POLITECNICO DI MILANO

ID 20 magnetic scattering

ESRF

Frustrated magnetism: the $Ca_3Co_2O_6$ paradigm

C. Mazzoli,
(ID20 Magnetic Scattering Beamline, ESRF)
Inelastic X-Ray Scattering group, Dip.to Fisica, Politecnico di Milano

NG-SCES 2011, Santiago de Compostela

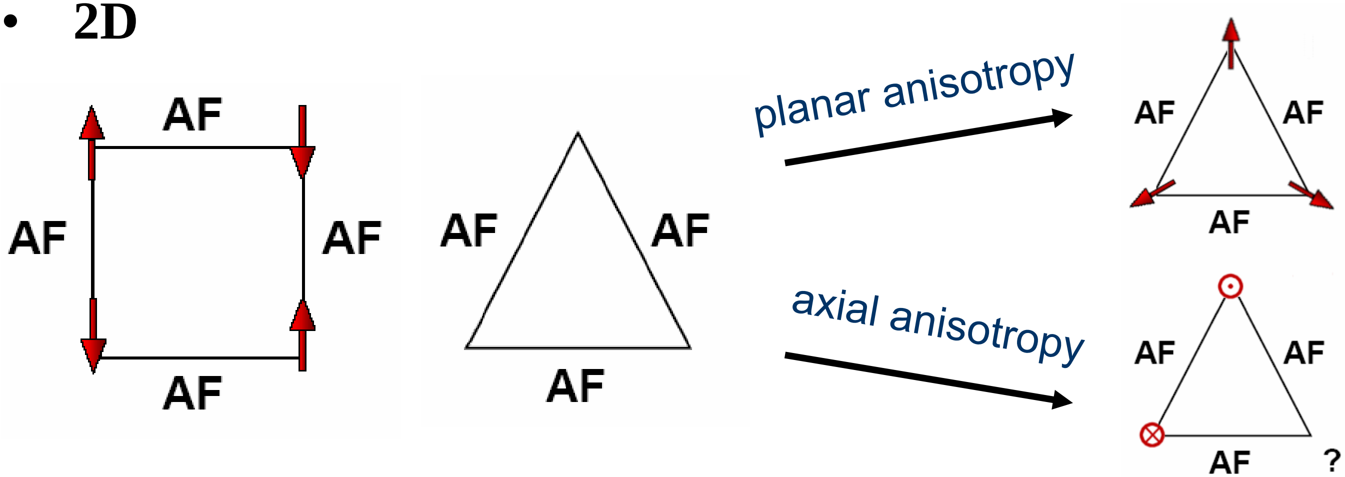
- Spin frustration
- $Ca_3Co_2O_6$
- What was known
- Our contributions (RXD, NPD and NMR) and current understanding of the system
- Conclusions

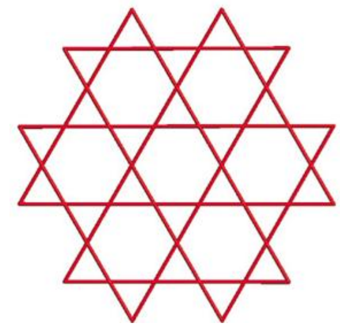
Collaborators

- S. Agrestini
  (Phys. Dep., Univ. of Warwick, Coventry, UK
  Lab. CRISMAT, CNRS UMR 6508, Caen, F)
  Max Planck Inst. for Chemical Physics of Solids, Dresden, D
- A. Bombardi
  Diamond Ltd, RAL, Didcot, UK
- L. Chapon
  ISIS facility, RAL, Didcot, UK
- G. Allodi and R. De Renzi
  Dip.to di Fisica, Univ. di Parma, Parma, Italy
- M. Lees and O. Petrenko
  Phys. Dep., Univ. of Warwick, Coventry, UK
- F. de Bergevin
  ESRF, Grenoble, F

