Magnetic Order and Magnetoelectric effect in the frustrated Fe langasites

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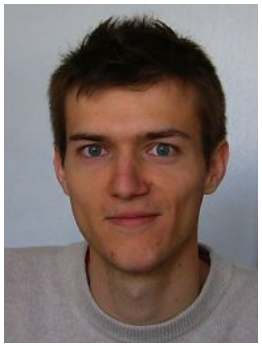

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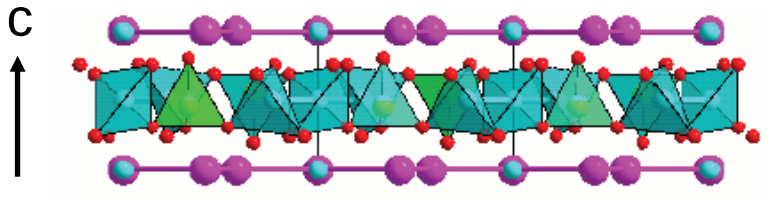
Outline : - Structure
- Magnetic properties
- Dielectric properties
- Conclusion

Fe-langasite : structure

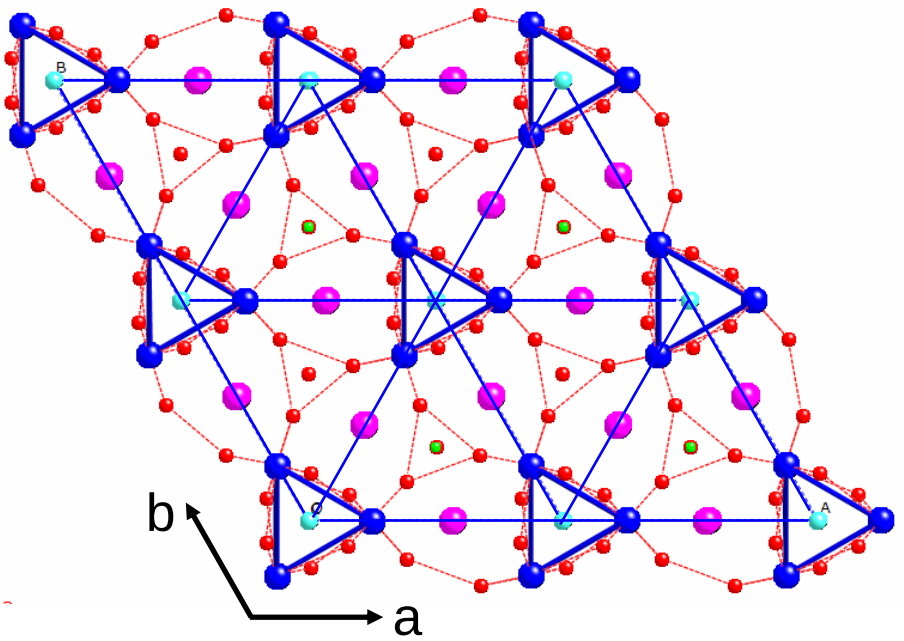
$Ba_3NbFe_3Si_2O_{14}$: space group P321

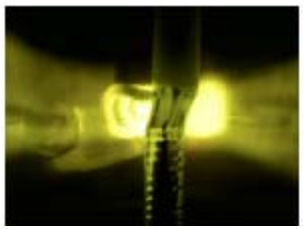
triangular lattice of Fe^{3+} triangles, S=5/2



Geometrical frustration
Acentric structure

⇒Magnetic properties?
⇒Magnetoelectric effect?
⇒Multiferroism?

