THz magneto-electric excitations in the chiral compound $Ba_3NbFe_3Si_2O_{14}$

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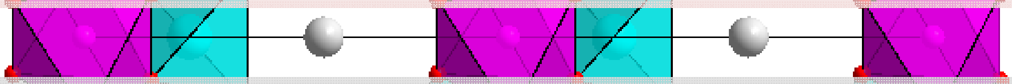

1. **THz spectroscopy**
2. **Dynamical magneto-electric effects: electromagnons**
3. **The langasite $Ba_3NbFe_3Si_2O_{14}$**
4. **THz spectroscopy on $Ba_3NbFe_3Si_2O_{14}$**
5. **Comparison with neutron inelastic measurements**
6. **Model for the magneto-electric excitation in $Ba_3NbFe_3Si_2O_{14}$**

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THz spectroscopy for condensed matter

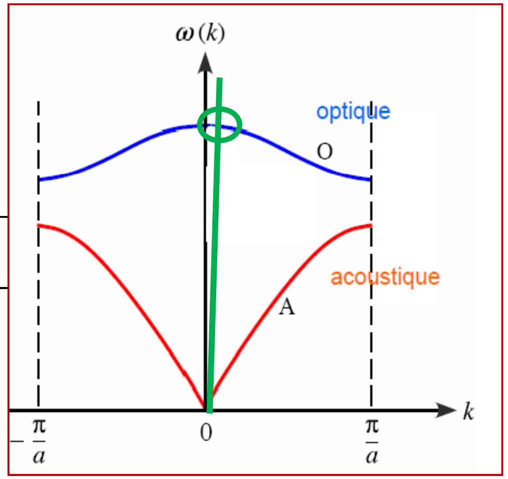
magnetic excitations

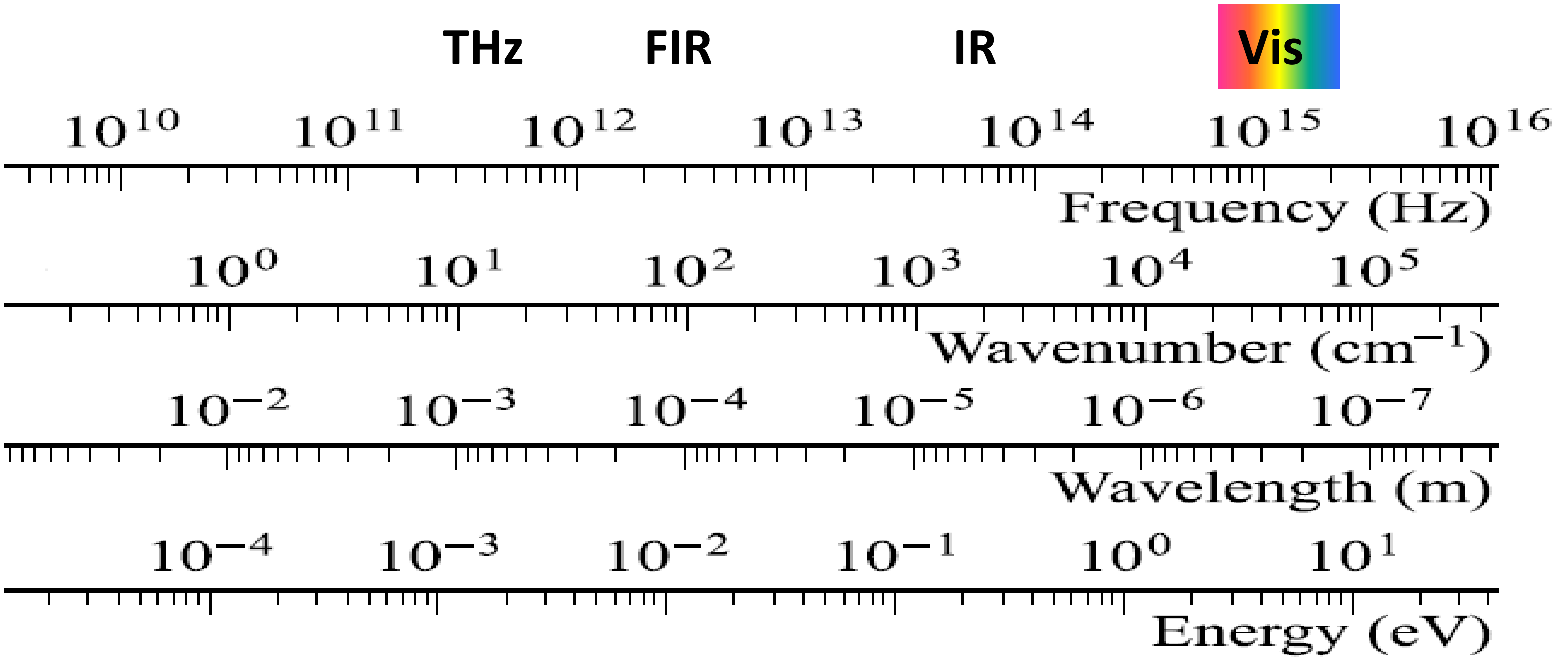
optical phonons

electronic excitations

superconducting gaps

Carrier scattering rates

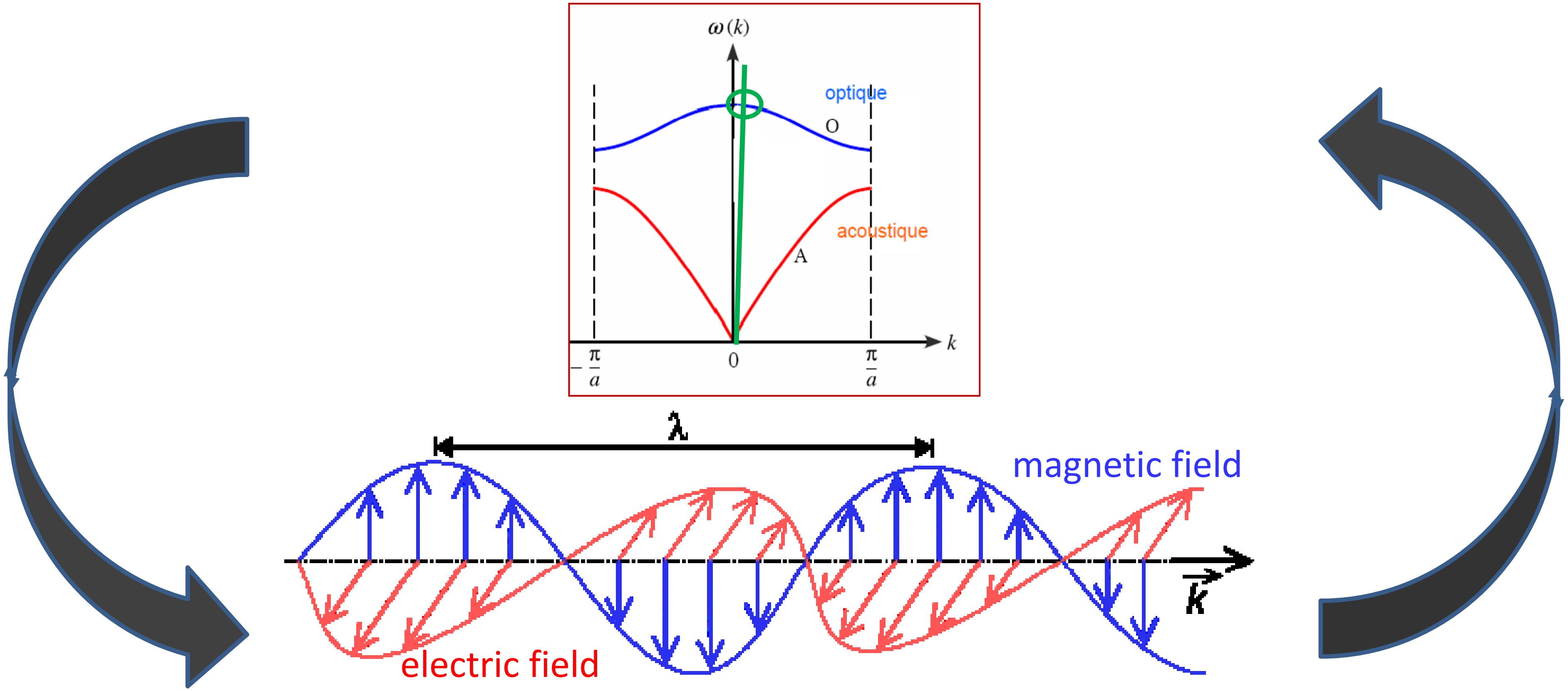




THz spectroscopy for condensed matter

magnetic excitations

optical phonons

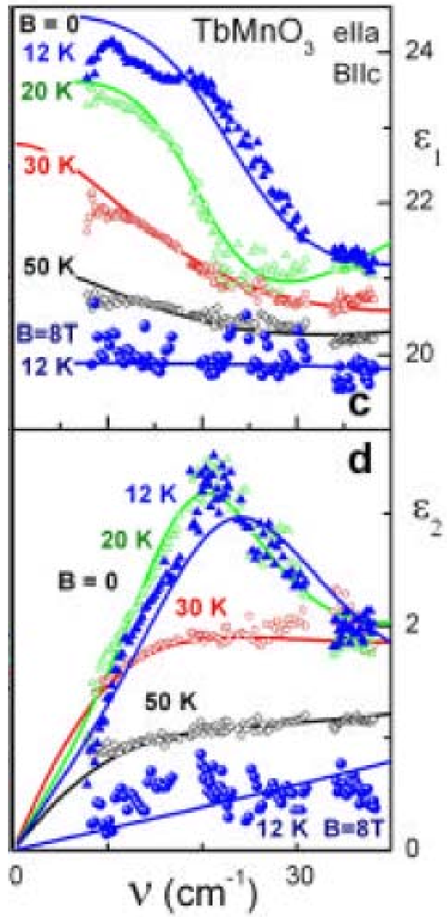


Dynamical magneto electric effects : electromagnons

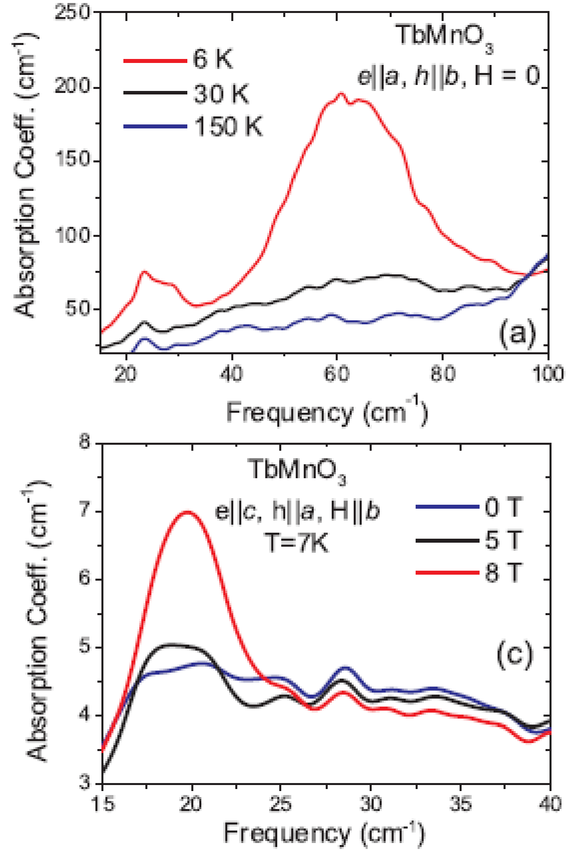
Pimenov et al:
possible evidence for electromagnons in multiferroic manganites
Nature 2006

$TbMnO_3$: excitation at 25 cm^{-1}
for *e//a* in the cycloidal phase.

+60 cm^{-1} for *e//a* in the cycloidal phase
+ … quite complex!?!



