Elliptical cycloidal phase and spin driven multiferroicity in Gd_2BaCuO_5

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Multiferroics

$CoFe_2O_4$

EuO

NiO

Ca_3CoMnO_6

$TbMnO_3$

$BaTiO_3$

$PbTiO_3$

PZT

$YMnO_3$

$BiFeO_3$

MFE

F

E

Applications

Four state ME-RAM

- Spin-wave generation
- Domain wall devices
- Amplification
- Frequency conversion

Type – II

- Correlation driven

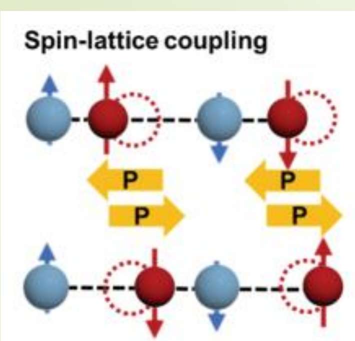
Type - I

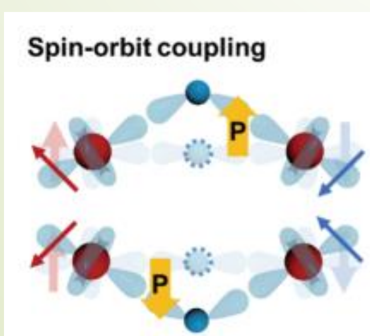
Magnetic

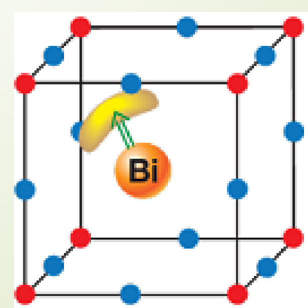
Dielectric

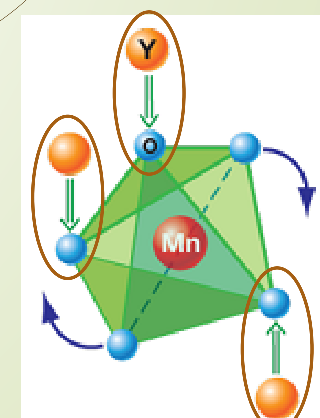
d^n vs. d^0

Chemical incompatibility



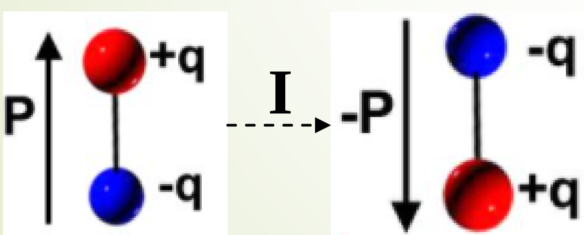


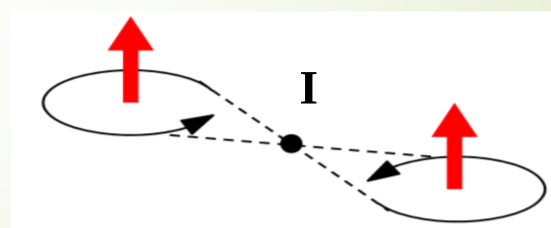


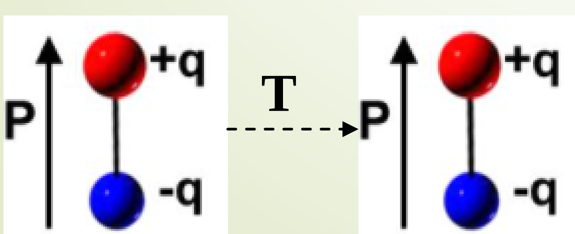


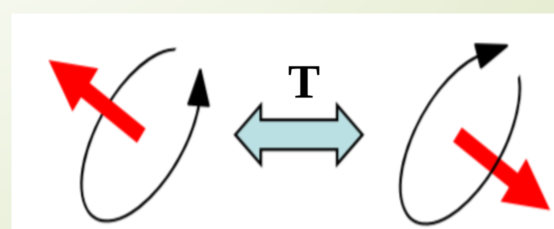
Symmetry requirements

Materials	Spatial inversion (I)	Time reversal (T)
FE	No	Yes
FM/AFM	Yes	No
MFE	**No**	**No**