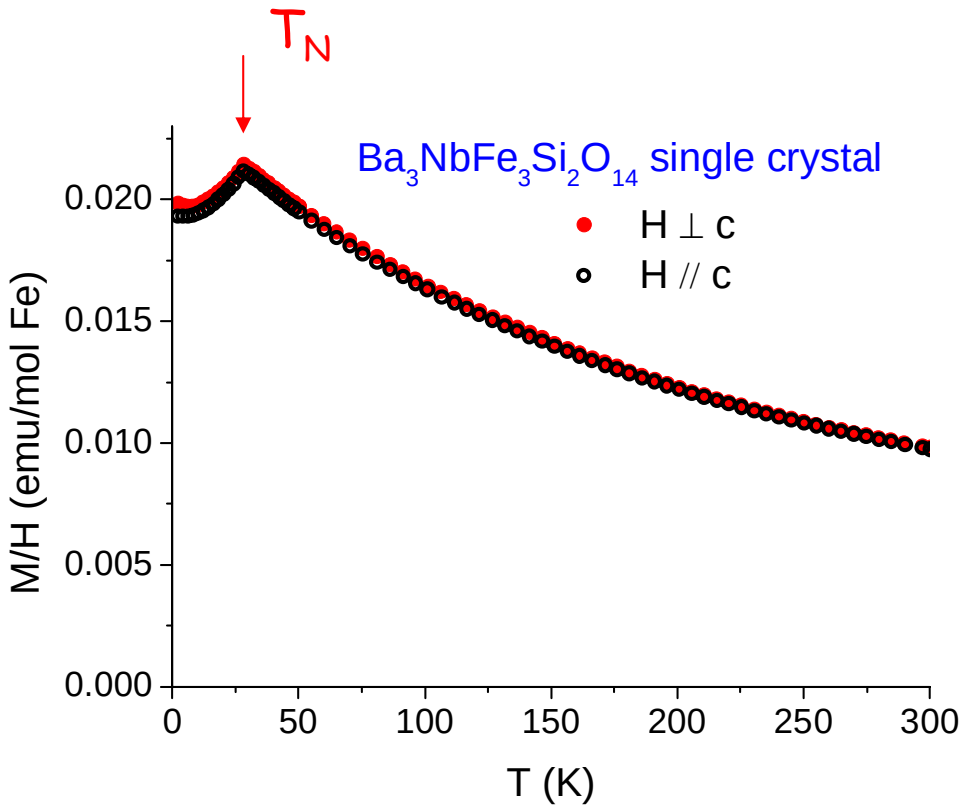




Crystal grown by floating zone method in image furnace

Fe-langasite : magnetic properties

Macroscopic measurements

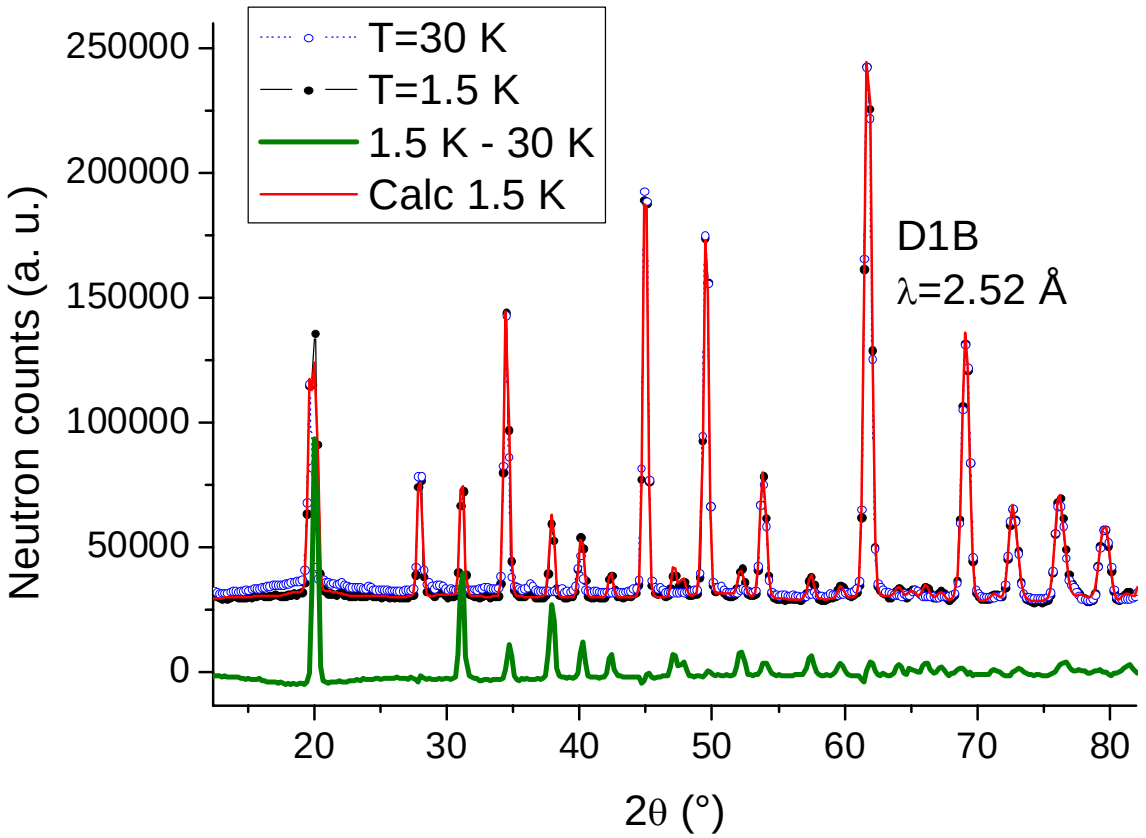


Magnetic transition → antiferromagnetic order at T_N~28 K

Fe-langasite: magnetic structure

Neutron diffraction
on powder (D1B),
and single crystal (D15) at ILL

Powder pattern

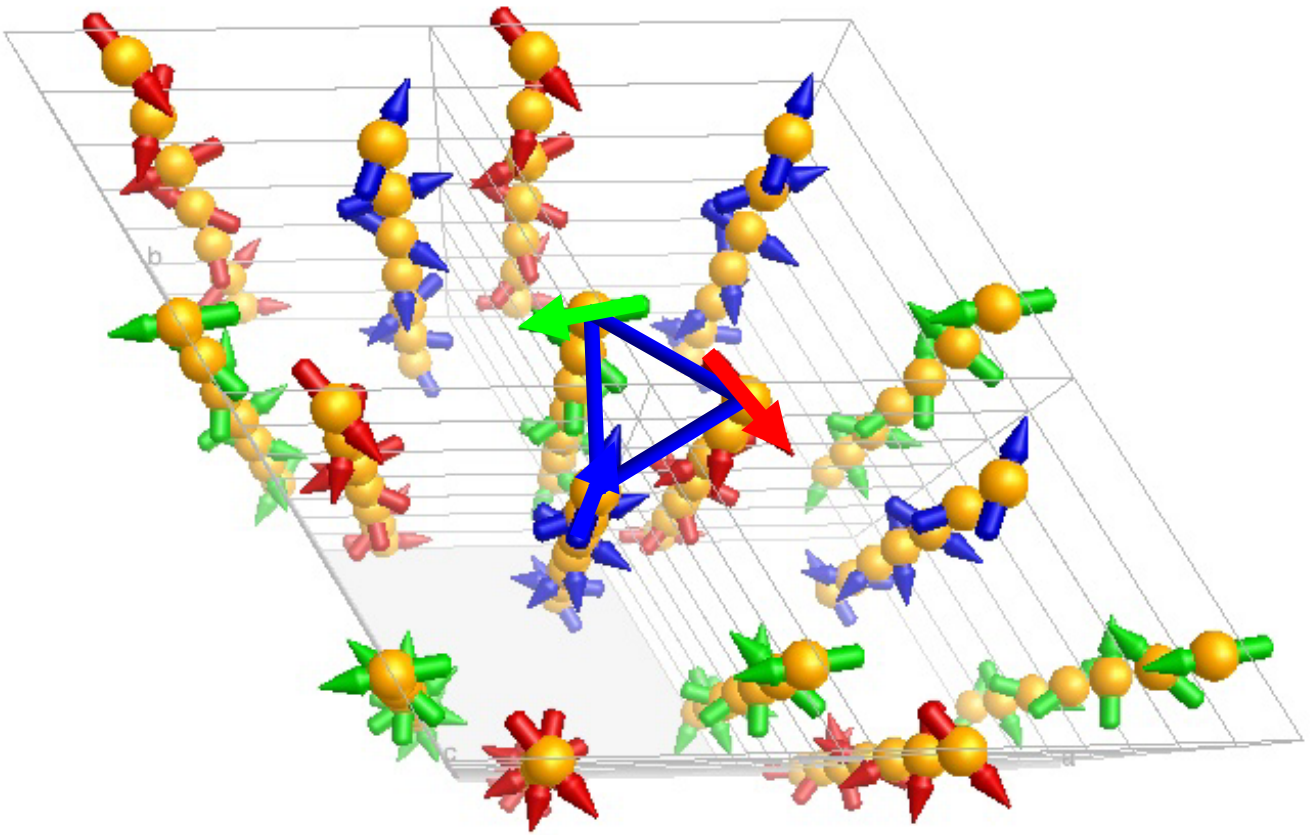


Below T_N=28 K,
AF order with
propagation vector
k=(0,0,~1/7)

Fe-langasite: magnetic structure

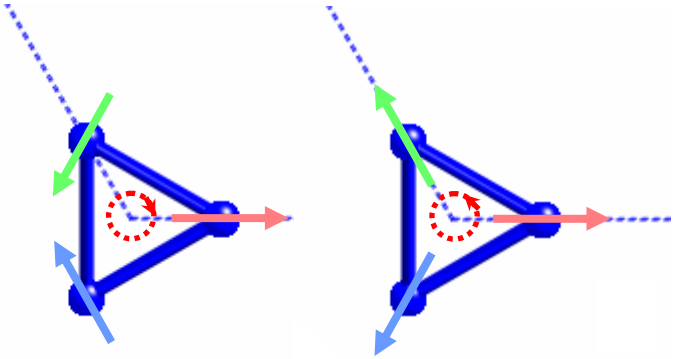
120° moments on triangles in the (a, b) plane
→ signature of frustration

Helices propagating along c



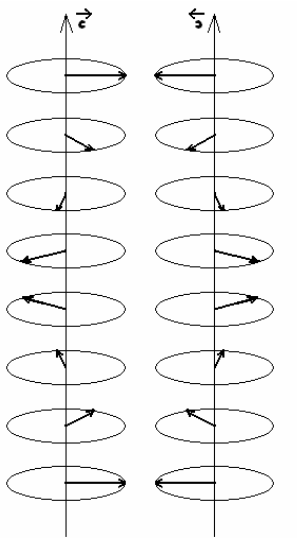
Chirality

- Structural chirality : acentric structure



X-ray (anomalous)
⇒ aenantiomorph

- Triangular chirality $\vec{C}_T = \frac{2}{3\sqrt{3}} \sum_{triangle} \left[\vec{S}_i \times \vec{S}_j\right]$



- Helicity $\vec{C}_H = \left[\vec{S}_i \times \vec{S}_j\right]$

- Chirality domains unbalanced by external constrains
- Single domain in non-centrosymmetric crystals

  e. g. MnSi - helical modulation and sense of rotation
  - due to Dzyaloshinskii-Moryia interaction

