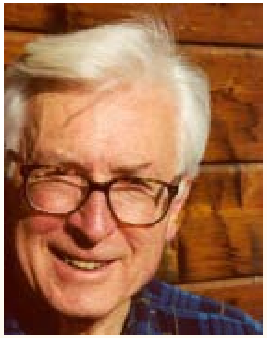
Multiferroics – coexistence of ferromagnetism and ferroelectricity

A. Szewczyk, Institute of Physics, Polish Academy of Sciences

- **Multiferroics** – definition
- **Origins of mutual exclusion** of magnetic and electric orderings
- Mechanisms that make such coexistence possible:
  - perovskites: $FeBiO_3$, $TbMnO_3$
  - manganites: $(Sr,Ba)MnO_3$
  - olivines (?)
  - hexagonal manganites
- **Ferrotoroidic** ordering as a new kind of long-range order in multiferroics
- Ideas of application of multiferroics

Multiferroics – coexistence of ferromagnetism and ferroelectricity

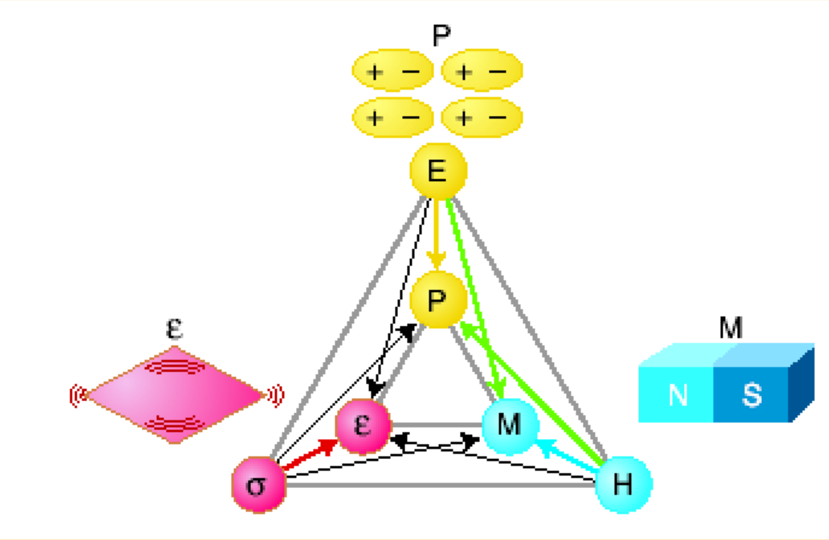
H. Schmid, *Multiferroic magnetoelectrics*, Ferroelectrics 162, 317 **(1994)**.



† 2 kwietnia 2015

Multiferroics: materials in which several (at least two) qualitatively different long-range orderings coexist:

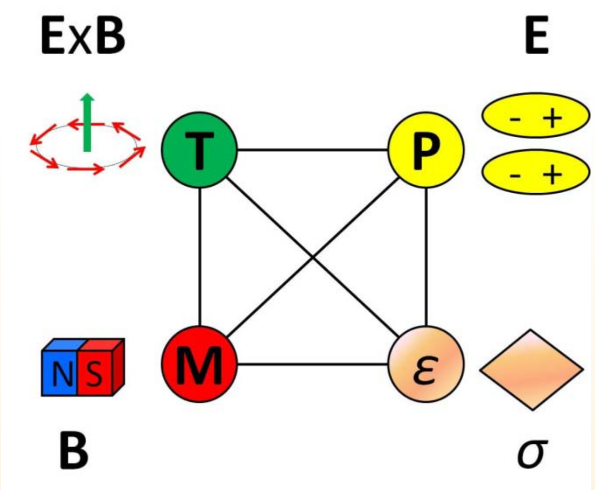
– ferromagnetic (or antiferromagnetic)
– ferroelectric
– ferroelastic (ferrodistorsive)

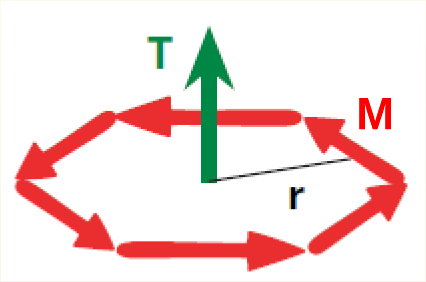


Spaldin & Fiebig, Science **309**, 391 (2005).

Multiferroics – coexistence of ferromagnetism and ferroelectricity

- **Multiferroics** – materials in which several long-range orderings coexist (H. Schmid - 1994), e.g.:
  - ferromagnetic (or antiferromagnetic)
  - ferroelectric
  - ferroelastic
  - **ferrotoroidic**

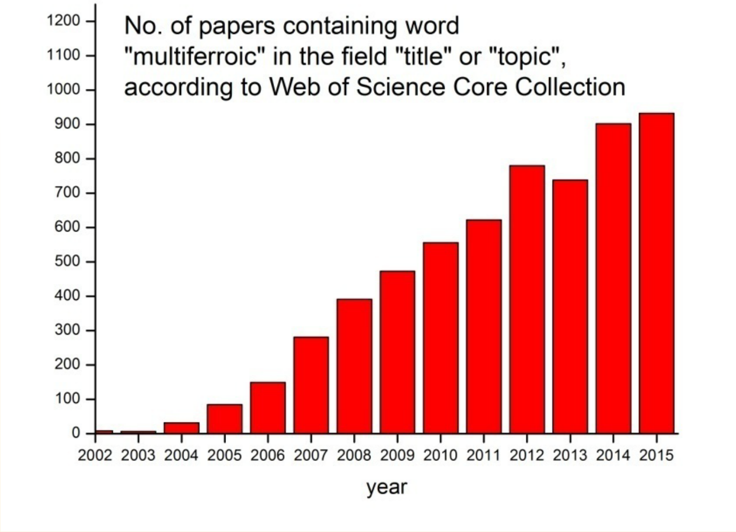




$$\mathbf{T} = \frac{1}{2}\sum_n \mathbf{r}_n \times \mathbf{M}_n \neq 0$$

toroidal moment

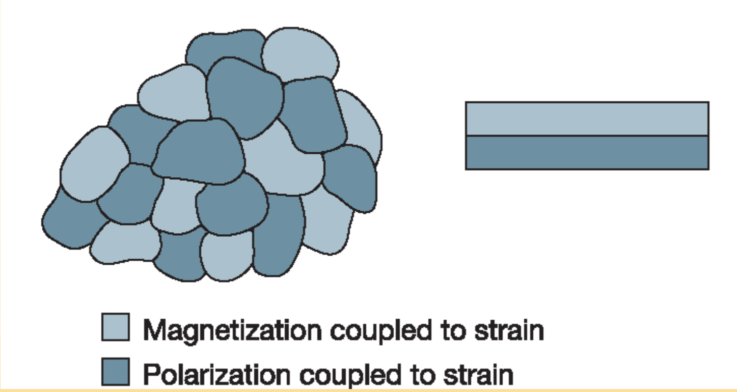
Multiferroics – coexistence of ferromagnetism and ferroelectricity



- **Multiferroics** – materials in which several long-range orderings coexist (H. Schmid - 1994), e.g.:
  - ferromagnetic (or antiferromagnetic)
  - ferroelectric
  - ferroelastic
  - **ferrotoroidic**
- **Origins of strong interest:**
  - **presence of „cross relations"**
  - ferromagnetic ferroelectics: **magnetoelectic effect** (electrically driven magnetic memories!)
  - coding of information in sign of the toroidal moment (no stray field around magnetic vortices, thus, minimization of cross-talking)
  - ferroelectic ferroelastics: (sonar – Peter and Jacob Curie)
  - ferromagnetic ferroelasticsi: (magnetostrictive devices)

Classification of multiferroics

1. **Proper (or of the 1st type)**
   Weak direct coupling between different order parameters, e.g., indirect coupling via elastic properties ($BiFeO_3$).
2. **Improper (or of the 2nd type)**
   Strong coupling between order parameters, e.g., compounds in which an electric order appears as the result of appearance of the magnetic order.
3. **Composites** – multiphase granular materials or multilayers (e.g., if we have a piezomagnetic and a piezoelectric component, the coupling between magnetization and polarization can be accomplished via elastic properties)

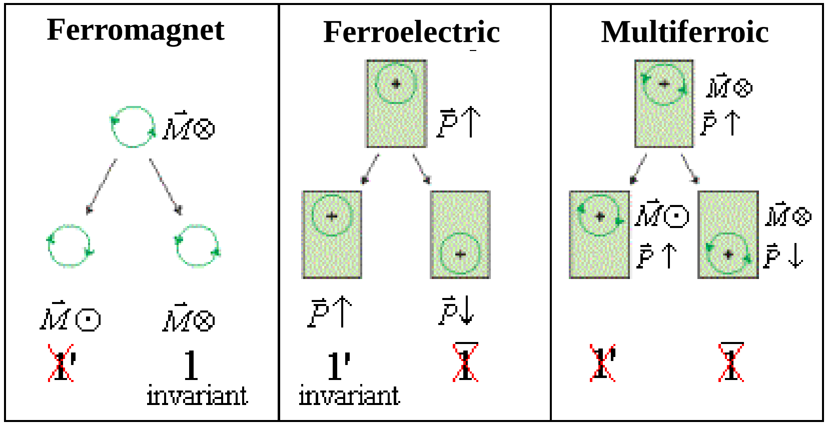


Why are there so few magnetic ferroelectrics?