Pimenov 2006



Valdes Aguilar 2009

MAGNONS DRESSED WITH ELECTRIC CHARGES EXCITABLE BY A DYNAMICAL ELECTRIC FIELD
different coupling mecanisms (static/dynamical) …

Dynamical magneto electric effects

MAGNONS DRESSED WITH ELECTRIC CHARGES EXCITABLE BY A DYNAMICAL ELECTRIC FIELD

Pimenov et al:
possible evidence for electromagnons in multiferroic manganites
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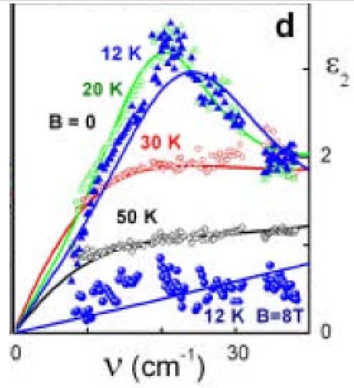
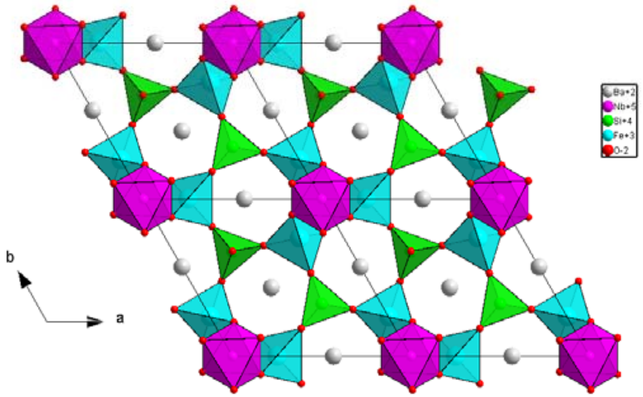
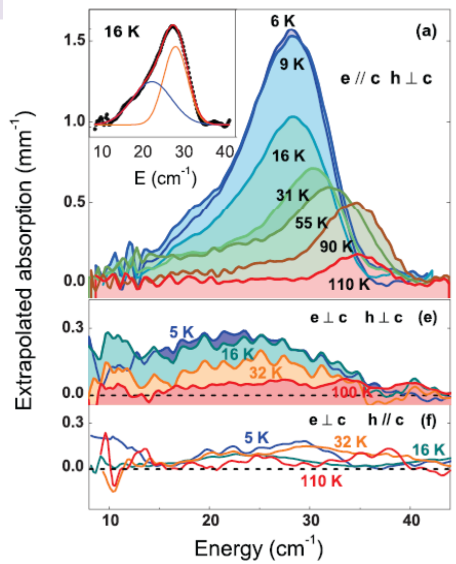


REVERSE EXCITATIONS : ATOMIC VIBRATIONS DRESSED WITH ELECTRICAL CURRENT HENCE MAGNETO ACTIVE

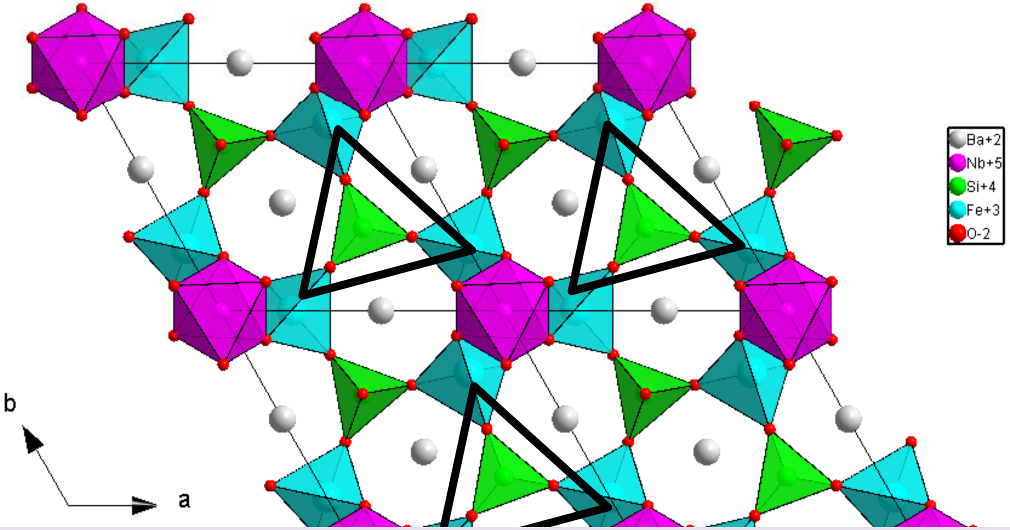
In the chiral compound $Ba_3NbFe_3Si_2O_{14}$

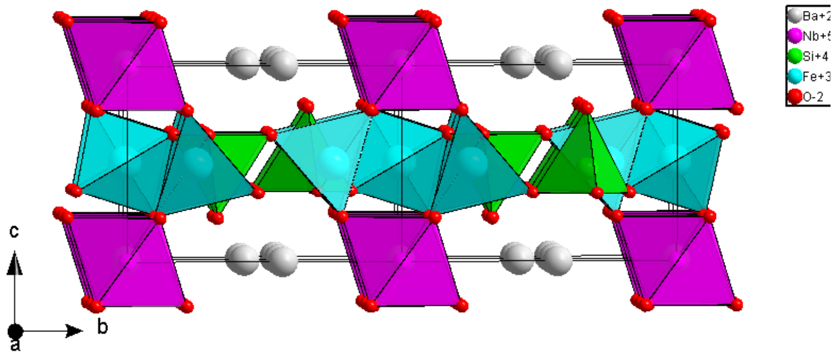




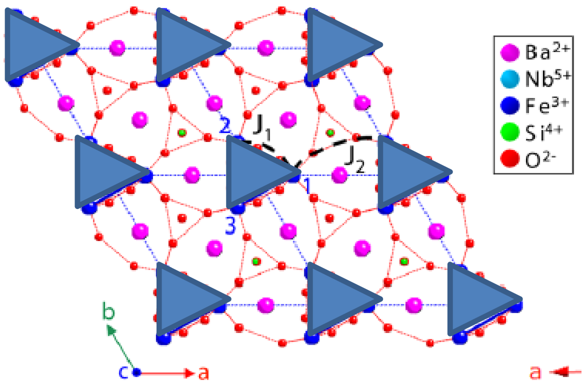
Fe langasites : structure

Non centro symetric Space group P321

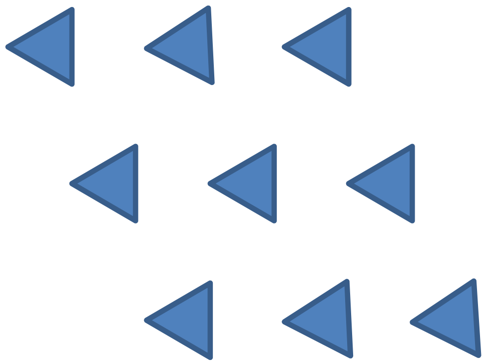




Triangular network of Fe^{3+} triangles (S = 5/2 L =0) stacked along *c* (D3 symetry)



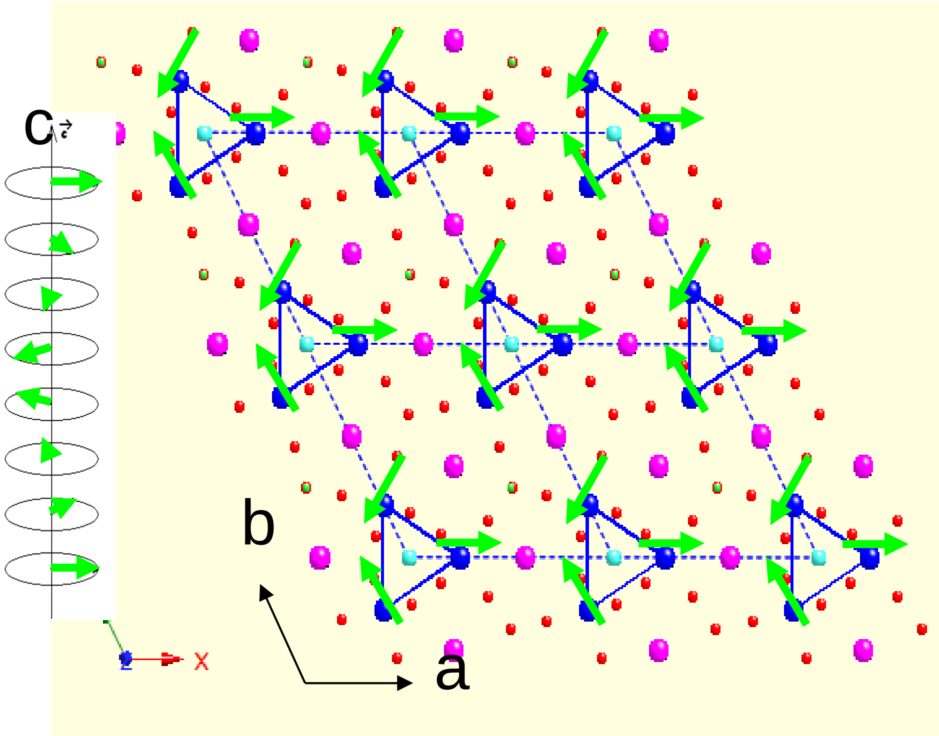
Crystal with **right** / left handed chirality



Fe langasites : magnetic structure below T_N=27 K

ILL (neutrons diffraction)

Helices propagating along c



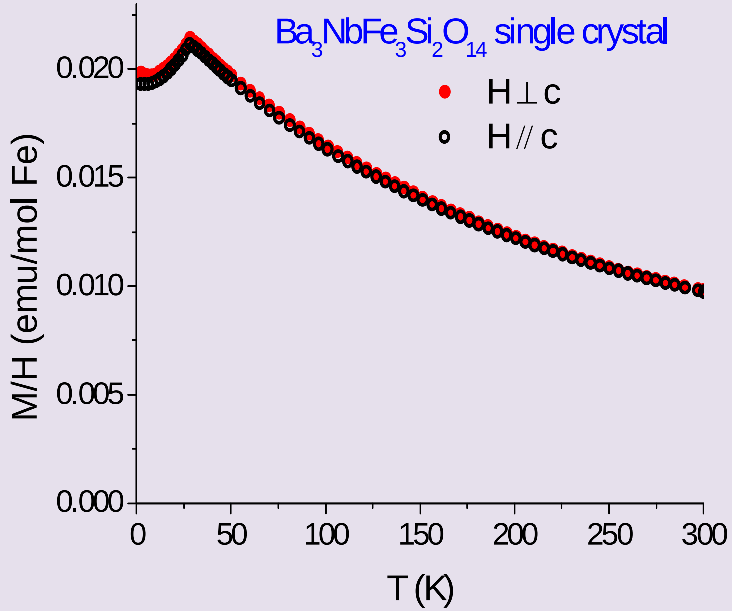
120° moments on triangles in the (a, b) plane
→ **magnetic frustration**

One single chirality within the triangles
One single chirality within the helices

K. Marty et al, PRL 2008

NbFe langasite

Magnetic properties

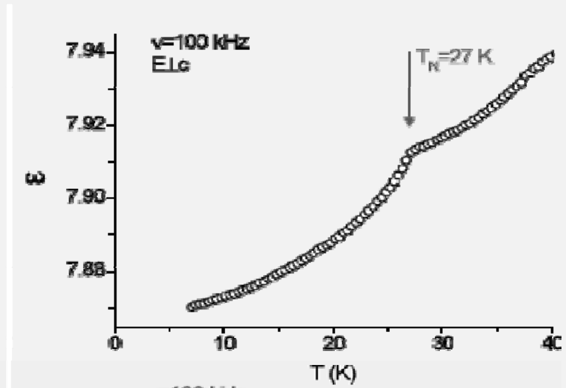


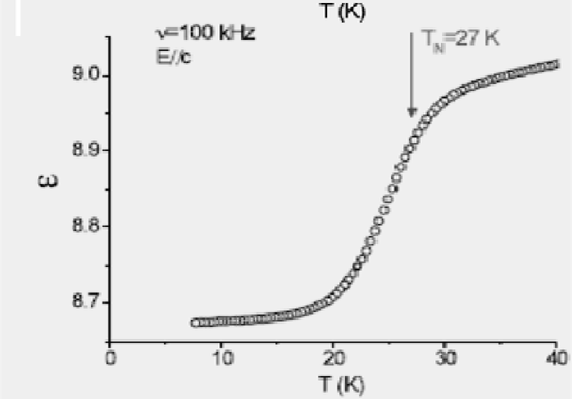
AFM order: $\mathbf{T_N = 27\ K}$

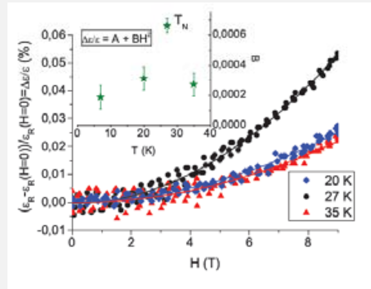
Curie-Weiss behavior :