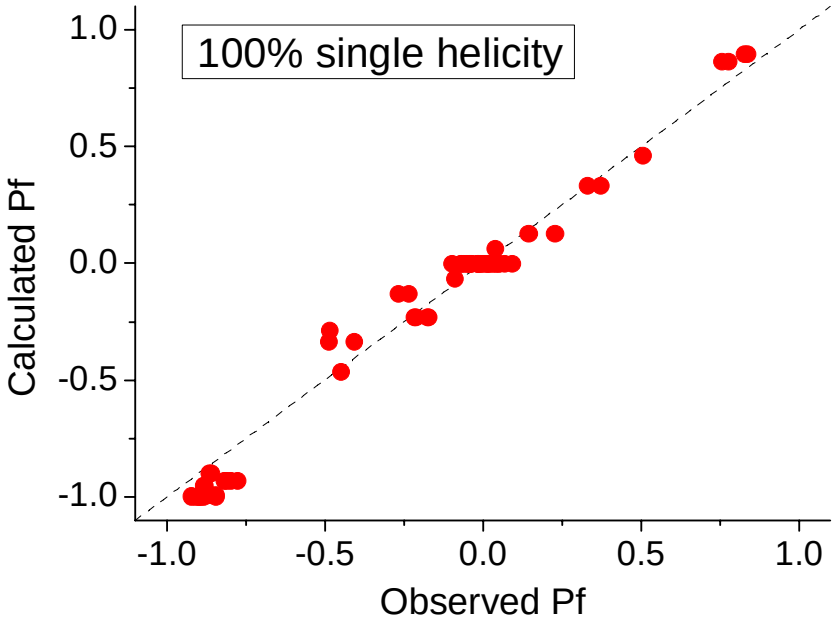
What about Fe-langasite?

Fe-langasite: magnetic domains

Diffraction on single crystal with unpolarized neutrons
&
with polarized neutron and spherical polarization analysis

CRYOPAD
(IN22, ILL)

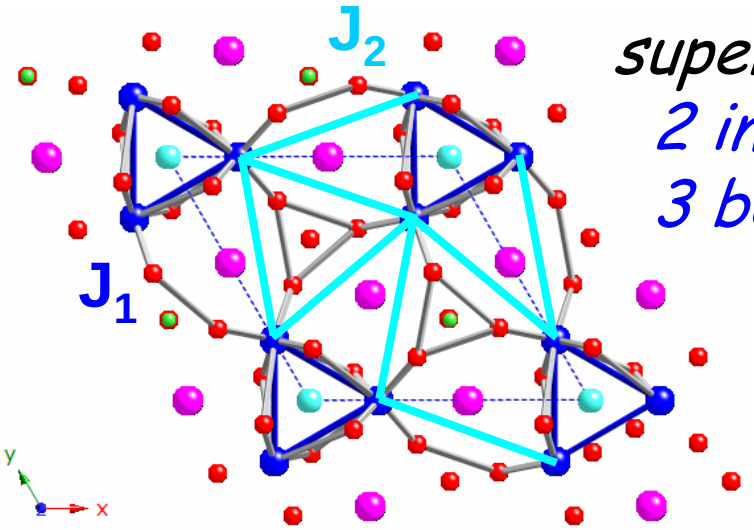
Calculated vs measured final polarization for several magnetic reflections



⇒Monodomain magnetic structure
(helicity & triangular chirality)

Fe-langasite: magnetic properties

Mean-field analysis: probe of the microscopic mechanisms



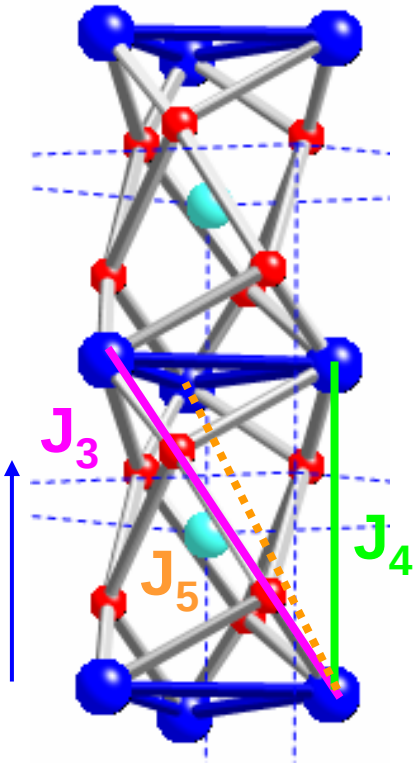
AF J_1, J_2

Heisenberg Hamiltonian :

super-exchange interactions

2 intra-plane

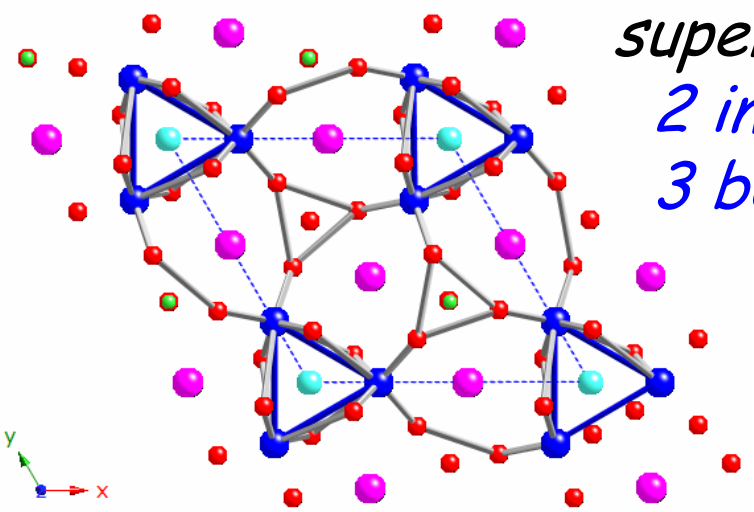
3 between adjacent planes



$J_3 \neq J_5$

Fe-langasite: magnetic properties

Mean-field analysis: probe of the microscopic mechanisms



Heisenberg Hamiltonian :