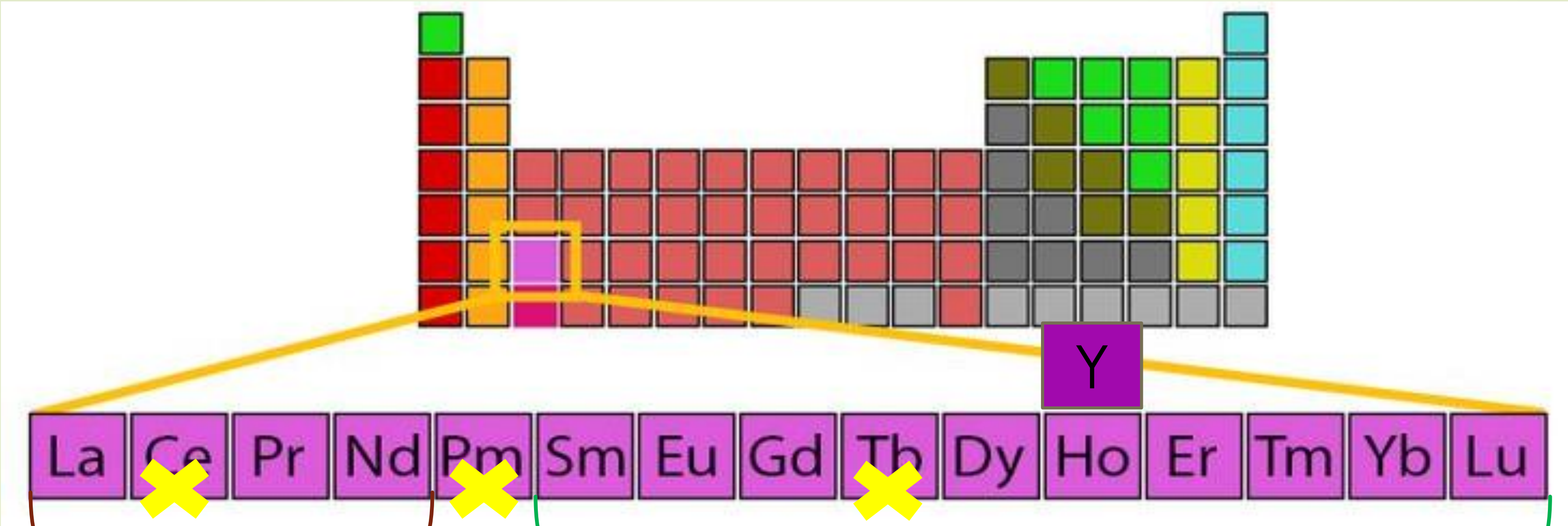




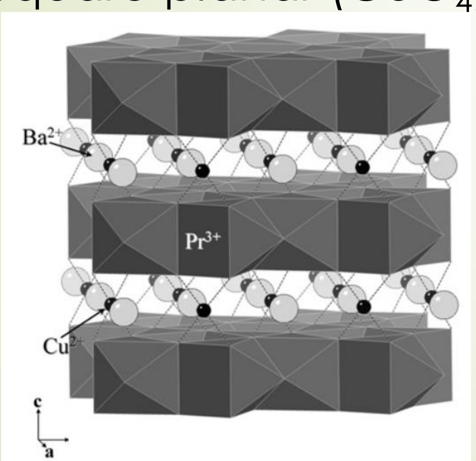




R_2BaCuO_5 (R = Rare earth)



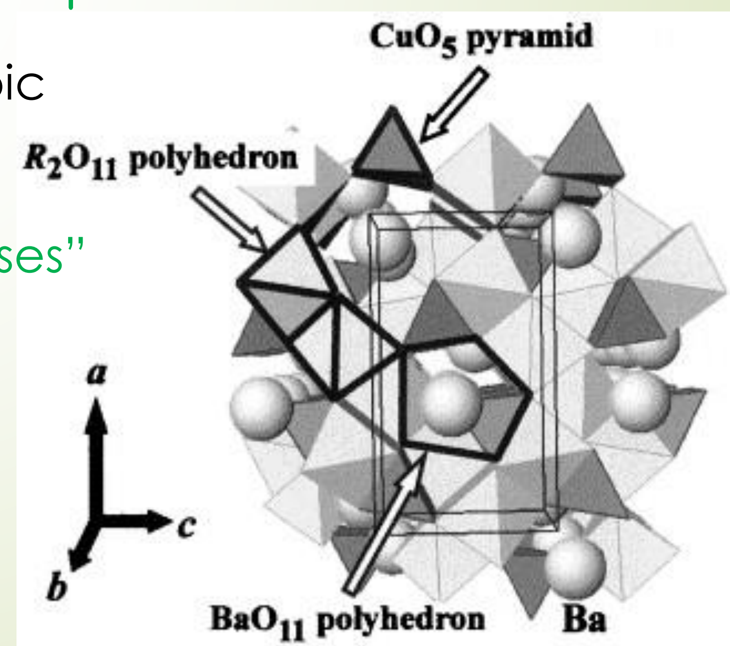
Tetragonal (*P4/mbm*)
Square planar (CuO_4)



"Brown Phases"

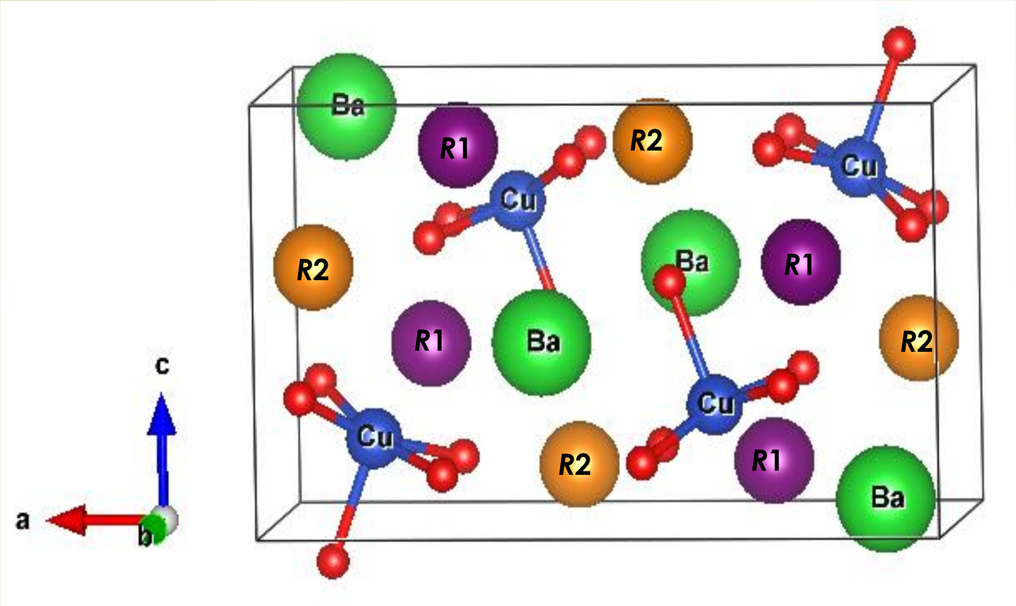
Orthorhombic
(*Pnma*)

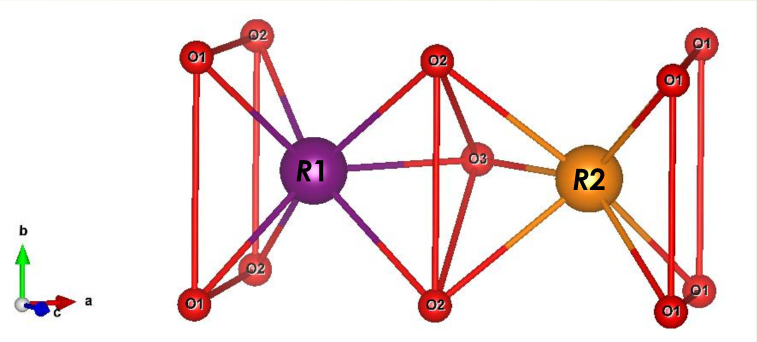
"Green Phases"