θ_{CW} = **- 170 K**

Dielectric properties



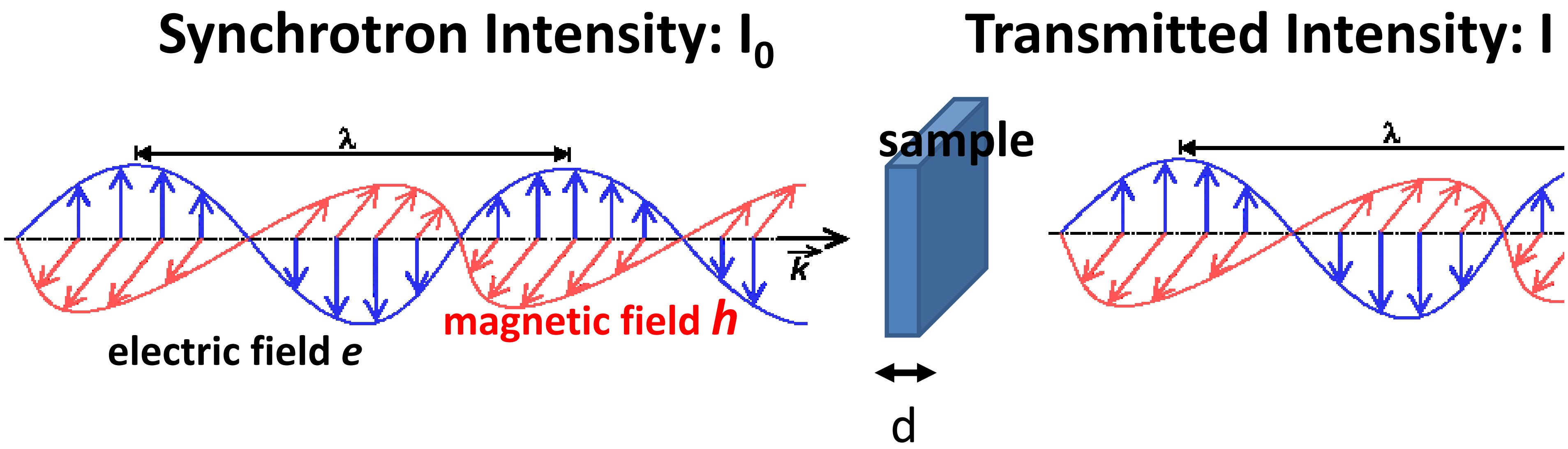




Weak satic magneto-electric coupling at TN

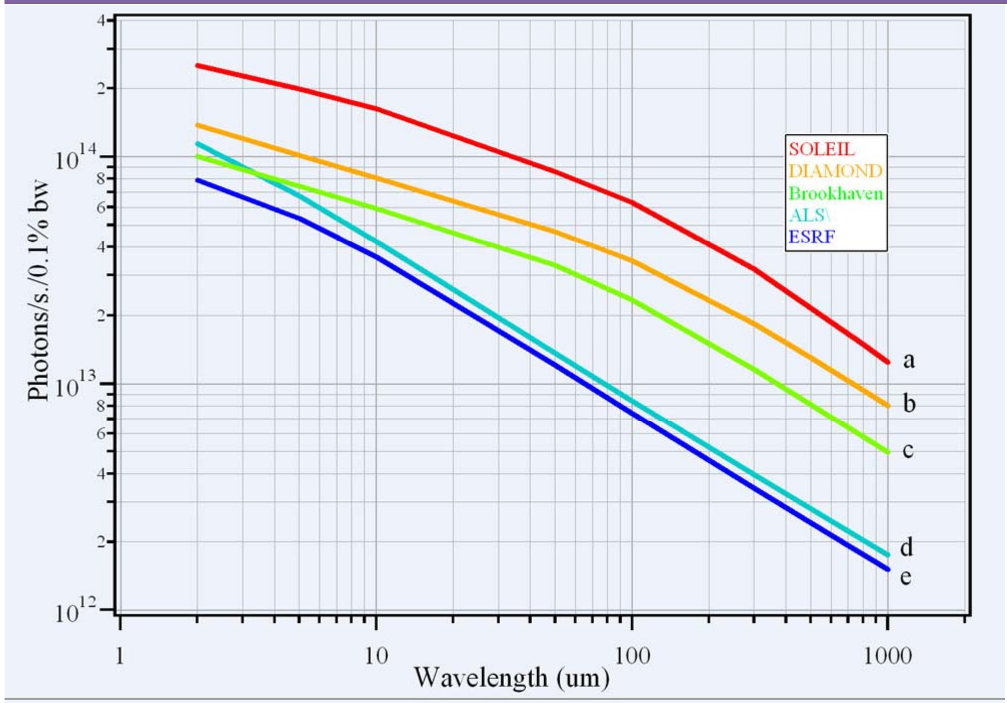
S. deBrion

Fe langasites : Dynamical properties in the THz range on the AILES beamline at SOLEIL



Transmission : T = I / I0 ≈ exp $(-\alpha d)$ Absorbance : Abs= - Ln (T) $\approx \alpha d$	White beam + Fourier Transform spectrometer : Abs (frequency) 8 cm^{-1} < frequency < 100 cm^{-1}

THz measurements



100 cm^{-1} 10 cm^{-1}

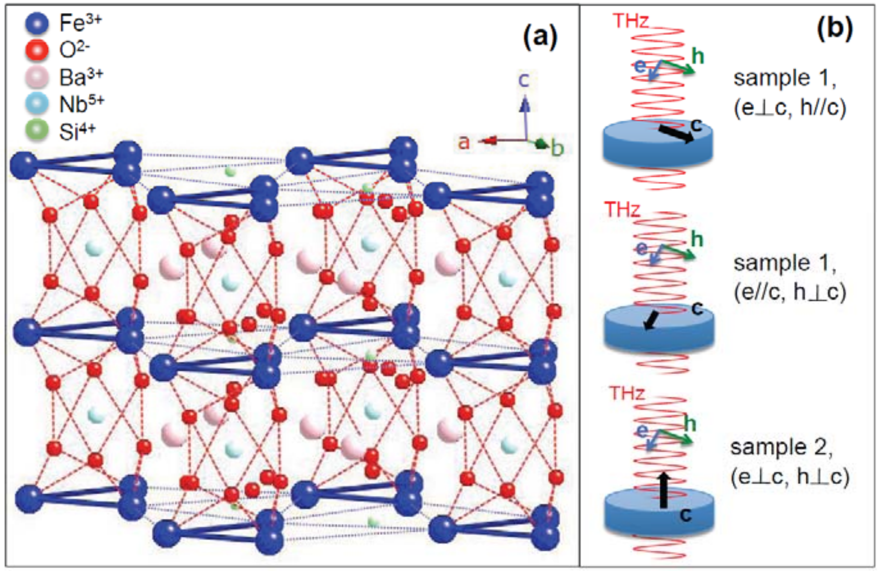
1 meV = 8 cm^{-1} = 0.24 THz

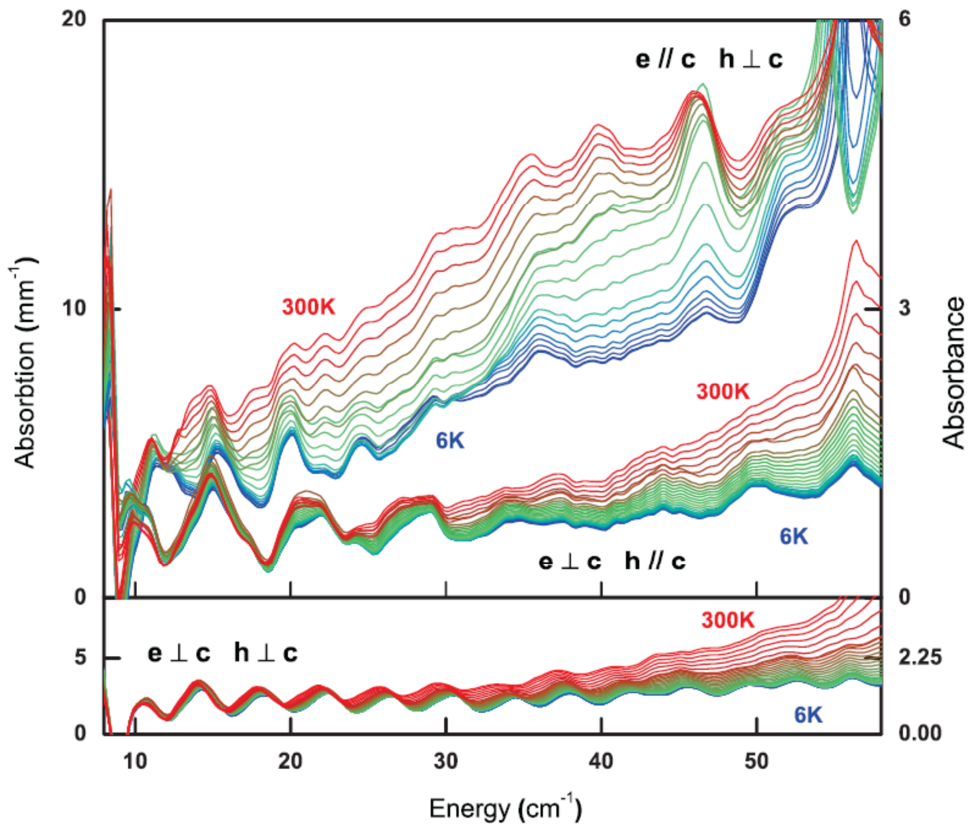
The AILES beam line at SOLEIL + IFS 125 FT spectrometer + He pumped bolometer + cryostat .