Spin frustration

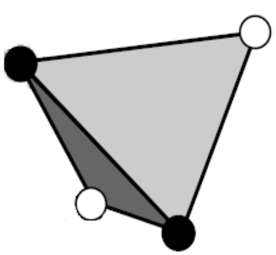
- **2D**

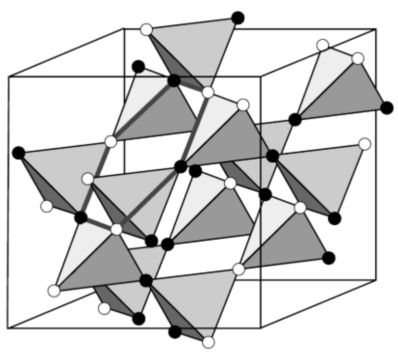


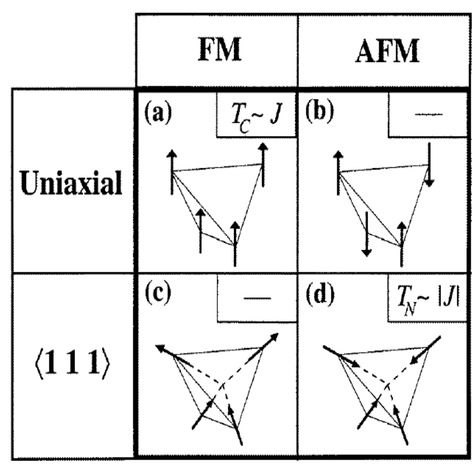


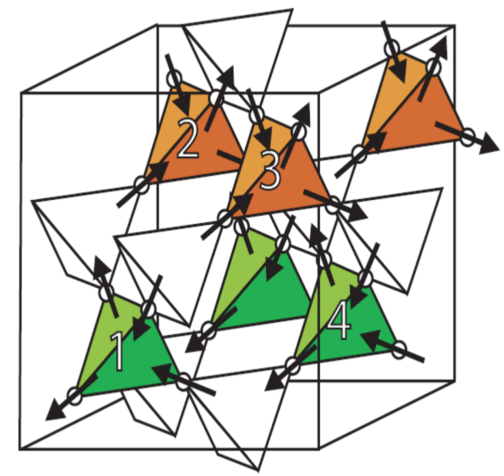
Nat. Mat.
1 (2002) 91–92

- **3D**



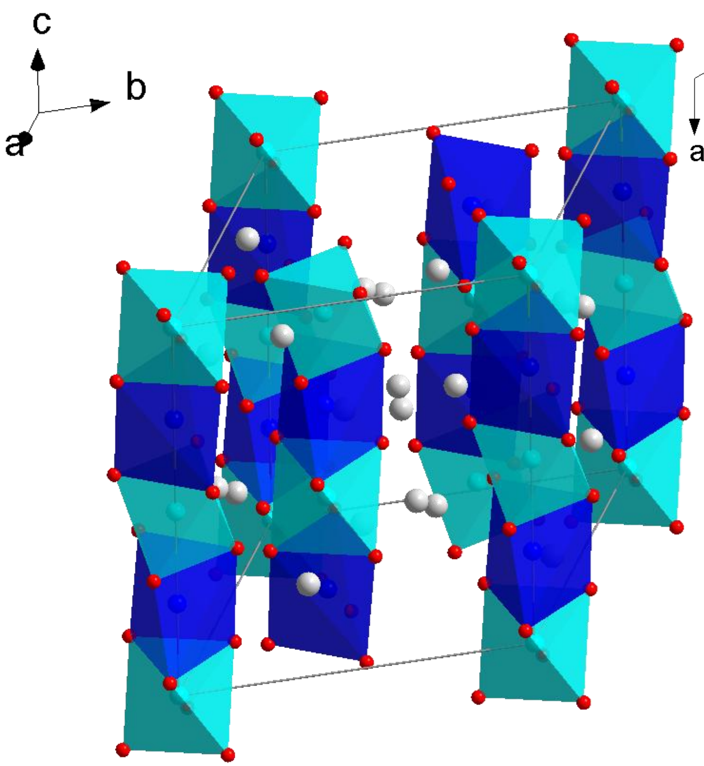


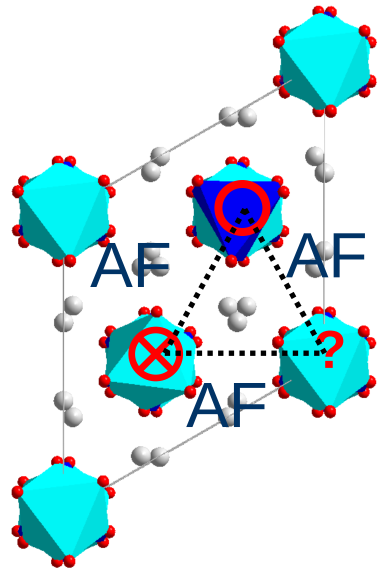


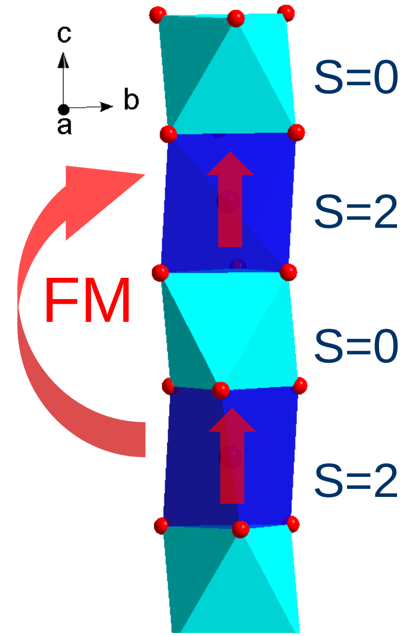


Science 294 (2001) 1495, JPCondMat 10 (1998) L215

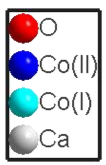
$Ca_3Co_2O_6$ structure







CF at trigonal prism: strong **magneto-crystalline uniaxial anisotropy**



} Co^{3+} ($3d^6$)

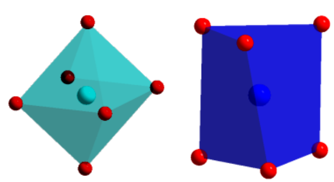
Burnus PRB74 (2006) 245111

R-3c

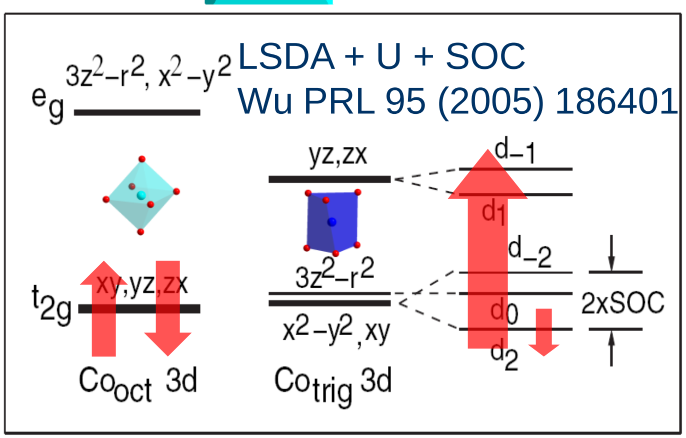
-h+k+l=3n

a = 9.08

c = 10.4

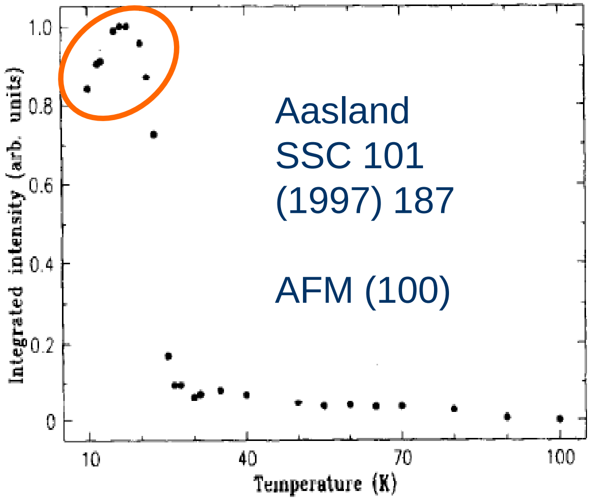


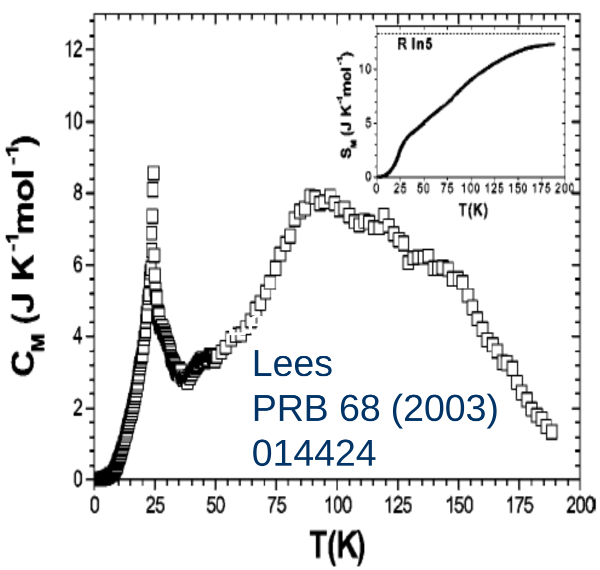
Fjellvag JSSChem 124 (1996) 190



S=0 **S=2**

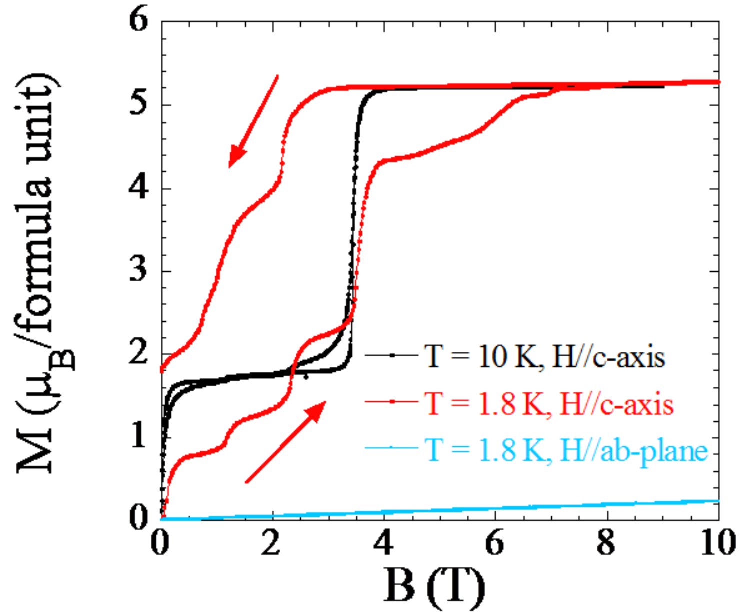
$Ca_3Co_2O_6$ static magnetic properties (I)





