson,[2] H. A. Dabkowska,[1] and B. D. Gaulin[1,6]



$$H = -J\sum_{\langle i,j\rangle}\vec{S}_i.\vec{S}_j - D\sum_i\left(\vec{S}_i.\vec{d}_i\right)^2$$

SPIN ICE structure

H Cao, A Gukasov I Mirebeau, PRL , 100, 22,227602, 2008

Low field and temperatures (*H* ≤ 1 T, 200 mK)

H // 110

- α chains:
- β chains

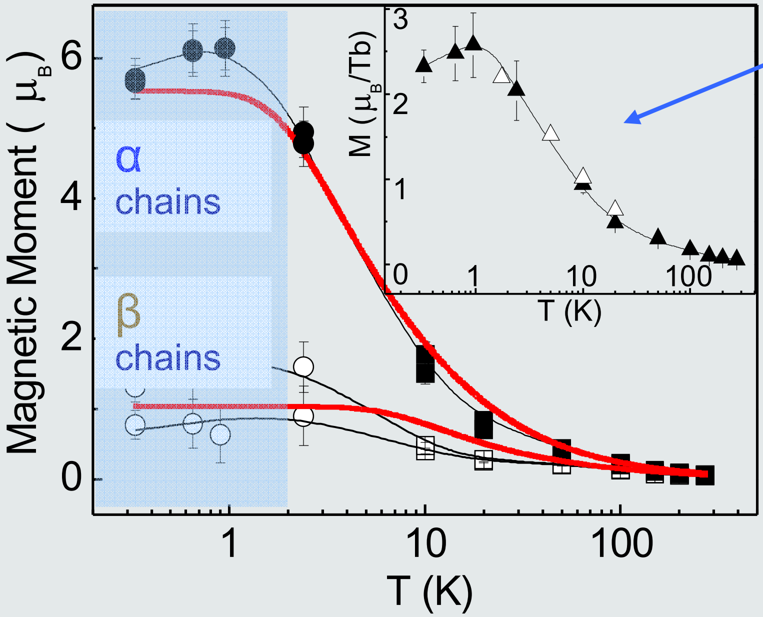
local <111> easy axis

close to ***H*** (36^0)

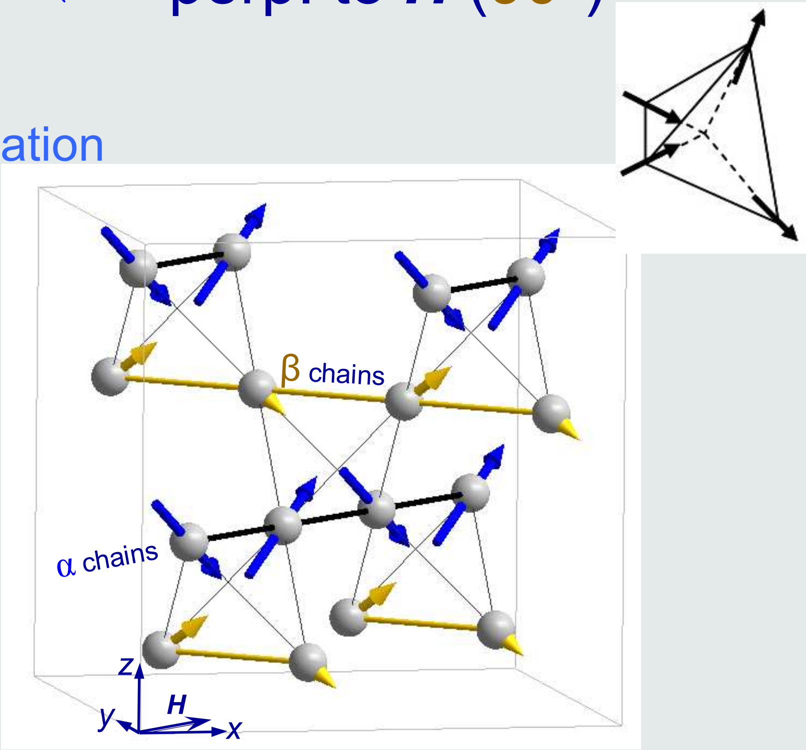
perp. to ***H*** (90^0)

Very different moments

Magnetization



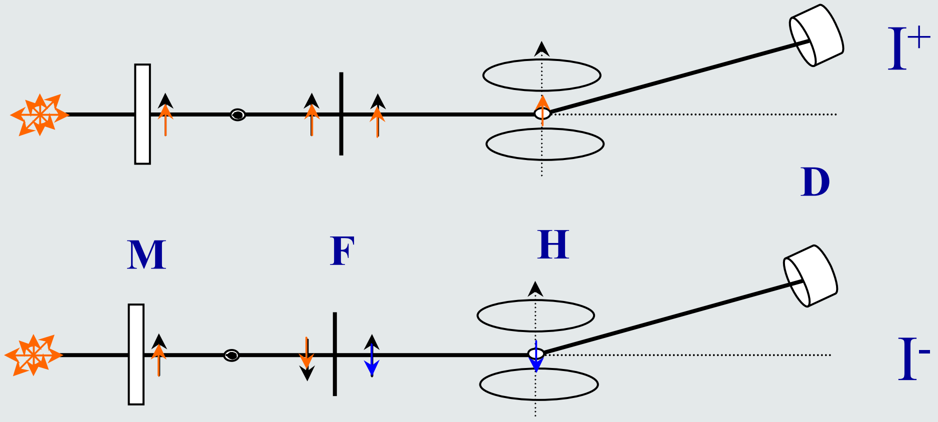
Red lines: CF calculations



***H* = 1 T & *T* = 200 m K**

Close to spin ice

POLARIZED NEUTRON DIFFRACTION



P_0 parallel to H

P_0 antiparallel to H

$$I^{+} \propto (F_N + F_M)^2 \qquad I^{-} \propto (F_N - F_M)^2$$

$$I^{\pm} \propto F^2_N \pm 2F_N(P_0 {*} F_M) + F_M^2$$

FLIPPING RATIO MEASUREMENTS

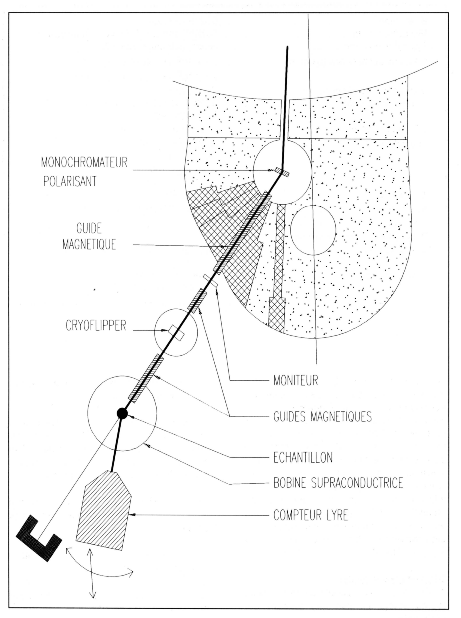
$$I^+ \propto (F_N + F_M)^2 \qquad I^- \propto (F_N - F_M)^2$$

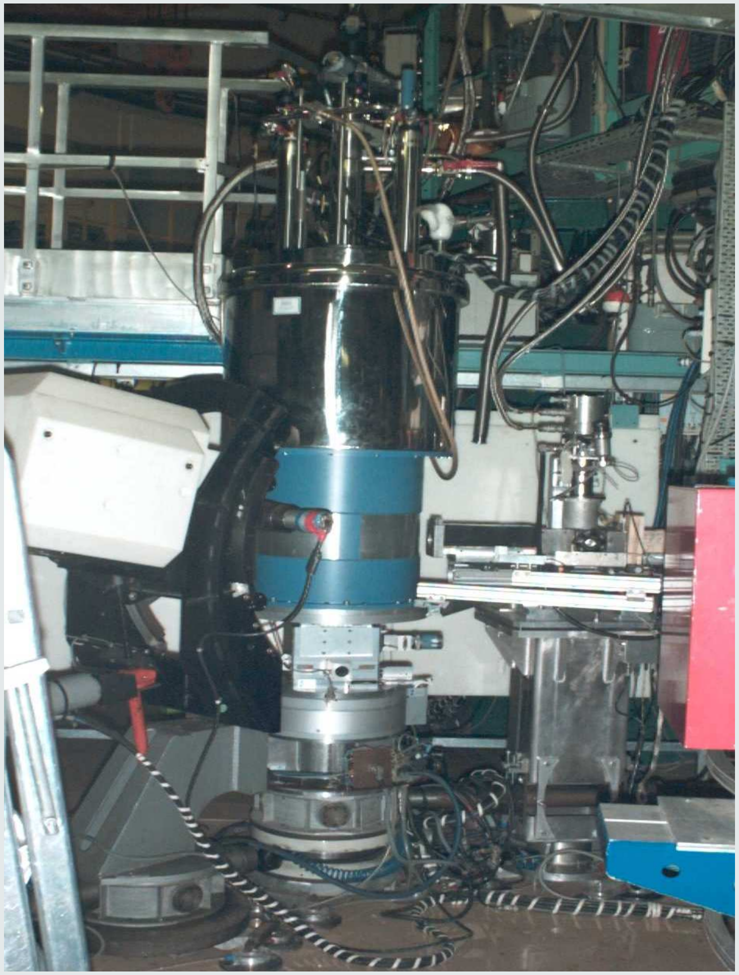
$$R = I^+ / I^- = (F_N + F_M)^2 / (F_N - F_M)^2$$

$$R = (1+\gamma)^2 / (1-\gamma)^2 \qquad F_M(\mathbf{q}) = \gamma * F_N(\mathbf{q})$$

5C1 DIFFRACTOMETER (LLB)

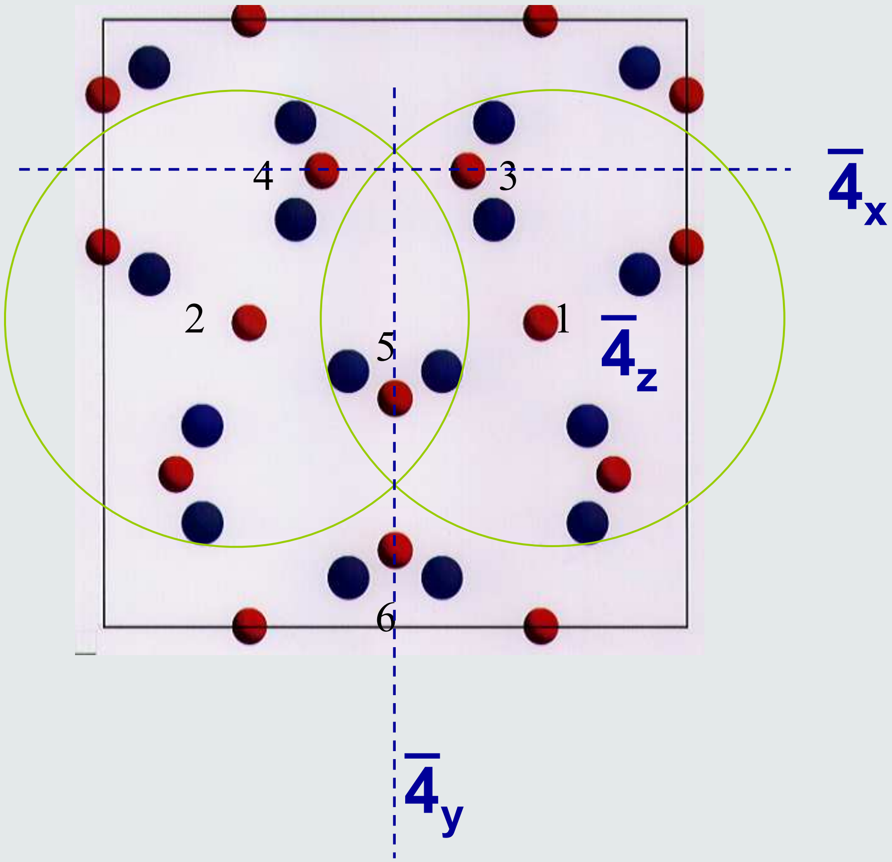
- 0.84 Å
- Heussler 50x50 mm^2 in transmission
- Cryoflipper
- Asymmetric 7.8 T magnet
- $-5^\circ < \nu < 20^\circ$
- lifting counter

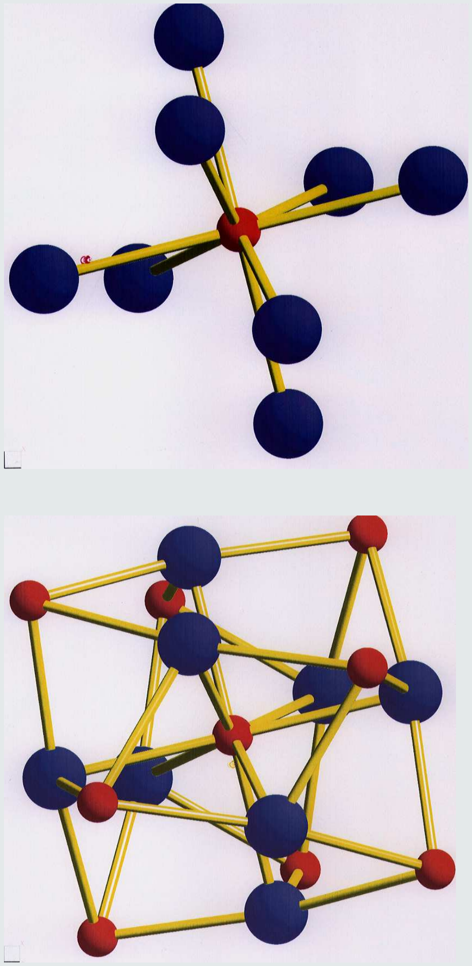




U_3P_4 (Th_3P_4 STRUCTURE I-43d)

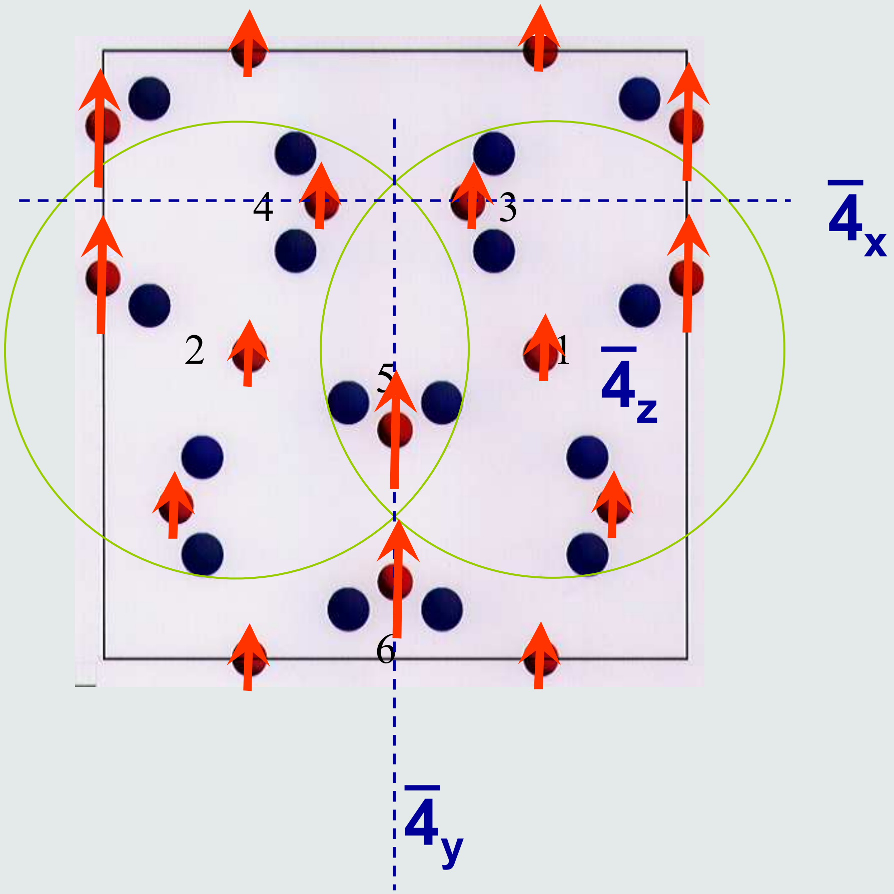
A. Gukasov P. Wisniewski and Z. Henkie J. Phys. C, 1998





U_3P_4 (Th_3P_4 STRUCTURE I-43d)

A Gukasov, P Wisniewski and Z Henkie. J of Phys, Cond Matt. 8, 10589,1996



$\mu_{5,6}=2.1\mu_B$

$\mu_{1,2,3,4}= 1.65\mu_B$

U_3P_4 (Th_3P_4 STRUCTURE I-43d)