An intense broadband THz source, 8 – 80 cm^{-1} , res 0.5 cm^{-1}

98 % vertical Polarisation

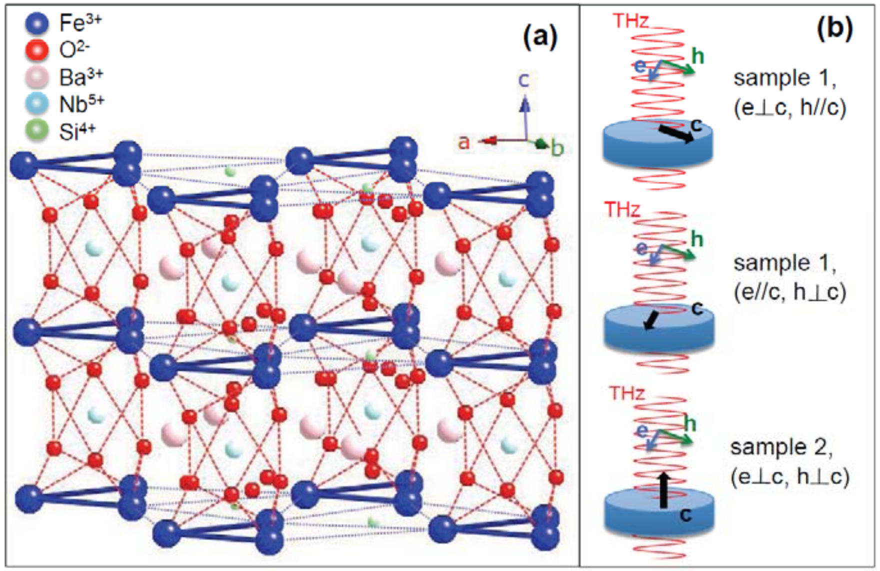
Fe langasite : THz absorbtion

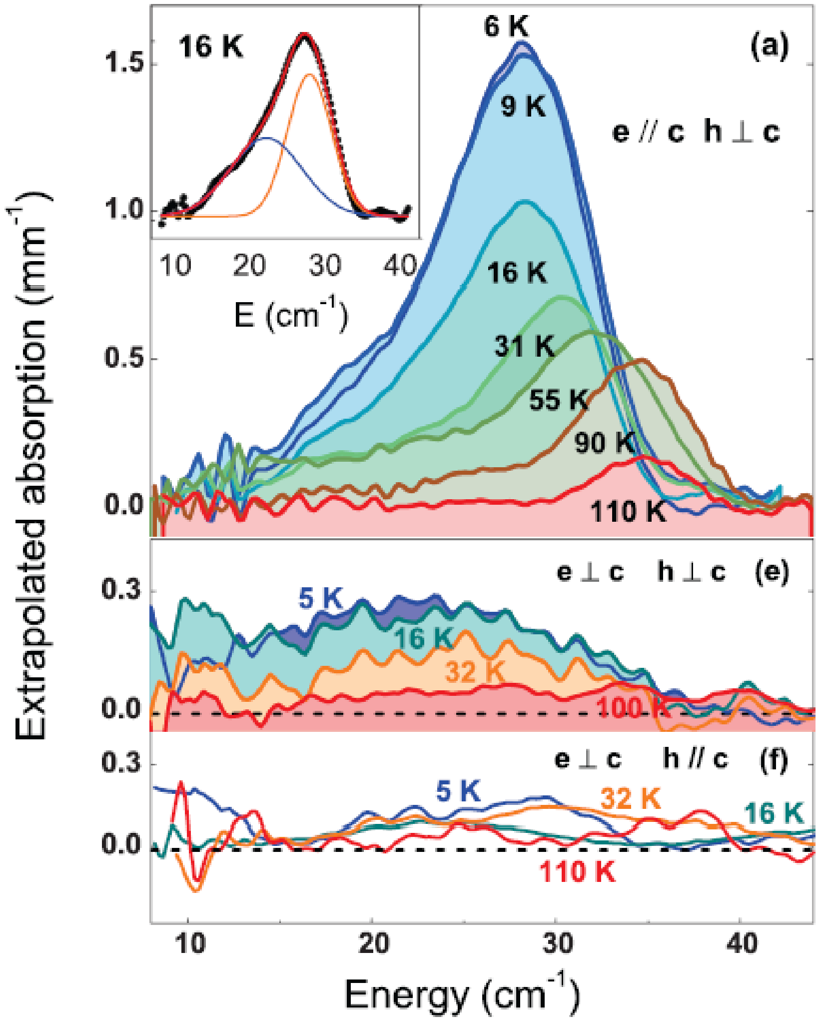




Larger overall absorption for e // c
that always increases with temperature
OK with low frequency dielectric response

THz relative absorbtion with 6 μm Si beam splitter :





Two excitations at low T:

i) Centered at 23 cm^{-1} width 10 cm^{-1}

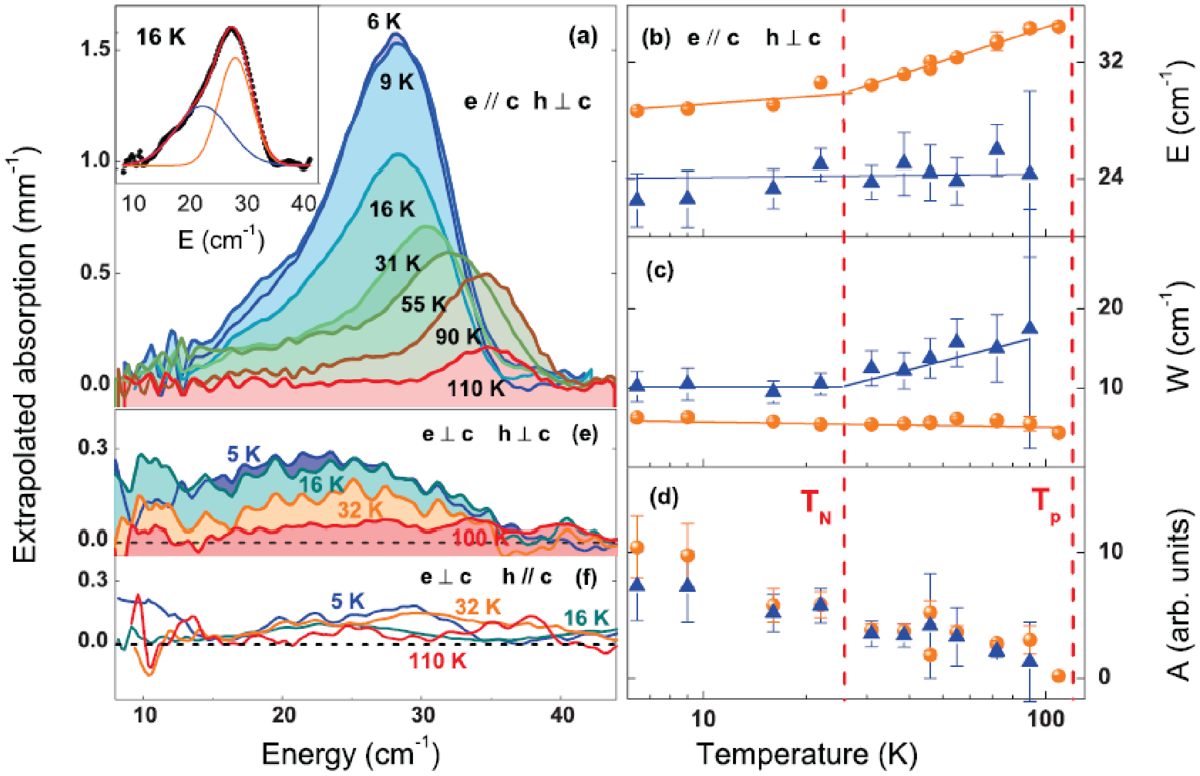
ii) Centered at 29 cm^{-1} width 5 cm^{-1}

Different selection rules:

i) h ⊥ c

ii) e // c

THz AILES measurements with 6 μm Si beam splitter



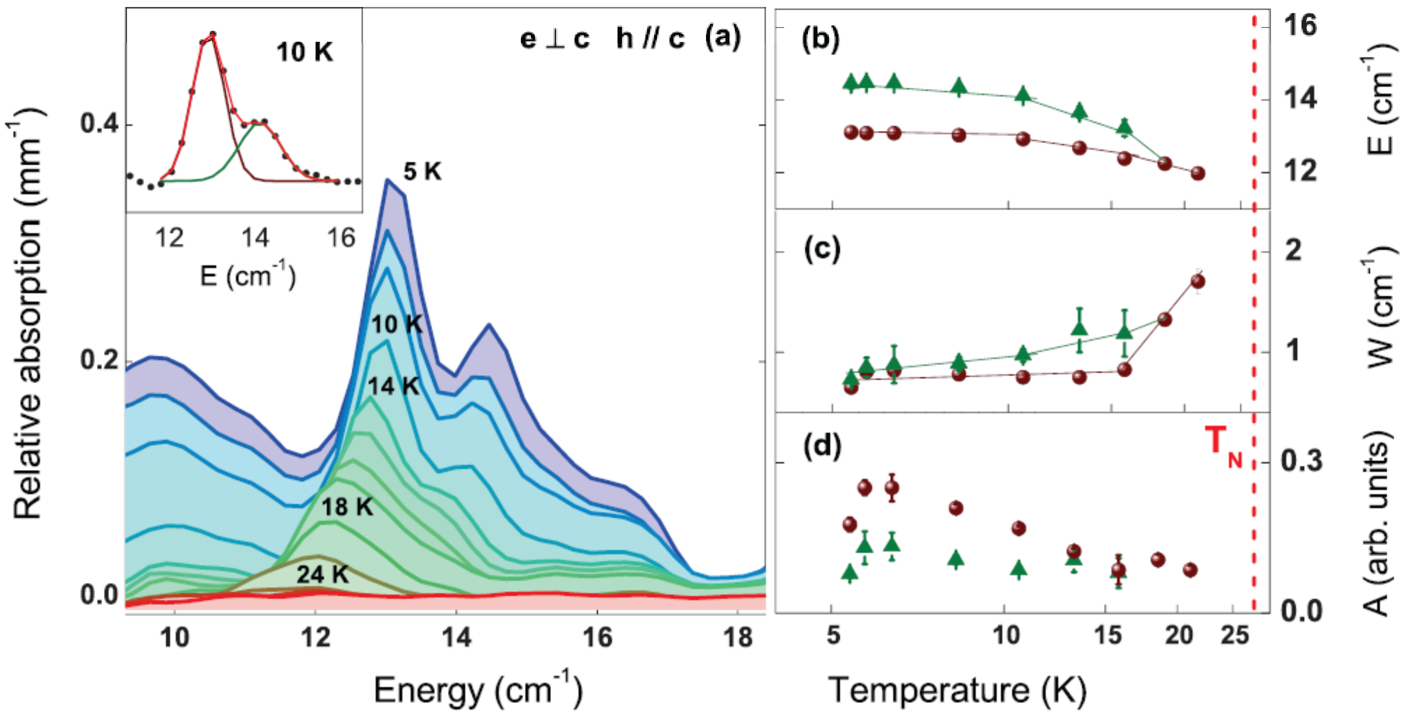
Two excitations at low T: at 23 cm^{-1} and 29 cm^{-1}, width 10 cm^{-1} and 5 cm^{-1}

Same spectral weight: persist up to T_p ≈ 110K ≈ 4T_N

Different temperature dependence for their position and line width

Different selection rules: h ⊥ c, e // c

THz AILES measurements with 125 μm Si beam splitter



Third excitation at low T :

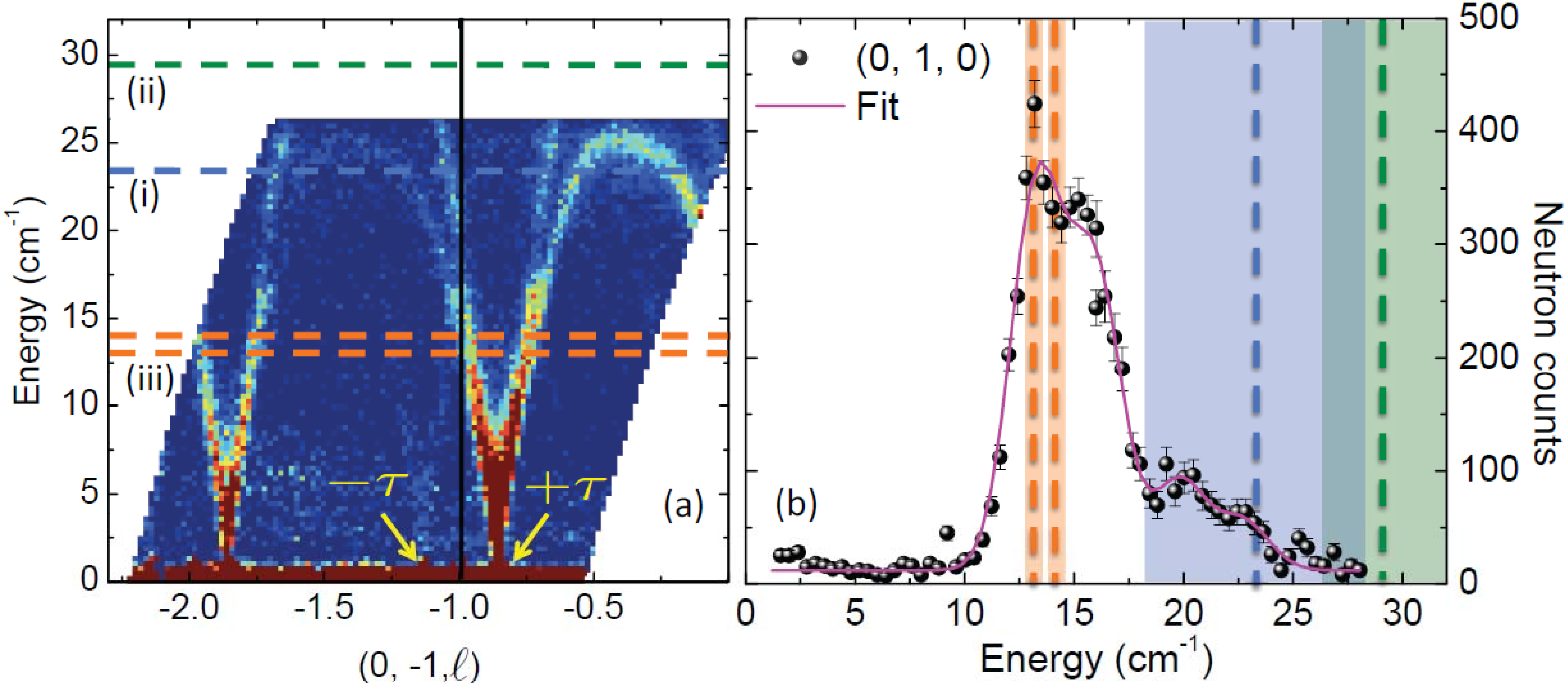
two Gaussian contributions at 12.9 and 14.1 cm^{-1}, same width 0.8 cm^{-1}