super-exchange interactions

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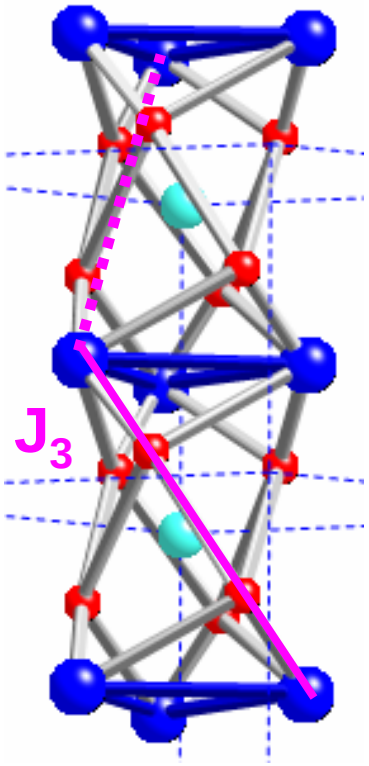


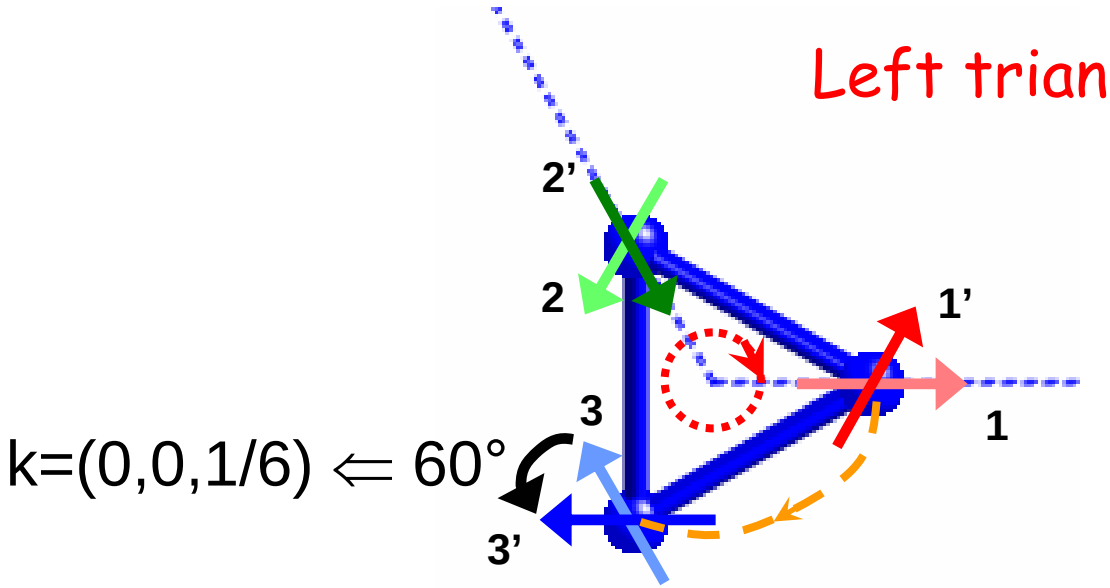
⇒ Describe the magnetic structure

⇒ selects two pairs of helicity and triangular chirality (C_H, C_T)

Fe-langasite: magnetic properties

Link between the magnetic chiralities (C_H, C_T) and the structural one

Structure : Left handedness



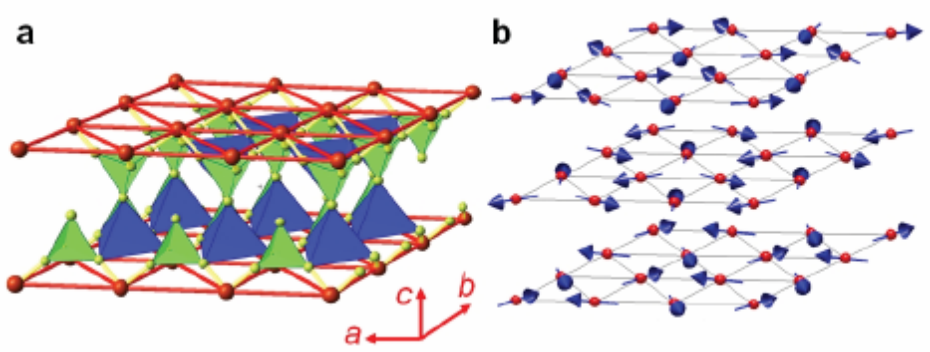
Left triangular chirality

Right helicity

Marty et al., Phys. Rev. Lett. (in press)