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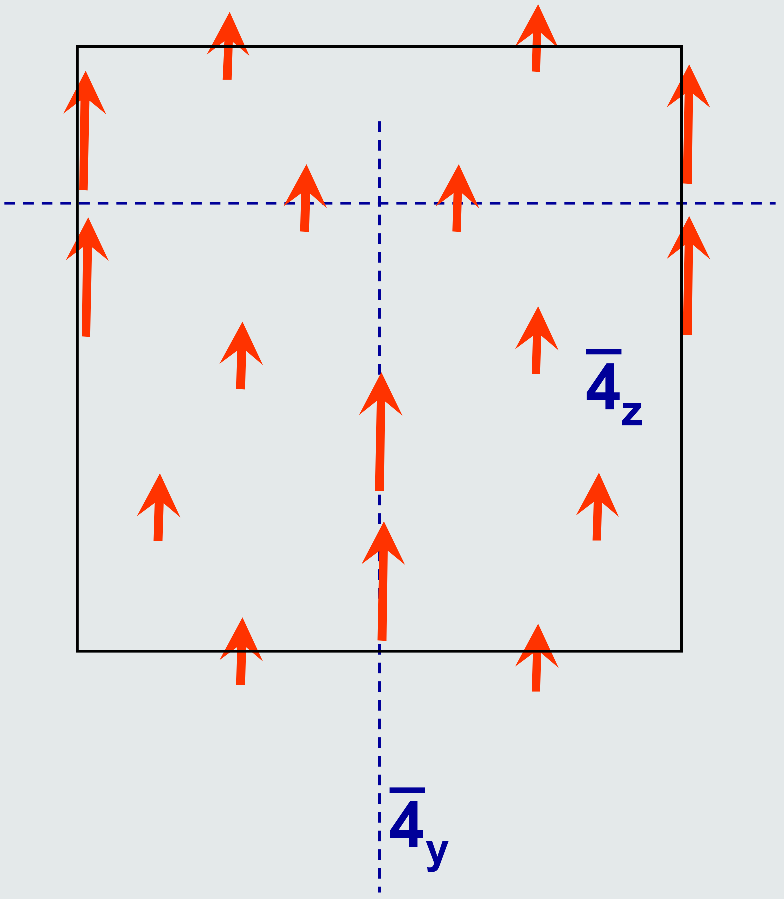


$\bar{4}_x$ $\mu_{5,6}=2.1\mu_B$

$\bar{4}_z$

$\mu_{1,2,3,4}= 1.65\mu_B$

$\bar{4}_y$

LOCAL ANISOTROPY OF SUSCEPTIBILITY IN CRYSTALS WITH Th_3P_4 STRUCTURE

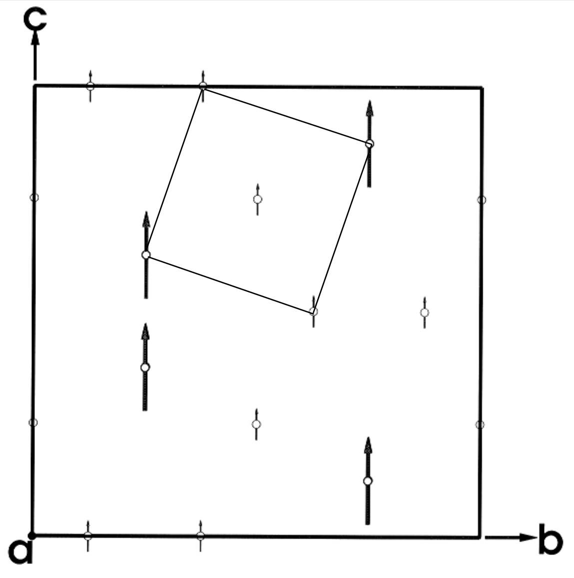
J. X. Boucherle et al. Physica B, 281-282, 139-140, 2000

A Gukasov and P J Brown, J Phys C, 14, 8831, 2002

H||(0 0 1) 5T

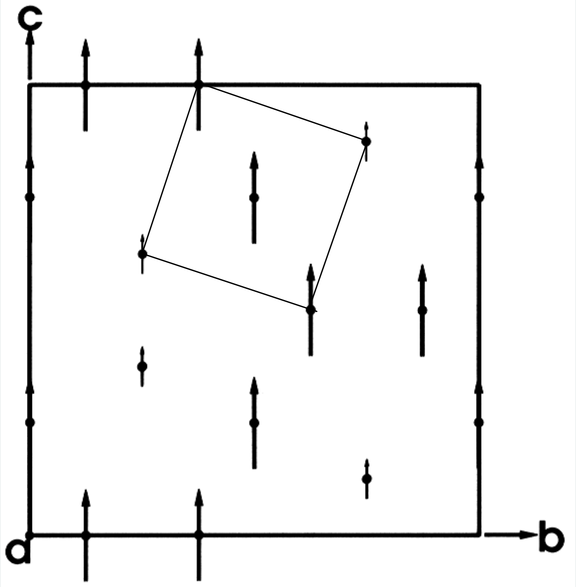
Sm_3Te_4 D3, ILL

$\mu_{1,2} = 0.083\mu_B$ $\mu_{3,4,5,6} = 0.033\mu_B$



$Nd_{3-x}S_4$ 5C1, LLB

$\mu_{12} = 0.62\mu_B$ $\mu_{3,4,5,6} = 1.44\mu_B$



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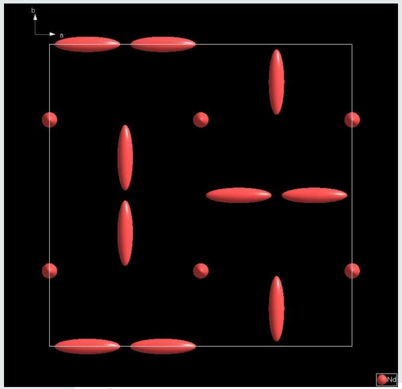
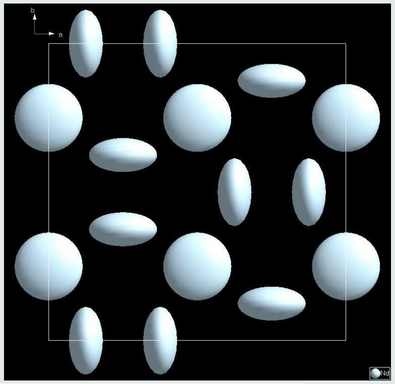
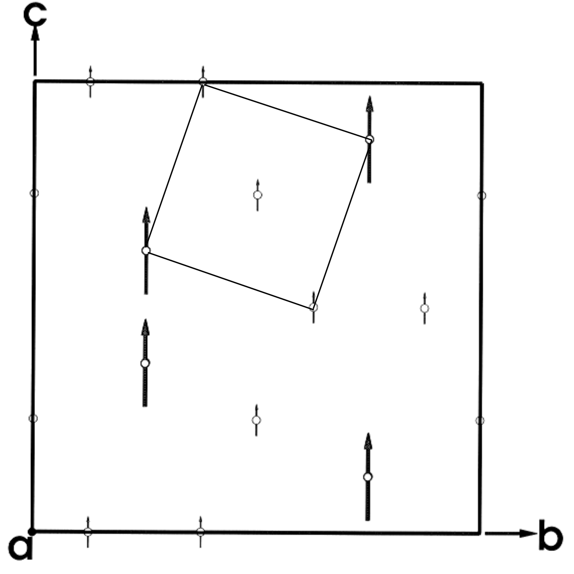
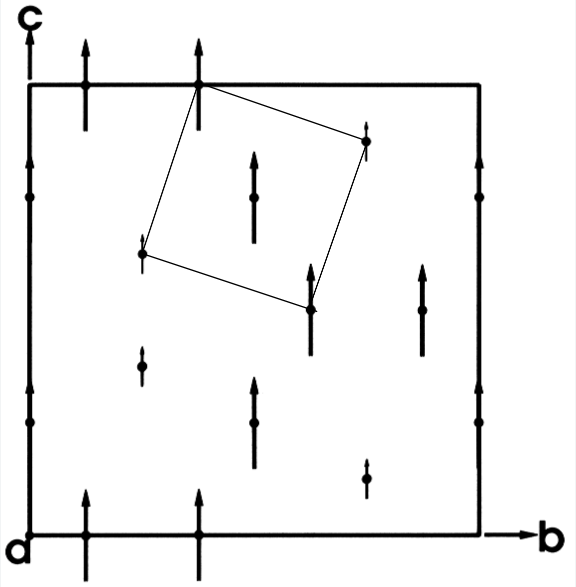


A Gukasov and P J Brown,
J Phys C, 14, 8831, 2002







ANISOTROPIC SUSCEPTIBILITIES

$$\chi_{ij} = \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ & \chi_{22} & \chi_{23} \\ & & \chi_{33} \end{pmatrix}$$

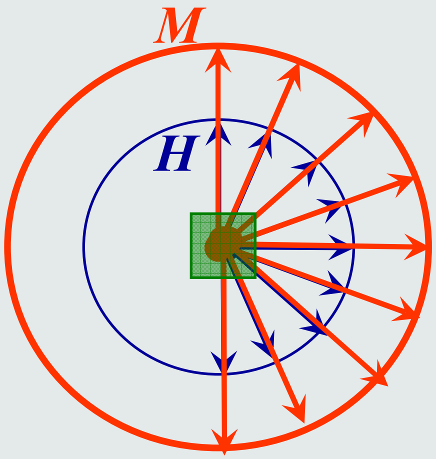
Bulk magnetisation $M_i\,(r) = \chi_{ij} H_j$

The number of independent components of χ_{ij} is determined by the crystal symmetry class:

cubic groups	**1 parameter**
all uniaxial groups	**2 parameters**
Orthorhombic	**3 ...**
Monoclinic	**4 ...**
Triclinic	**6**

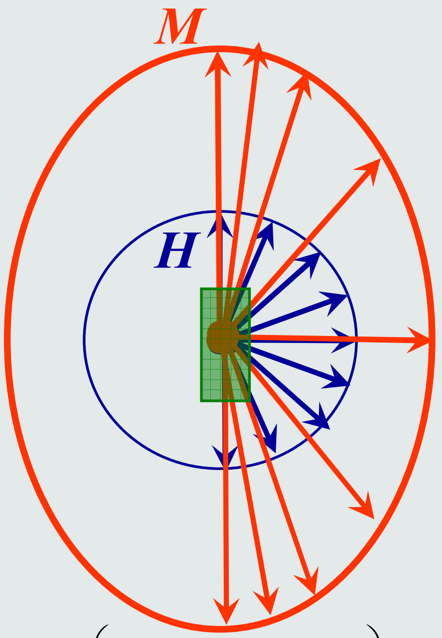
ANISOTROPIC BULK SUSCEPTIBILITY

CUBIC, $\chi_{11} = \chi_{22} = \chi_{33}$



$$\chi_{ij} = \begin{pmatrix} \chi_{11} & 0 & 0 \\ 0 & \chi_{11} & 0 \\ 0 & 0 & \chi_{11} \end{pmatrix}$$

UNIAXIAL $\chi_{11} = \chi_{22} < \chi_3$



$$\chi_{ij} = \begin{pmatrix} \chi_{11} & 0 & 0 \\ 0 & \chi_{11} & 0 \\ 0 & 0 & \chi_{33} \end{pmatrix}$$

ANISOTROPIC SITE SUSCEPTIBILITIES

A Gukasov and P J Brown, J Phys C, 14, 8831, 2002

Bulk magnetisation

$$M_i = \Sigma\, M^a{}_i = \Sigma\ \chi^a{}_{ij} H_j$$

$$M^b{}_i = \Sigma\, R_g(t)\, \chi^a{}_{ij}\, R_g(t)^{-1} H_j$$

$R_g(t)$ **is the symmetry operator** $\quad r_b = R_g(t)\, r_a$

$$M_i = \Sigma_g\, R(t)\, \boxed{\chi^a{}_{ij}}\, R(t)^{-1} H_j$$

REFINEMENT OF ANISOTROPIC SUSCEPTIBILITIES

$$I^{\pm} \propto N^2 \pm 2\, P_{0z}\, N\, M_z + M_z^2$$

$$M_i = \Sigma_a\, \chi^a_{ij} H_j$$

$$I^{\pm} \propto N^2 \pm 2F_N(P_0{}^{*}\, \Sigma\chi^a_{ij} H_j) + |\Sigma\chi^a_{ij} H_j|^2$$

$R = I^+ / I^-$, CHILSQ (CCSL)

A Gukasov and P J Brown, J Phys C, 14, 8831, 2002

ANISOTROPIC SUSCEPTIBILITY PARAMETERS (ASPs)

$$\chi_{ij} = \begin{pmatrix} \chi_{11} & 0 & 0 \\ 0 & \chi_{22} & 0 \\ 0 & 0 & \chi_{22} \end{pmatrix}$$

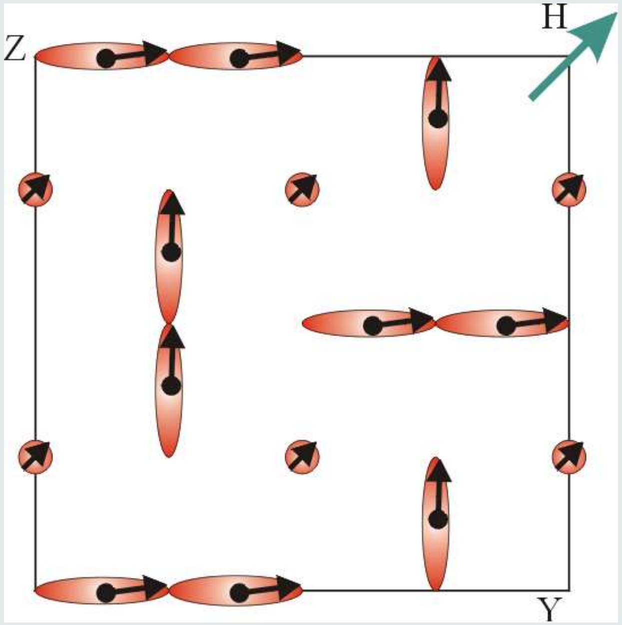
A Gukasov and P J Brown, J Phys C, 14, 8831, 2002