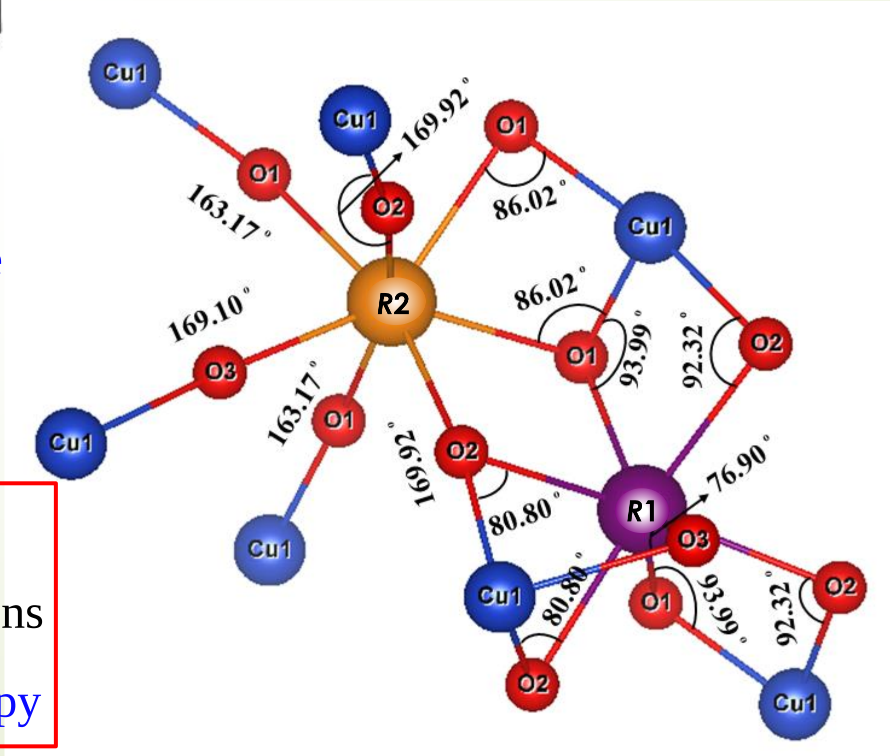
C. Michel and B. Raveau, J. Solid State Chem. **43**, 73 (1982)
A. Salinas-Sanchez, *et. al.*, J. Solid State Chem. **100**, 201 (1992)

Crystal Structure

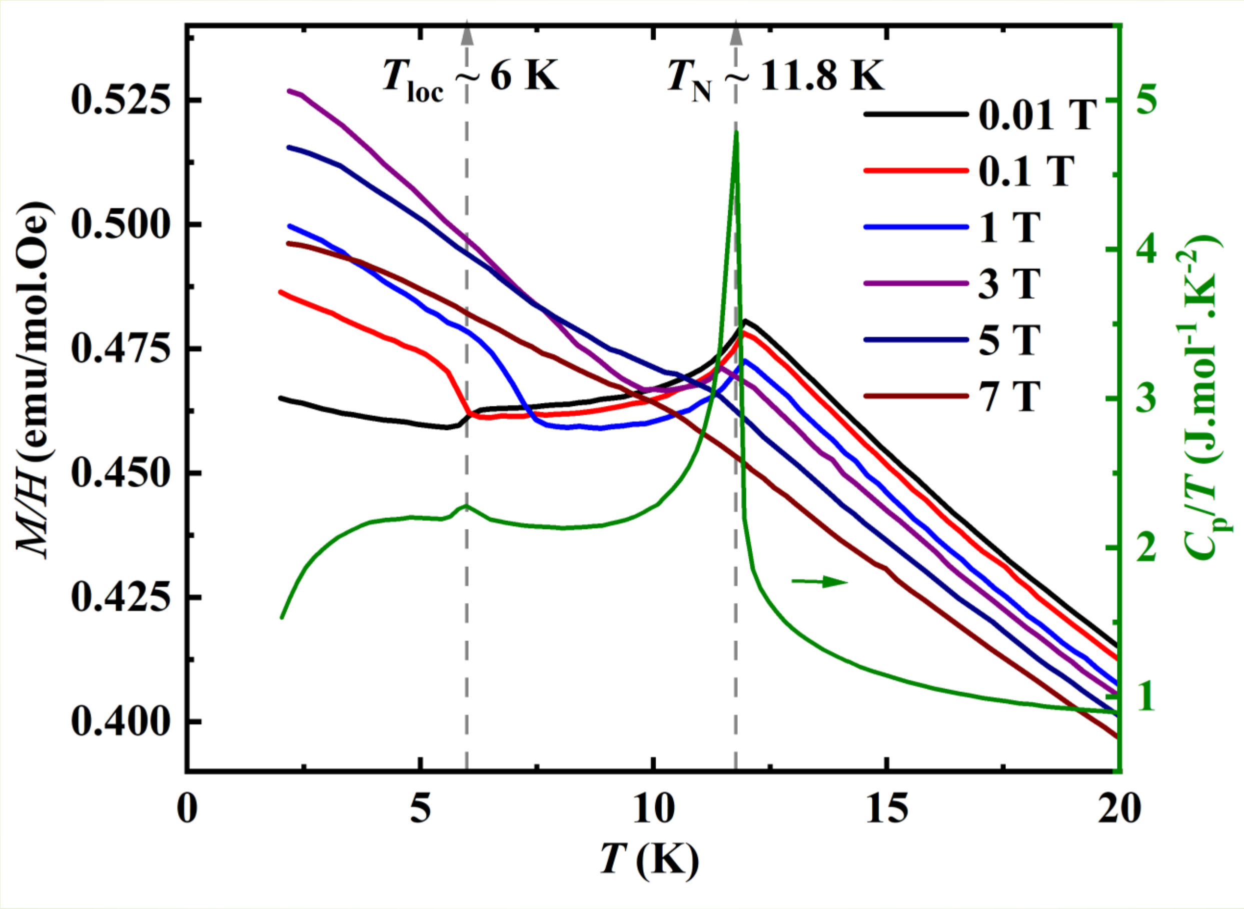


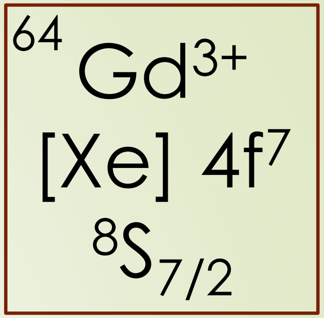


- The copper ions are situated in distorted square pyramid CuO_5
- No Cu-Cu, Cu-O-Cu, Cu-O-O-Cu bonds
- Interaction path: Cu^{2+}-O^{2-}-R^{3+}-O^{2-}-Cu^{2+}
- Cu-Cu, *R*-*R*, ***R*-Cu (4f-3d coupling)** interactions
- Crystal-field excitations and magnetic anisotropy