Chirality and ferroelectricity

Case of $RbFe(MoO_4)_2$:
triangular lattice of Fe^{3+}

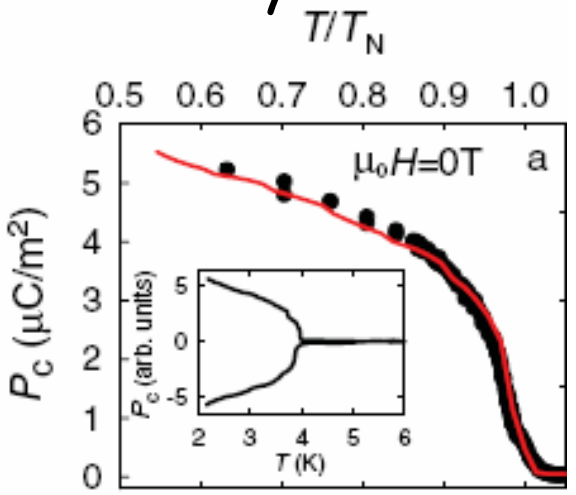


Kenzelmann et al. PRL (2007)

120° magnetic structure
but centro-symmetric ⇒ 2 domains of magnetic chiralities

$\exists P_E$ at T_N ⇒ symmetry based phenomenological theory

Proportional to the chirality unbalance

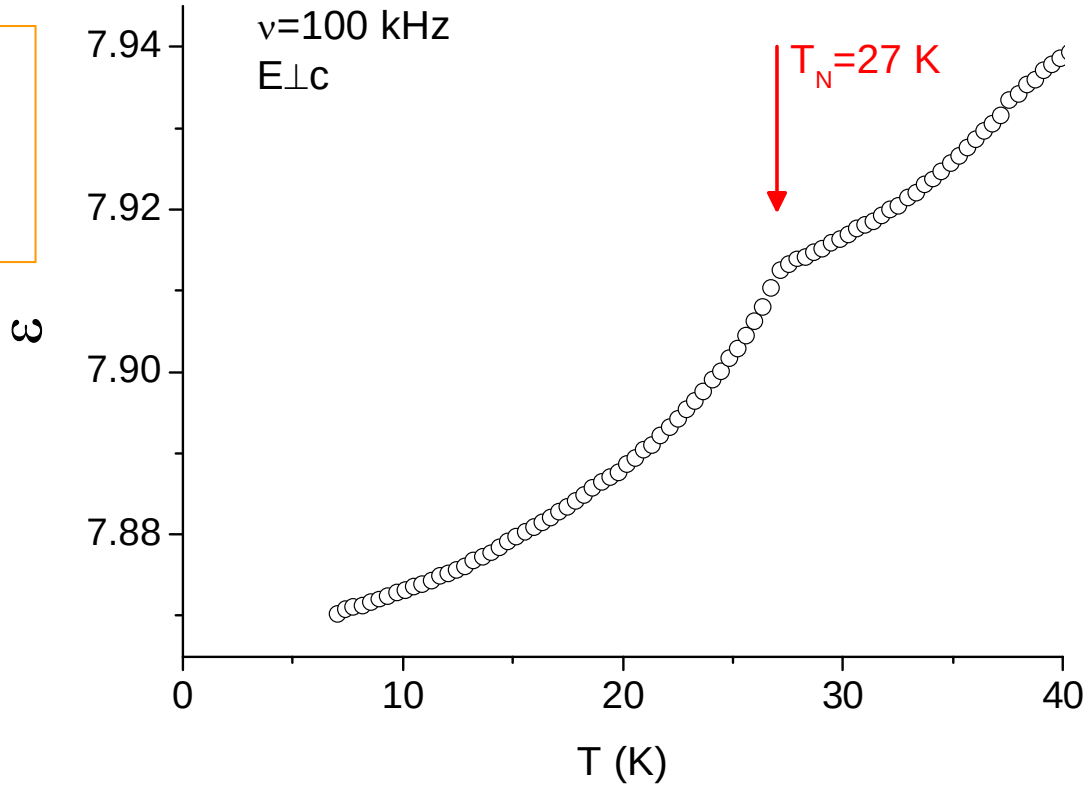


Conclusion :
«trigonal stacked triangular antiferromagnets with 120° structure are multiferroics.»