NPD and C_M:

- AFM state
- S//c-axis
- T_N~25K
- loss of intensity at low T

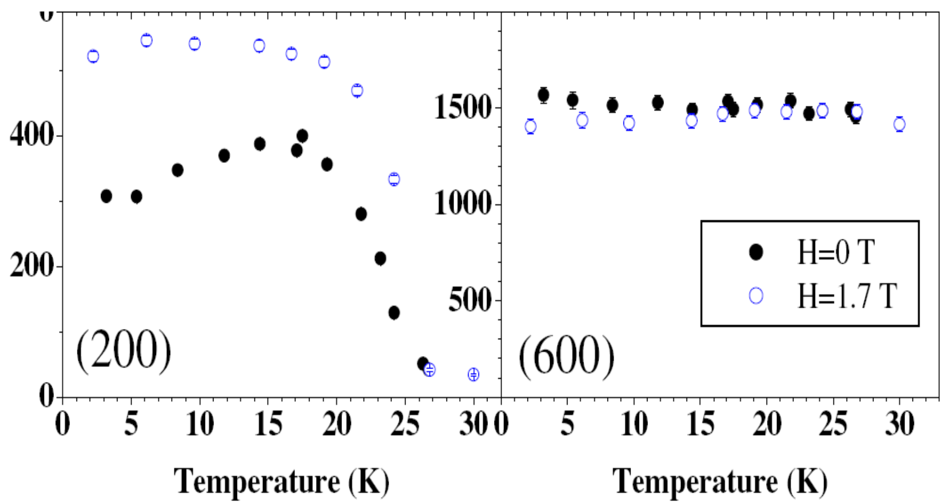


Magnetization:

- Ising
- $M_S > 5\ \mu B \rightarrow L \neq 0$
- $M_P \sim ⅓\ M_S \rightarrow$ fM
- Steps at low T

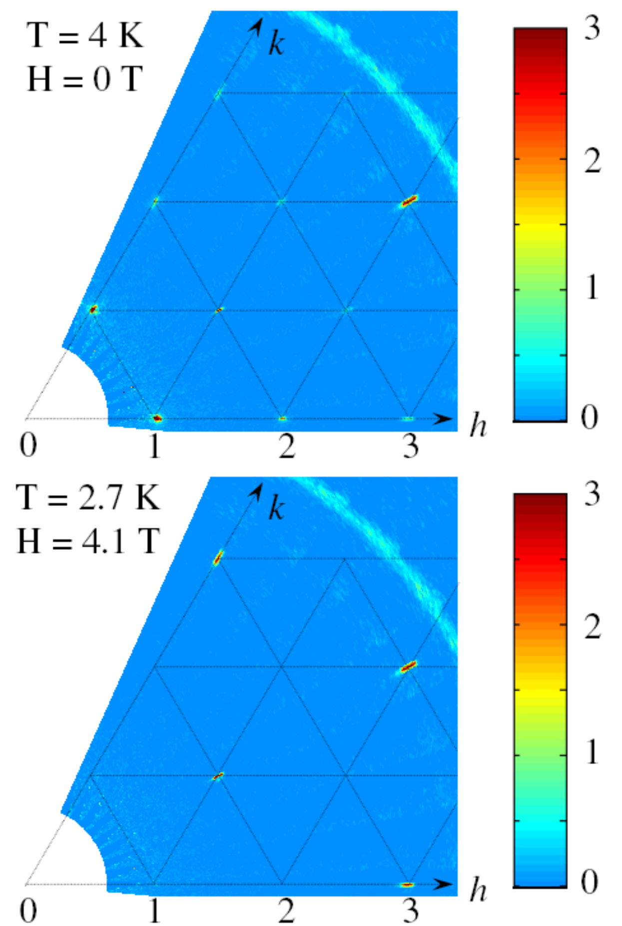
Hardy
PRB 70 (2004)
064424

$Ca_3Co_2O_6$ static magnetic properties (II)



single crystal ND

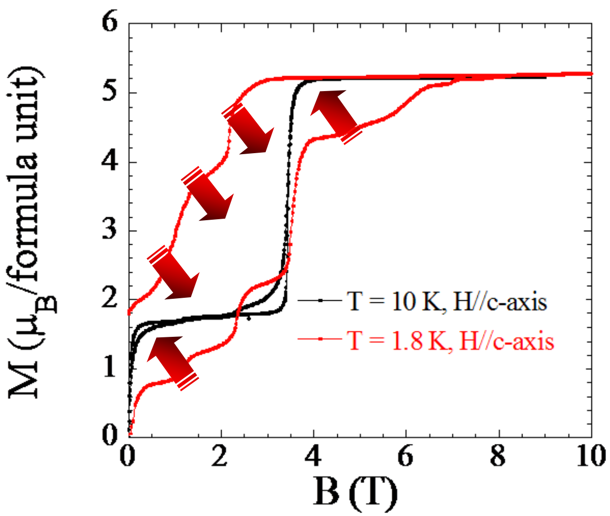
Petrenko EPJB47 (2005) 79

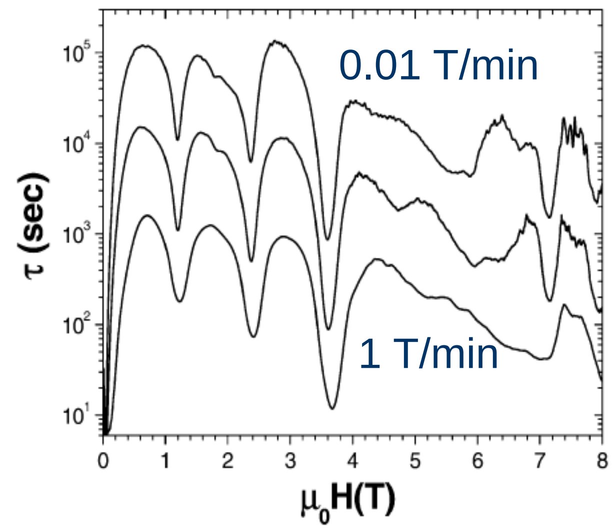


- No superlattice reflections, magnetic SG: P-3
- FM phase for B > 4T
- fM structure for ~1.2 < B < 4T
- for B < ~1.2T: strong reduction of AFM intensity at low T → dynamic contributions

Magnetic ground state? Step region??