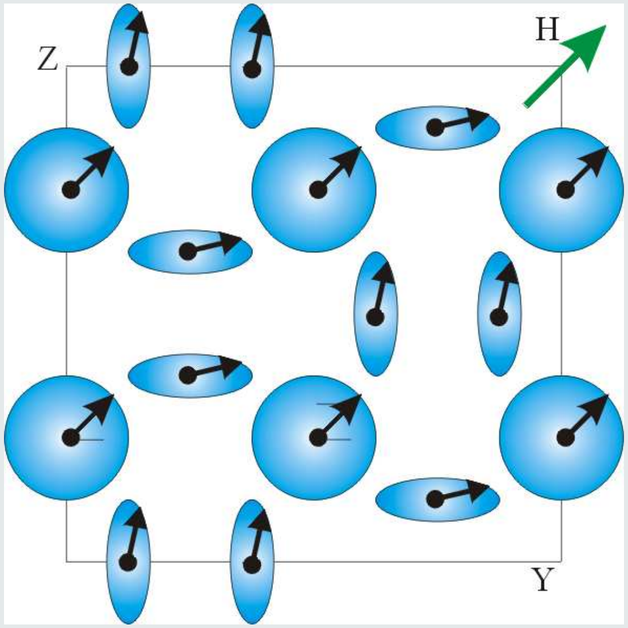
Prolate Sm_3Te_4

$\chi_{11} < \chi_{33}$

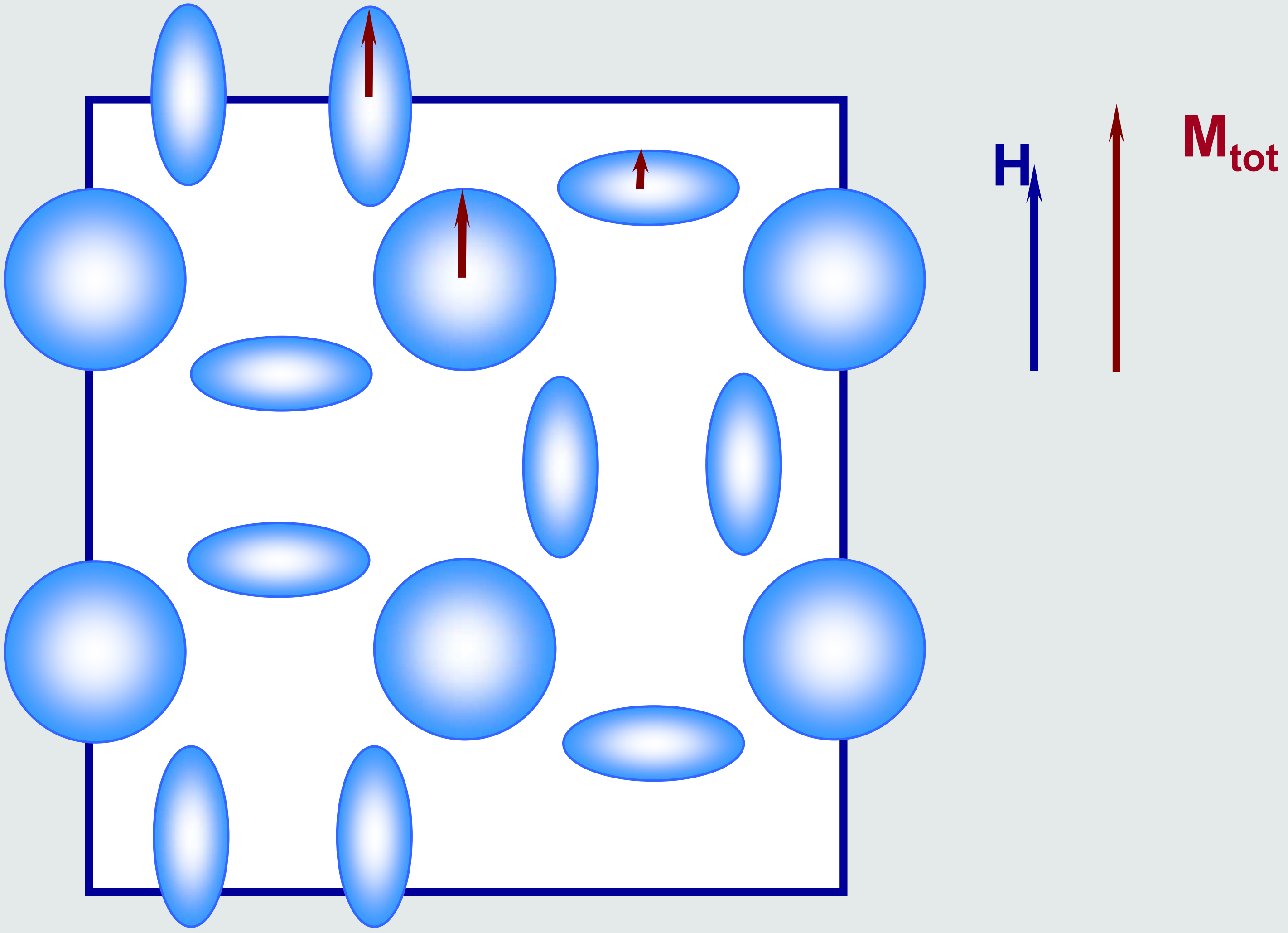
Oblate $Nd_{3-x}S_4$

$\chi_{11} > \chi_{33}$





OBLATE MAGNETIC ELLIPSOIDS IN $Nd_{3-x}S_4$



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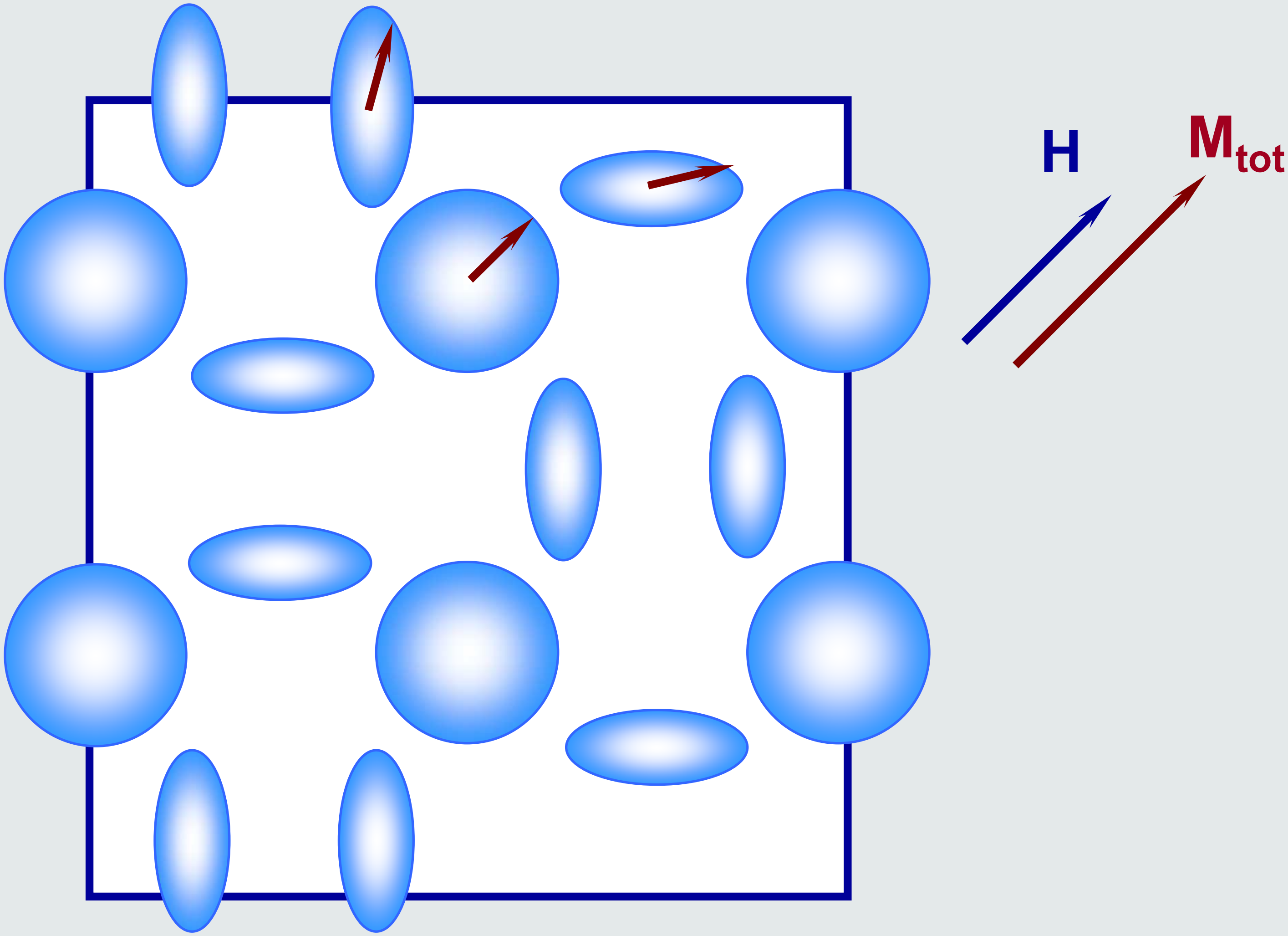
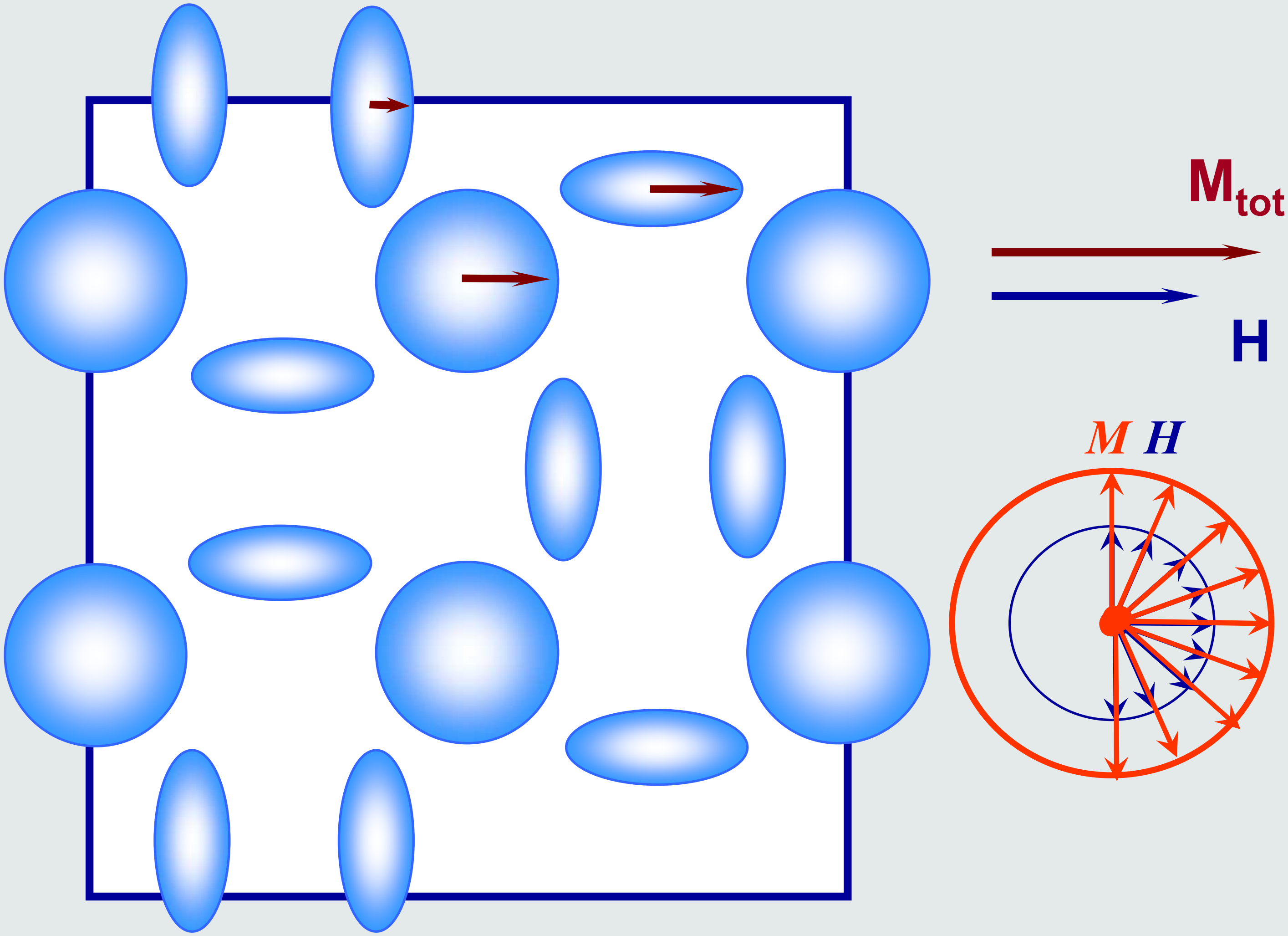


A Gukasov and P J Brown, J Phys C, 14, 8831, 2002

OBLATE MAGNETIC ELLIPSOIDS IN $Nd_{3-x}S_4$



A Gukasov and P J Brown, J Phys C, 14, 8831, 2002

ATOMIC DISPLACEMENT PARAMETERS (u_{ij}) AND ATOMIC SUSCEPTIBILITY PARAMETES (χ_{ij})

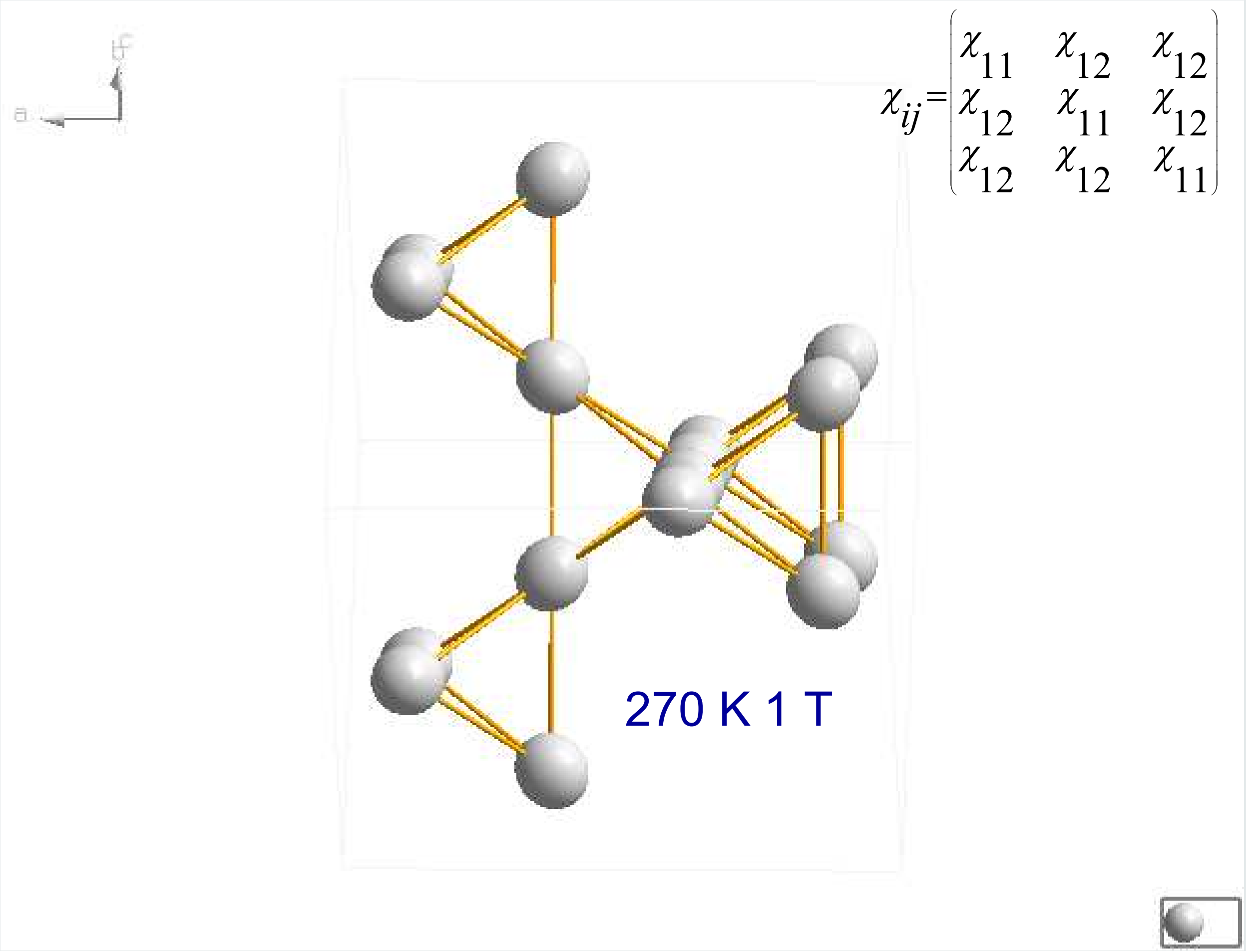
- u_{ij} are probes of the shape of elastic potential well
- u_{ij} give informaton about atomic vibration, dynamics, disorder ...