Fe-langasite: dielectric properties

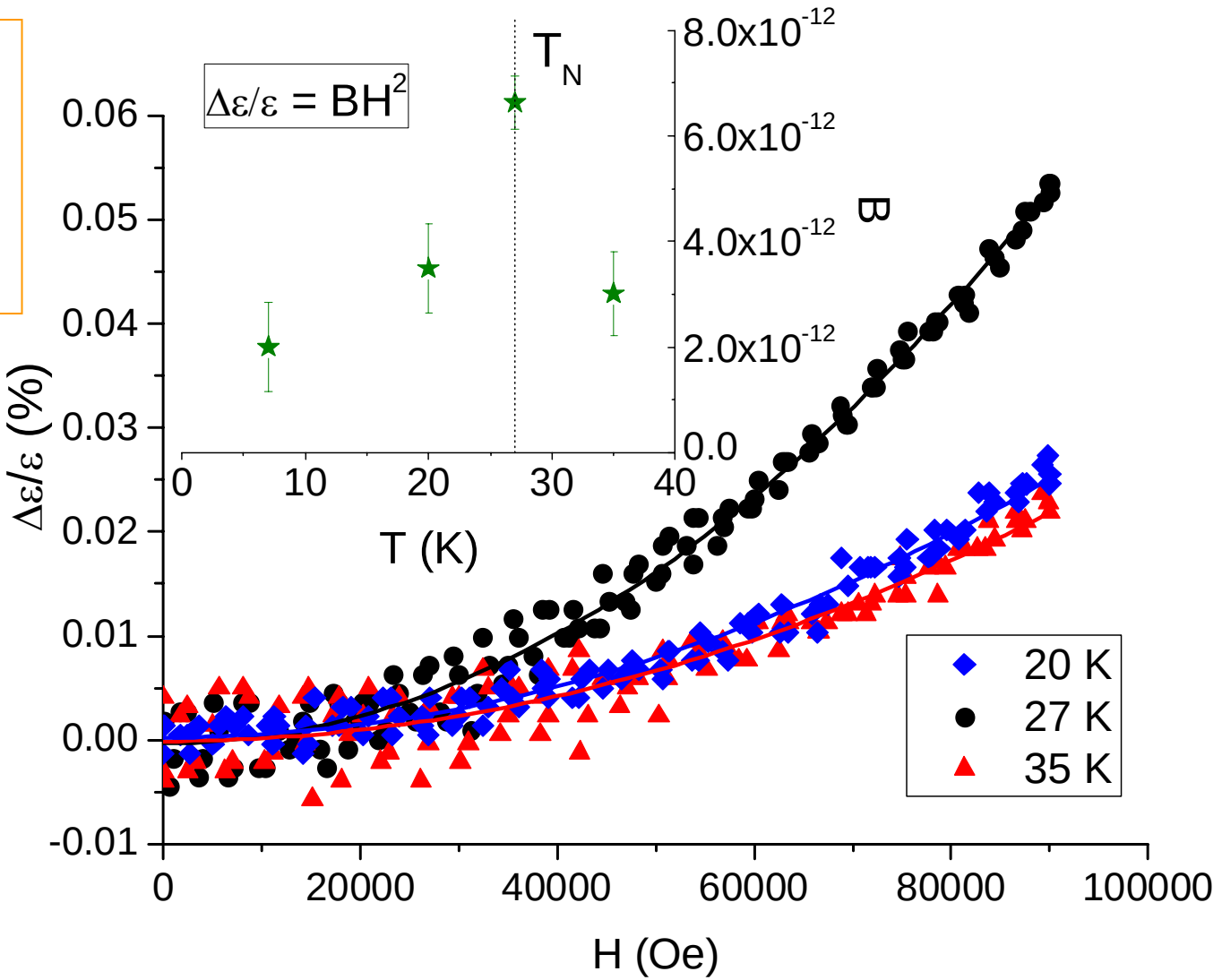
Coll. with B. Zawilsky, J. Marcus, Institut Néel
and C. Simon, B. Kundys, CRISMAT, ENSICAEN

Dielectric constant
via capacity measurements
Anomaly near T_N for $E \perp c$



Fe-langasite: dielectric properties

Magnetodielectric coupling
quadratic in H
Maximum at T_N



Fe-langasite: dielectric properties

Free Energy : $$F = F_0 - P_i E_i - M_i H_i - \frac{1}{2}\varepsilon_{ij} E_i E_j - \frac{1}{2}\chi_{ij} H_i H_j + \ldots$$

$$- \alpha_{ij} E_i H_i - \frac{1}{2}\beta_{ijk} E_i H_j H_k - \frac{1}{2}\gamma_{ijk} E_i E_j H_k - \frac{1}{2}\delta_{ijkl} E_i E_j H_k H_l$$

Results similar to $YMnO_3$, ferroelectric at T_E= 930 K and 120° antiferromagnetic order at T_N=74 K *Bellido et al.*

Pyroelectric current measurement
No evidence of spontaneous electrical polarization?

Why?

Conclusion

Non-centrosymmetric $Ba_3NbFe_3Si_2O_{14}$ langasite :

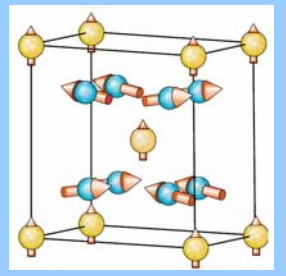
- Very original magnetic structure below T_N=28 K
  single triangular chirality and helicity linked by the twist of the magnetic interactions
- Magnetodielectric coupling/ No ferroelectricity (?)

→Further investigation of dielectric properties
→Many other possible chemical substitutions
→Probe of the excitations

ECOLE THÉMATIQUE

Apport des symétries en matière condensée

Presqu'île de Giens, 11-17 mai 2009



Organisation

thgroupes@ill.fr

B. Grenier *UJF&CEA, Grenoble*
V. Simonet *CNRS, Grenoble*
H. Schober *ILL, Grenoble*

Site WEB opérationnel courant juin
→ **http://www.ill.eu/news-events/workshops-events/ecole-theorie-des-groupes/**

Programme

- Formalisme de la théorie des groupes et des représentations, théorie de Landau
- Applications : groupes d'espace et transitions de phase cristallographiques, structures magnétiques, excitations (phonons, champ cristallin, …)
- Exemples : multiferroïques et supraconductivité