N.Hill/Spaldin, J. Phys. Chem. B 104, 6694 (2000)

These phenomena appear in materials of different properties and symmetry, thus, they exclude mutually:

- **ferromagnetic ferroelectrics do not have:**
  - time inversion symmetry $(1')$
  - space inversion symmetry $(\bar{1})$.



- **ferroelectrics**
  - dielectrics, **P** is a “normal” vector, no inversion symmetry, invariant to time inversion

- **ferromagnets**:
  - conducting materials or insulators having the inversion symmetry, **M** is a pseudovector (axial vector), which changes sign under time inversion and is invariant to space inversion.

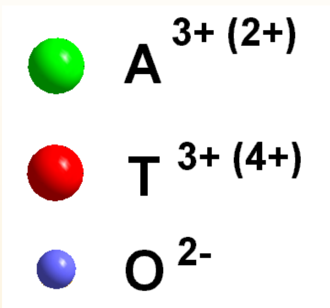
Promising materials: perovskites (ATO_3), T – transition metal

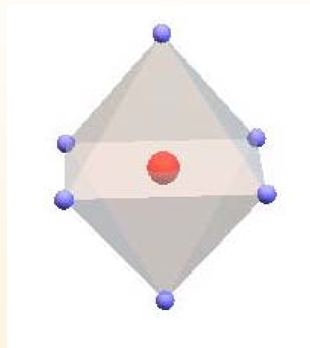
Why are there so few magnetic ferroelectrics?

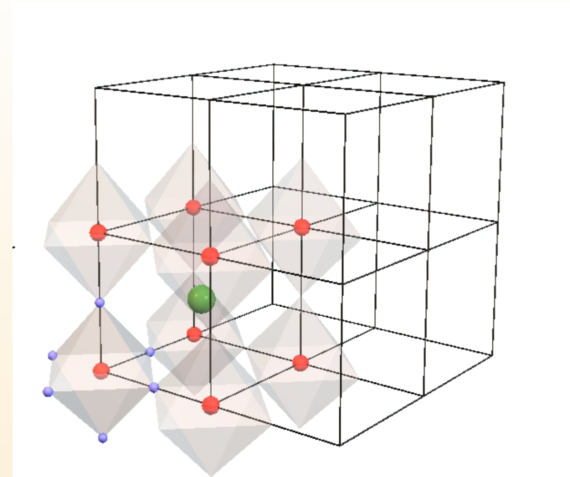
Perovskites – ATO_3

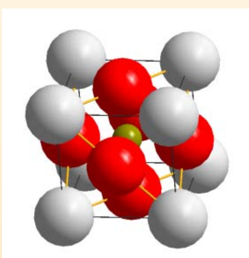
T – transition metal; *A* – RE, Y, alkaline earth (Ca, Sr, Ba)

Cubic structure of the ideal perovskite $(Pm\bar{3}m)$









$$t = \frac{(r_A + r_O)}{\sqrt{2}(r_B + r_O)}$$ tolerance factor (Goldschmidt)

~ 100 magnetic perovskites; ~ 100 frroelectric perovskites

a few perovskites with coexisting magnetic and electric orderings

($BiFeO_3$, $BiMnO_3$, $PbVO_3$)

Why are there so few magnetic ferroelectrics?

Perovskites – ATO_3

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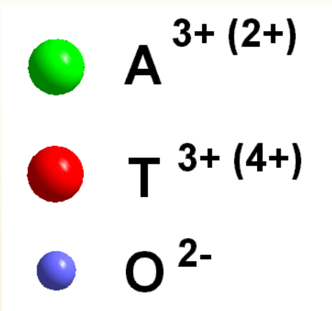
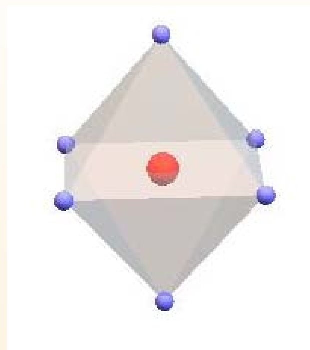
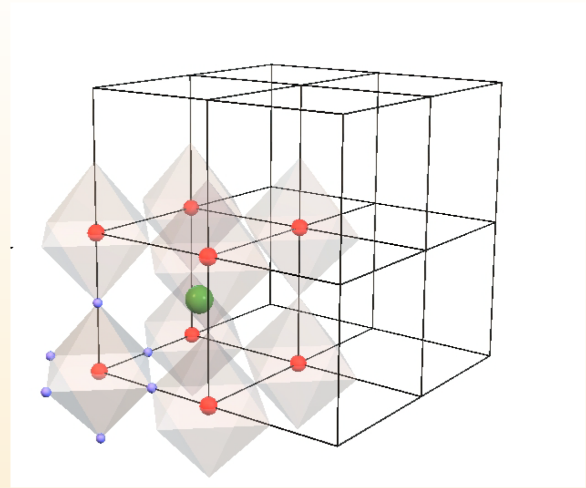




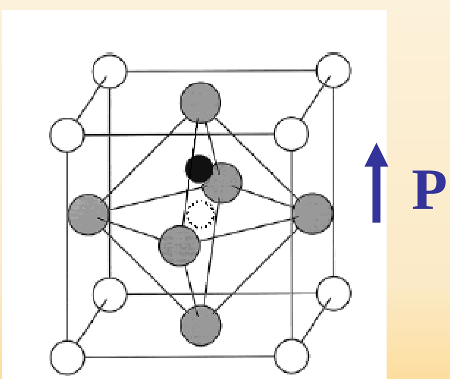


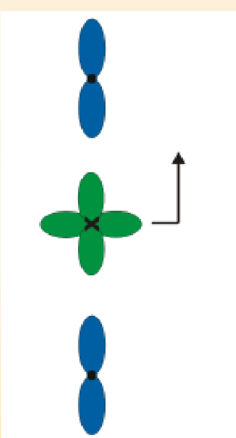
- **experimental finding:**
  - in **ferroelectric** perovskites, the *d* shell of the transition metal *T* is empty (d^0)
  - in **ferromagnetic** perovskites, the *T* ions have a partialy filled *d* shell.

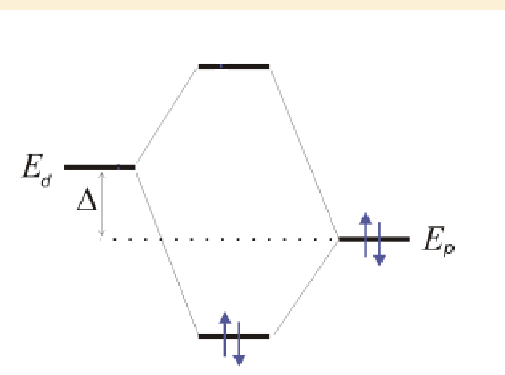
Why are there so few magnetic ferroelectrics?

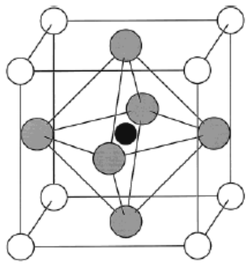
Perovskites – ATO_3 , T – metal przejściowy

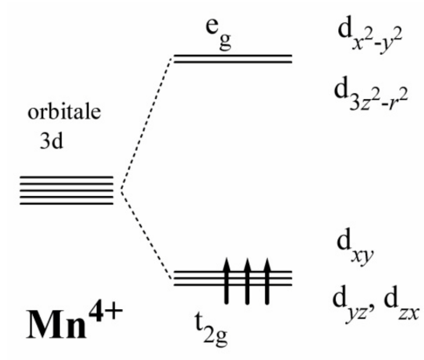
- **experimental finding:**
  - in **ferroelectric** perovskites, *d* shell of the *T* ion is empty (d^0)
  - w **ferromagnetycznych** - jony T mają częściowo zapełnioną powłokę d.
- explanation given by ***ab initio* calculations** (Spaldin/Hill, Khomski, Ederer**):**
  - **LSDA + U** – method taking into account Coulomb interactions between some of localized electrons
  - **LSDA + SIC** – method with correction for self interactions
  - for ***T*** with ***d*** ^{0}, displacement of *T* towards O^{2-} and creation of a covalent bond (*d* orbitals of T and *p* orbitals of O) is energetically convenient. Thus, nonzero **P** appears (ferroelectric perovskites with z Ti^{4+}, Ta^{5+}, W^{6+})