- χ_{ij} are probes of magnetic interaction
- symmetry constraints of χ_{ij} the same as of u_{ij}
- Anomalous χ_{ij} indicate strong local anisotropy
- and a possible channel of ordering

A Gukasov and P J Brown, J Phys C, 14, 8831, 2002

VISUALIZATION OF ASP BY *“MAGNETIC ELLIPSOIDS” IN $Tb_2Ti_2O_7$*

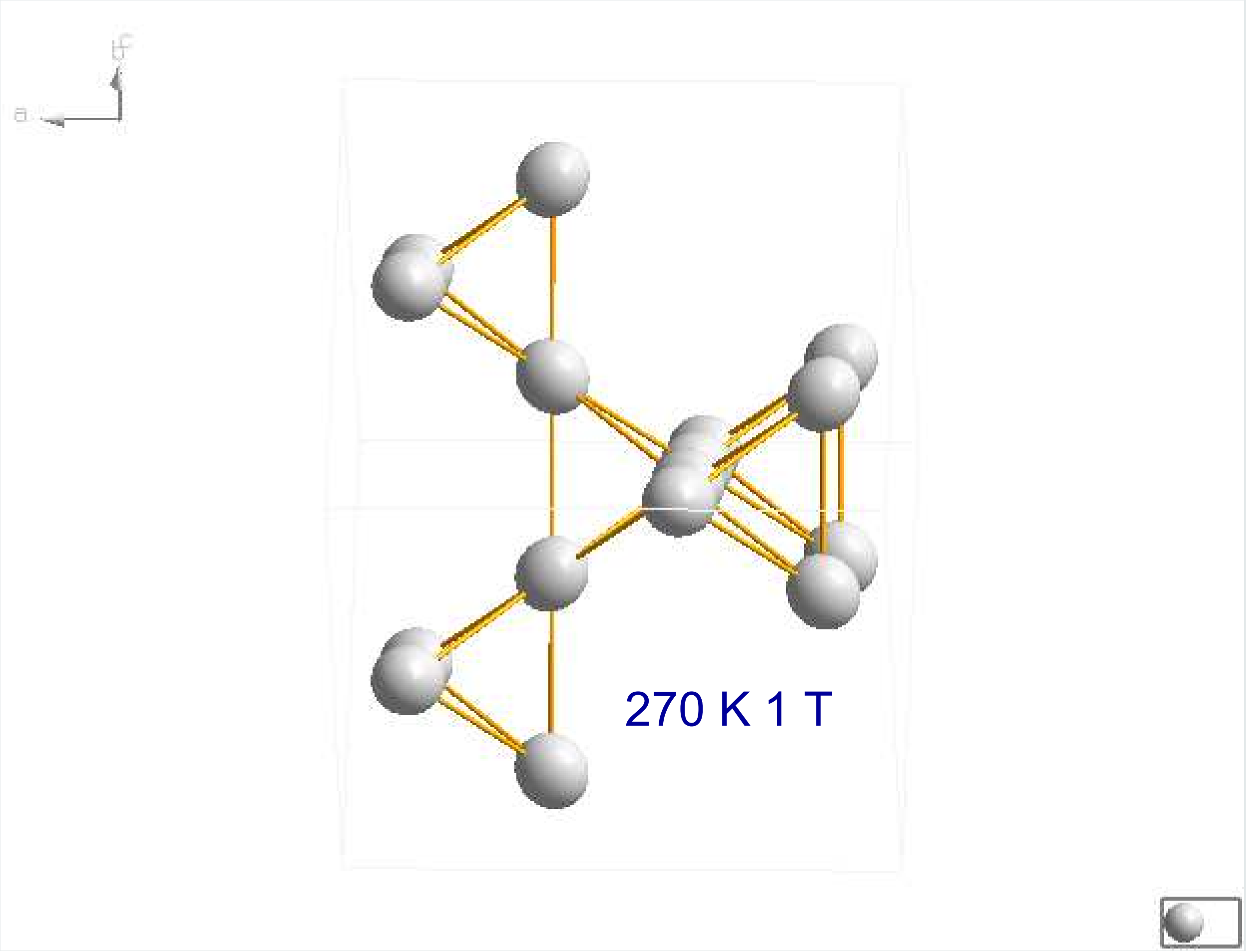
102 K gives χ_{11}=0.056 μ_B/T and $\chi_{12=}$=0.002μ_B



$$\chi_{ij}=\begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{12} \\ \chi_{12} & \chi_{11} & \chi_{12} \\ \chi_{12} & \chi_{12} & \chi_{11} \end{pmatrix}$$

VISUALIZATION OF ASP BY "*MAGNETIC ELLIPSOIDS" IN Tb2Ti2O7*

102 FR gives χ_{11}=0.056 μ_B/T and $\chi_{12=}$=0.002μ_B



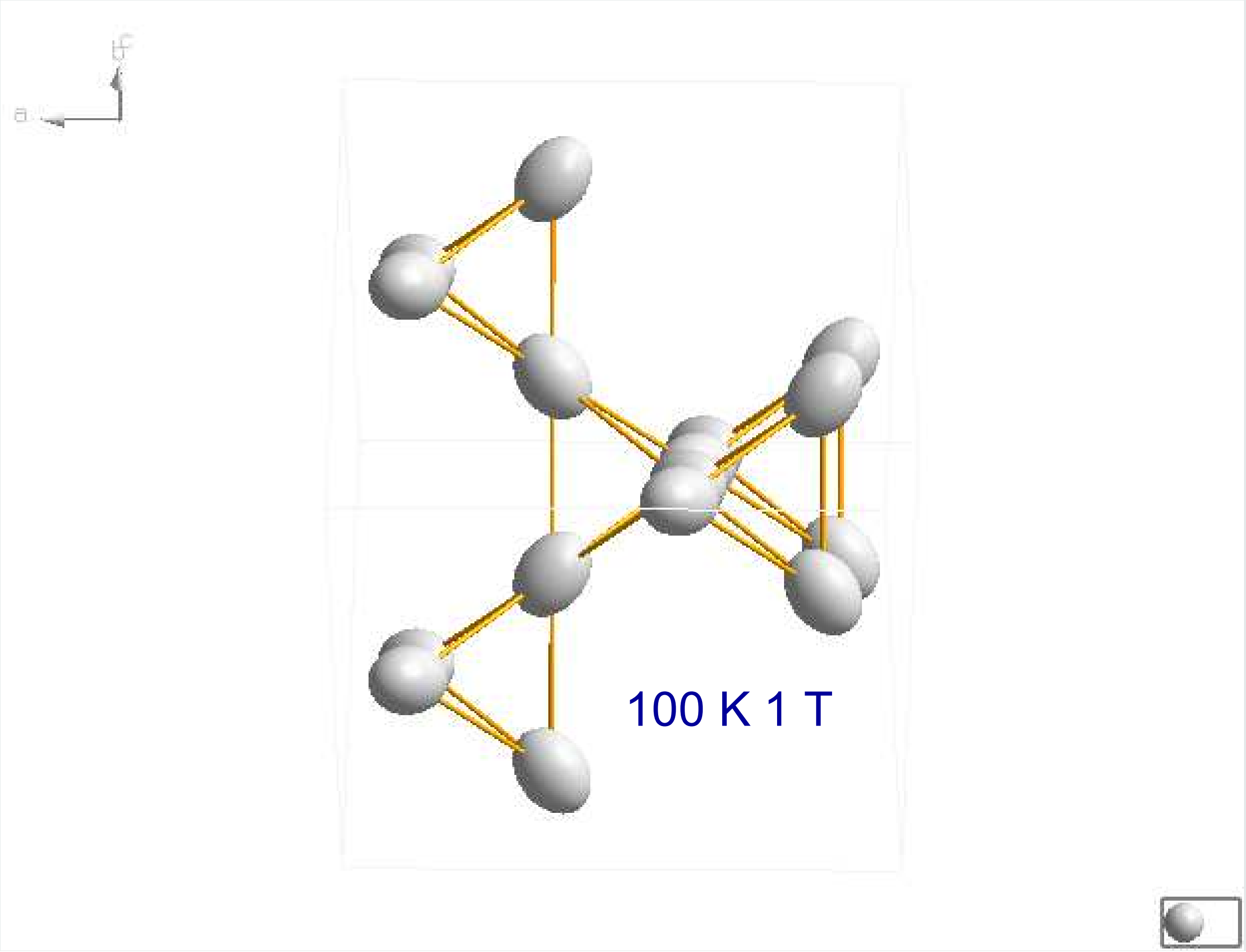
$$\chi_{ij}=\begin{pmatrix}\alpha & \beta & \beta\\ \beta & \alpha & \beta\\ \beta & \beta & \alpha\end{pmatrix}$$

$$\chi_{ij}=\begin{pmatrix}\alpha & 0 & 0\\ 0 & \alpha & 0\\ 0 & 0 & \beta\end{pmatrix}$$

VISUALIZATION OF ASP BY "*MAGNETIC ELLIPSOIDS" IN Tb2Ti2O7*

196 FR gives χ_{11}=0.17 μ_B/T and $\chi_{12=}$=0.04μ_B/T

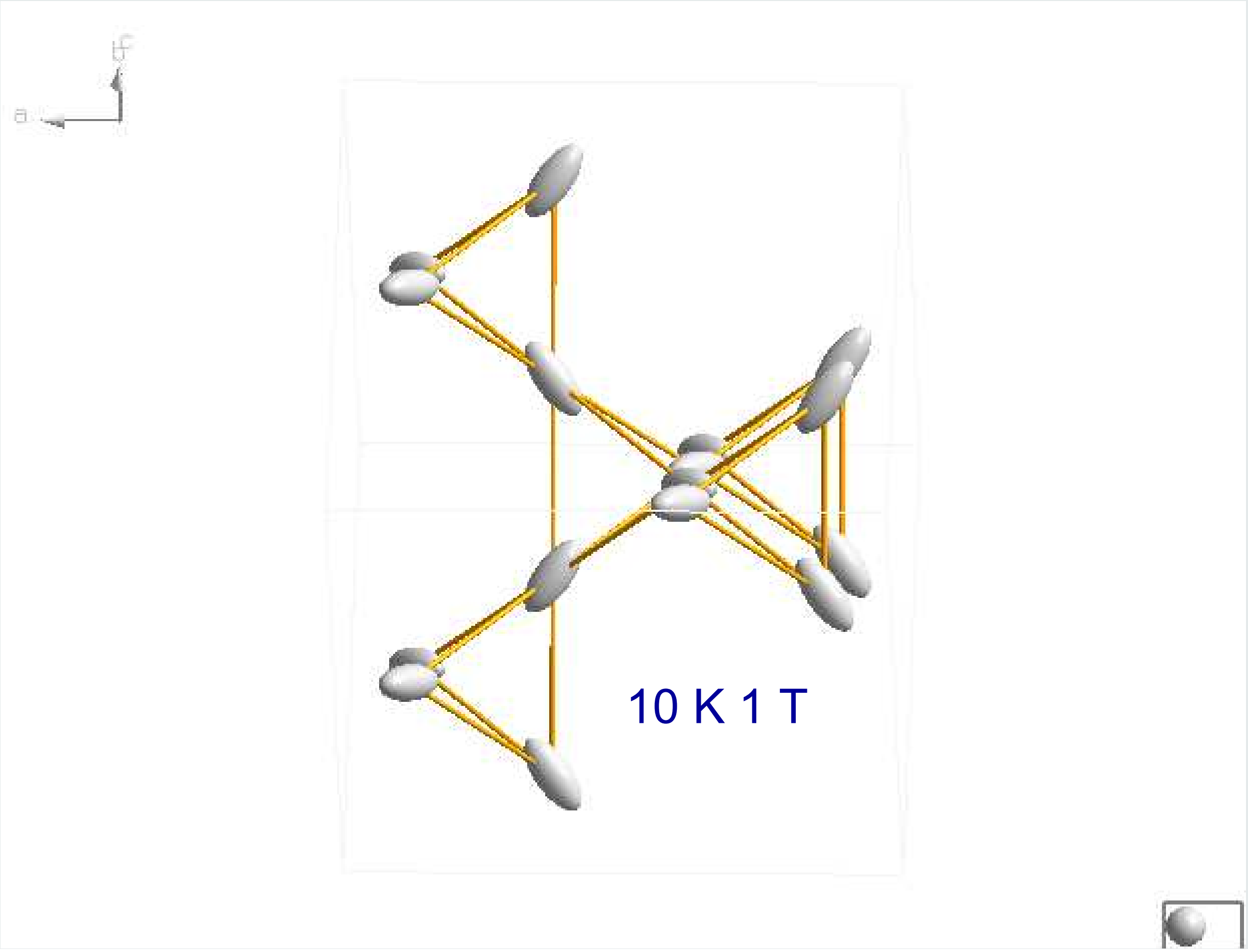
Ellipsoids are multiplied by T to compensate Curie-Weiss behavior



VISUALIZATION OF ASP BY “*MAGNETIC ELLIPSOIDS” IN Tb2Ti2O7*

423 FR gives χ_{11}=0.94 μ_B/T and $\chi_{12=}$=0.53μ_B/T

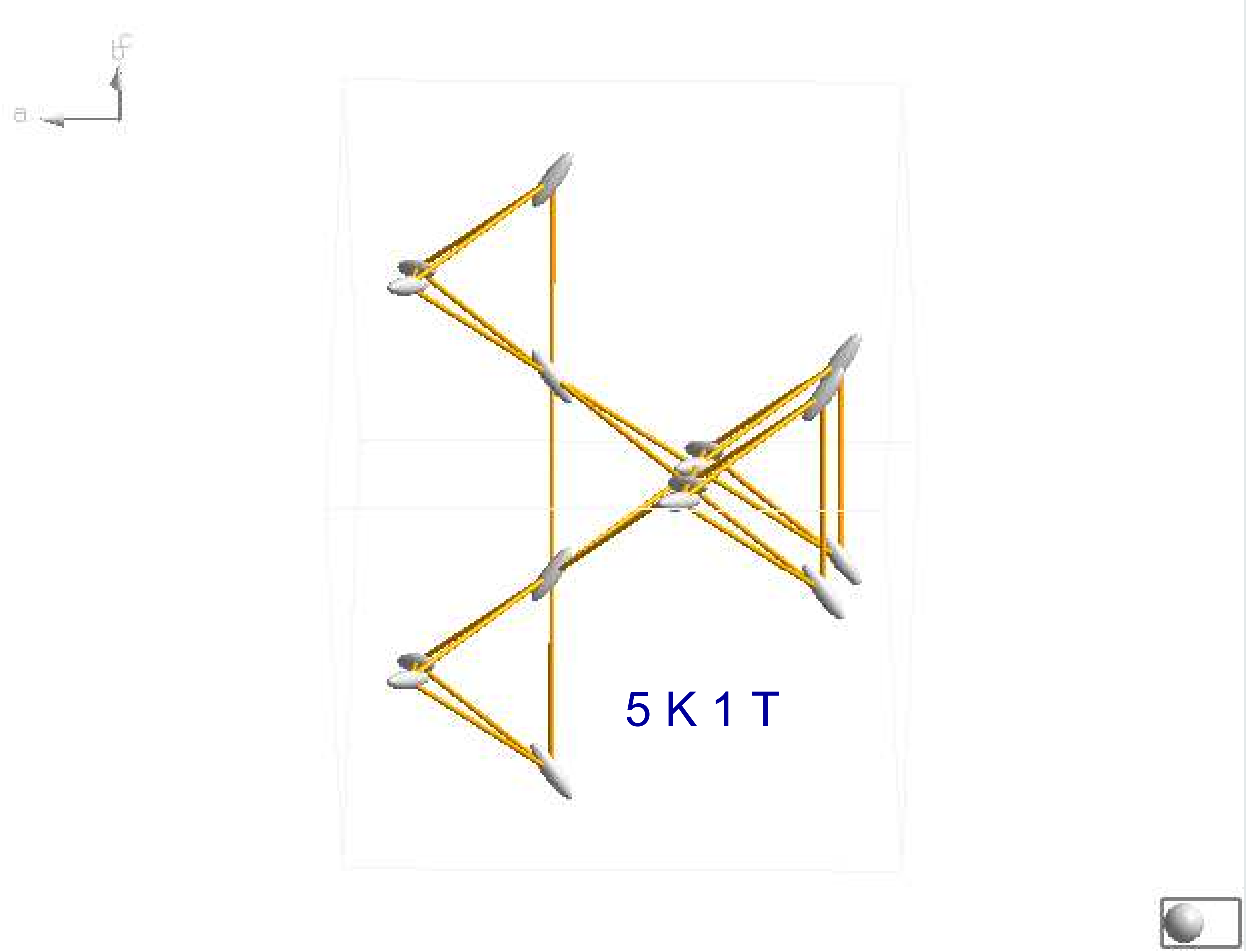
Ellipsoids are multiplied by T to compensate Curie-Weiss behavior



VISUALIZATION OF ASP BY "*MAGNETIC ELLIPSOIDS" IN Tb2Ti2O7*

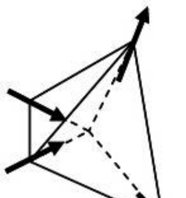
196 FR gives χ_{11}=0.98(2) μ_B/T and χ_{12}=0.76(1)μ_B/T, chi^2=2.9

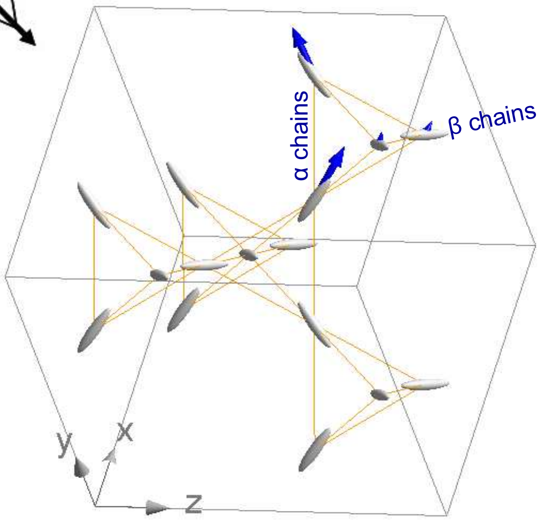
Ellipsoids are multiplied by T to compensate Curie-Weiss behavior