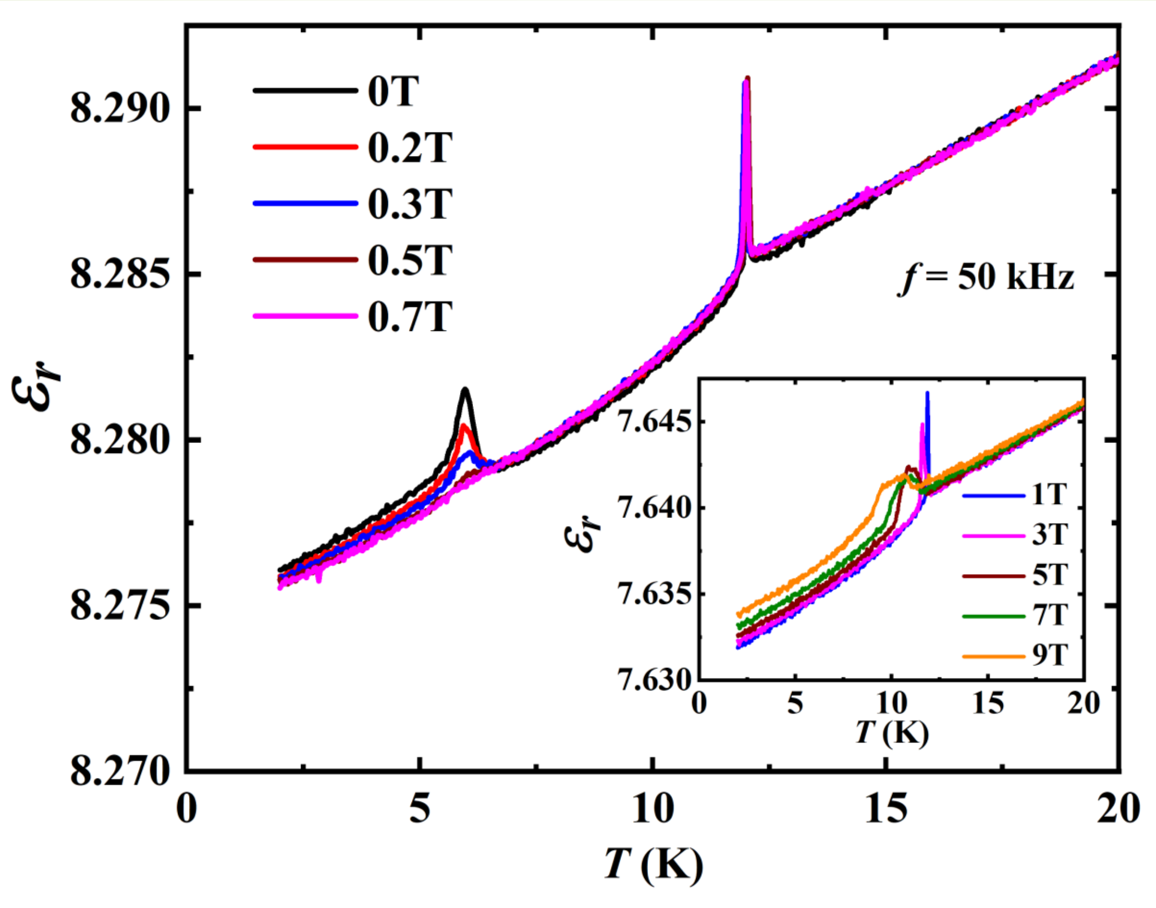
Gd_2BaCuO_5 ~ Magnetic properties





- ➢ Long-range AFM ordering of Gd^{3+} and Cu^{2+} spins at T_N = 11.8 K and another magnetic transition at T_{loc} = 6 K (lock-in transition)
- ➢ Simultaneous ordering of Gd^{3+} and Cu^{2+} spins unlike other R_2BaCuO_5 compounds

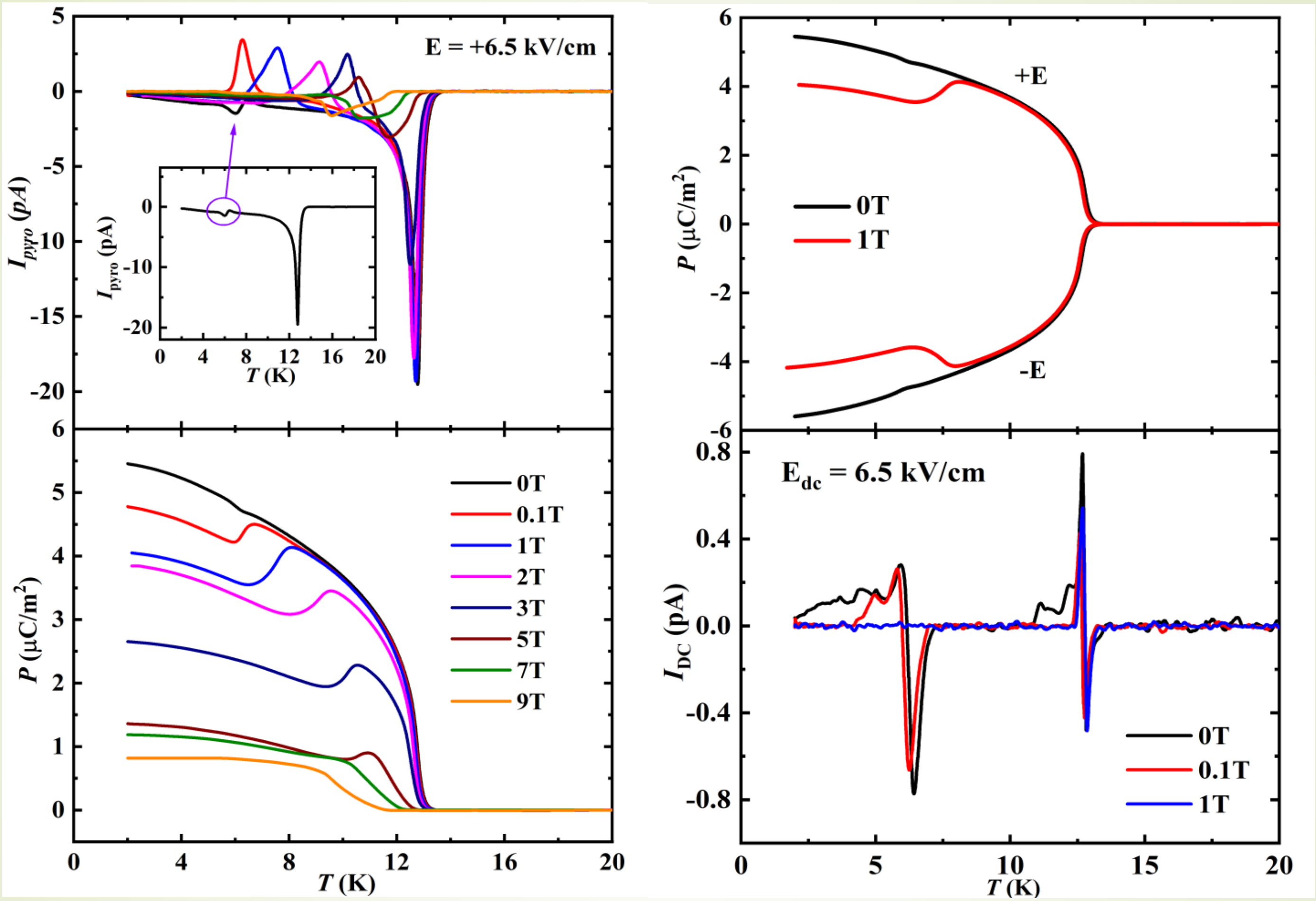
Dielectric properties



- It shows dielectric anomalies at both T_N and T_{loc} in the absence of magnetic field
- The anomaly at T_{loc} suppressed above H = 0.7 T