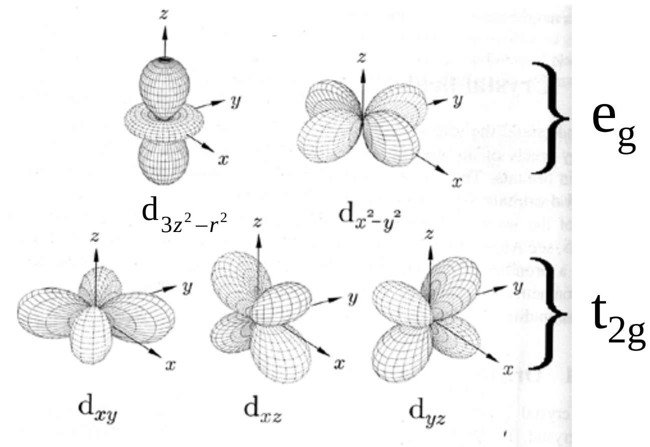




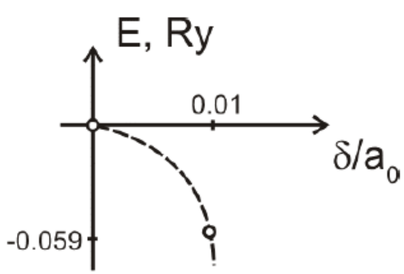




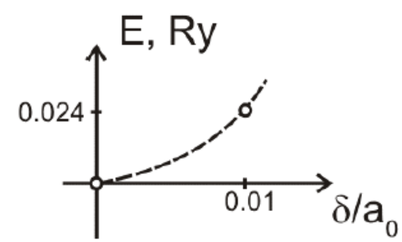




$J_H = 0$



$J_H = 0.8$ eV

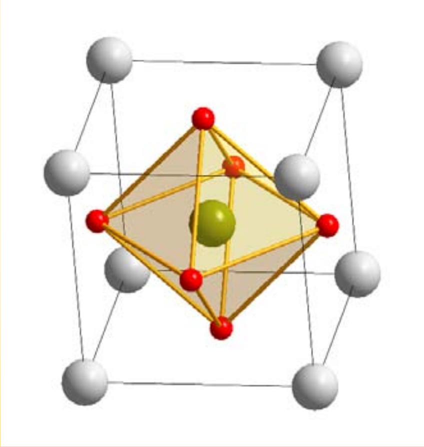


– for ***T*** with **partially filled *d* shell**, intraatomic exchange interaction (Hund's coupling) eliminates this mechanism of appearance of nonzero polarization and **ferroelectric ordering** (np. $CaMnO_3$, $RCrO_3$)

Why do magnetic ferroelectrics exist at all?

- **Perovskites (*AT*O$_3$):**
  1. **„Paramagnetic doping"** – G. Smolenskii, Y.Venevtsev, partial replacement of *T* d^0 ions with magnetic d^n ions

     $PbFe^{3+}_{1/2}Nb^{5+}_{1/2}O_3$ T_{FE}= 387 K, T_N=134 K, weak **FE**-**AM** coupling



Why do magnetic ferroelectrics exist at all?

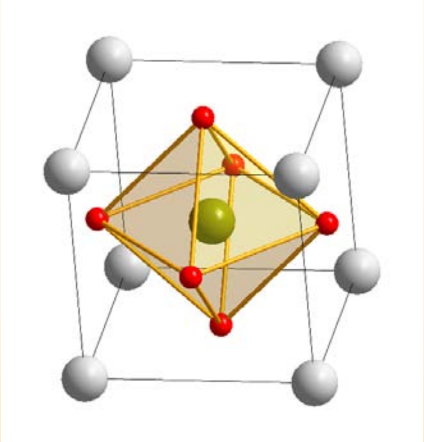
- **Perovskites (*A*TO_3):**

2. „Lone pair of s^2 electrons" – Bi (dangling bond)

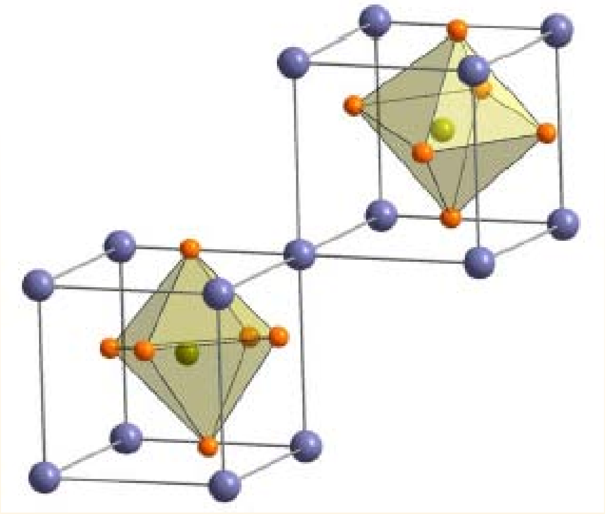
$BiMnO_3$ T_{FE}= 760 K, T_C=105 K

$BiFeO_3$ T_{FE}= 1100 K, T_N=643 K

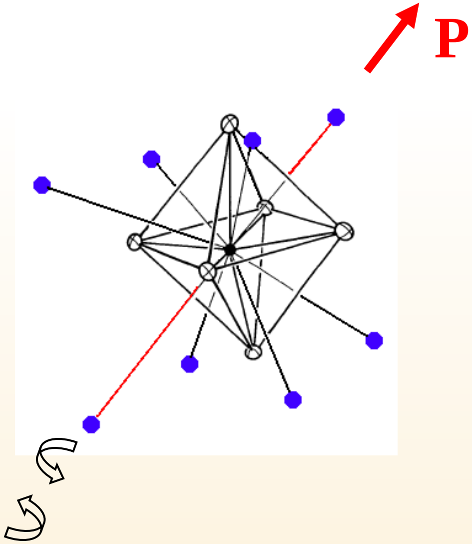
- Ionic bonds are made of 6p^3 electrons of bizmuth
- Remain 6s^2 electrons that do not participate in bonds
- Hybridization the *s* states and (empty) *p* states is the source of a large polarizability of Bi.
- It facilitates distortions of the crystalline structure and the appearance of **FE** ordering (Spaldin)
- Bi sublattice is responsible for the appearance of **FE** ordering $P \sim 6 - 150$ μC/cm^2
- 3*d* ions sublattice is responsible for the magnetic ordering.
- Thus, **m-e** coupling is not especially strong.



$BiFeO_3$ - crystalline structure – Kubel&Schmid, Acta Cryst. B46, 698 (1990)



$$Pm\bar{3}m \quad \rightarrow \quad R3c$$



- rotation of octahedra around <111> axis by ±13.8°
- distortion of the octahedra
- **displacement of Fe ions** from the centers of octahedra
- displacement of Bi ions along <111> axis
- **ferroelectric polarization** – result of existence of two different Bi-Fe distances along the <111> axis and of electron contribution

Why do magnetic ferroelectrics exist at all?

- **Perovskites (*A*TO$_3$):**

  3. **„Extension (stretching) of the oxygen octahedron"** - **$Sr_{1-x}Ba_xMnO_3$**

     - Sakai, …, Y. Tokura, *Phys Rev. Lett.* 107, 137601 (2011)
     - Pratt, …, B. Dabrowski, *Phys. Rev. B* 90, 140401 (2014)

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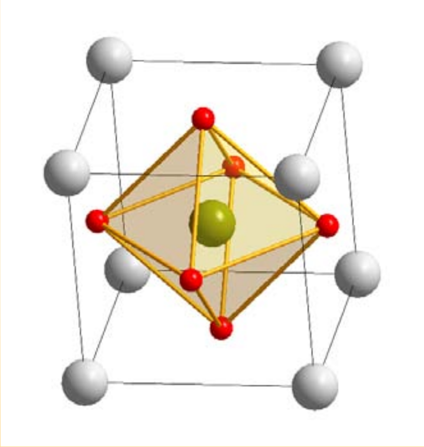

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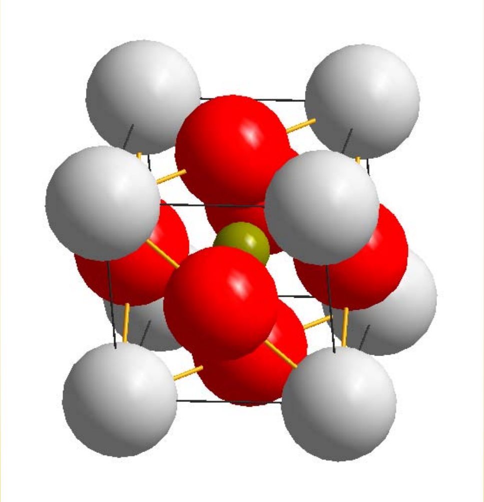

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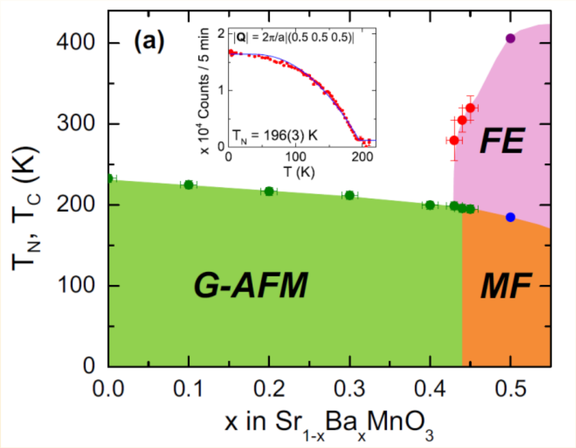

High temperature phase – cubic Pm-3m
FE phase – tetragonal

Results:

- Multiferroic phase (Pm-3m), MF, appears at ~285 K for $\boldsymbol{x \geq 0.44}$.
- The phase transition (~400 K) to the ferroelectric, FE, phase, smeared over ca 30 K, is not noticeable in the temperature dependence of specific heat
- The phase transition to the antiferromagnetic, AF, phase damps the tetragonal deformation and diminishes the polarization.