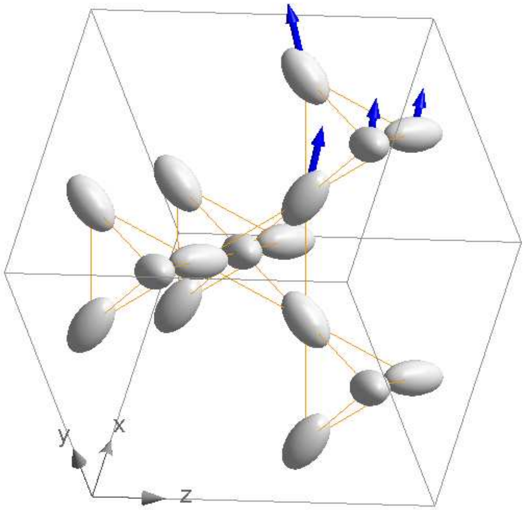
FROM HEISENBERG TO ISING BEHAVIOR

H // 110

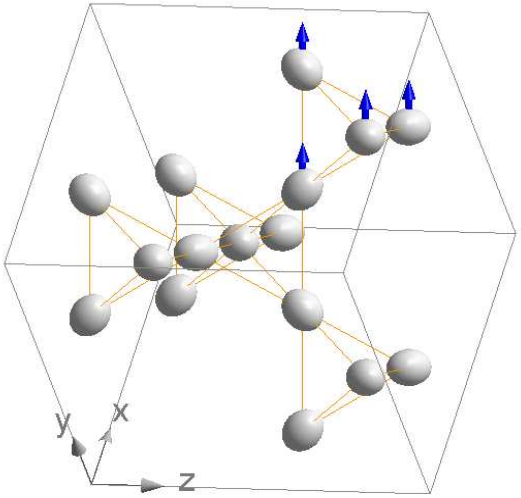




10 K



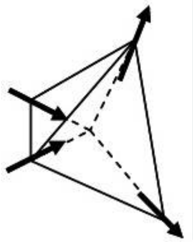
100 K

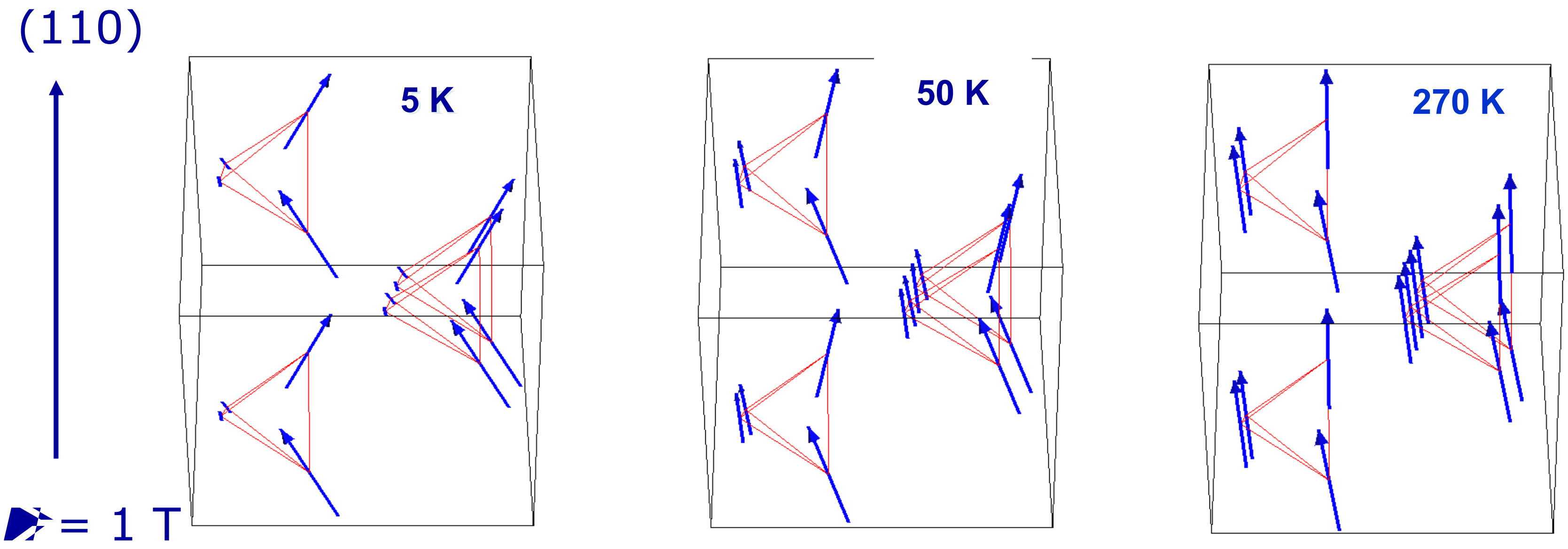


270 K

(BAD) SPIN ICE (k=0)