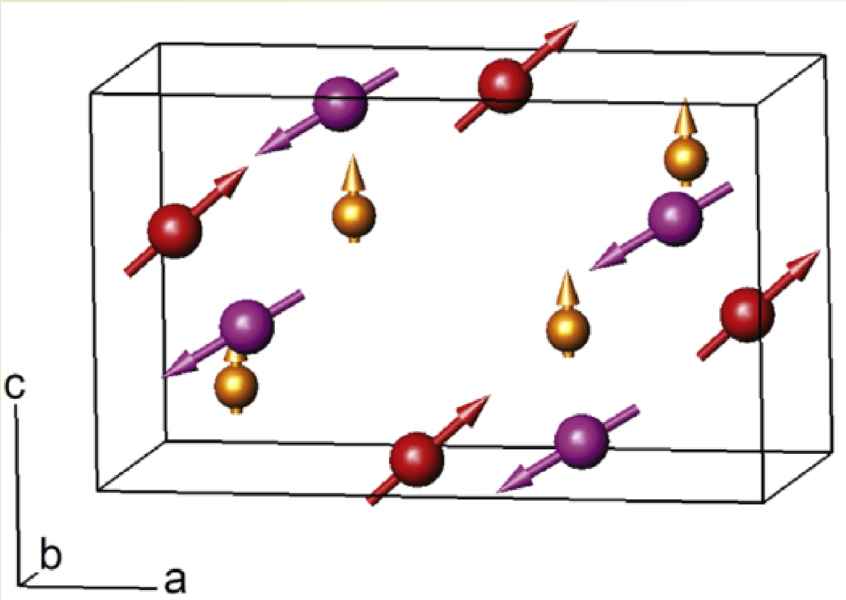
Multiferroic?

Spin-induced multiferroicity



- ➢ Spontaneous ferroelectric (FE) polarization at T_N = 11.8 K and T_{loc} = 6 K
- ➢ Under magnetic fields > 0.7 T, ferroelectricity at T_{loc} disappears
- ➢ Switching and DC bias supports the multiferroicity

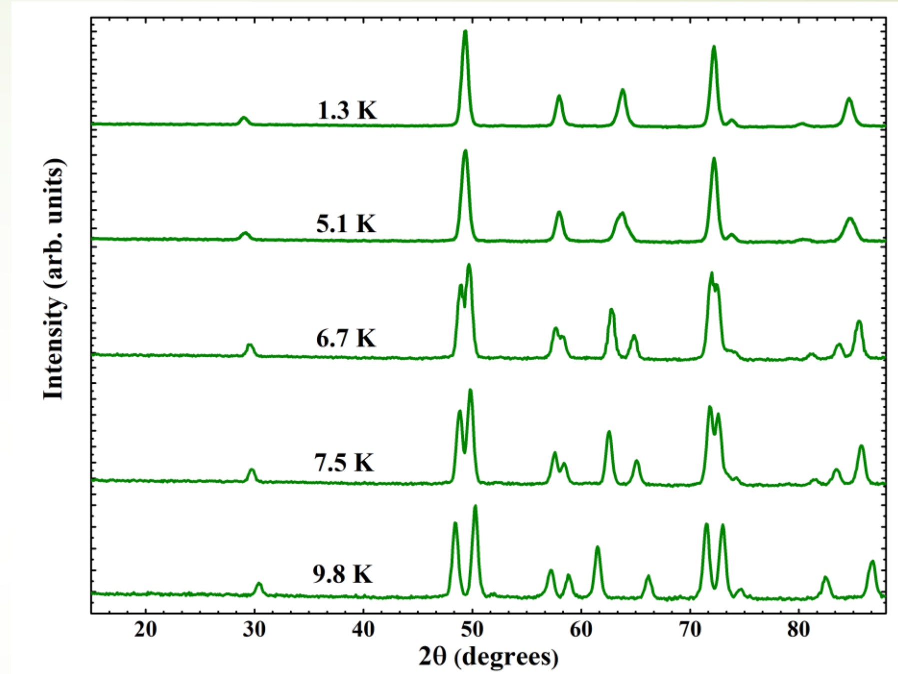
Reported magnetic structure



- Magnetic space group: P_S-1
- *Pnma* and **k** = (0, 0, 1/2)
- The observed multiferroicity is inconsistent with the reported magnetic structure