Why do magn

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     - Sakai, …, Y. Tokura
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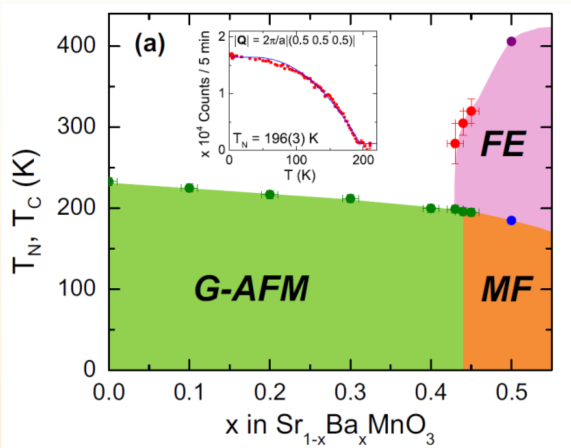
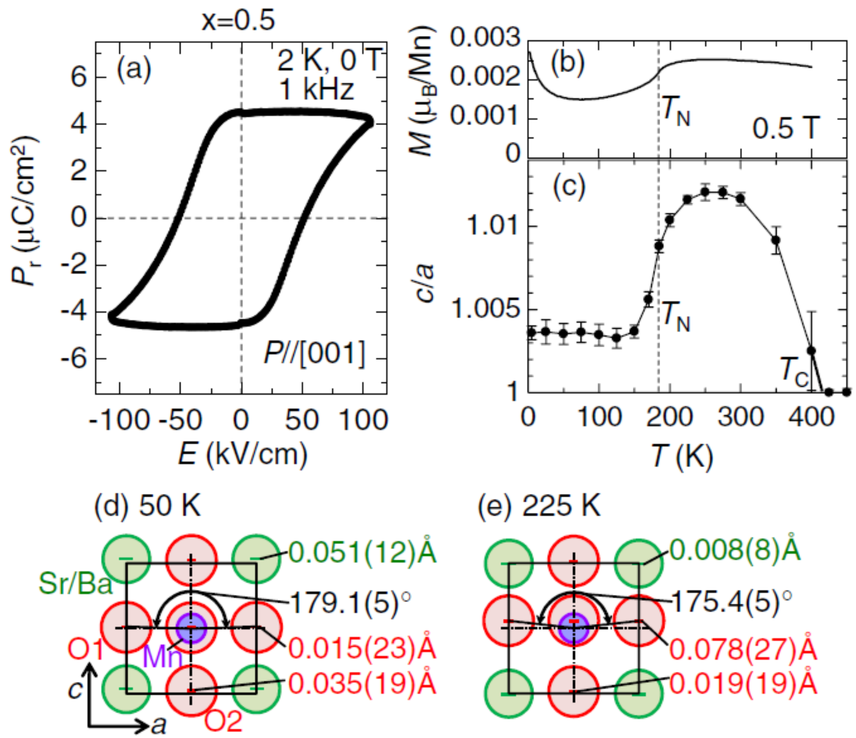


Sr$_{0.5}$Ba$_{0.5}$MnO$_3$



Sakai *et al*.., *Phys Rev. Lett.* 107, 137601 (2011)

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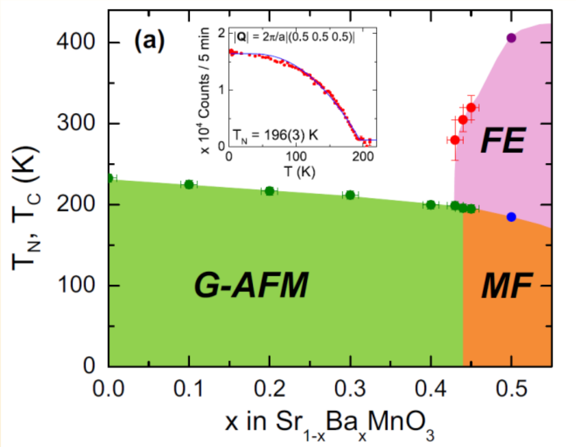

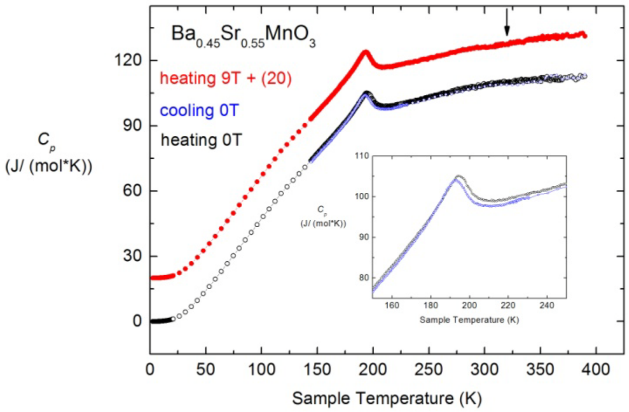

J. Więckowski, M. U. Gutowska, A. Szewczyk i in.

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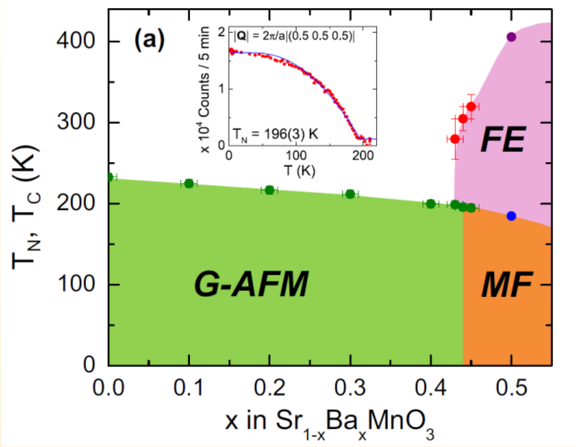

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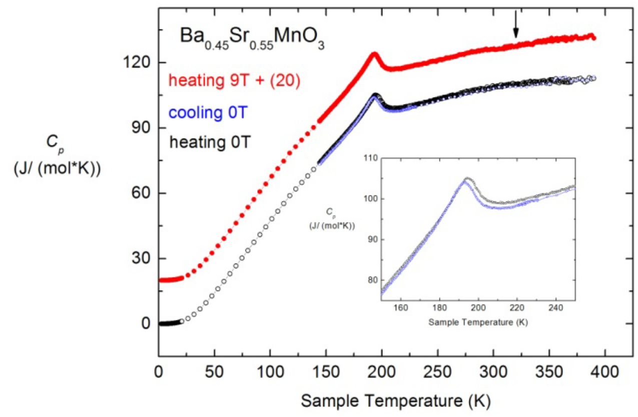

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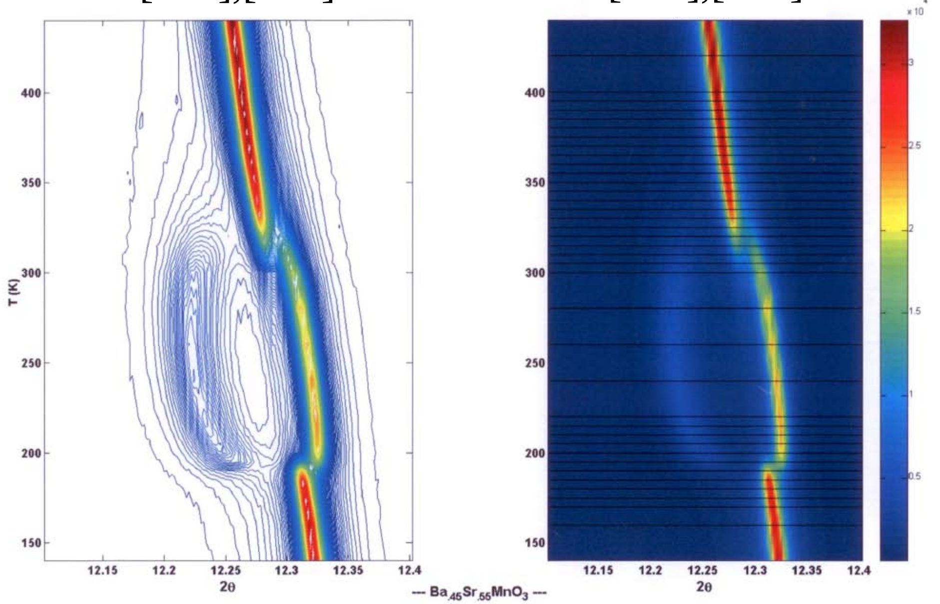