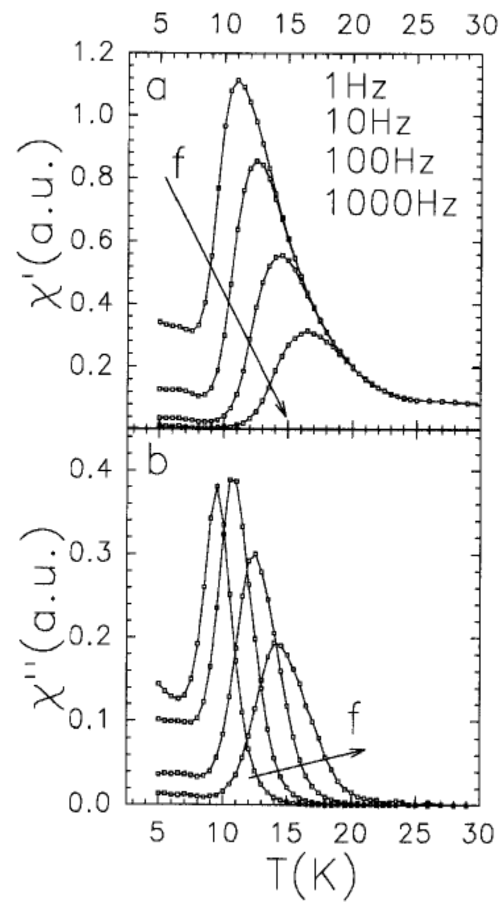
$Ca_3Co_2O_6$ dynamical magnetic properties

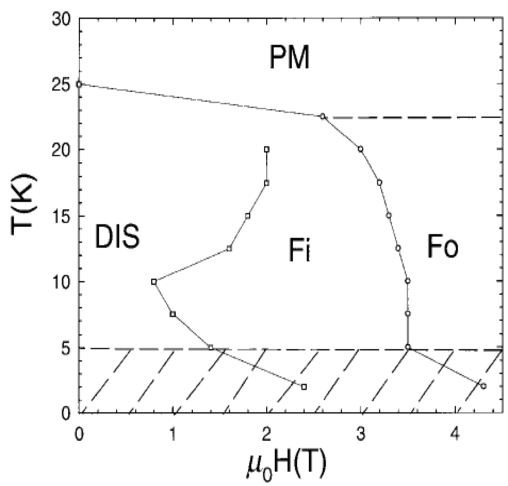


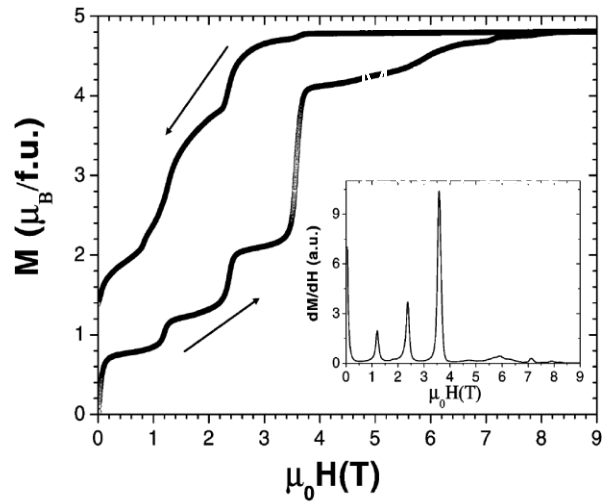


AC susc.

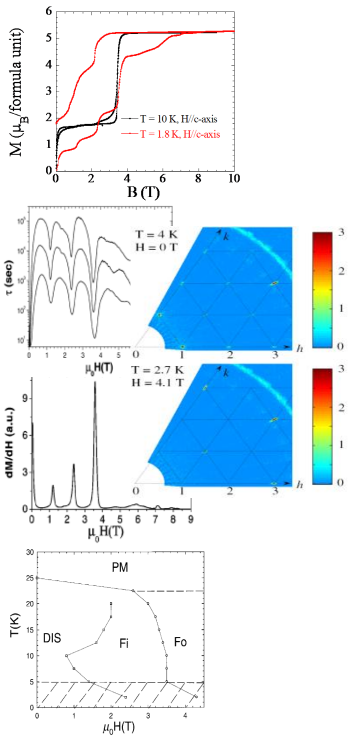
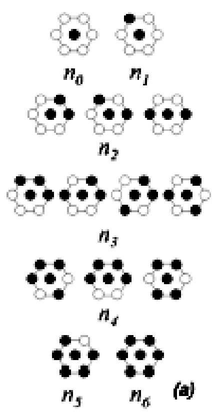
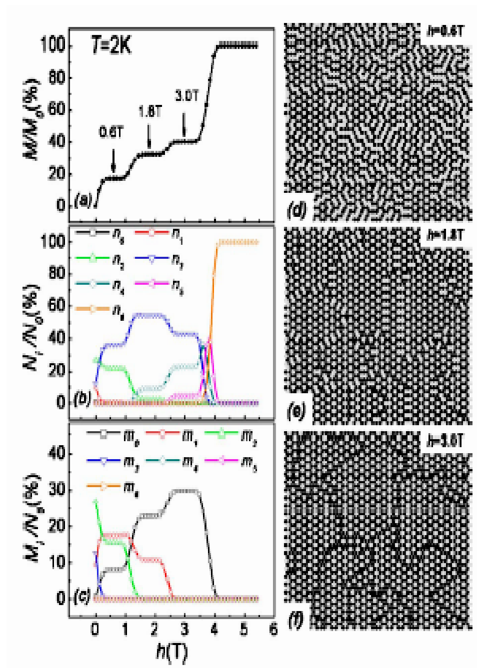
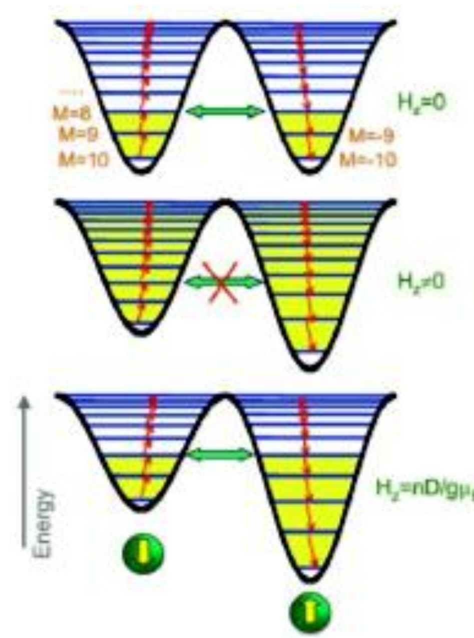
Maignan EPJB 15 (2000) 657