broaden and disappears at T_N

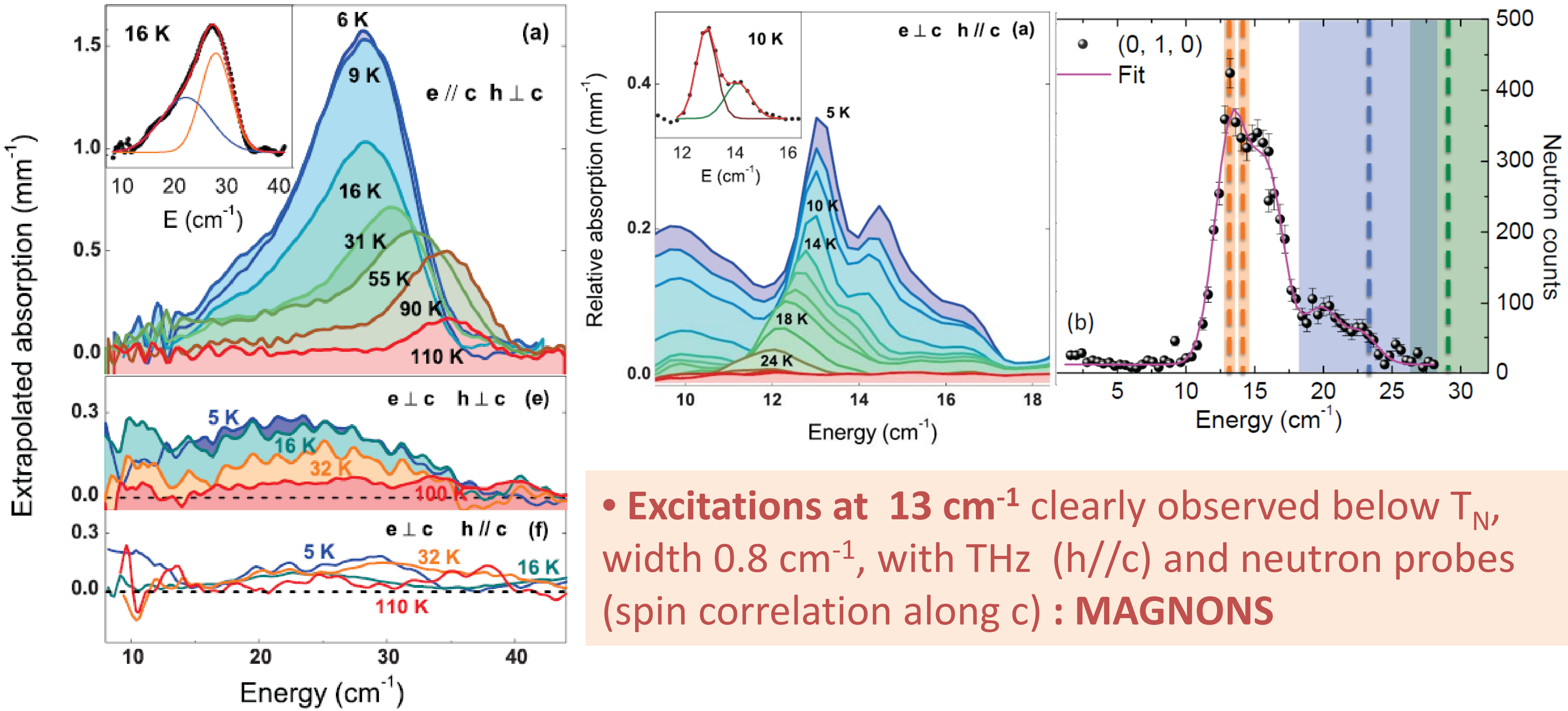
excited for h //c : MAGNONS, see neutron data.

THz / neutron measurements

THz / neutron : same magnetic excitations at the Brillouin zone center (0,0,0) equivalent to (0,-1,-1) or (0,1, 0)



THz excitations in $Ba_3NbFe_3Si_2O_{14}$



• **Excitations at 13 cm^{-1}** clearly observed below T_N, width 0.8 cm^{-1}, with THz (h//c) and neutron probes (spin correlation along c) **: MAGNONS**

- **excitation at 23 and 29 cm^{-1}**, width 10 and 5 cm^{-1},
- **observed up to T_p ≈4 T_N, with THz probe only: PHONONS**
  appearing below Tp ≈ 110 K: structural changes.
- **With different selection rules : one excited for e // c, one for h ⊥ c !!!**

origin of the THz phonon modes

D3 symetry:

rotations R_z about the z-axis (c axis) **may produce an electric polarization along c**

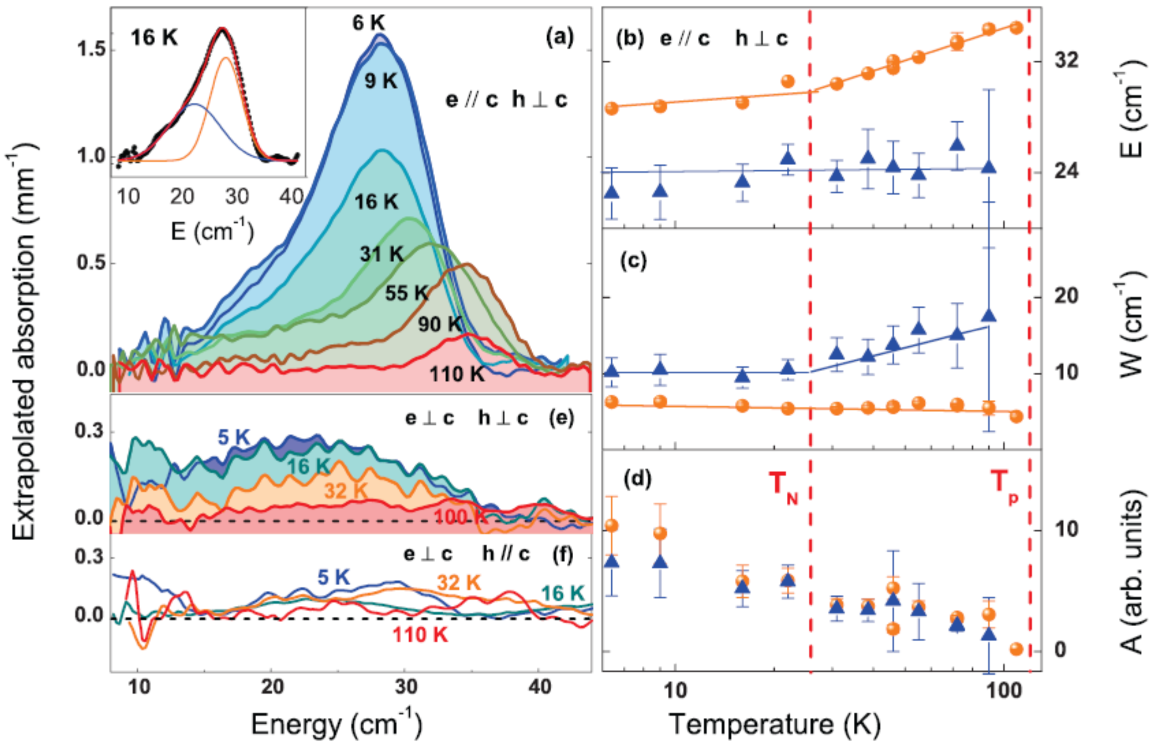
$$\delta P_z = (\alpha/\omega_p^2) R_z$$

α: coupling constant

ω_p : frequency of the phonon associated to P_z

That can **be excited by e // c :**

$\mathcal{L}_e = -\,\delta P_z\, e_z$



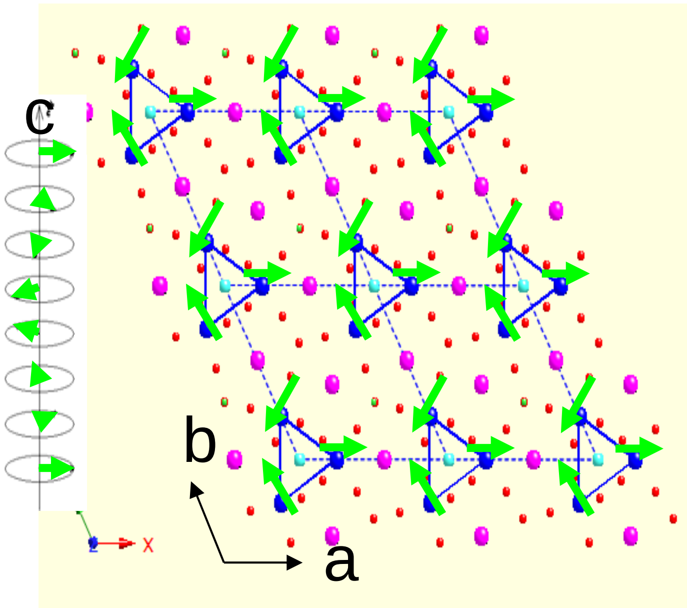
Coupling to the magnetic field: (Dzialoshinskii and Mills 2009)

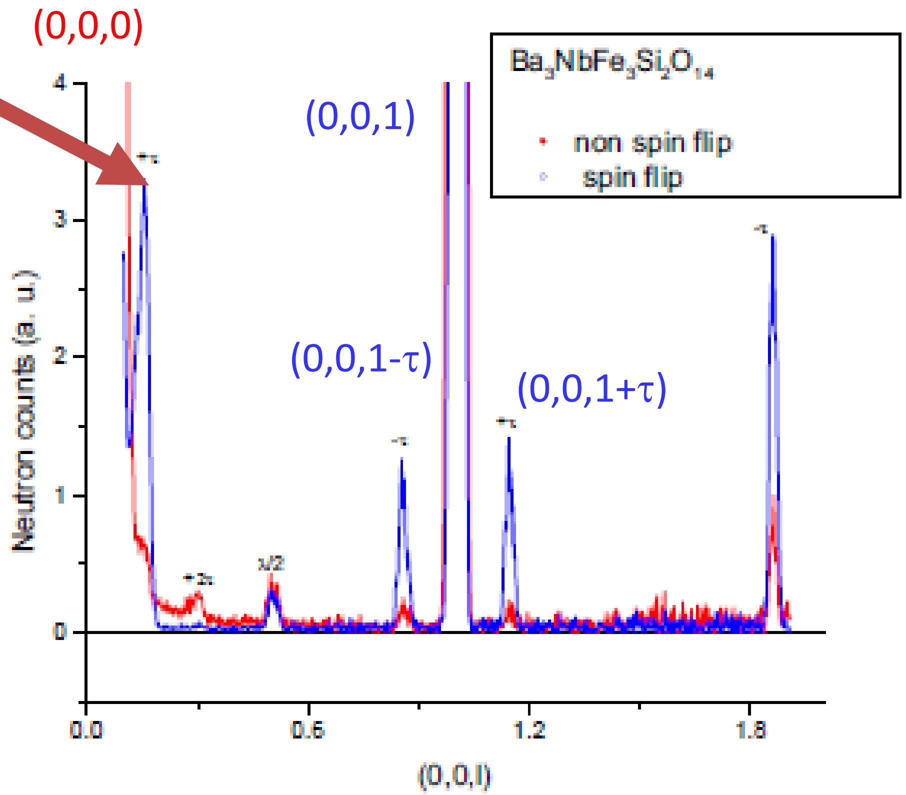
$\mathcal{L}_H = g\mathbf{h} \cdot (\mathbf{P} \times d\mathbf{P}/dt) = -g\,(\alpha/\omega_p^2)(dh_\perp/dt \times P_\perp)_z R_z$

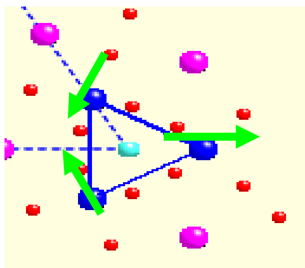
$P_\perp$ non zero: lost of D3 symetry → **helicoidal polarisation.**