I. V. Golosovsky, *et. al.*, J. Magn. Magn. Mater. **353**, 71 (2014)

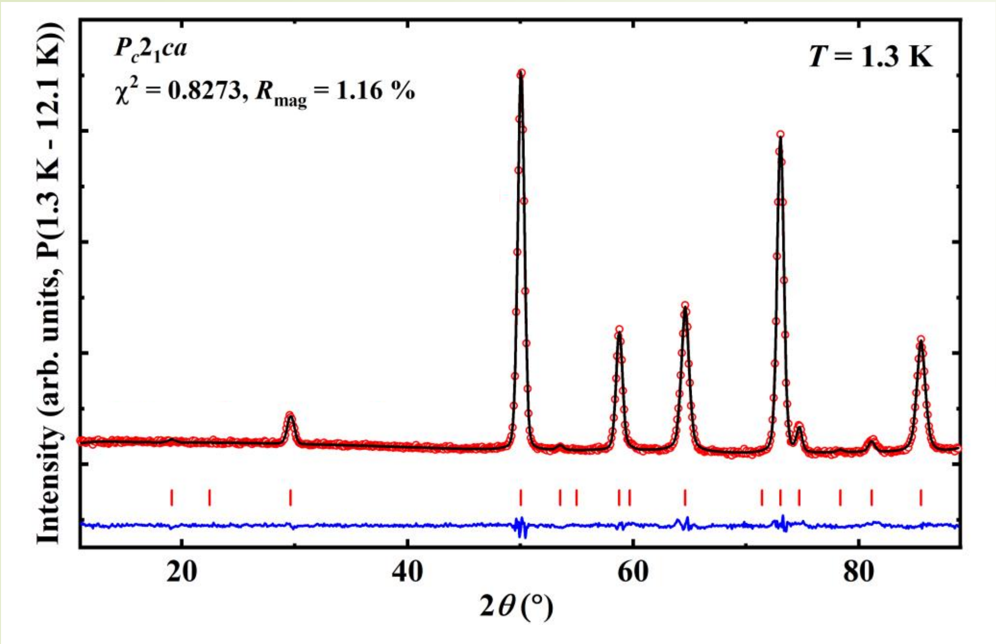
Neutron diffraction data



Magnetic contribution only

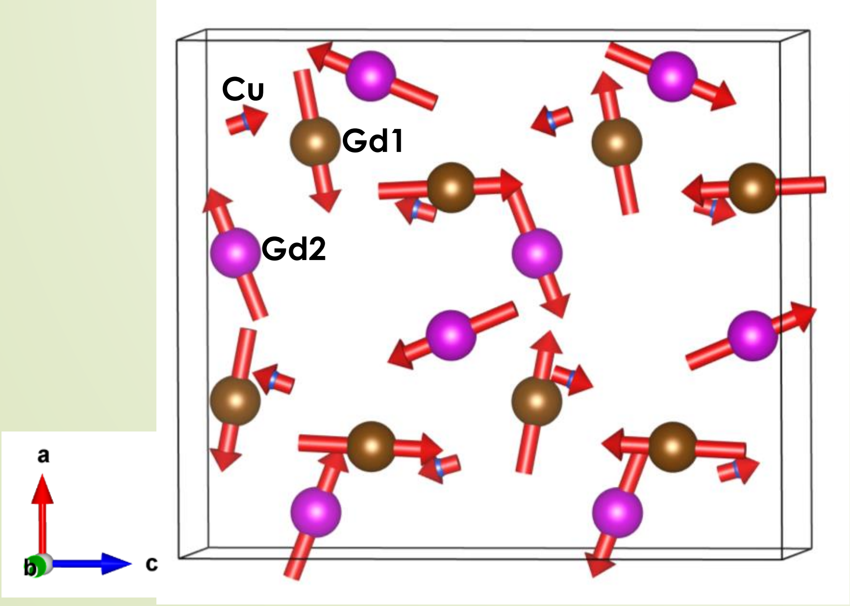
- Incommensurate magnetic ordering with **k-vector (0, 0, *g*)** below T_N = 11.8 K
- Commensurate with **k-vector (0, 0, ½)** below T_{loc} = 6 K

Ground state magnetic structure at 1.3 K



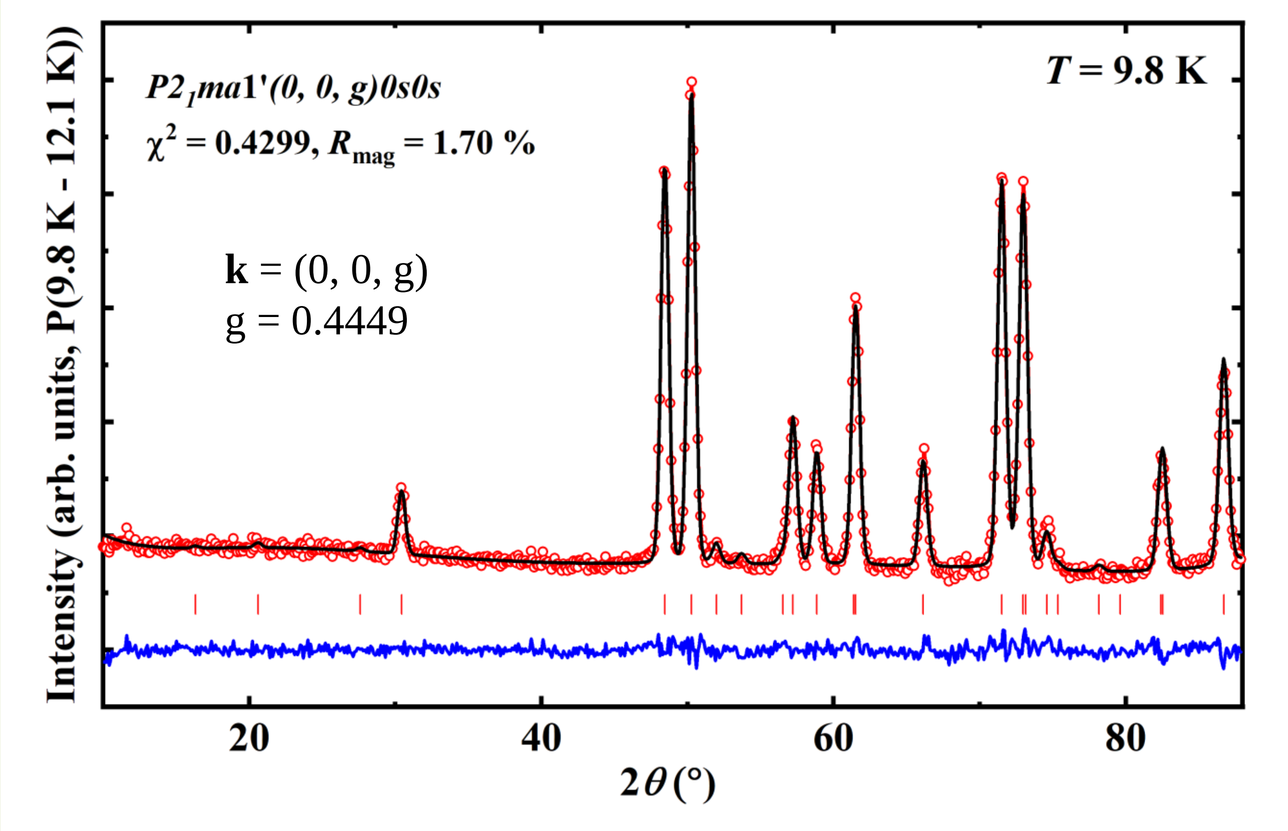
- **Commensurate k-vector (0, 0, ½)** and *Pnma*
- Two *irreps:* mZ1 and mZ2