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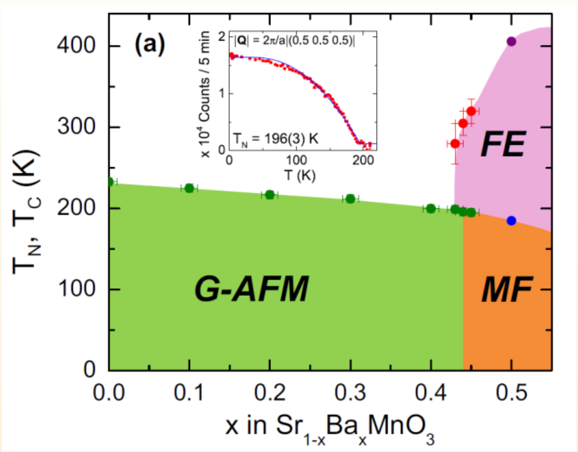

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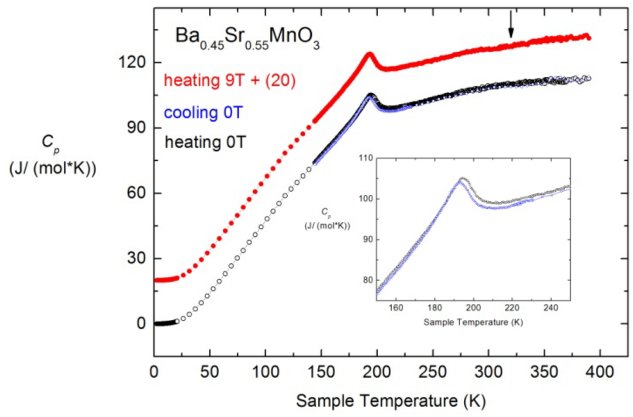

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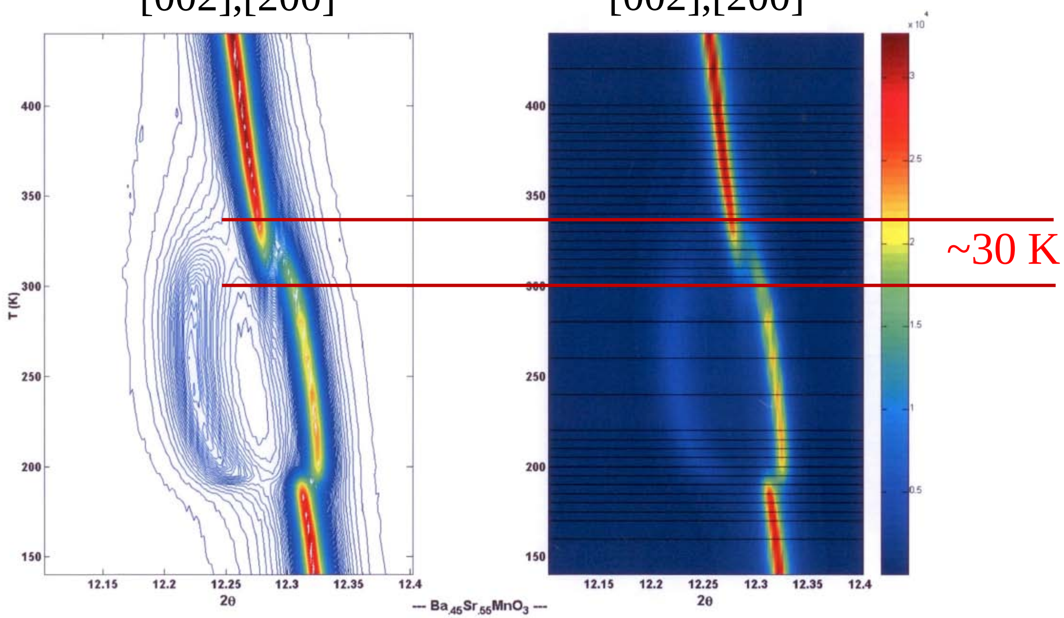


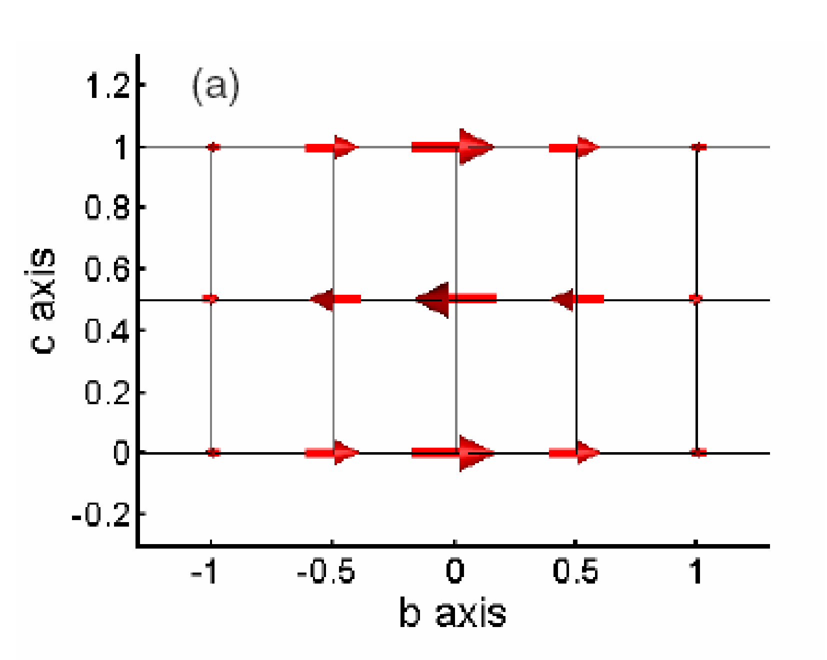
B. Dabrowski i in.

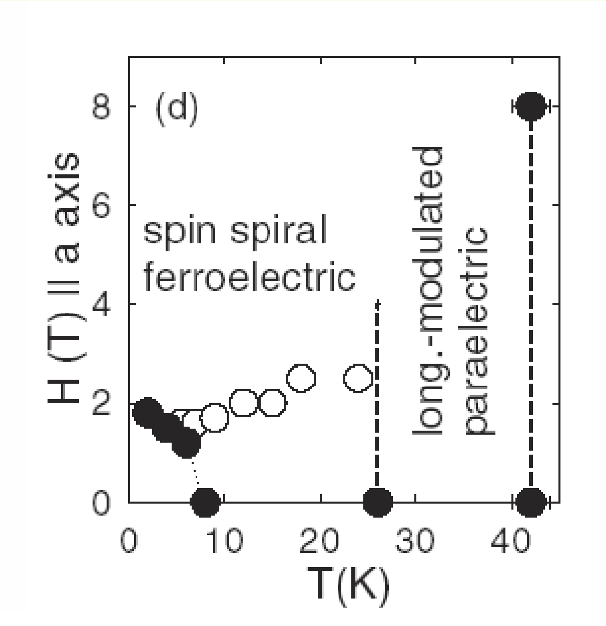
Why do magnetic ferroelectrics exist at all?

- **Perovskites (*ATO*$_3$):**
  1. **„Paramagnetic doping"** - **$PbFe^{3+}_{1/2}Nb^{5+}_{1/2}O_3$**
  2. **„Lone pair of s^2 electrons"** - **$BiFeO_3$**
  3. **„Stretching of the oxygen octahedron"** - **$Sr_{1-x}Ba_xMnO_3$**
  4. **„FE state induced by magnetic ordering"** - **$TbMnO_3$**

$TbMnO_3$

- at room temperature, paraelectric, paramagnetic phase;
- at T_N = 41 K – incommensurate, longitudinally modulated, antiferromagnetic, sinusoidal structure; inversion symmetry present, thus, paraelectric phase;





Kenzelmann et al., PRL **95**, 87206 (2005).
Cheong et al., Nature Mat. **6**, 13 (2007)