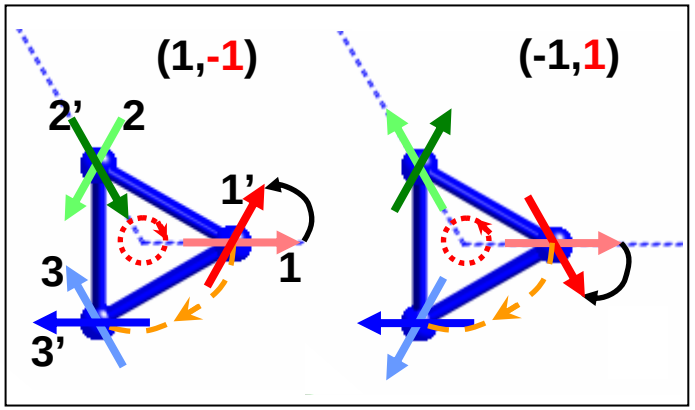
Orateurs

J. Villain, D. Fruchart, R. Ballou, B. Canals, B. Grenier, V. Simonet, P. Toledano, J. M. Perez-Mato, M. I. Aroyo, J. Rodriguez-Carvajal, F. Bourée, J. Schweizer, M. Amara, B. Ouladdiaf, G. Eckold, J. Kreisel, S. Raymond, L. Chapon, M. Houzet

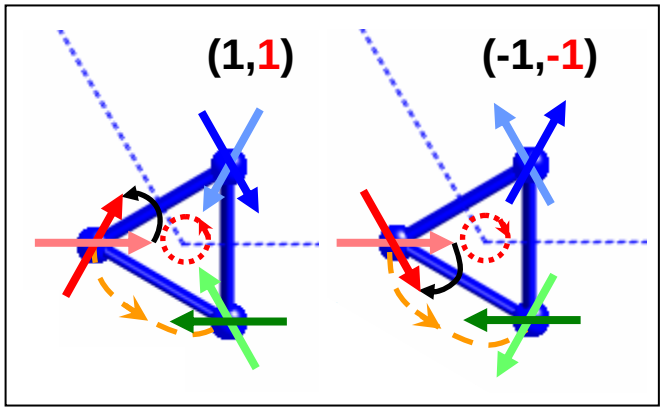
Fe-langasite: magnetic properties

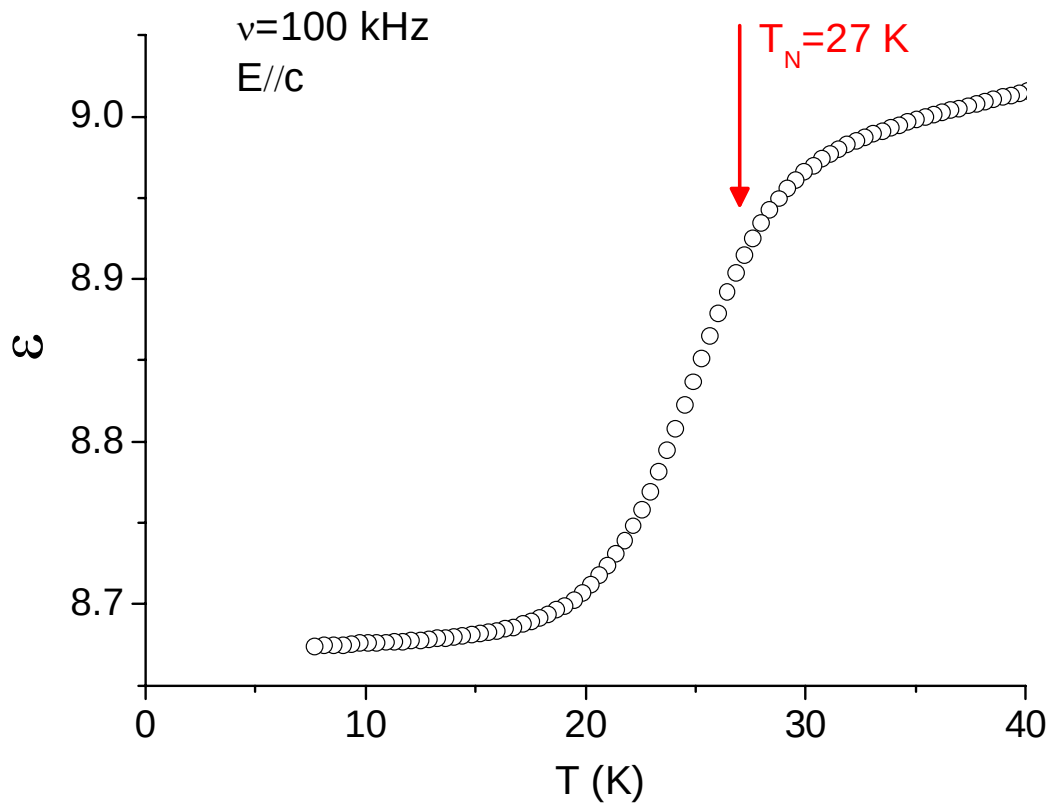
Link between the magnetic chiralities
and the structural one

Left handedness



right handedness




ν=100 kHz
E//c
T_N=27 K
9.0
8.9
8.8
8.7
ε
0
10
20
30
40
T (K)