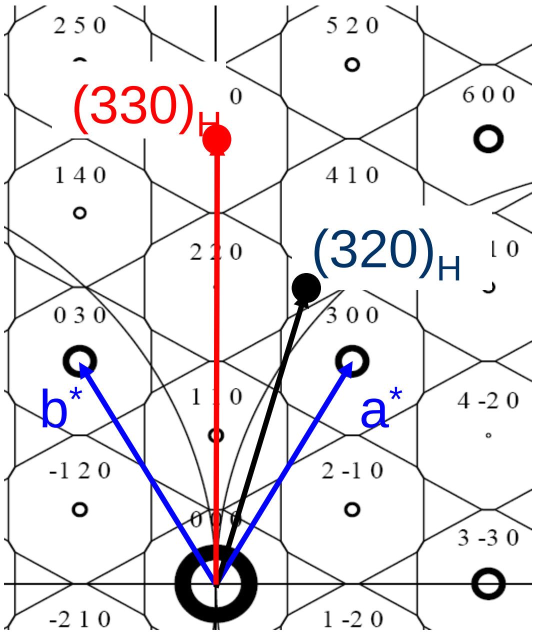


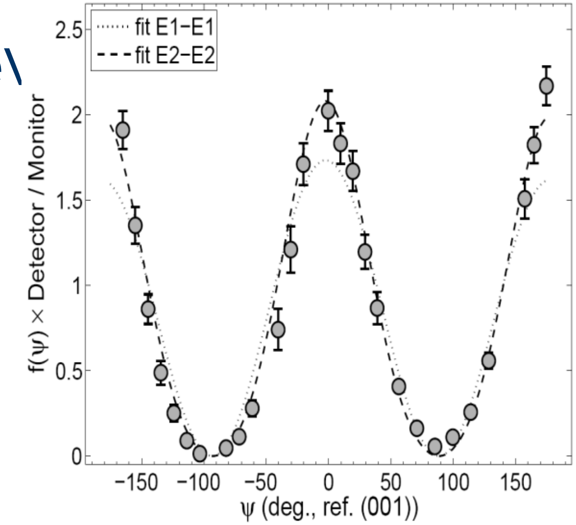
Magnetic models

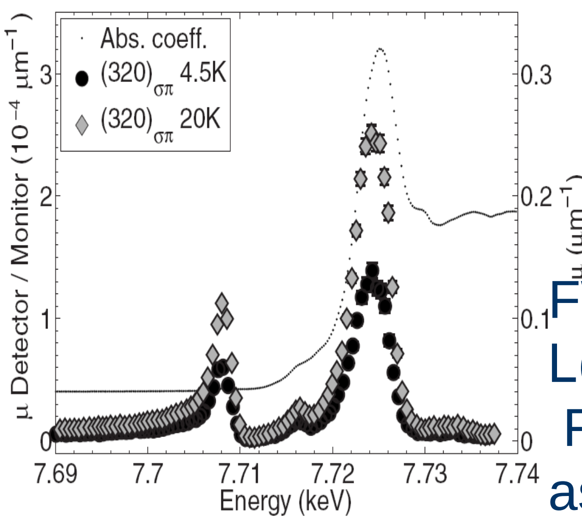
Exp. evidence	Proposed theoretical models	
	  Kudasov PRL 96 (2006) 027212 Burnus PRB 74 (2006) 134421	Hardy PRB 70 (2004) 214439 
Drawbacks	Step number	Short spin units along c-axis

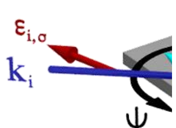
Magnetic-RXD (I)

Co K-edge E ~ 7.725 keV



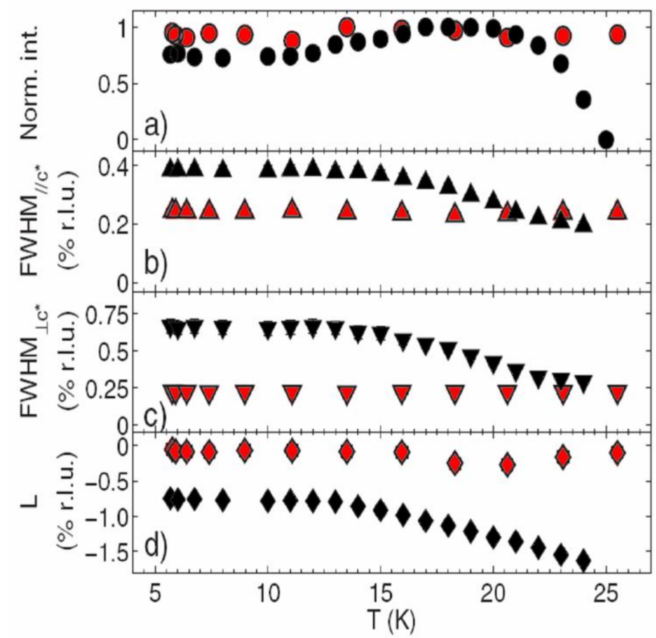






Magnetic (320)$_H$

Charge (330)$_H$

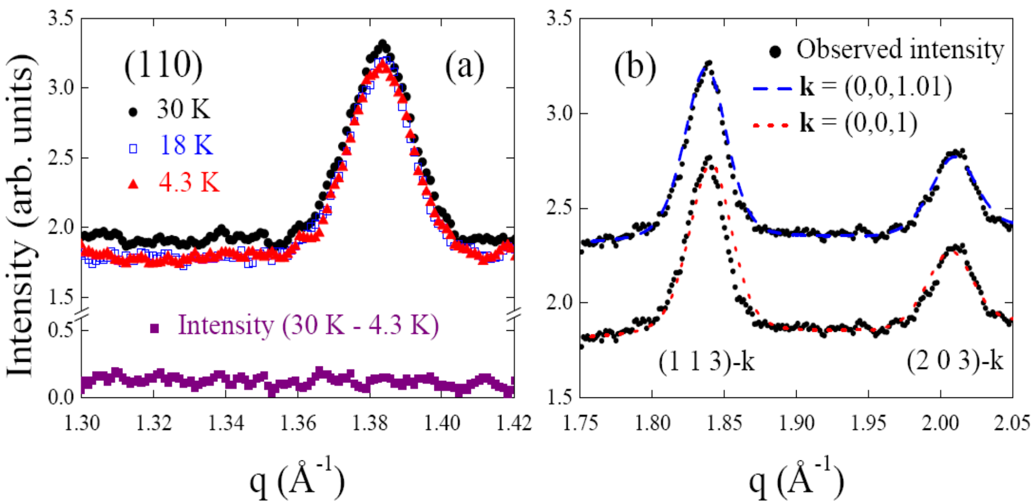


S. Agrestini, CM et al.
PRB 77 (2008)
140403R

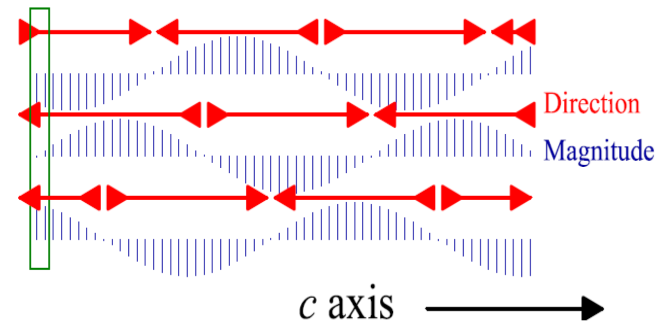
FWHM$_{Mag}$ ~ FWHM$_{Ch}$ → no QTM
L(T), L ≠ 0 → incommensurate
FWHM(T) → residual interaction
as source of the frustration

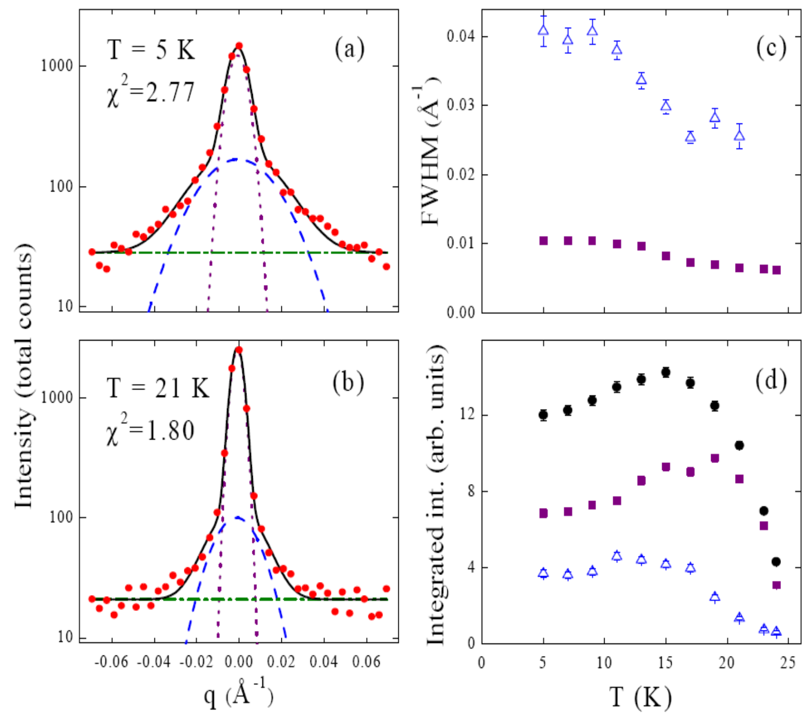
1st experimental
evidence of
modulated PDA