$TbMnO_3$

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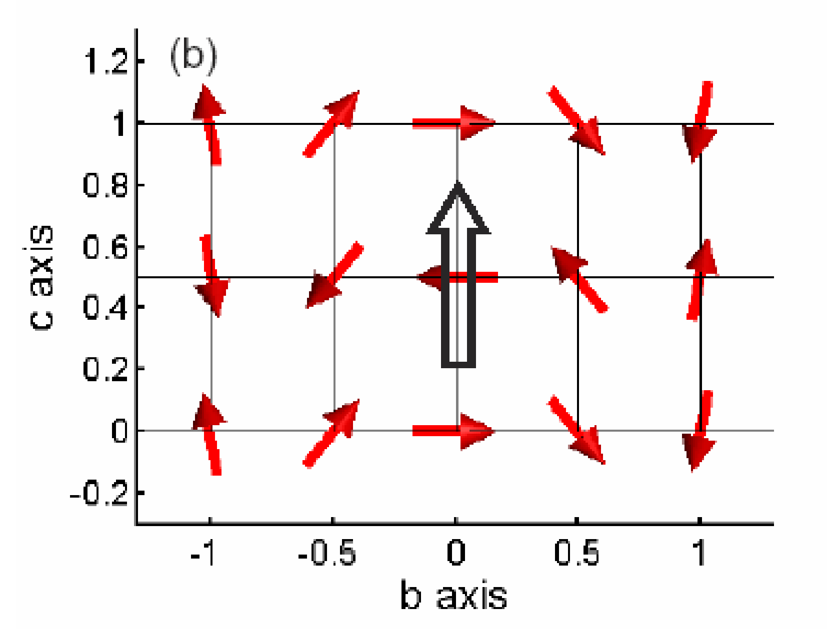

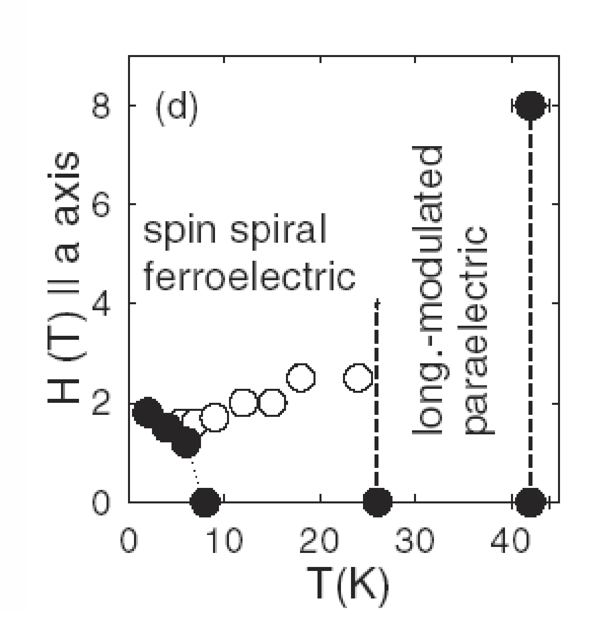

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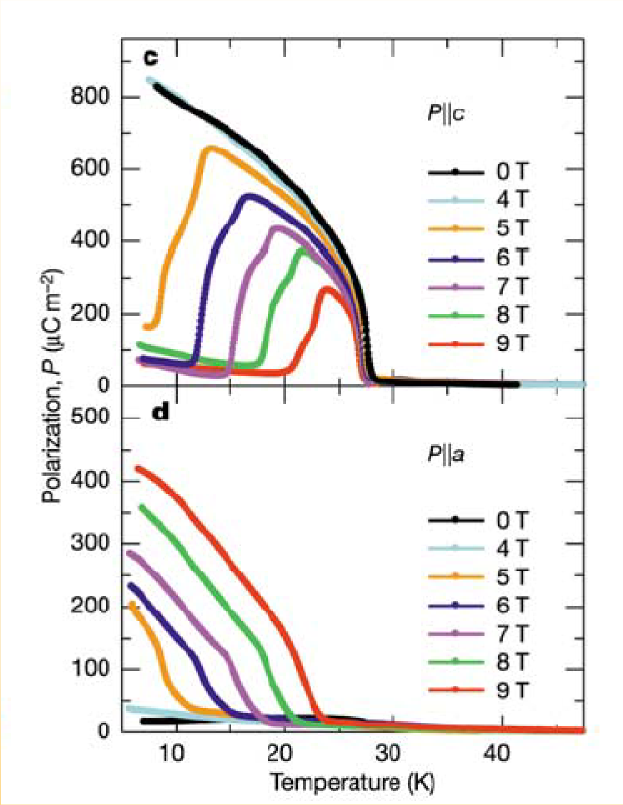


B||b

Kimura et al., Nature **426**, 55 (2003).

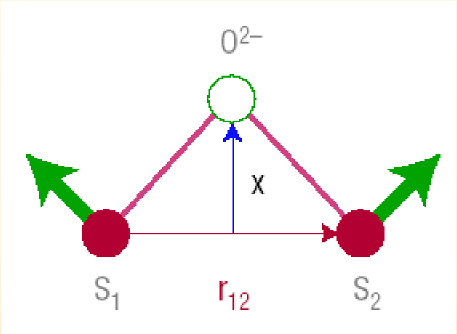
$TbMnO_3$

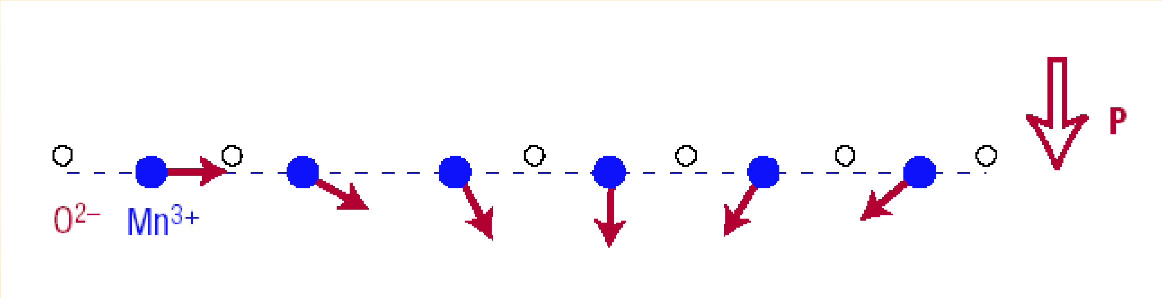
- at room temperature, paraelectric, paramagnetic phase;
- at T_N = 41 K – incommensurate, longitudinally modulated, antiferromagnetic, sinusoidal structure; inversion symmetry present, thus, paraelectric phase;
- at T_{loc}= 27 K, the phase transition to the incommensurate spiral structure without inversion; ferroelectric polarization (~0.08 $\mu C/cm^2$ w T =10 K) found.

What is a physical mechanism responsible for the appearance of the polarization?

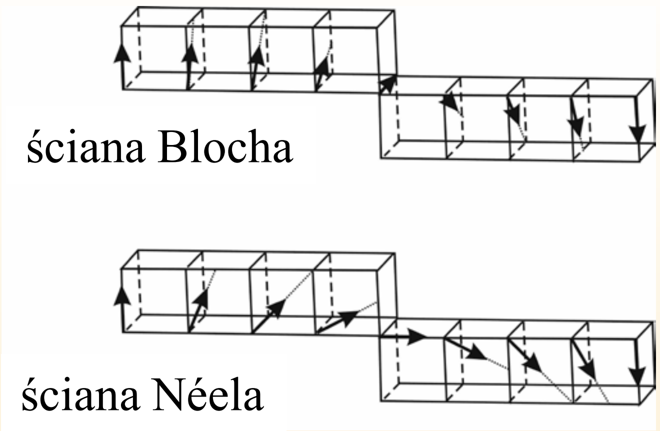
Dzyaloshinski-Moriya coupling

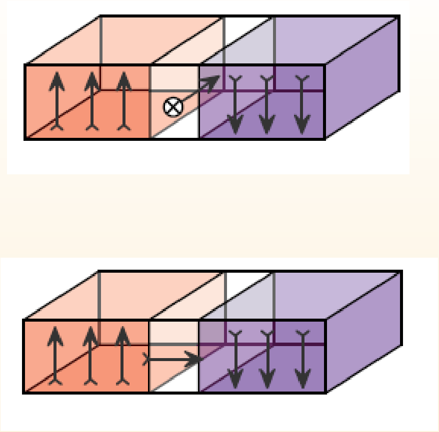
$$\vec{D} \cdot \left(\vec{S}_i \times \vec{S}_j \right) \qquad \vec{D} \sim \vec{r}_i \times \vec{r}_j$$