mZ1	mZ2
P_a2_1/m	P_c2_1/c
P_bmc2_1	$\boldsymbol{P_c2_1ca}$
P_am	P_cc



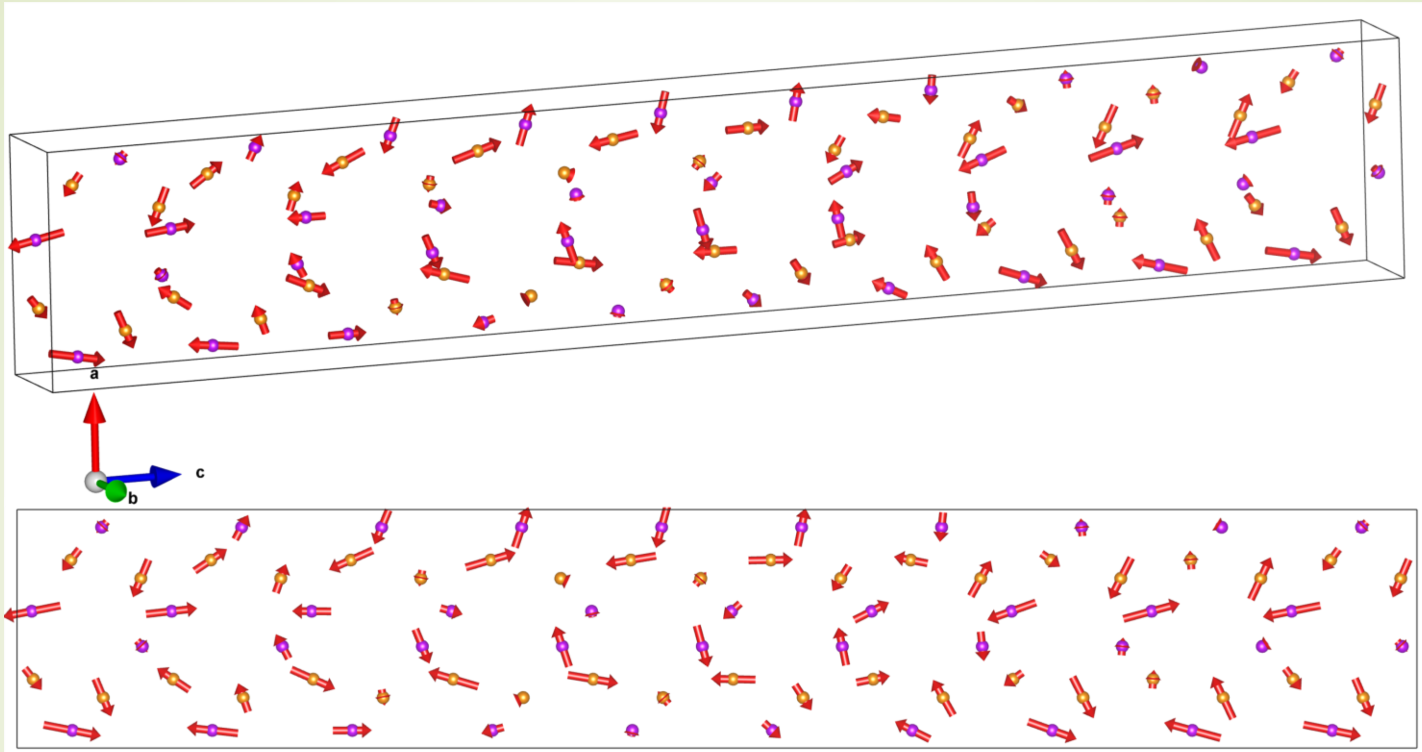
- Magnetic symmetry 2*mm* is polar and allows ferroelectricity.
- $P = (p_x, 0, 0)$
- Strongly non-collinear magnetic structure

Neutron diffraction (T_{loc}<T<T_N)



- Four possible *irreps: mLD1*, *mLD2*, *mLD3*, *mLD4*
- Mixing of *irreps*: *mLD2*⊕*mLD3*
- Magnetic super space group: $P2_1ma1'(0, 0, g)0s0s$
- Point group: $2mm1'$ is polar

Cycloidal structure ($T_{loc}<T<T_N$)

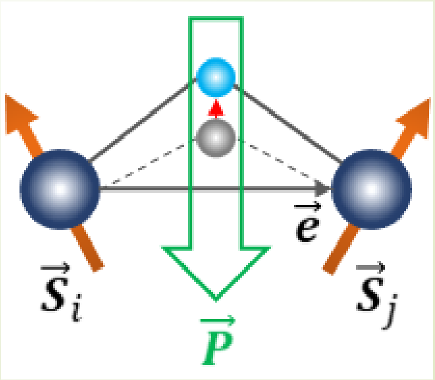


Magnetic structure viewed along b-axis

- Elliptical cycloidal structure
  (which breaks the inversion symmetry)