PND (I)



S. Agrestini et al.
PRL 101 (2008) 097207

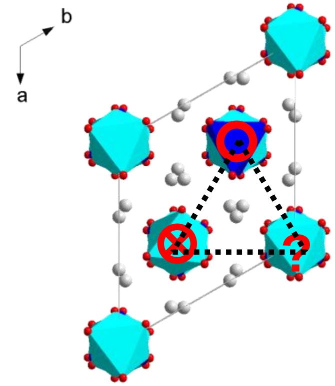




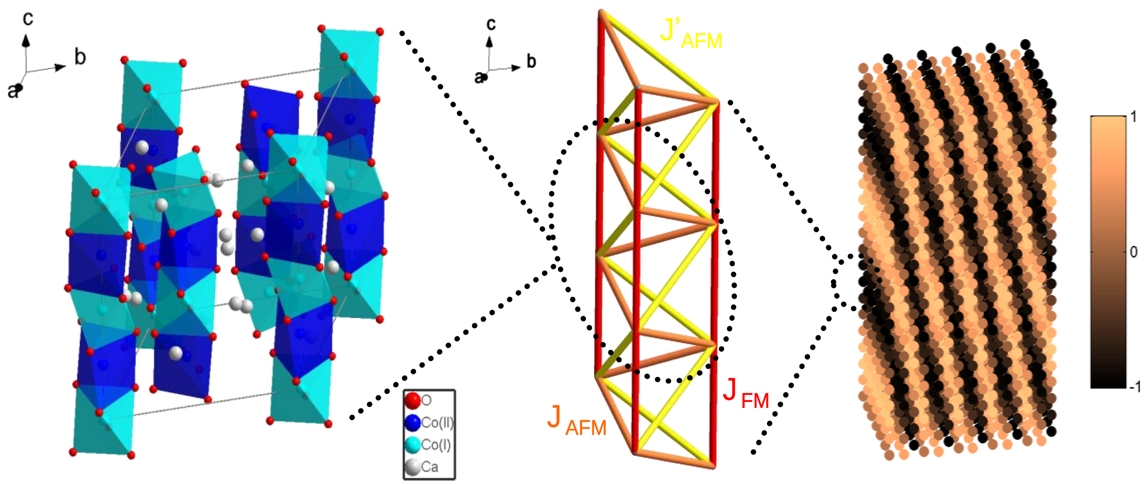
- No distortions
- Incommensurate magnetism confirmed + new description of magnetic phase
- Short correlation phase → metastable phases, maximum contribution at low T

Conclusions (after scattering experiments)

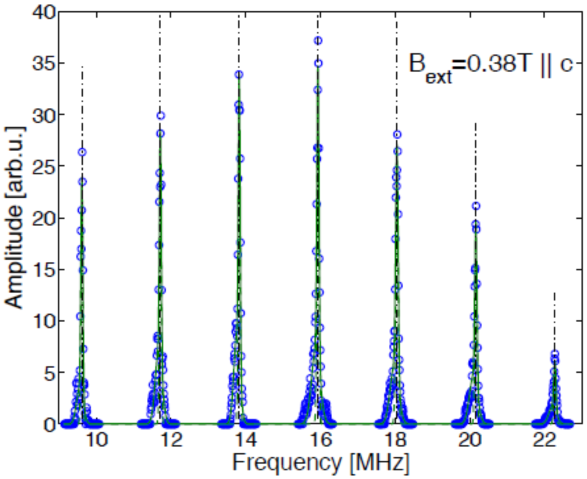
- no evidences supporting QTM model
- incommensurate magnetism → i-PDA state
- 3D character of the magnetic coupling is fundamental → new magnetic exchange pattern proposed