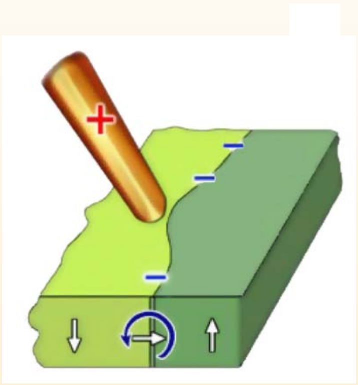




Spiral structure is present *per se* in domain walls







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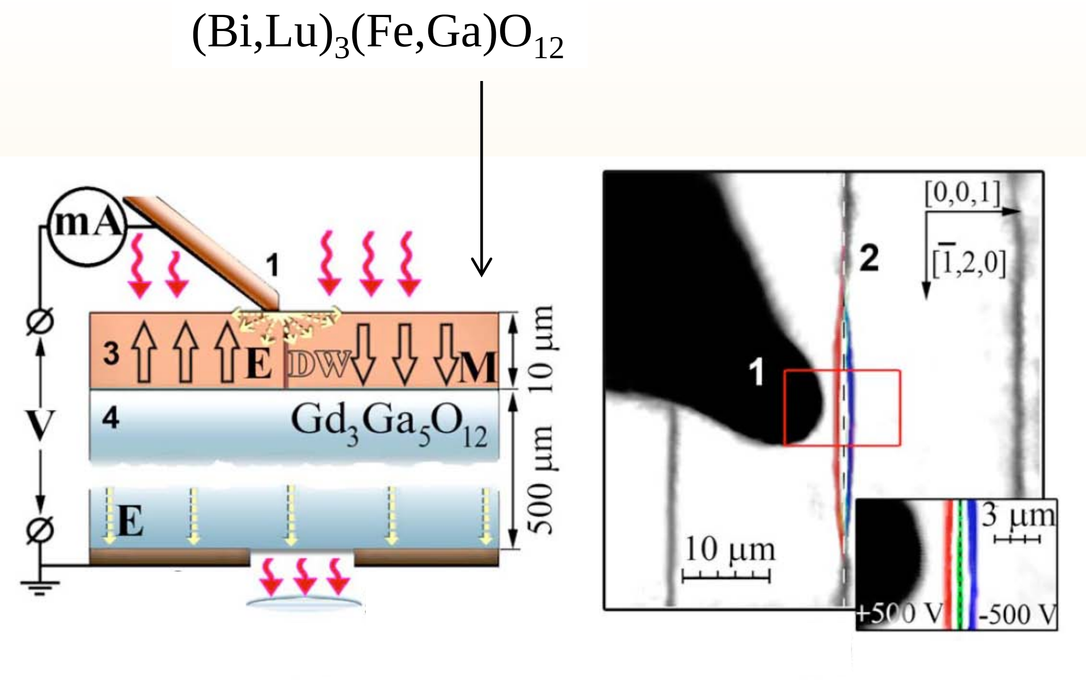


A.S. Logginov, G.A. Meshkov, A.V. Nikolaev, E.P. Nikolaeva, A.P. Pyatakov, and A.K. Zvezdin *Appl. Phys. Lett.* **93**, 182510 (2008).

Olivine $LiNiPO_4$

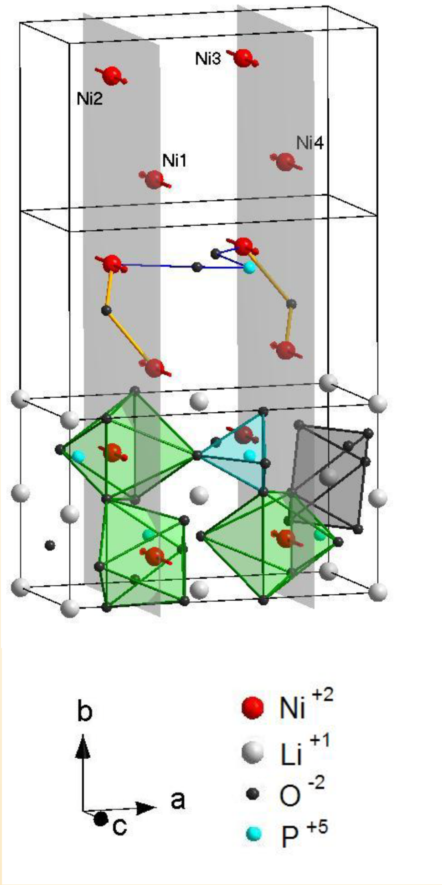
Properties of olivines $LiTPO_4$

- Strong coupling of the *T* ions in (100) planes (superexchange *T*-O-*T*)
- Weak coupling between (100) planes (superexchange *T*-O-P-O-*T*)
- Strong magnetocrystalline anisotropy (easy axis different for different *T* ions)
- Quasi – two-dimensional Ising system
- Antiferromagnetic order appears at low temperatures ($T_N < 50$ K)
- Large ionic conductivity due to Li^+ (promising cathode materials for Li-ion batteries)
- **Very strong linear magnetoelectric effect**
  ($P_a = \alpha_{ab} H_b$ dla T = Co $|\alpha_{yx}(4.2\text{ K})| = 30.6$ [ps/m]),

  but

no spontaneous dielectric polarization was found till now

Structure of olivines, space group Pnma



Olivine $LiNiPO_4$

- Quasi – two-dimensional Isinga system – ***c*** is the easy magnetization axis
- Antiferromagnetic order develops in two steps, on lowering temperature :
  - 2nd order transition: paramagnetic – incommensurate phase (modulated , antiferromagnetic) T_{N1}=21.8 K
  - 1st order transition: incommensurate – antiferromagnetic phase, T_{N1}=20.9 K
- **Specific heat** studies *(M. Gutowska, S. Lewińska, A. Szewczyk, T. Zajarniuk et al.)*

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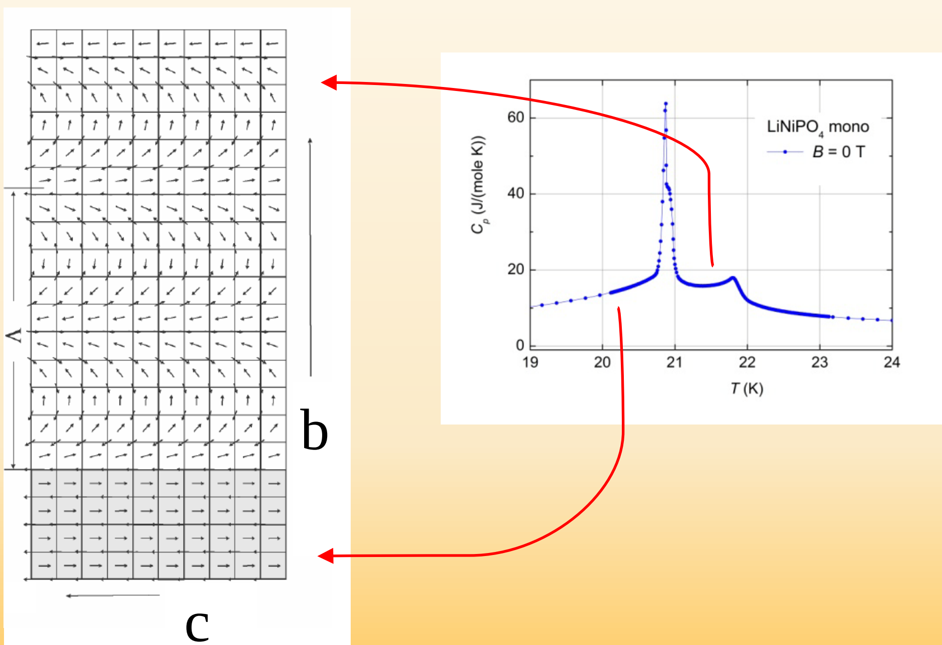
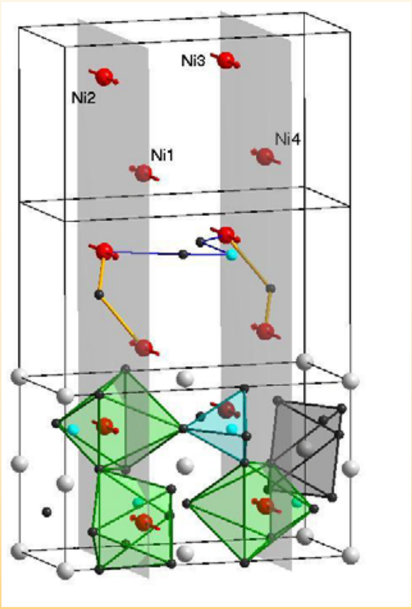
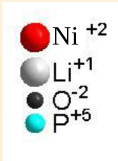
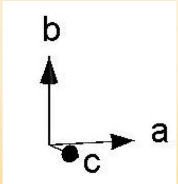