Hints for the lost of D3 symmetry

Forbidden in perfect triangular arrangement of the spins



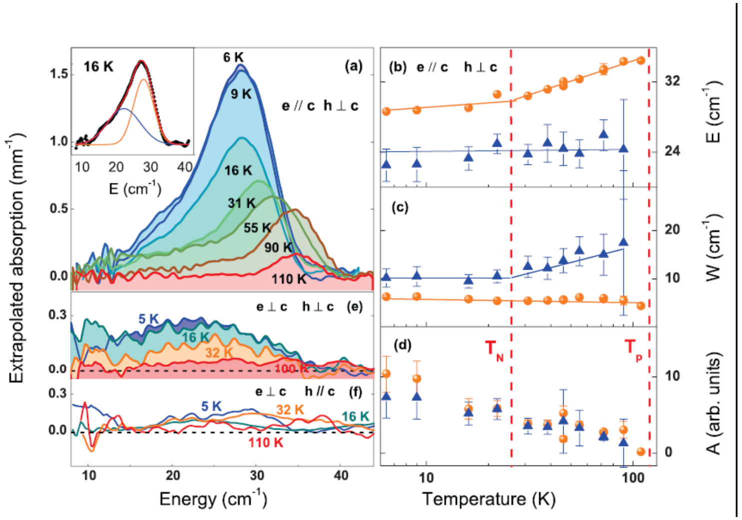




Lost of 3 fold axis

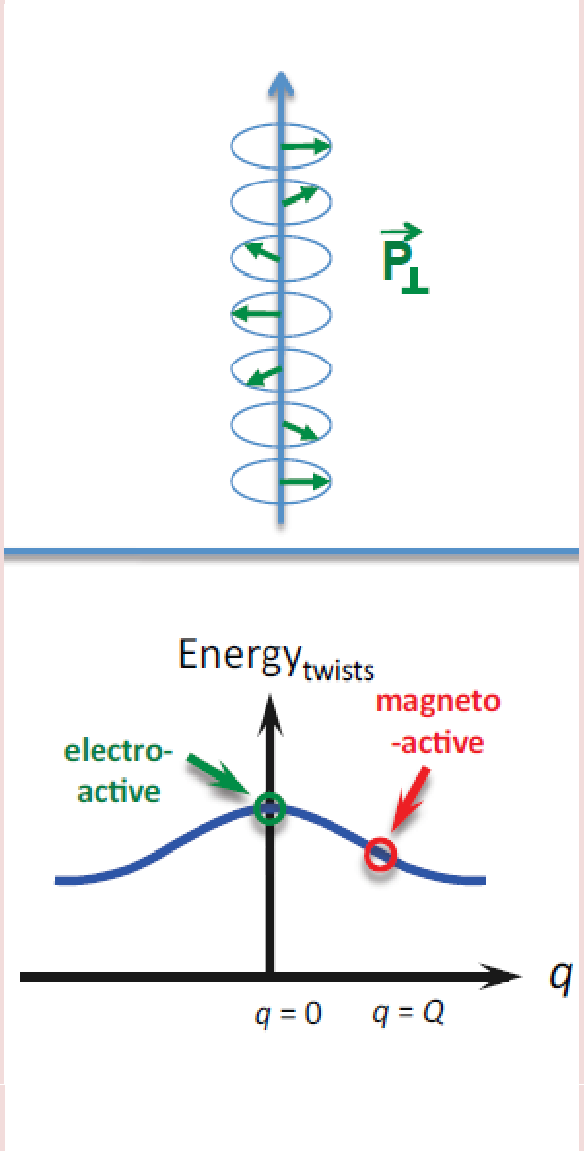
D3 → C2

Dispersion law and selection rules



- excitation at 23 cm^{-1} excited for e //c

•Excitation at 29 cm^{-1}, excited for for h ⊥ c



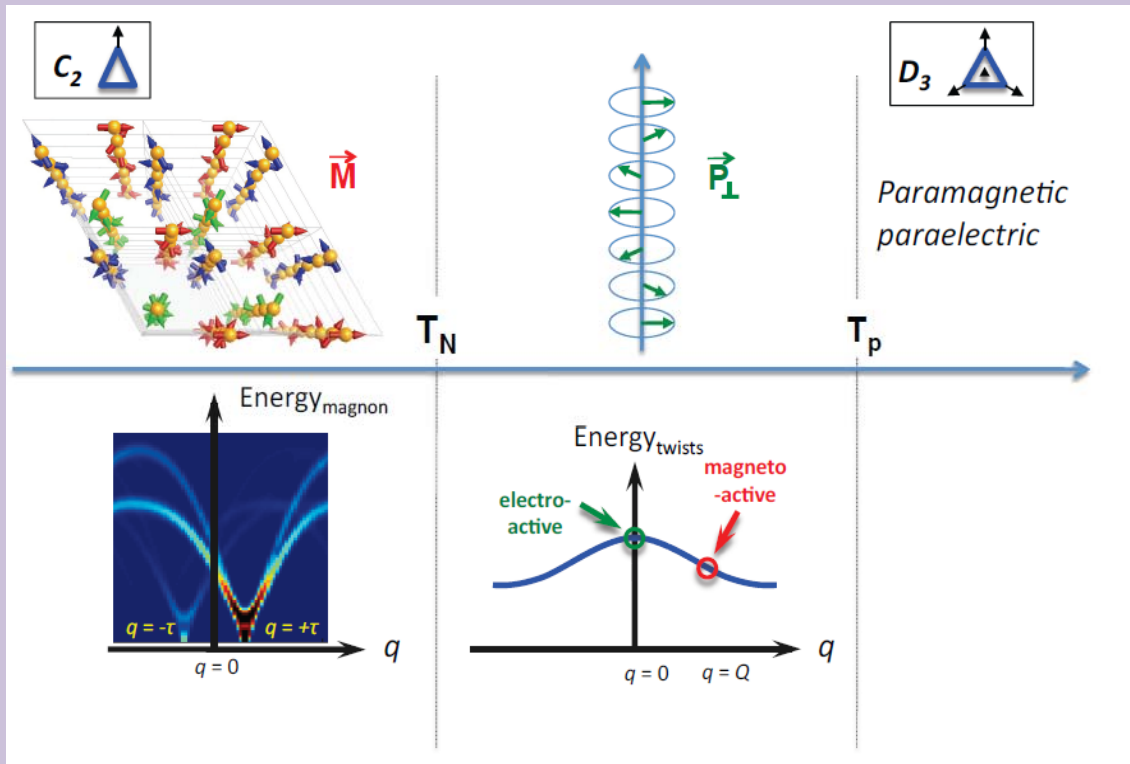
suggested phase diagram

Ginzburg Landau free energy:

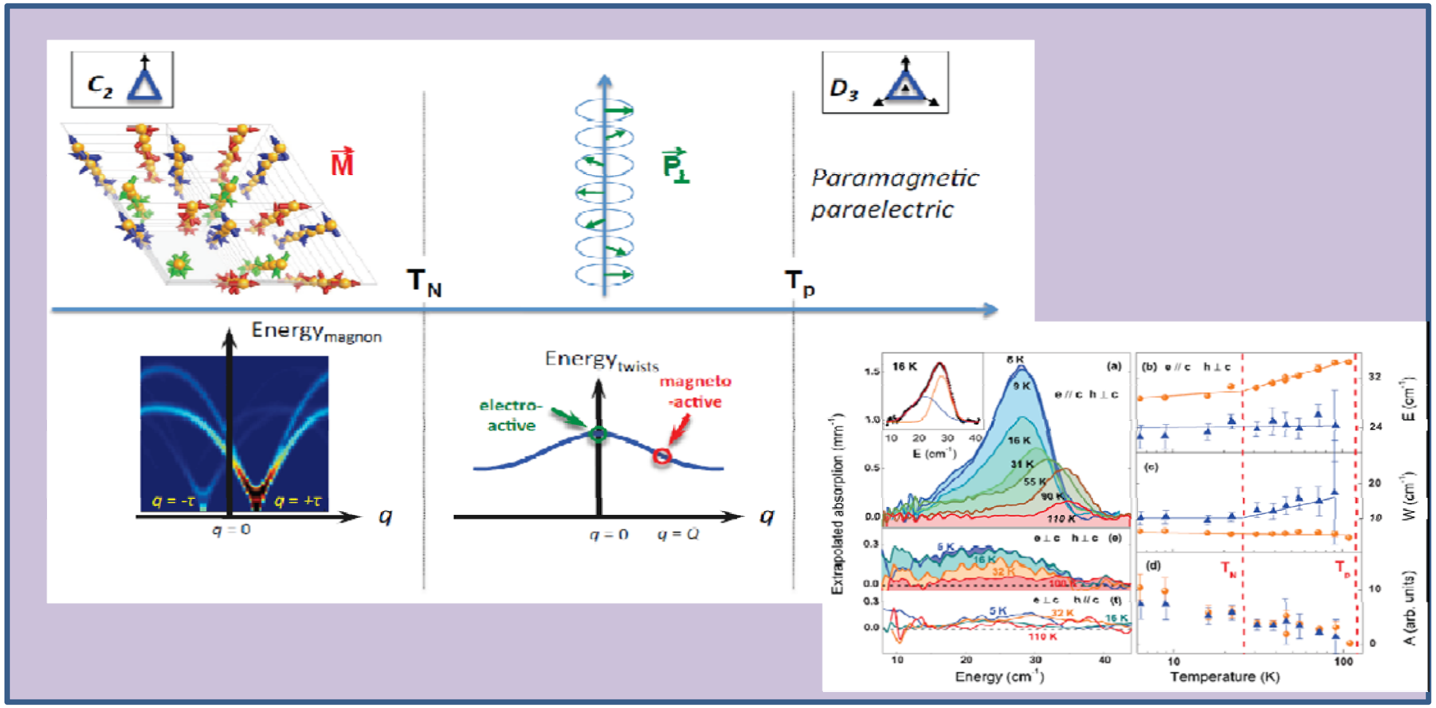
$$F = \frac{a}{2}\rho^2 + \gamma\rho^3\cos 3\theta + \frac{b}{4}\rho^4 - \lambda\rho^2(\partial_z\theta) + \frac{c}{2}[(\nabla\rho)^2 + \rho^2(\nabla\theta)^2].$$

$$P_\perp \quad [\omega^2_{P_z}(P_\perp) = \omega^2_{P_z}(0) - \beta P_\perp^2 + \ldots]$$