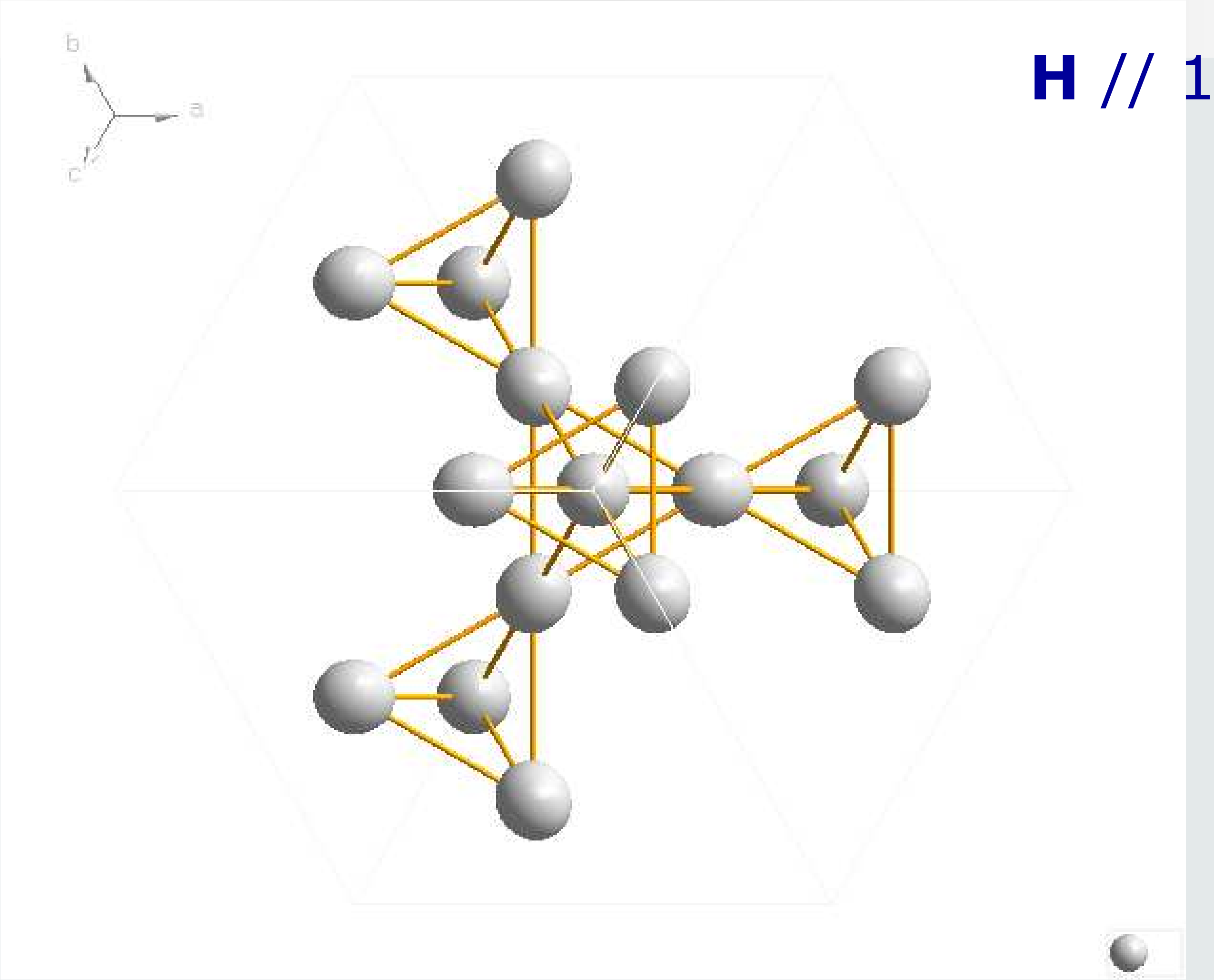
H // 110





Moments are multiplied by T to compensate Curie-Weiss behavior

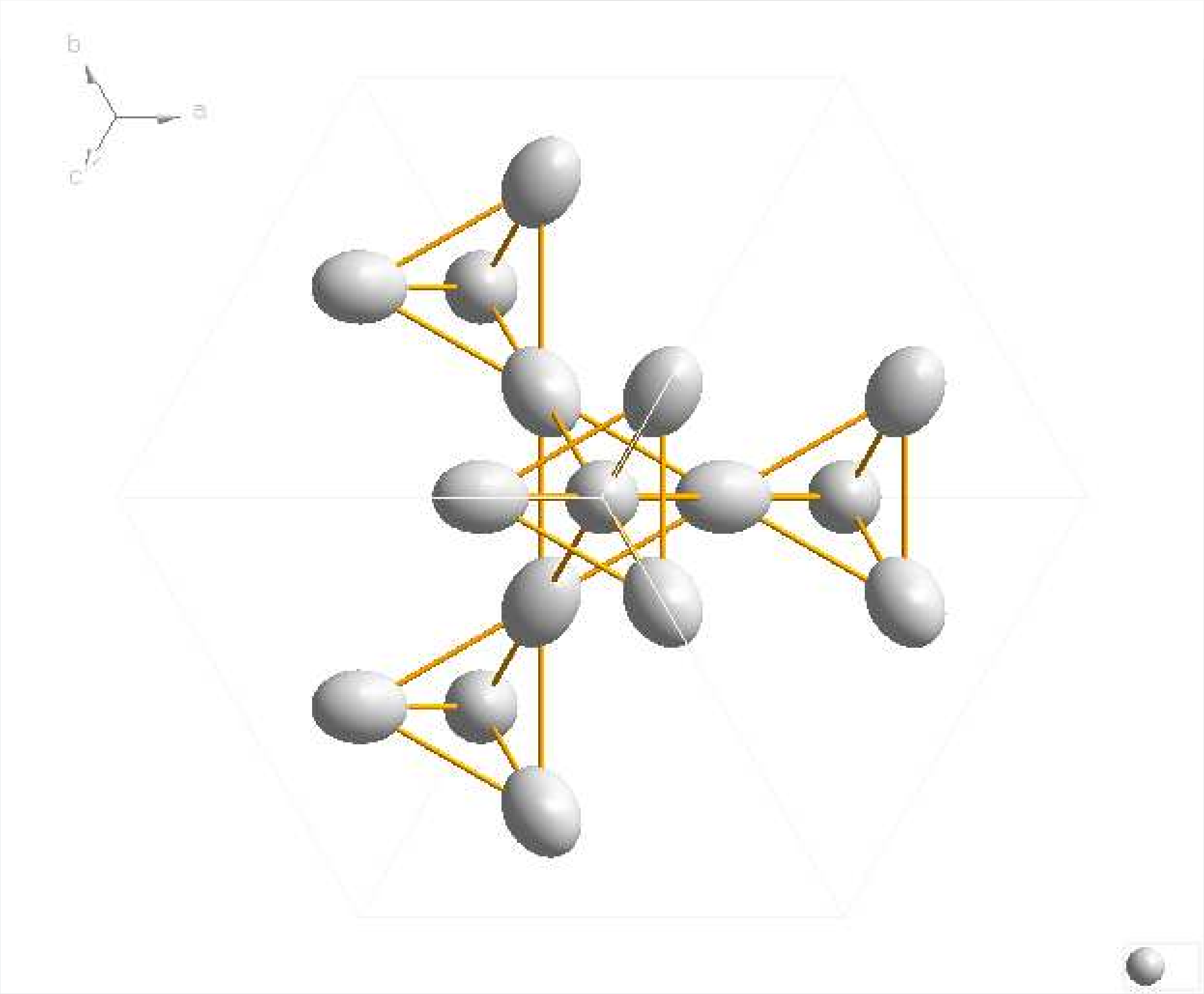
VISUALIZATION OF ASP BY “*MAGNETIC ELLIPSOIDS*” *H II [111]*



H // 110

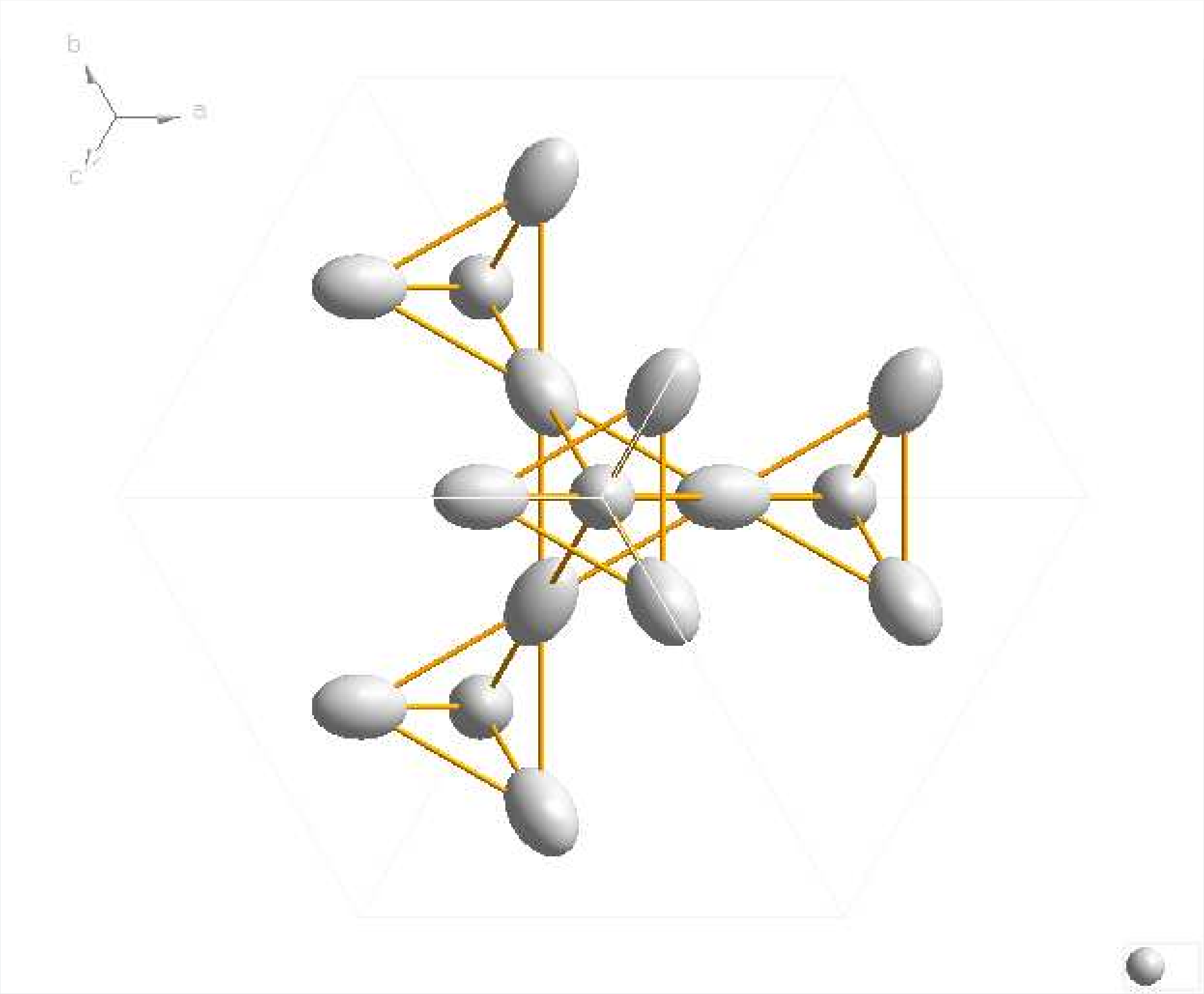
270 K 1 T, 100 FR

VISUALIZATION OF ASP BY “*MAGNETIC ELLIPSOIDS*” *H II [111]*



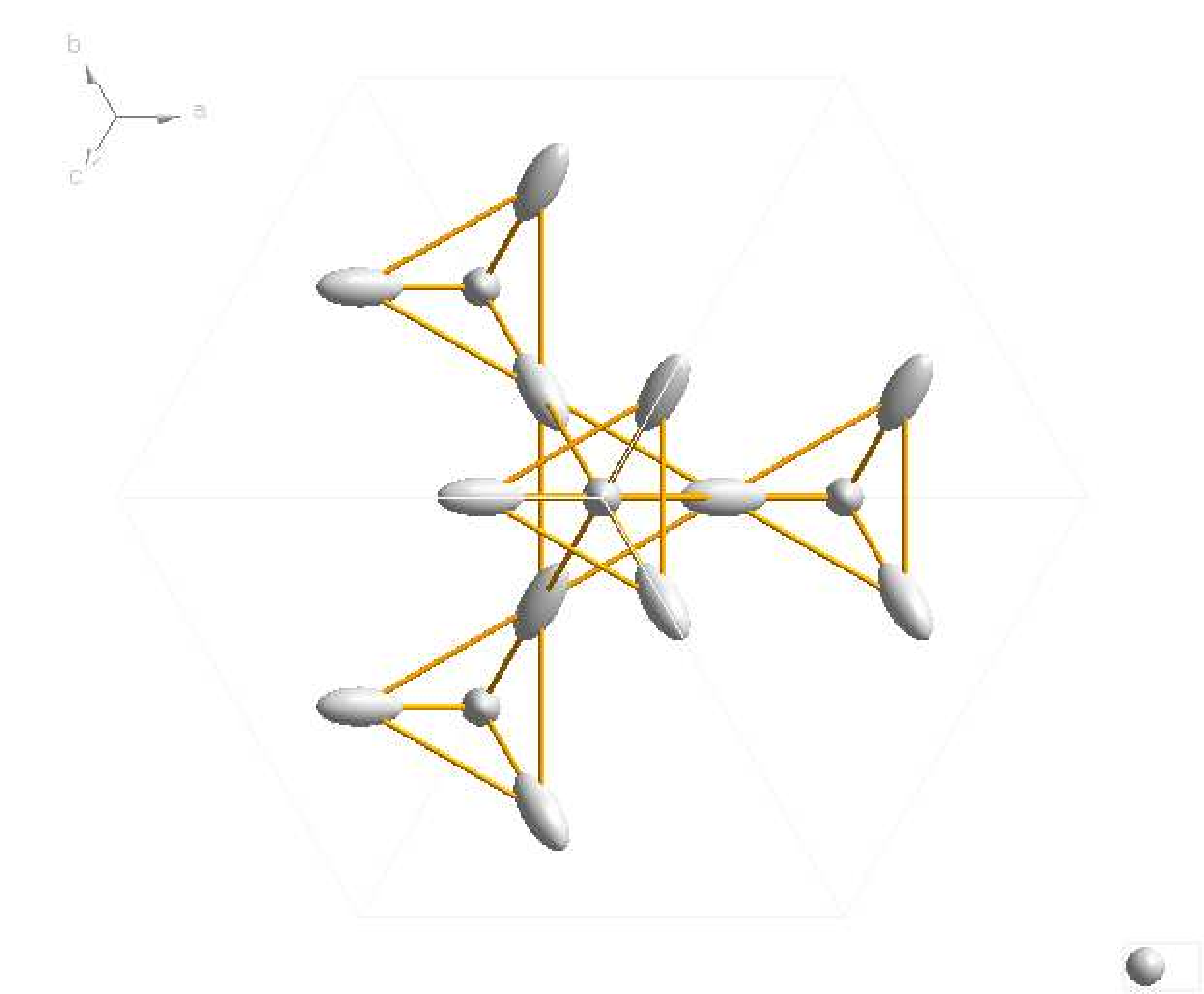
100 K 1 T, 100 FR

VISUALIZATION OF ASP BY *"MAGNETIC ELLIPSOIDS"* *H II [111]*



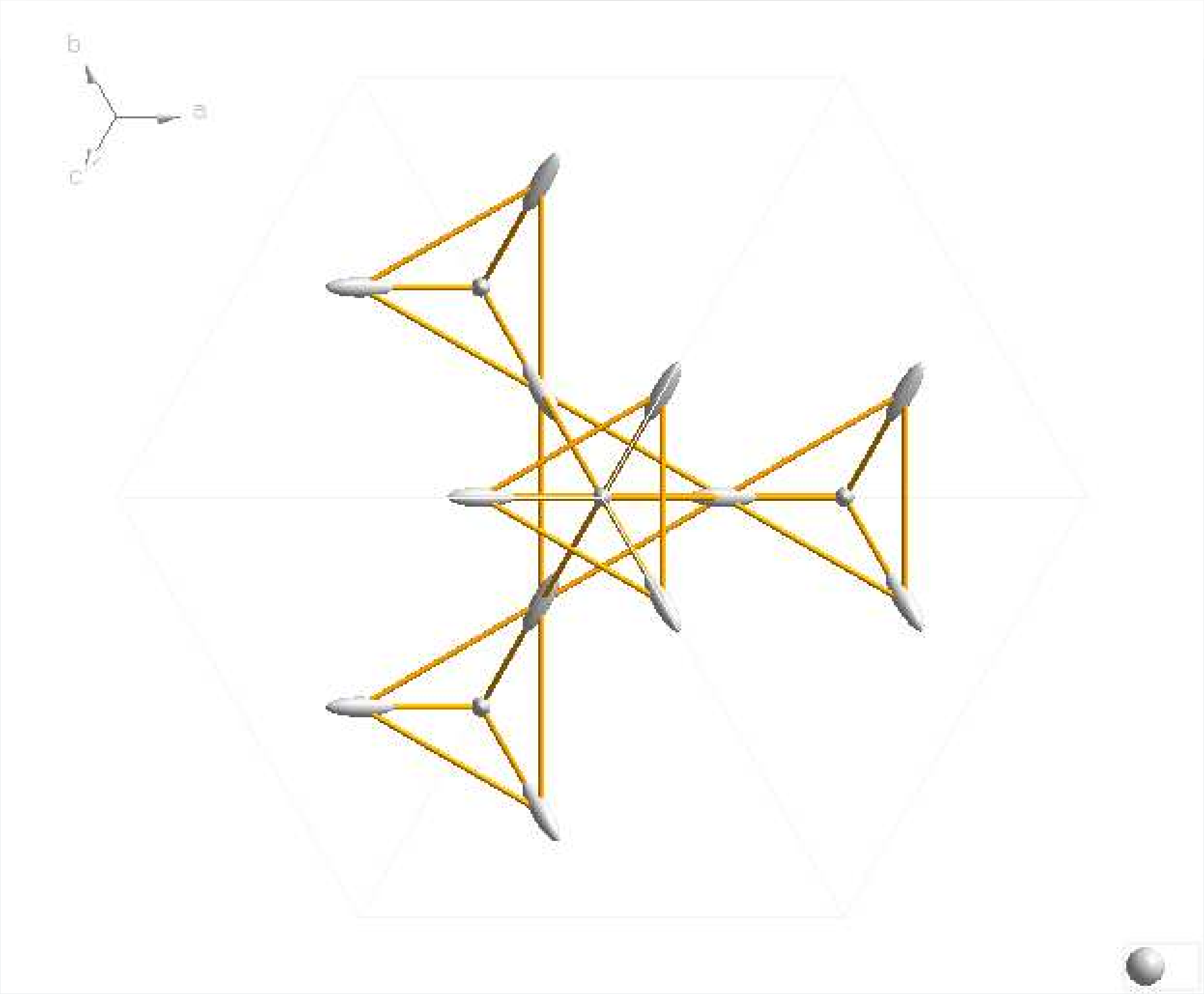
50 K 1 T, 100 FR

VISUALIZATION OF ASP BY "*MAGNETIC ELLIPSOIDS*" *H II [111]*



10 K 1 T, 150 FR

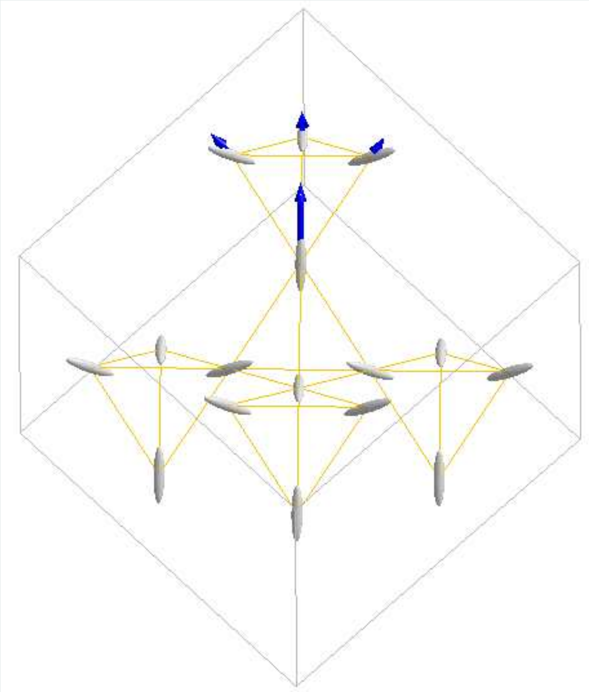
VISUALIZATION OF ASP BY "*MAGNETIC ELLIPSOIDS*" *H II [111]*

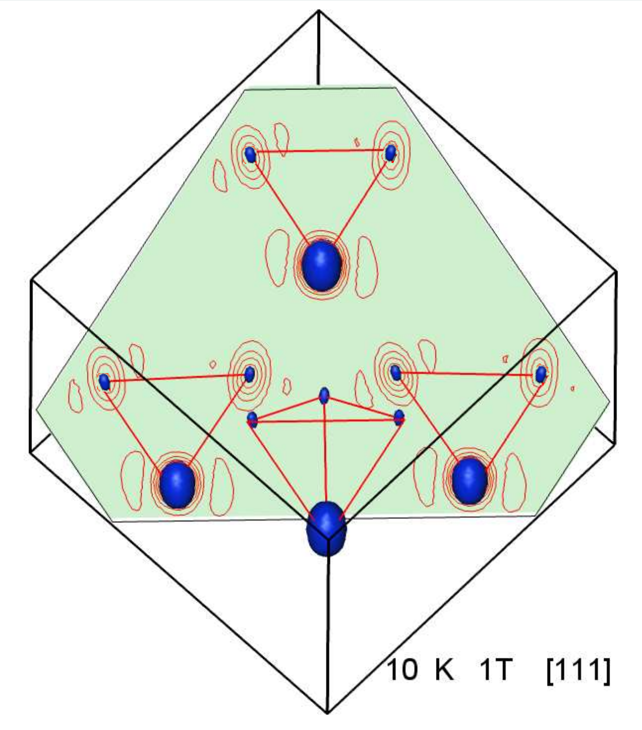


5 K 1 T, 150 FR

(BAD) “One-in three-out” spin ice

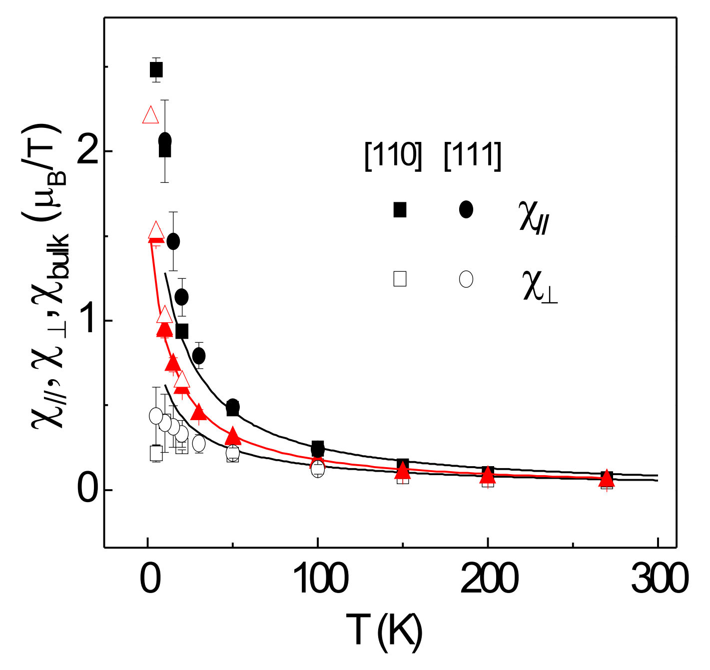
H // 111

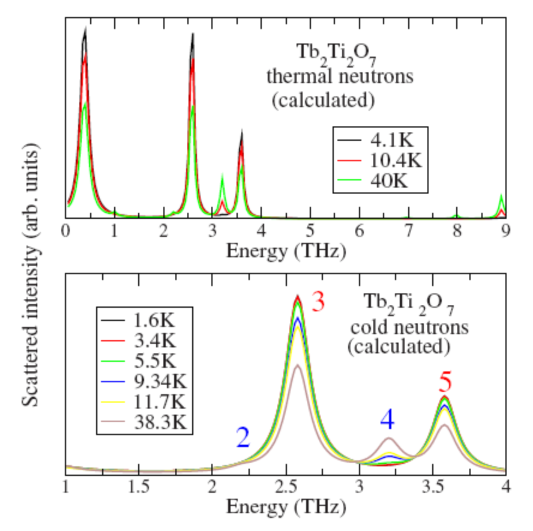




CF calculation of susceptibility tensor

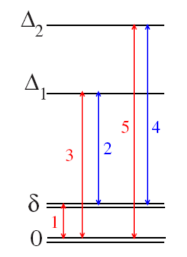
$Tb_2Ti_2O_7$





$B_2^0 = 315\ \mathrm{cm}^{-1}$, $B_4^0 = 1820\ \mathrm{cm}^{-1}$, $B_4^3 = -580\ \mathrm{cm}^{-1}$,

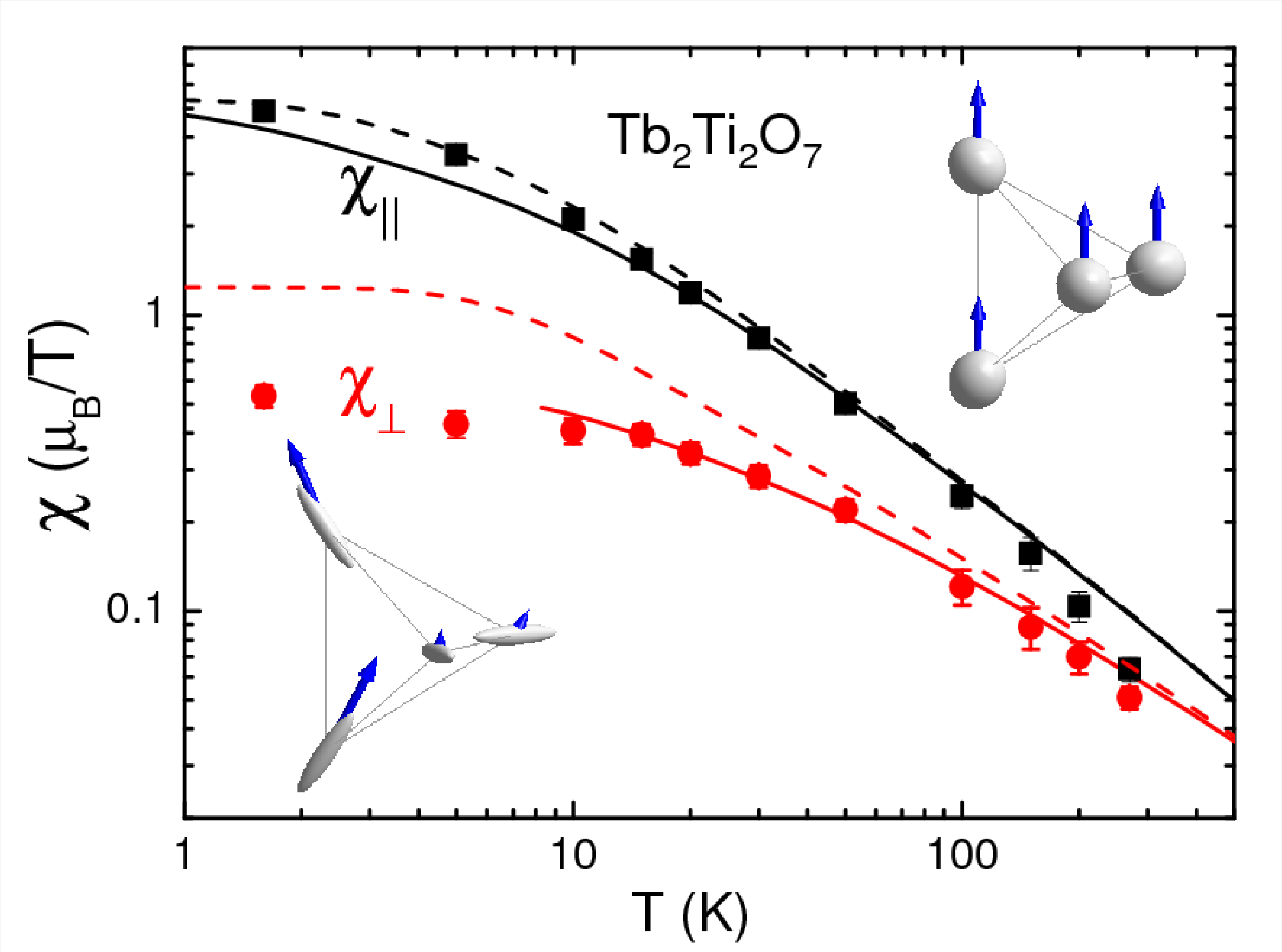
$B_6^0 = -300\ \mathrm{cm}^{-1}$, $B_6^3 = 228\ \mathrm{cm}^{-1}$, $B_6^6 = 900\ \mathrm{cm}^{-1}$.