- Inverse Dzyaloshinskii-Moriya interaction

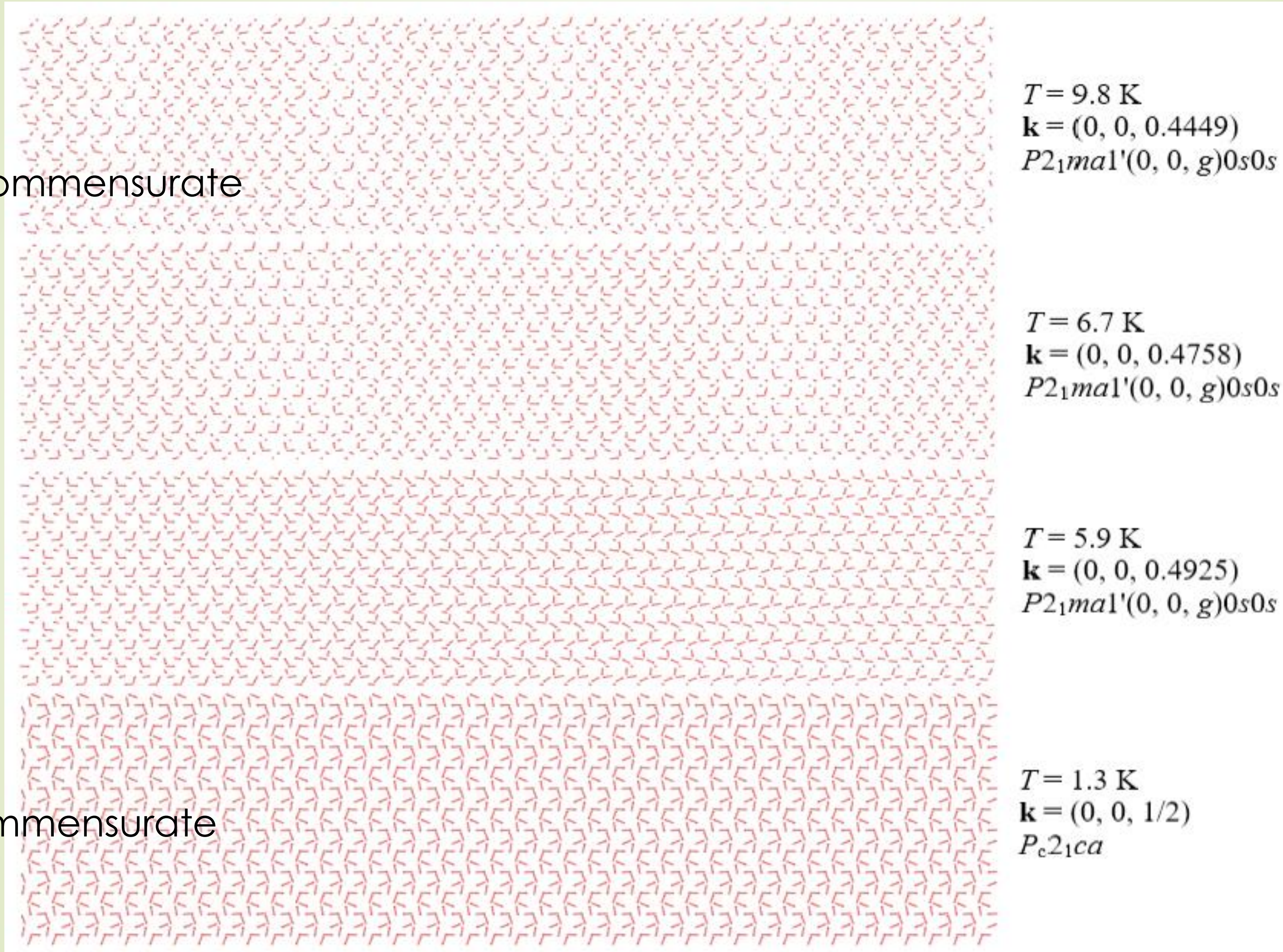
$$\vec{P}_{ij} \propto \vec{e}_{ij} \times (\vec{S}_i \times \vec{S}_j)$$



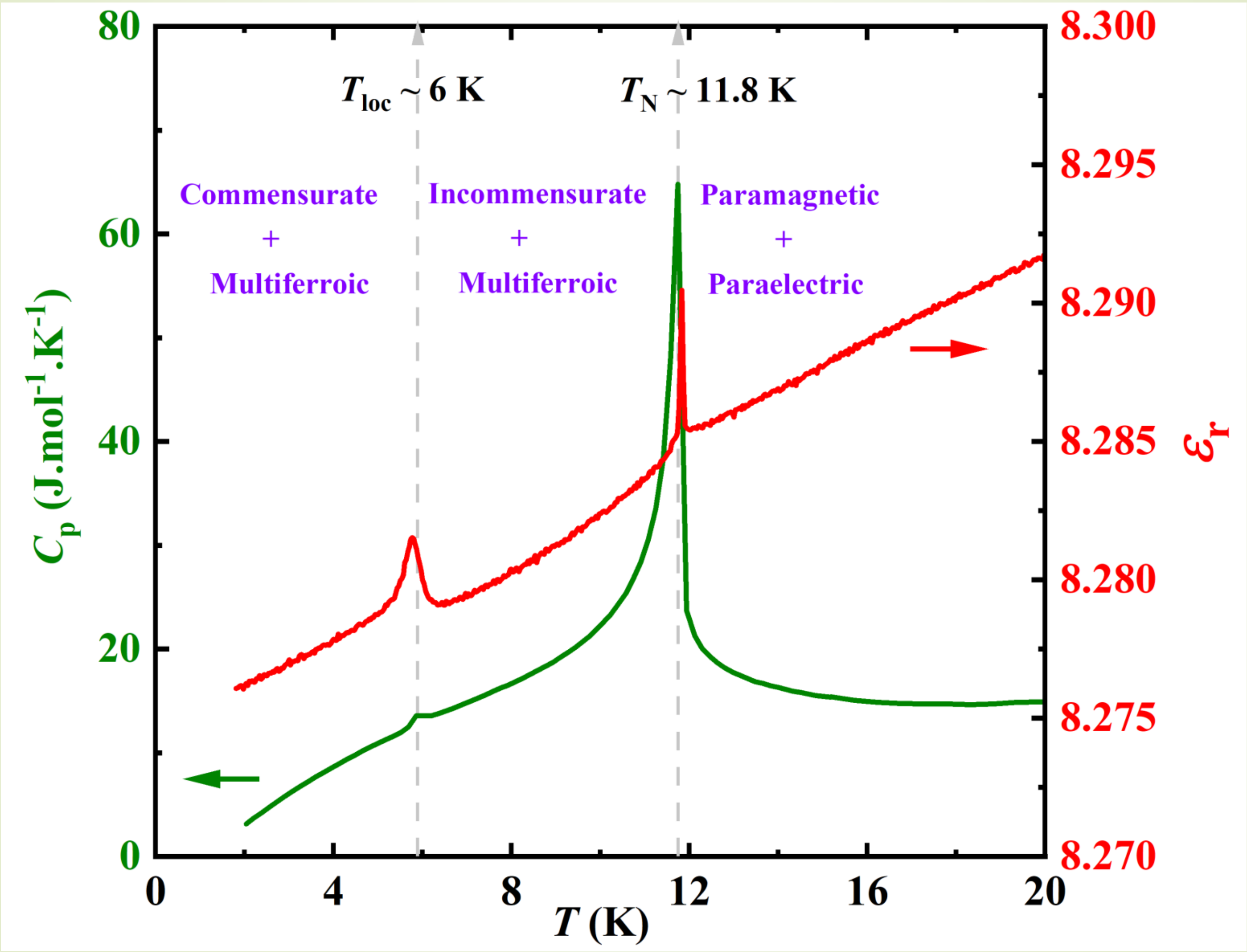
Global view of magnetic structure

Incommensurate

Commensurate



Summary



T_loc ~ 6 K
T_N ~ 11.8 K
Commensurate
+
Multiferroic
Incommensurate
+
Multiferroic
Paramagnetic
+
Paraelectric
C_p (J.mol^-1.K^-1)
ε_r
T (K)
0
20
40
60
80
4
8
12
16
8.270
8.275
8.280
8.285
8.290
8.295
8.300

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Thank You!