$$(P_x, P_y) = (\rho\cos\theta, \rho\sin\theta)$$

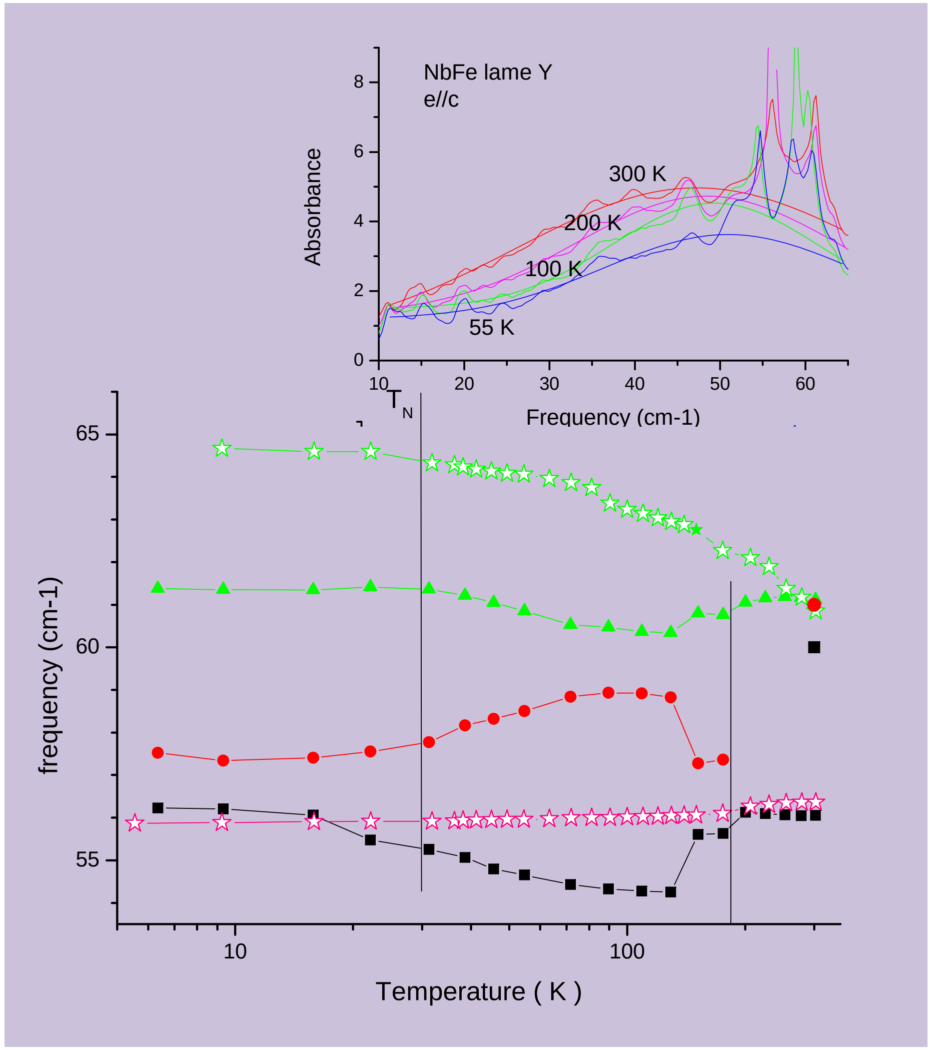


CONCLUSION



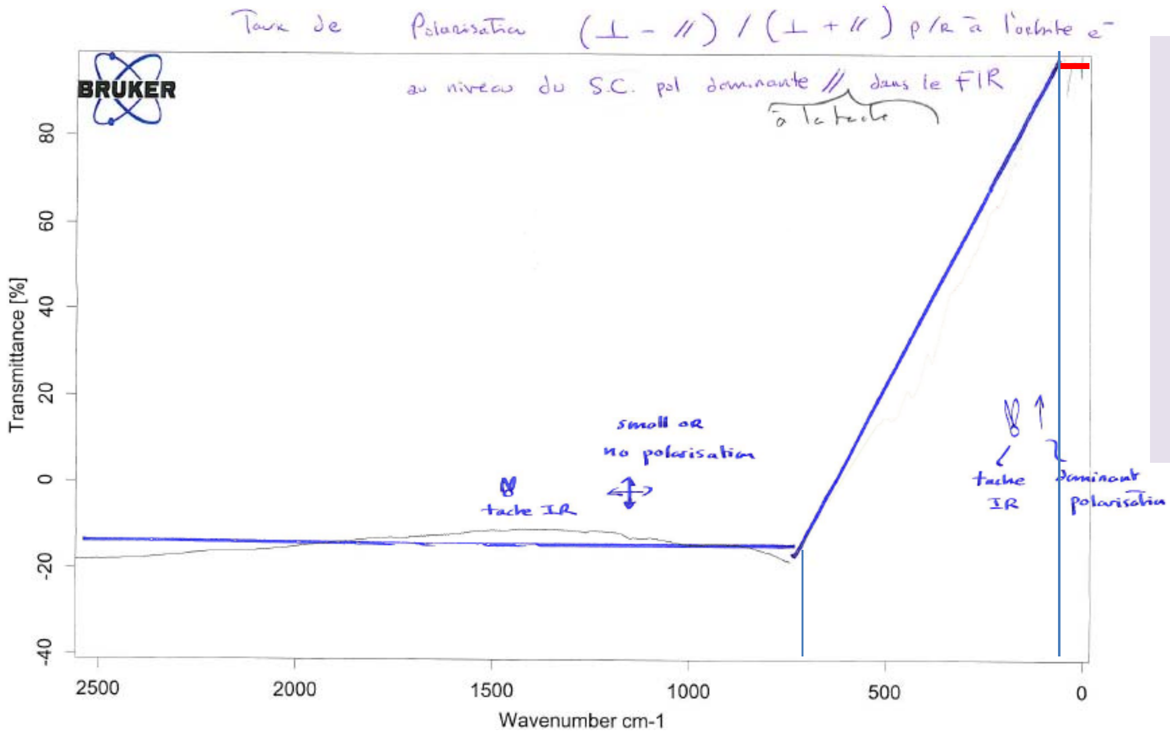
New dual excitation in the chirl langaiste: rotation mode of the lattice whose magneto-electric activity reveals a transition towards a state of helical polarisation

Hints for the lost of D3 symmetry



THz measurements : two localised phonon modes split below ≈200 K

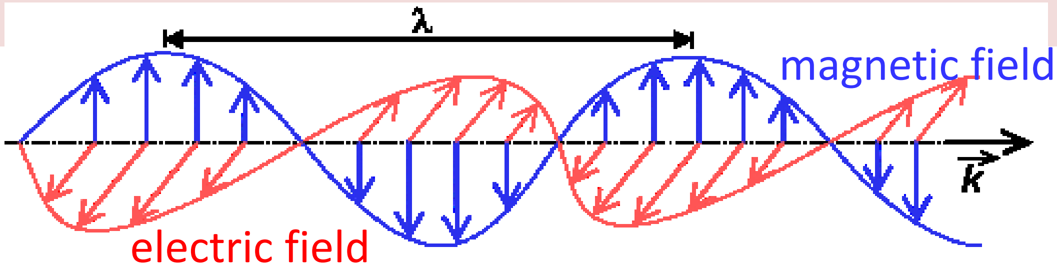
Manip SOLEIL : polarisation du faisceau

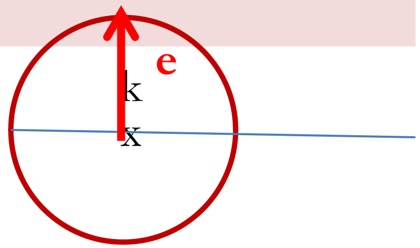


Polarisation:

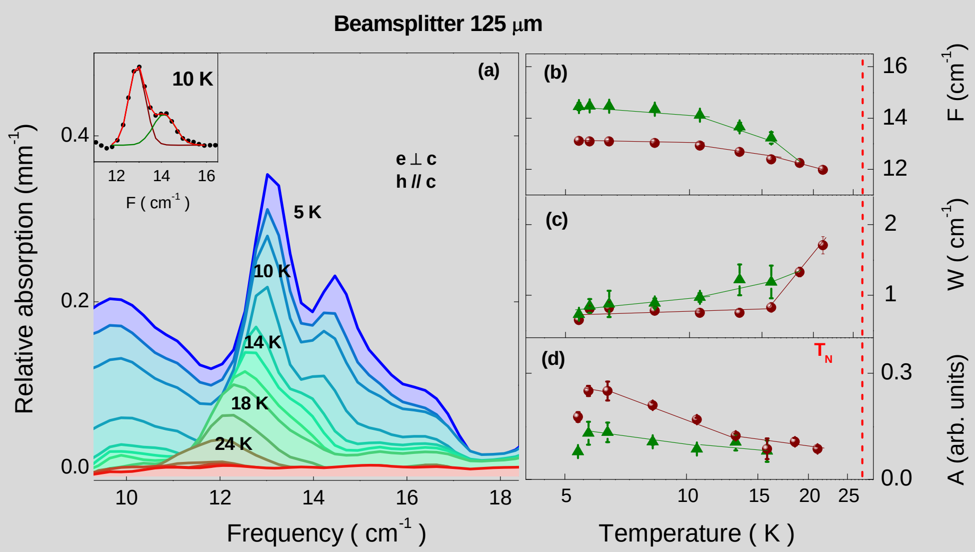
- **verticale (95 – 98 %) dans le THz**
- **faible (15% horizontale) au dessus de 700 cm^{-1}**
- **décroit linéairement entre 100 et 700 cm^{-1} de 2% tous les 10 cm^{-1} environ**

Lumière polarisée à 98 % dans le THz, champ électrique excatement vertical, perpendiculaire à l'orbite des électrons dans l'anneau:

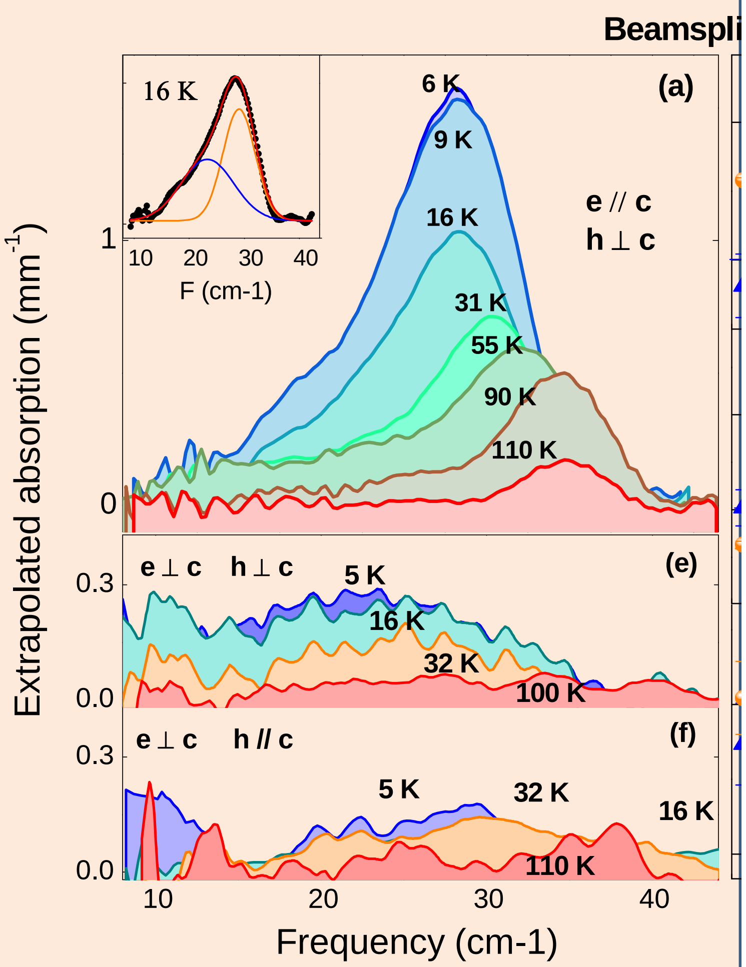




THz absolute / relative absorbtion :



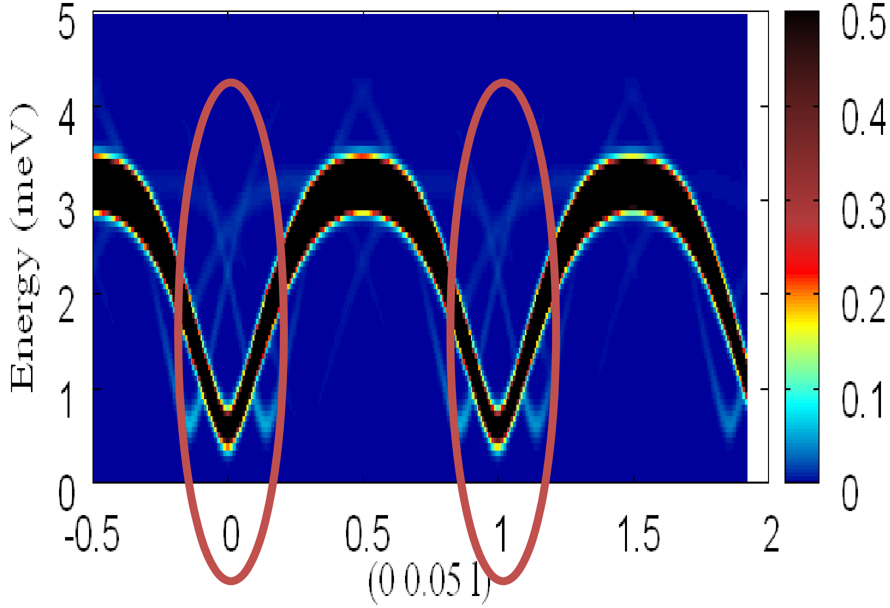
Abs (T) – Abs (45 K)