Lines shows fit using CF parameters from inelastic neutrons for Tb2Ti2O7.
I. Mirebeau, M. Hennion and P. Bonville . Phys Rev. B 184436, 2007

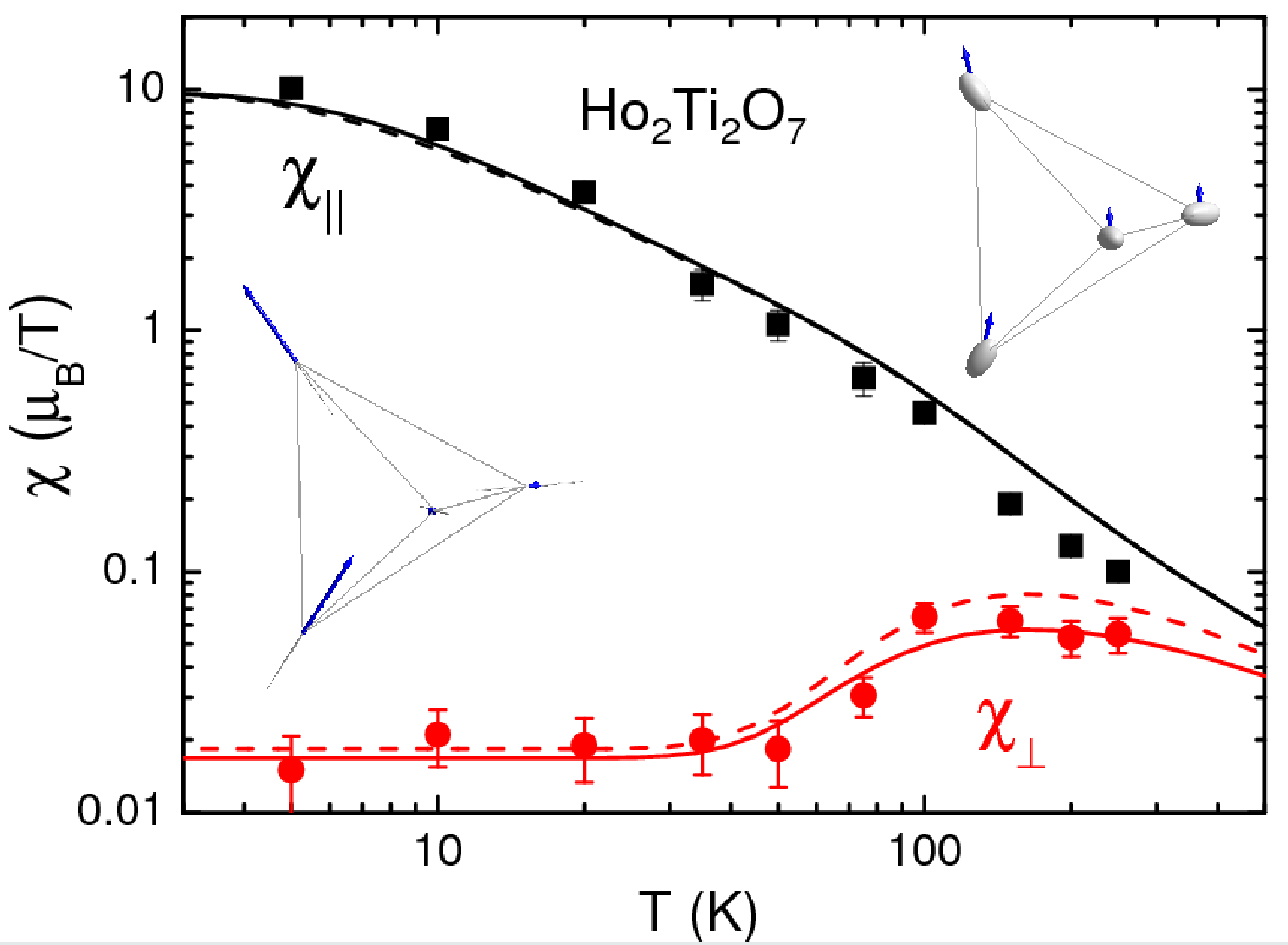
Quasi-Ising Type, Spin Liquid

Lines shows fit using CF parameters from inelastic neutrons for Tb2Ti2O7.
I. Mirebeau, M. Hennion and P. Bonville . Phys Rev. B 184436, 2007



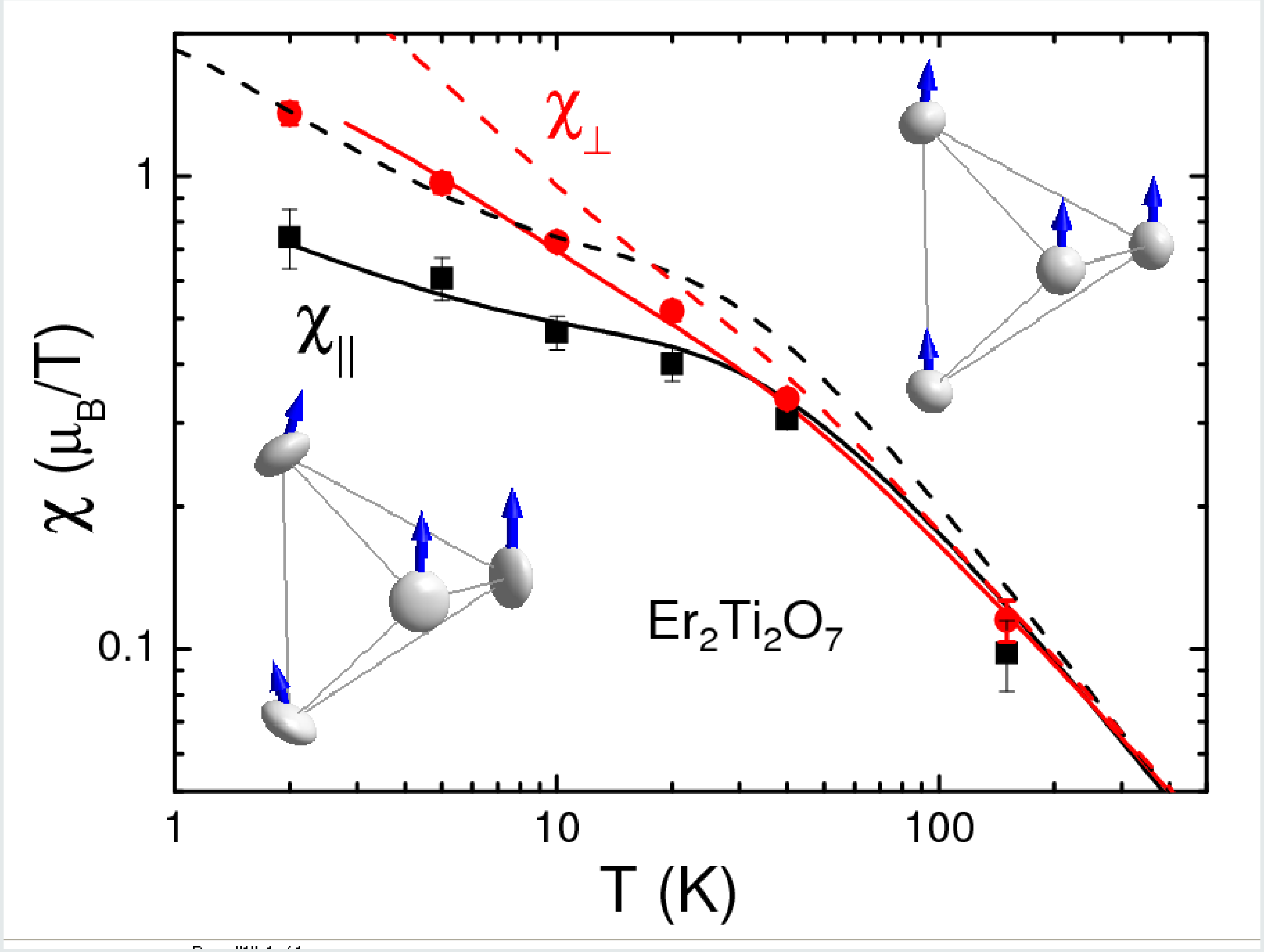
Ising Type (Spin Ice)

Lines shows fit using CF parameters from inelastic neutron data



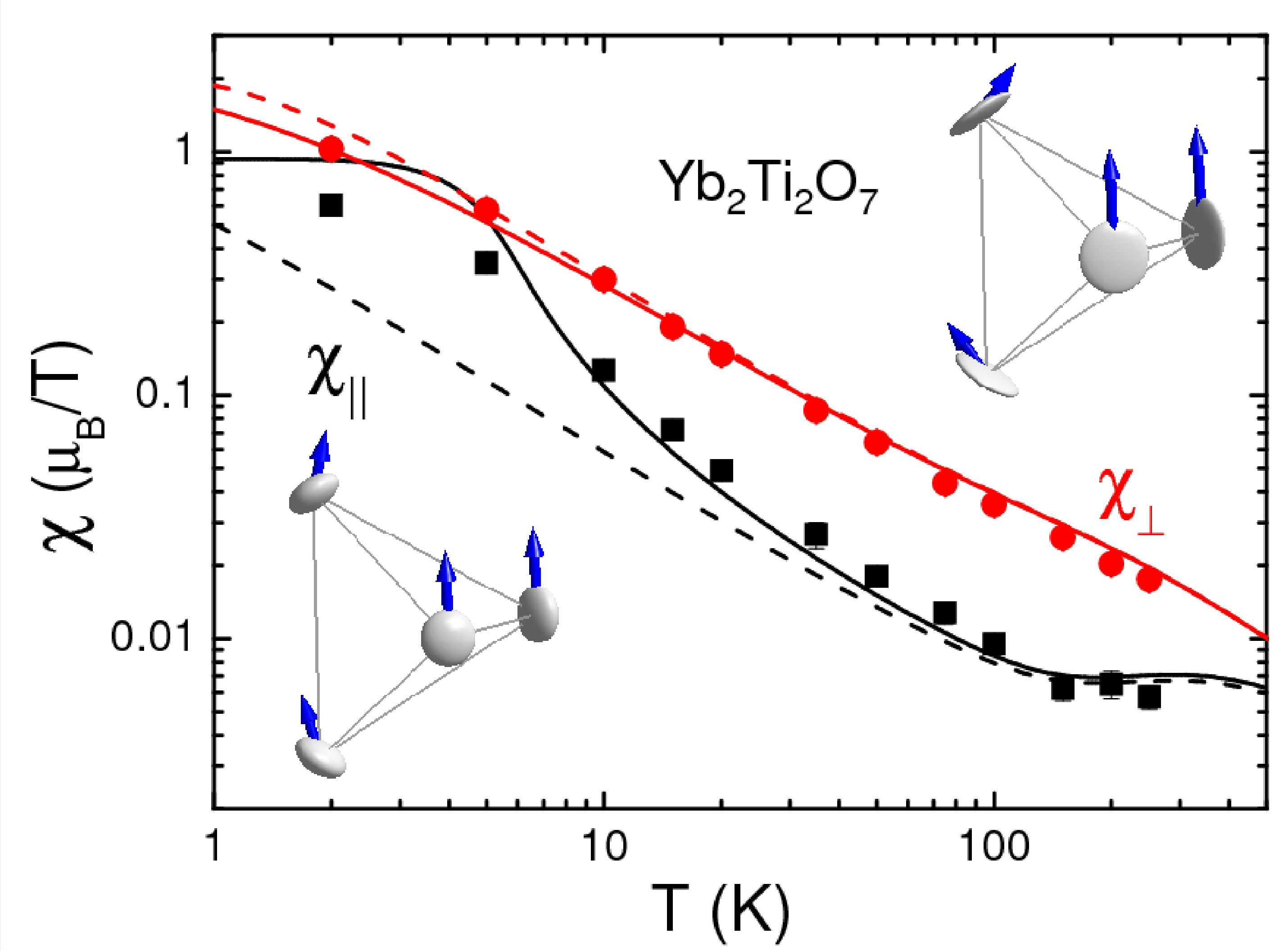
XY Type 3D-Antiferromagnet

Lines shows fit using CF parameters extrapolated from Ho2Ti2O7 and Tb2Ti2O7

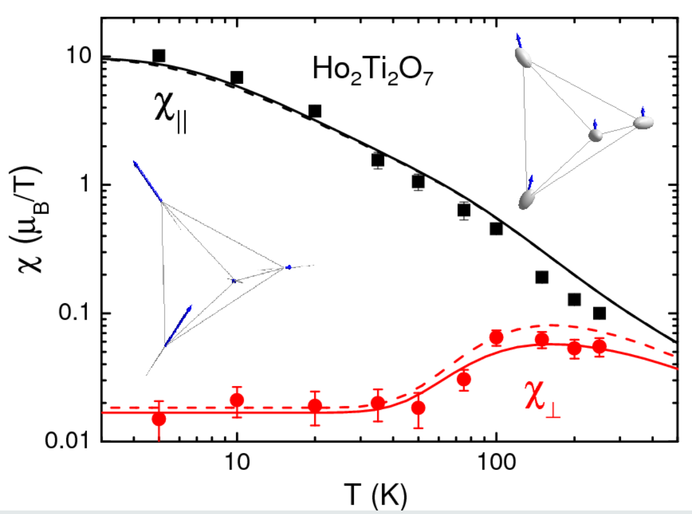


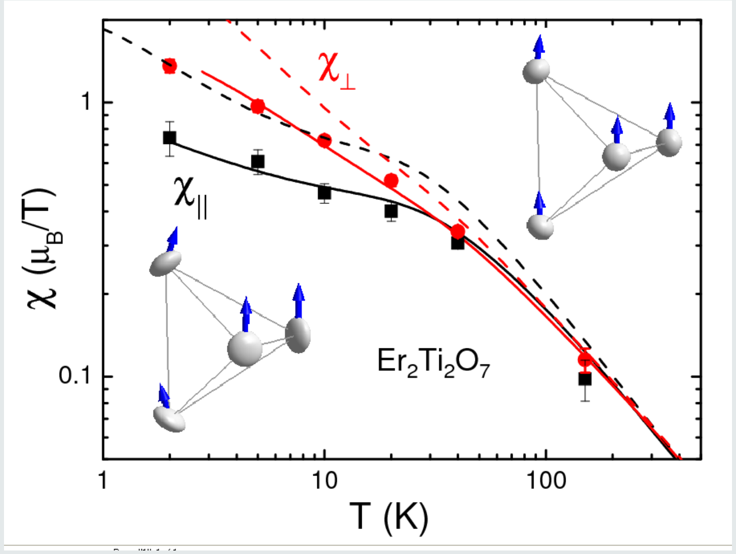
XY Type ???