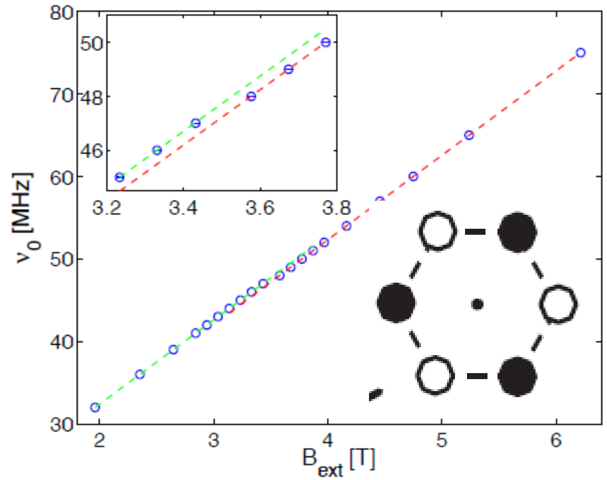


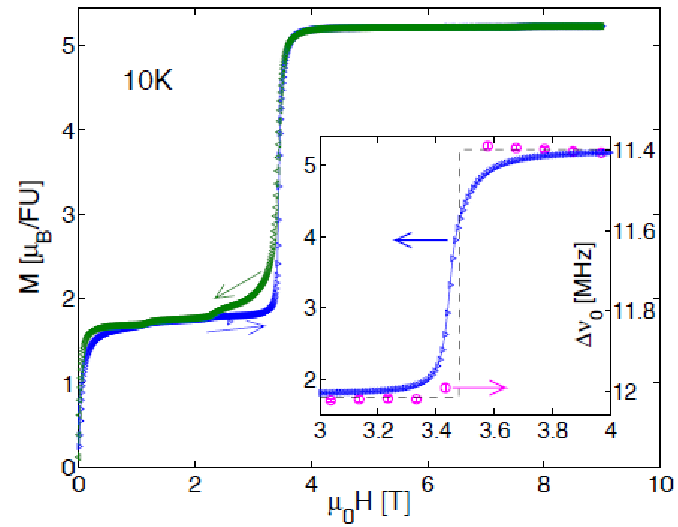
3D interactions ⇐ 2D Ising



^{59}Co (I=7/2) NMR in applied B







$B_{nuc} = 2\pi\, \nu_0 / {}^{59}\gamma$

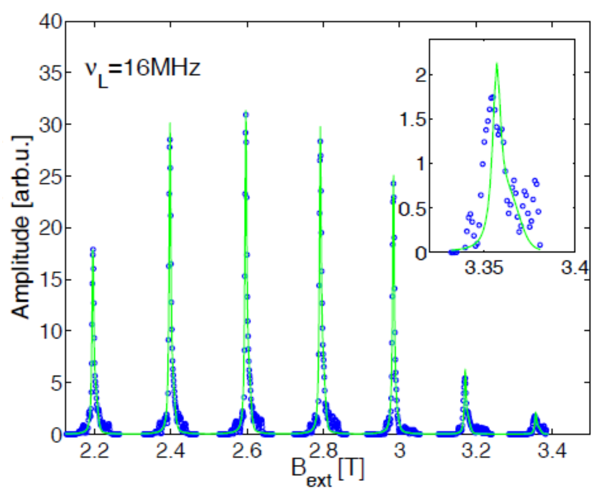
${}^{59}\gamma = 2\pi\ 10.10$ MHz/T

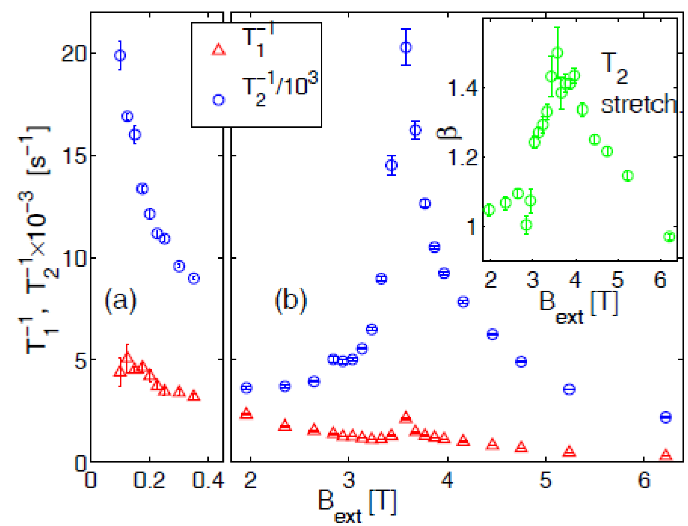
+

$$\mathcal{H}_Q = \frac{h\nu_Q}{6}\left[3I_z^2 - I\left(I+1\right) + \frac{\eta}{2}\left(I_+^2 + I_-^2\right)\right]$$

as a perturbation

Just S=0 is visible





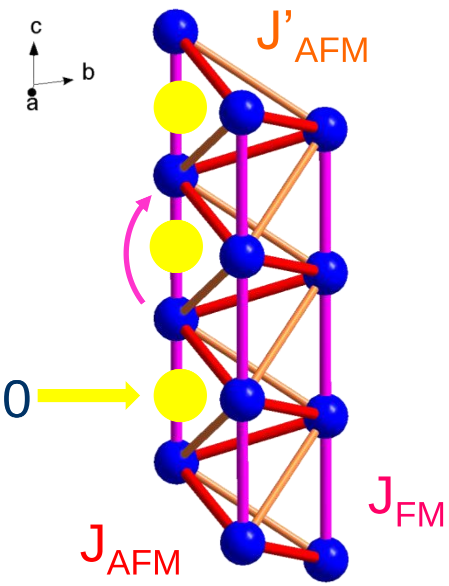
G. Allodi et al.
PRB 83 (2011) 104408
and further publication
in progress

Conclusions (after NMR experiments)

- super-exchange path linking 2 Co(II) vertically: no direct overlap through Co(I)
- transition is abrupt, smoothing detected by bulk measurements has to be due to "available fractions of magnetic moment"
- T_2 divergence and low T data suggest glassy behavior at meta-magnetic transitions (steps)
- no small magnetic units are detected. T_1 relaxation deals with thermally activated objects resembling domain walls motions