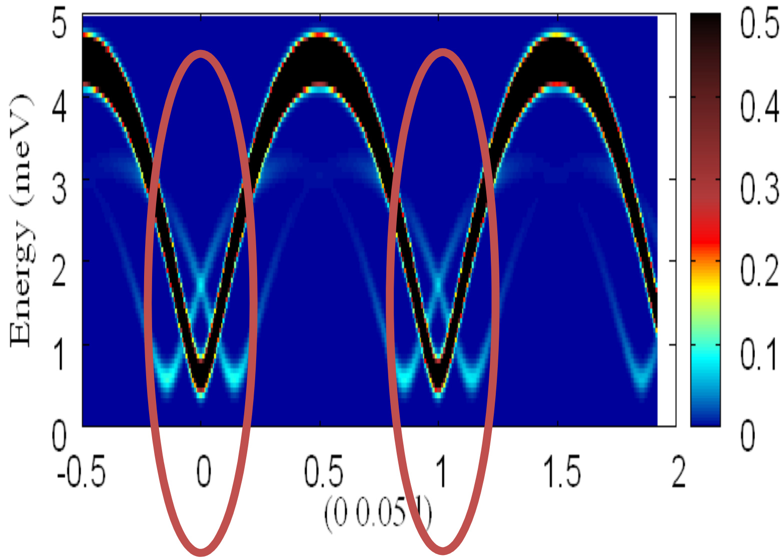
Abs (T0) – Abs (T) + FFT band block filter→ extrapolated Abs(T0)

Fe langasites : spin wave modeling

Magnetic model: spin wave theory using 5 magnetic interactions



Spin correlations along a or b

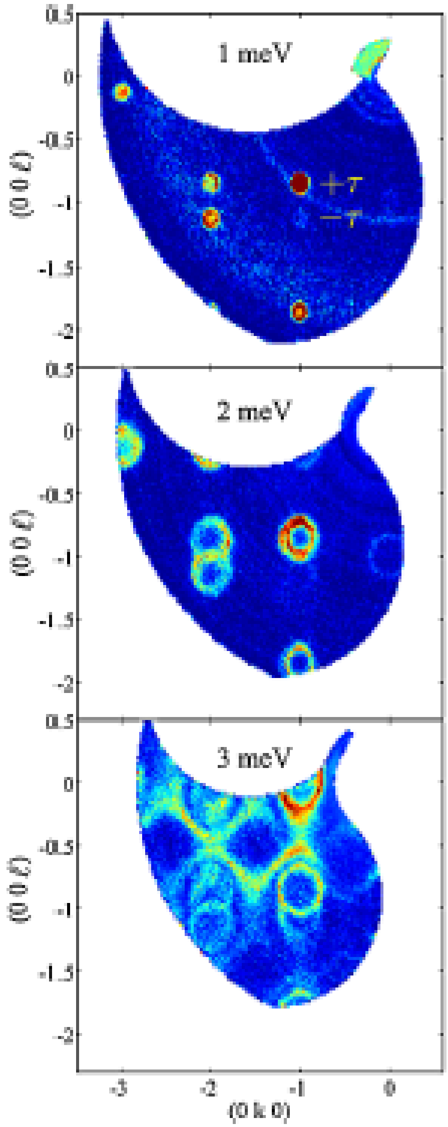


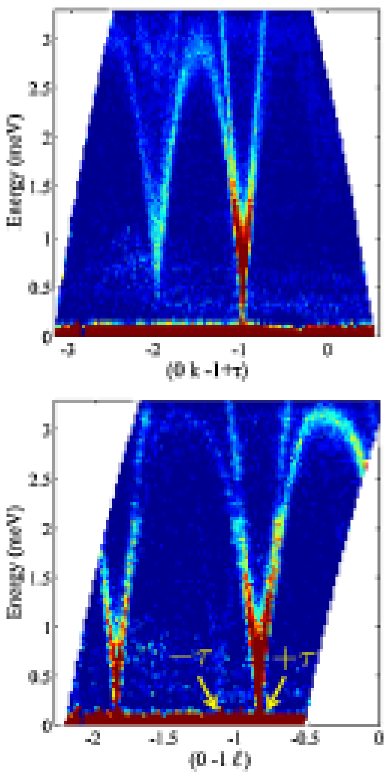
Spin correlations along c

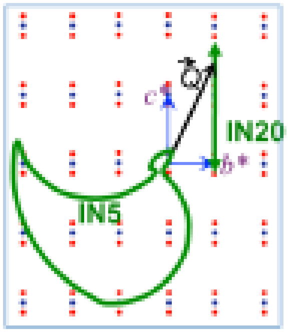
expected magnons at zone center : from 2.1 to 2. 8 meV , from 17 cm-1 to 23 cm-1

expected magnon at zone center : 1.7 meV = 13.5 cm-1

NbFe langasite : inelastic neutron measurements





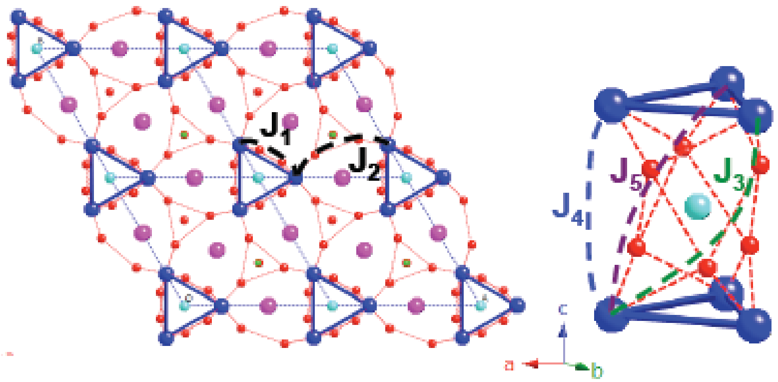


at 2 K :

2 magnetic branches, symetric at $+\tau, -\tau$ with $\tau = 1/7$

spectrum extended up to 3.5meV and 5 meV (28 and 40 cm-1)

Magnetic model: spin wave theory using 5 magnetic interactions

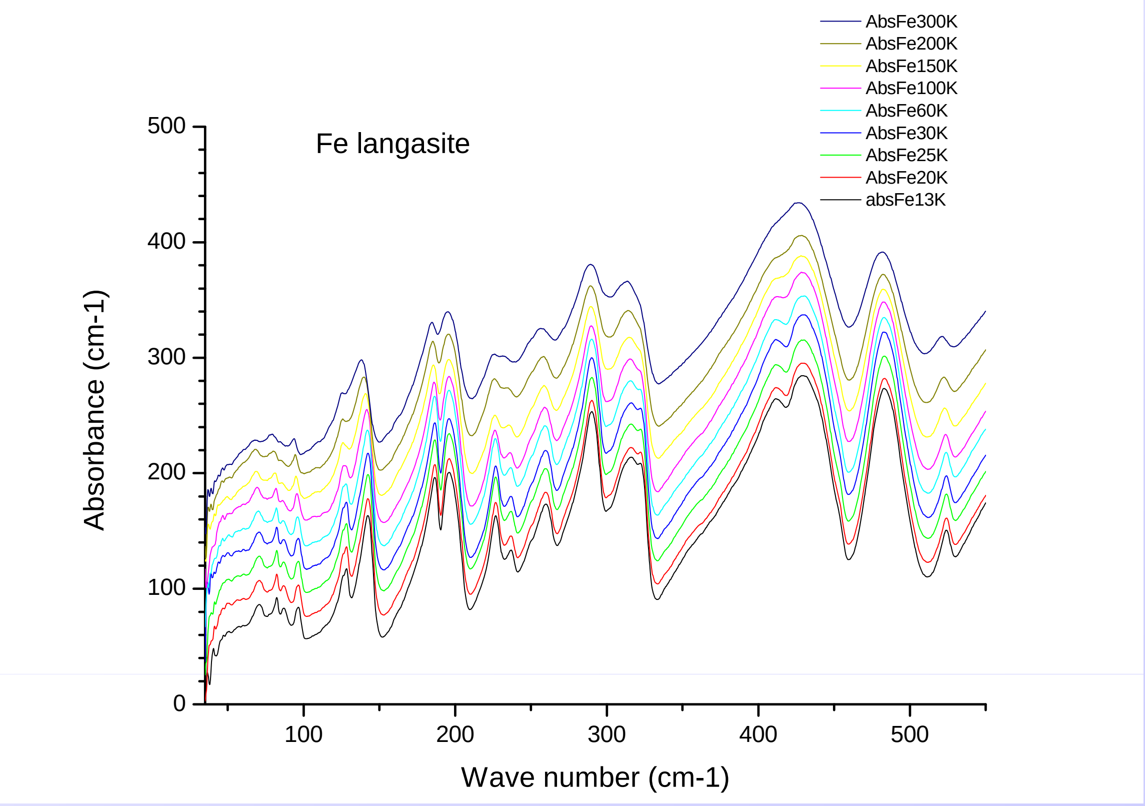


M. Loire et al, PRL 2011

The langasite $Ba_3NbFe_3Si_2O_{14}$

Dynamical properties

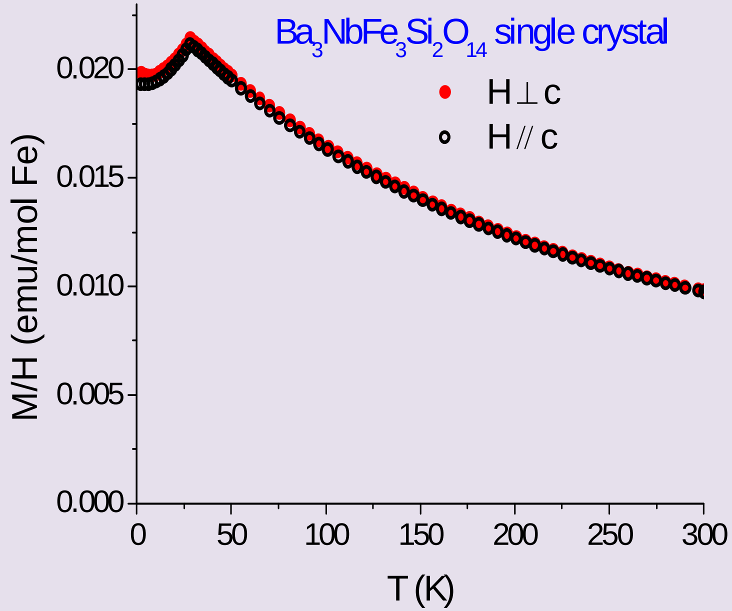
SOLEIL FIR measurements



Multiple phononic bands …

NbFe langasite

Magnetic properties

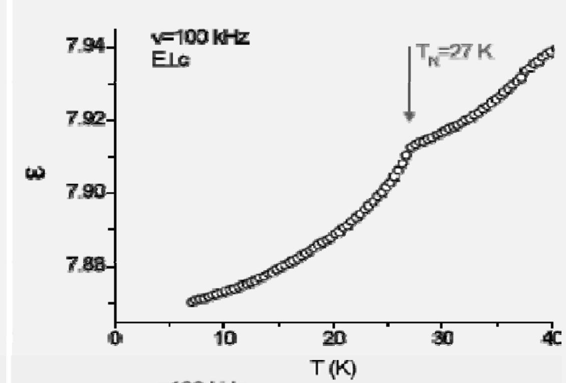


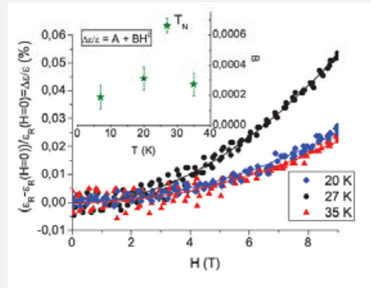
AFM order: $\mathbf{T_N = 27\ K}$

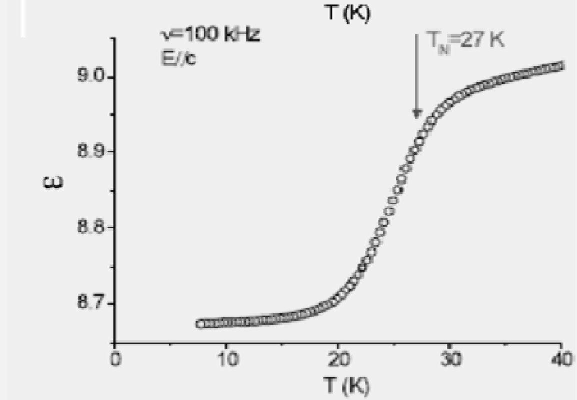
Curie-Weiss behavior :

θ_{CW} = **- 170 K**

Dielectric properties







Weak satic magneto-electric coupling at TN

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