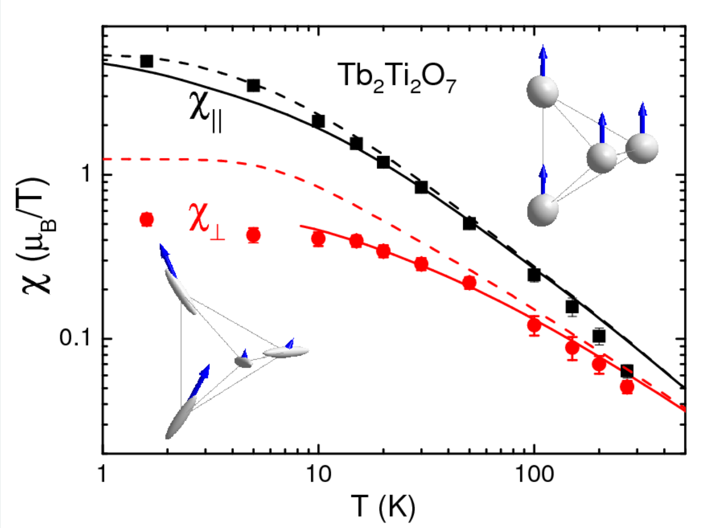
Lines shows fit using L=3, S=1/2 model

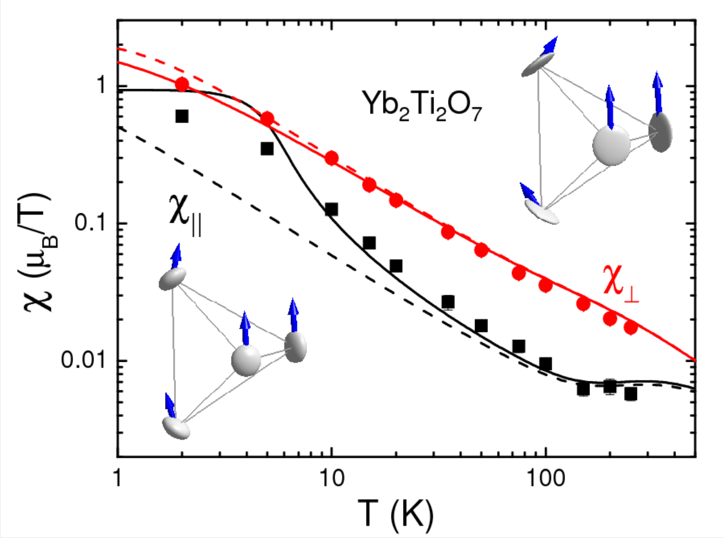


Ising and XY(?) pyrochlores

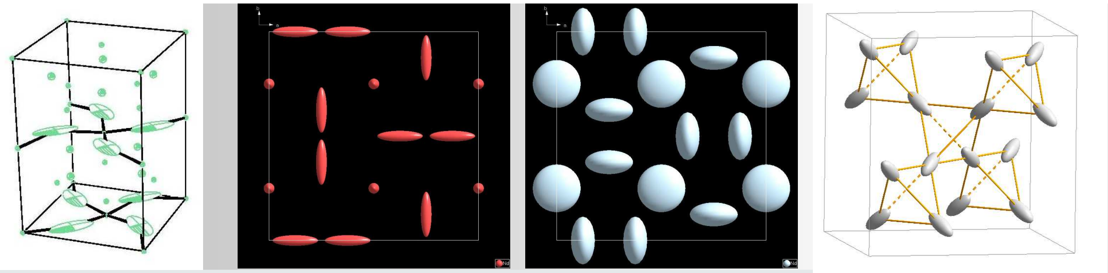








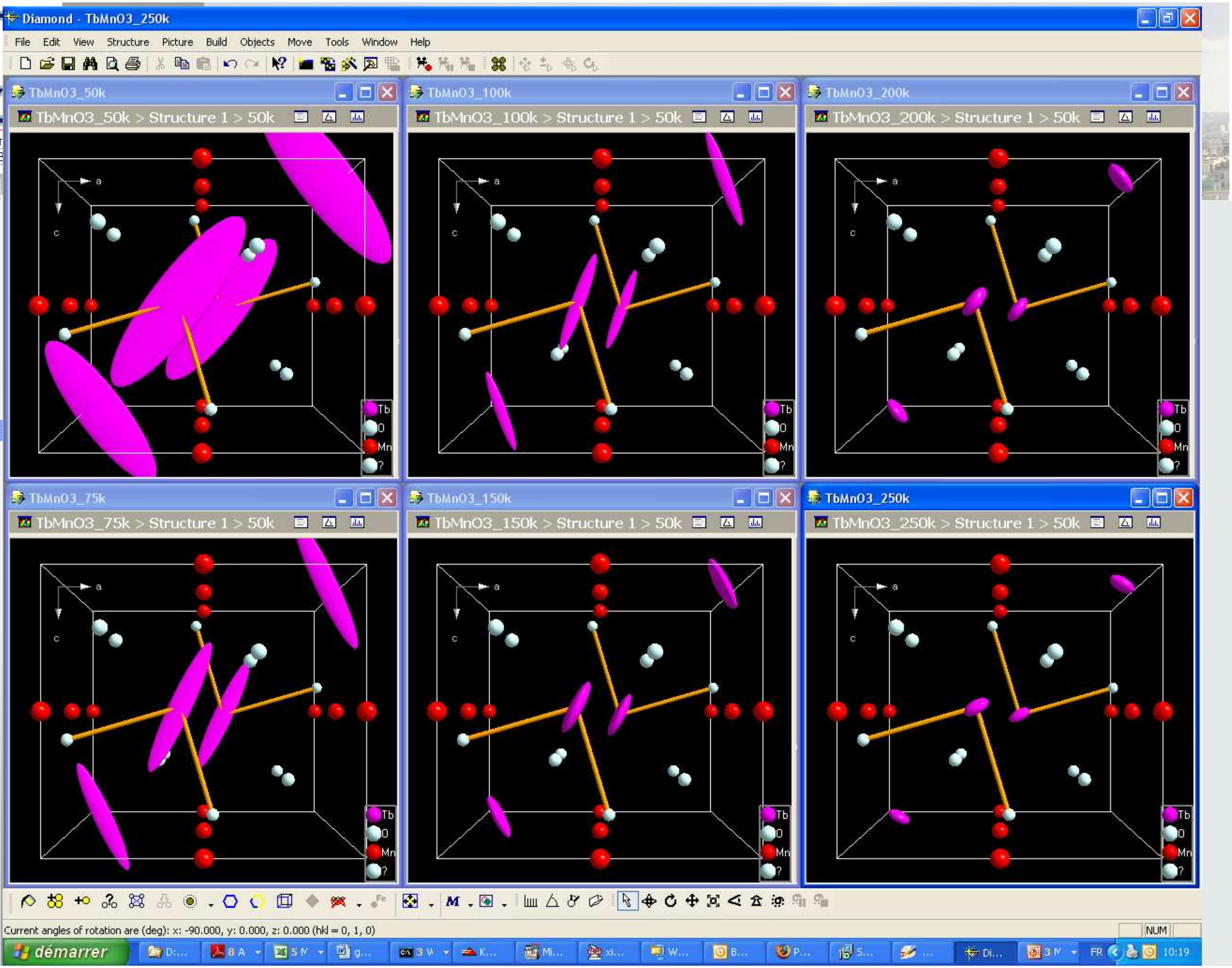
SUMMARY



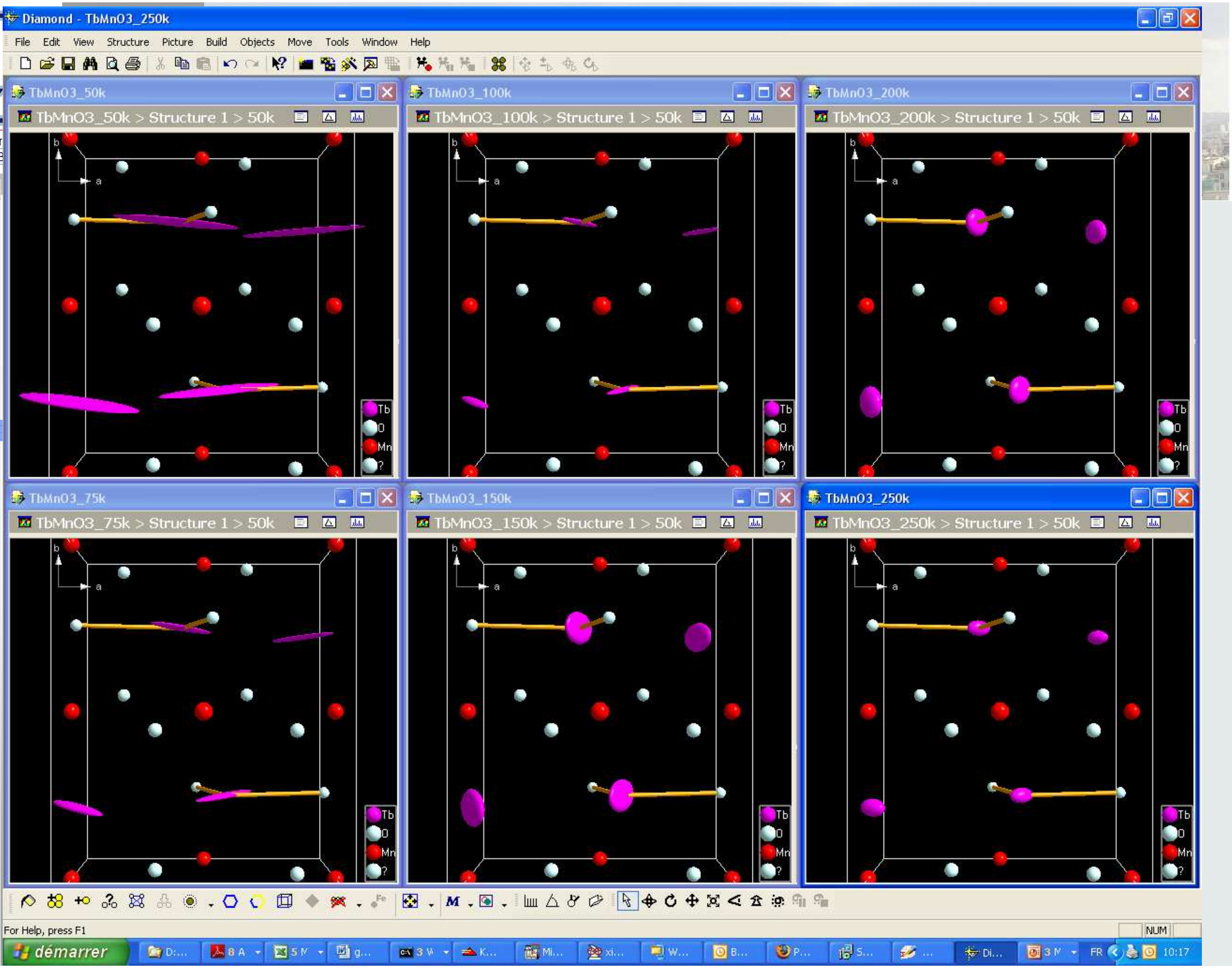
χ_{ij} tensor

- gives universal description of large T and H range with 2 parameters for any field direction
- evidence Heisenberg- Ising or XY evolution
- Can be calculated using CF parameters
- Reconstruction of the noncollinear density ?

- *H. Cao, A. Gukasov, I. Mirbeau and P. Bonville ,PRL , 100, 22,227602, 2008*
- Physica B *PNCMI 2008 Proceeding*
- *J. Phys.: Conf. Ser. 145 012021*, 2008



Current angles of rotation are (deg): x: -90.000, y: 0.000, z: 0.000 (hkl = 0, 1, 0)


Diamond - TbMnO3_250k
File Edit View Structure Picture Build Objects Move Tools Window Help
TbMnO3_50k
TbMnO3_50k > Structure 1 > 50k
TbMnO3_100k
TbMnO3_100k > Structure 1 > 50k
TbMnO3_200k
TbMnO3_200k > Structure 1 > 50k
TbMnO3_75k
TbMnO3_75k > Structure 1 > 50k
TbMnO3_150k
TbMnO3_150k > Structure 1 > 50k
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TbMnO3_250k > Structure 1 > 50k
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PRL **100,** 227602 (2008) PHYSICAL REVIEW LETTERS week ending
6 JUNE 2008

Electric-Field-Induced Spin Flop in $BiFeO_3$ Single Crystals at Room Temperature

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[1]*Service de Physique de l'Etat Condensé, DSM/IRAMIS, CEA Saclay, F-91191 Gif-Sur-Yvette, France*
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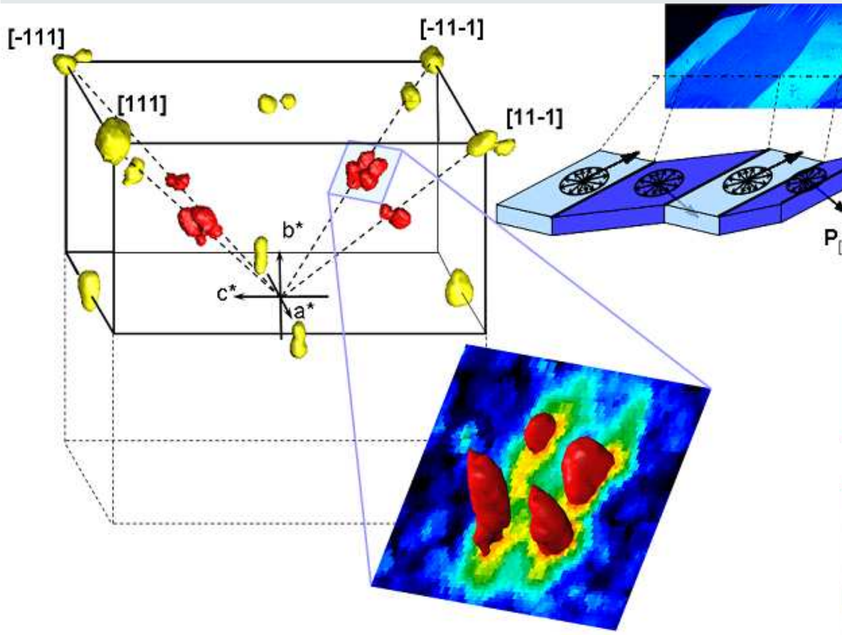
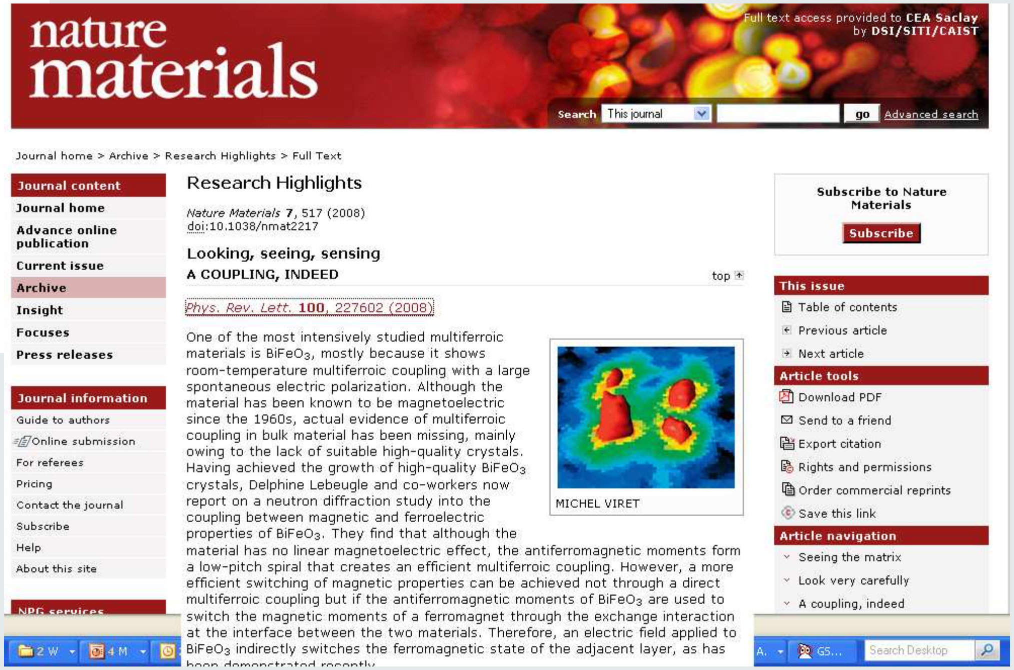
(Received 24 January 2008; published 2 June 2008)





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Research Highlights

Nature Materials **7**, 517 (2008)
doi:10.1038/nmat2217

Looking, seeing, sensing

A COUPLING, INDEED

Phys. Rev. Lett. **100**, 227602 (2008)

One of the most intensively studied multiferroic materials is $BiFeO_3$, mostly because it shows room-temperature multiferroic coupling with a large spontaneous electric polarization. Although the material has been known to be magnetoelectric since the 1960s, actual evidence of multiferroic coupling in bulk material has been missing, mainly owing to the lack of suitable high-quality crystals. Having achieved the growth of high-quality $BiFeO_3$ crystals, Delphine Lebeugle and co-workers now report on a neutron diffraction study into the coupling between magnetic and ferroelectric properties of $BiFeO_3$. They find that although the material has no linear magnetoelectric effect, the antiferromagnetic moments form a low-pitch spiral that creates an efficient multiferroic coupling. However, a more efficient switching of magnetic properties can be achieved not through a direct multiferroic coupling but if the antiferromagnetic moments of $BiFeO_3$ are used to switch the magnetic moments of a ferromagnet through the exchange interaction at the interface between the two materials. Therefore, an electric field applied to $BiFeO_3$ indirectly switches the ferromagnetic state of the adjacent layer, as has been demonstrated recently.

MICHEL VIRET