Co(I), S=0

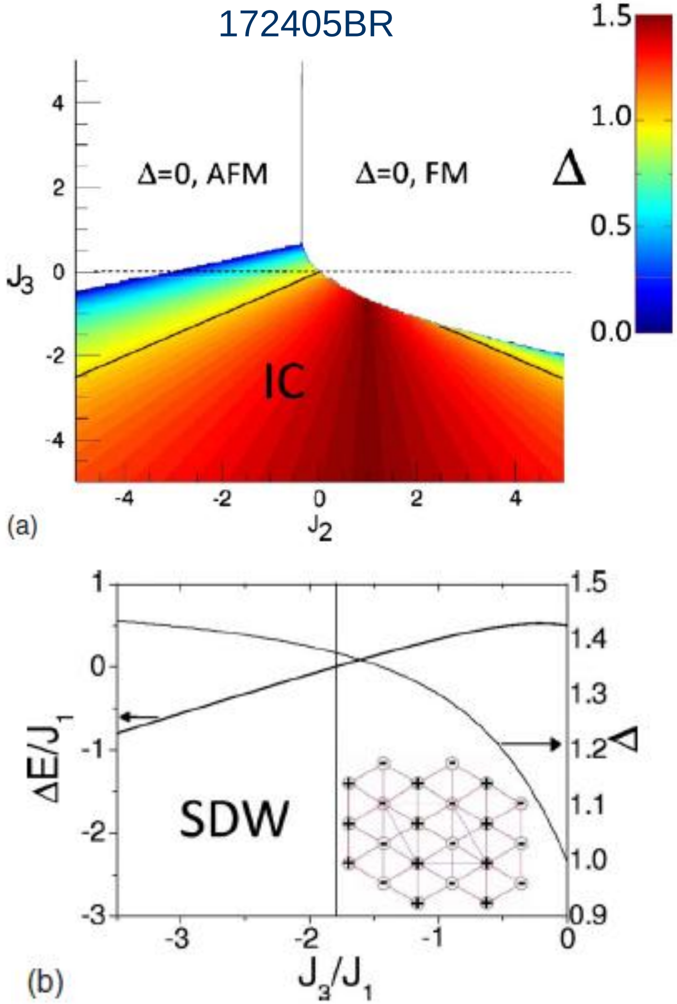
3D interactions

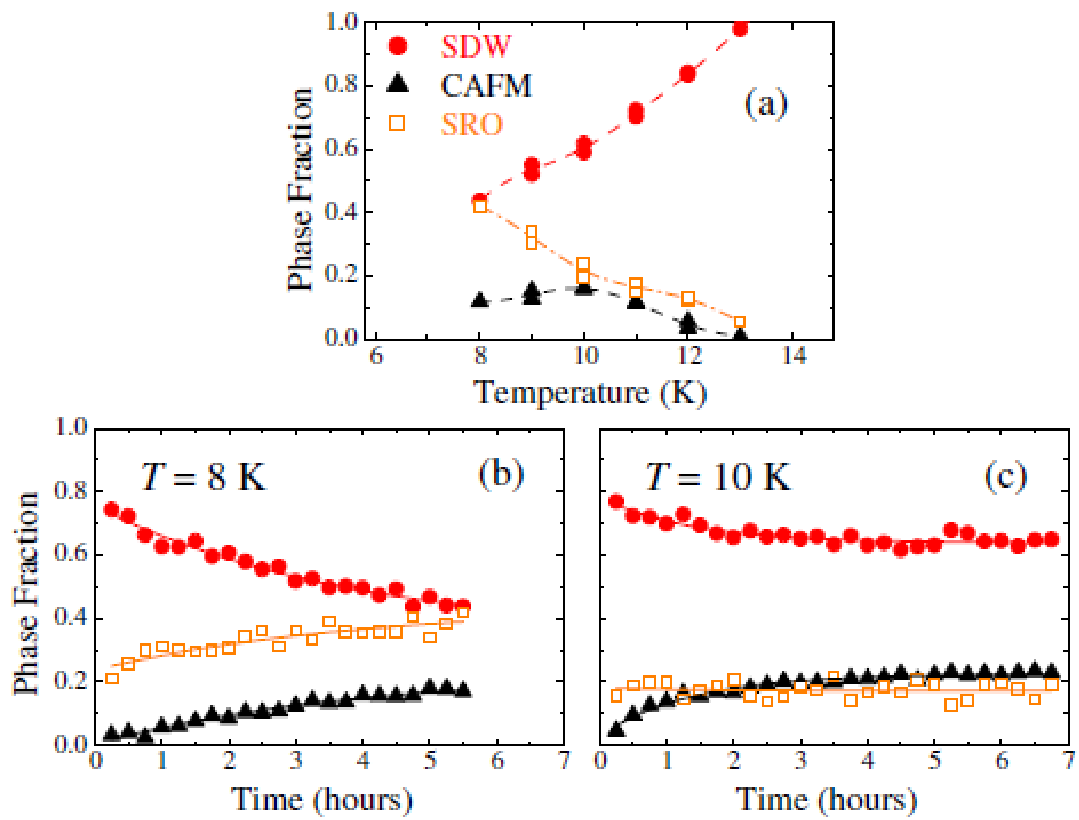


PND (II): Slow Magnetic Order-Order transition

L. Chapon
PRB 80 (2009)
172405BR

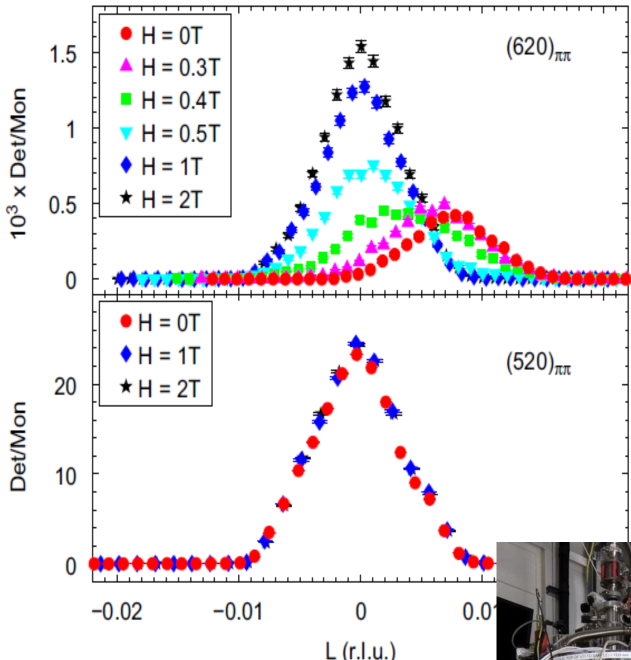
S. Agrestini et al.
PRL 106 (2011)
197204

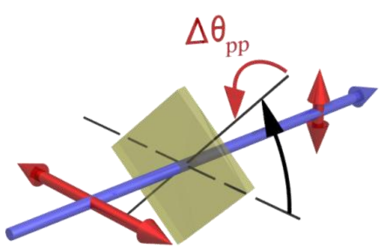


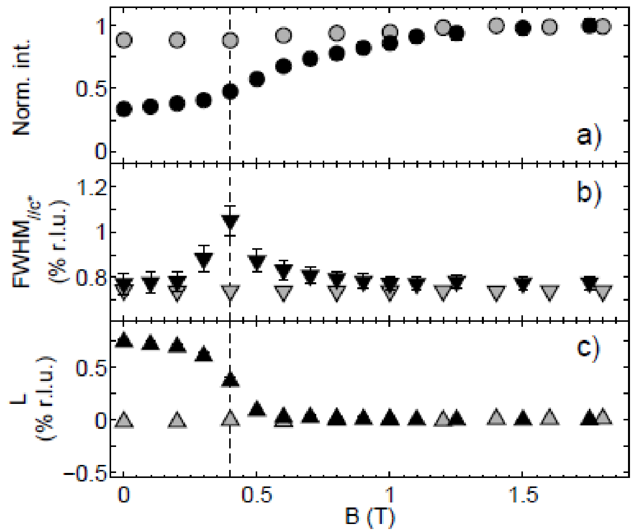


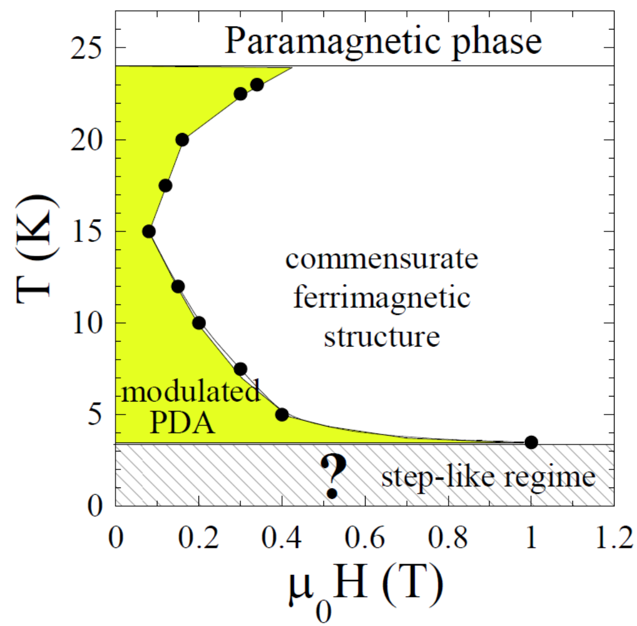
Magnetic-RXD (II): field measurements

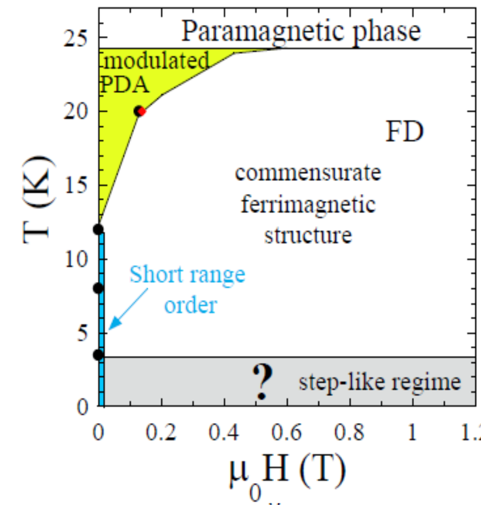
C. Mazzoli et al.
PhysicaB 404 (2009) 3042

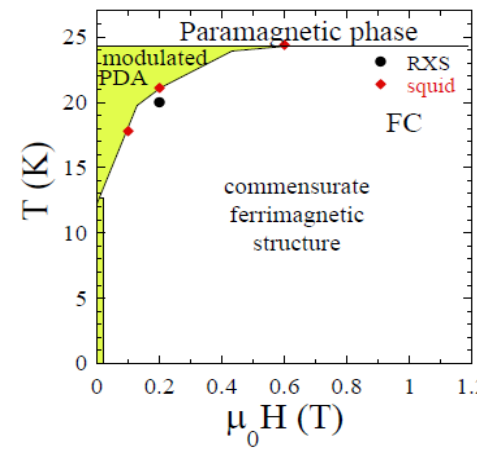












C. Mazzoli et al.
publication in progress

Summary

By using complementary techniques (magnetic-REXS, PND, NMR,…) to investigate this magnetically frustrated system

- Magnetic interaction: exchange pattern 2D $\rightarrow$ 3D
- Model system $\rightarrow$ complex and extremely rich situation
- New magnetic phase discovered
- Intrinsic dynamics of Order-Order phenomenon, very long time scale
- Now, magnetic-RXD in magnetic field accesses the step region. Further analysis is in progress to understand the contribution of the two magnetic phases to magnetic bulk properties and their interplay