Pnma - space group of olivines

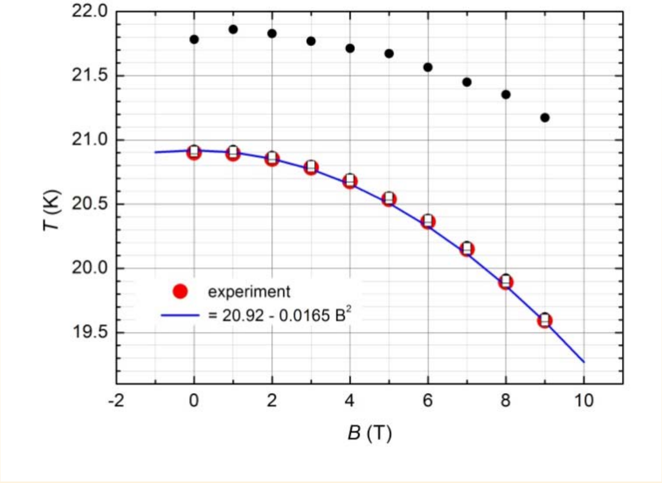




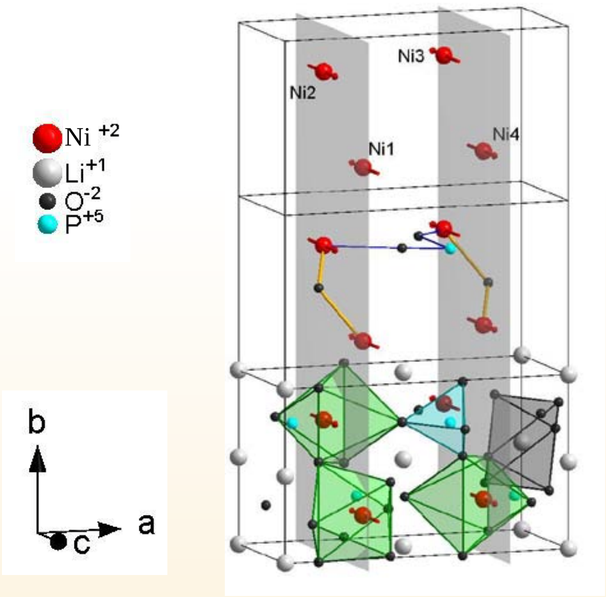


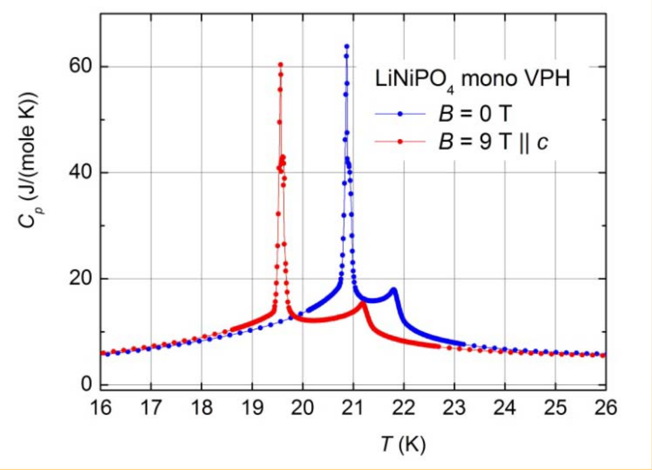
Olivine $LiNiPO_4$ specific heat

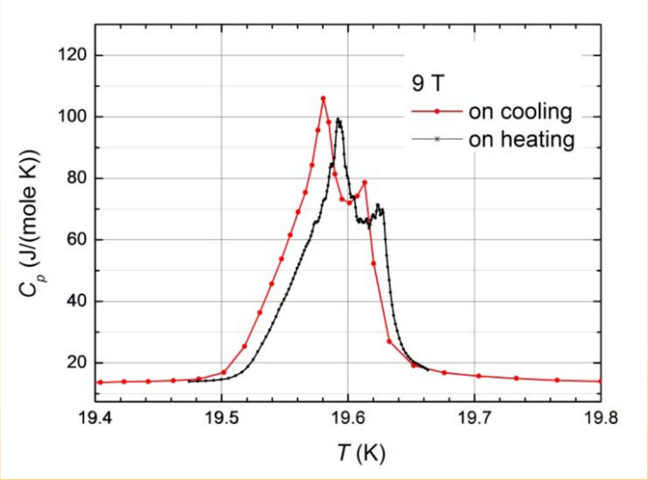
Magnetic field || to *a* and to *b* does not influence position of the anomalies (!)



Field || do *c*







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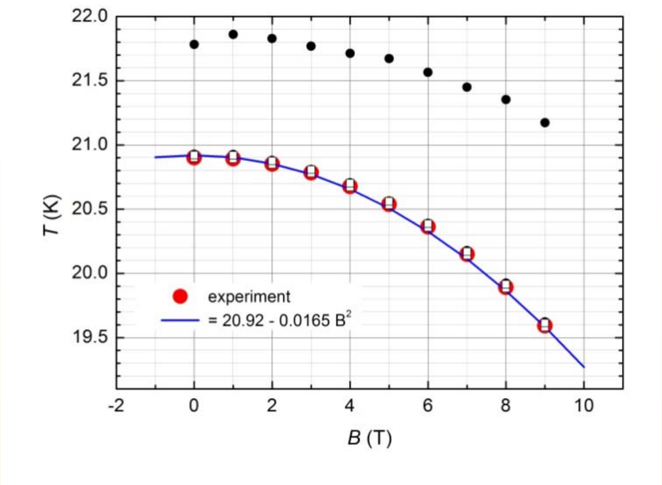

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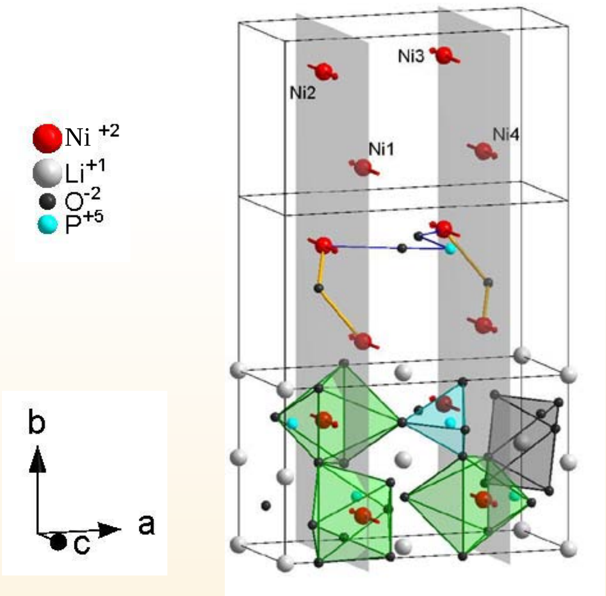
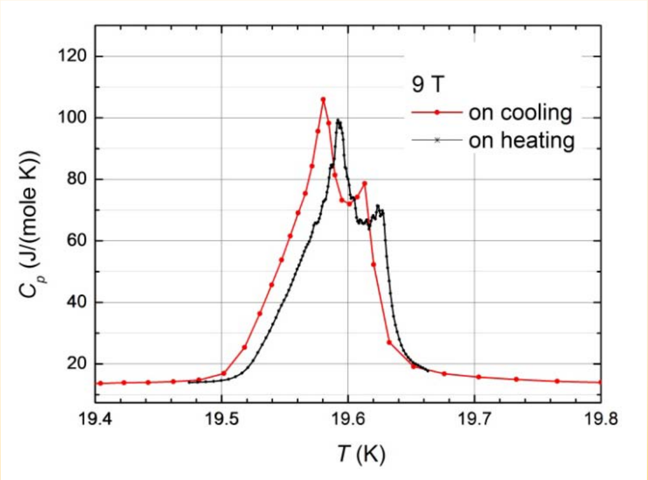


Are these **two coupled transitions in two coupled subsystems?**



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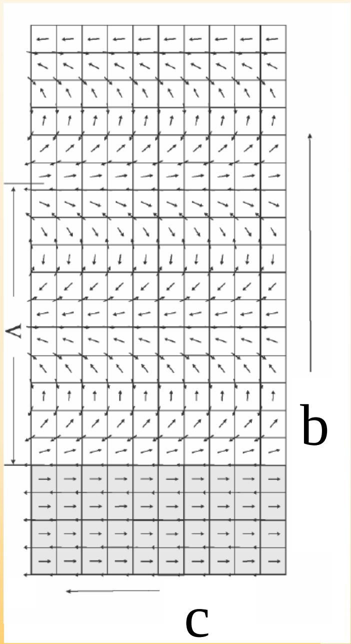
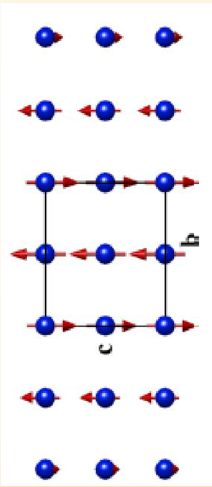
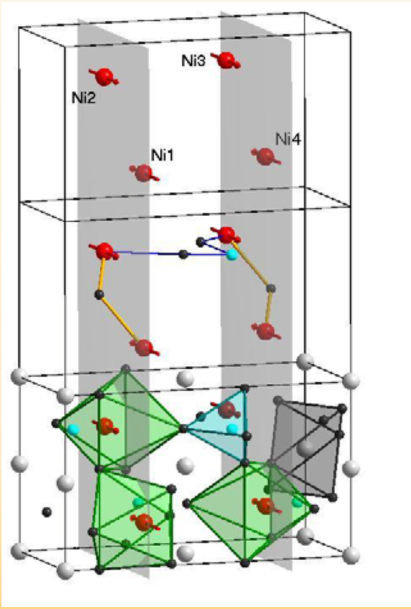
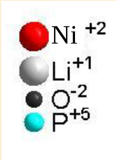


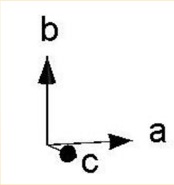
Jensen et al. Phys. Rev. B 79, 092412 (2009).





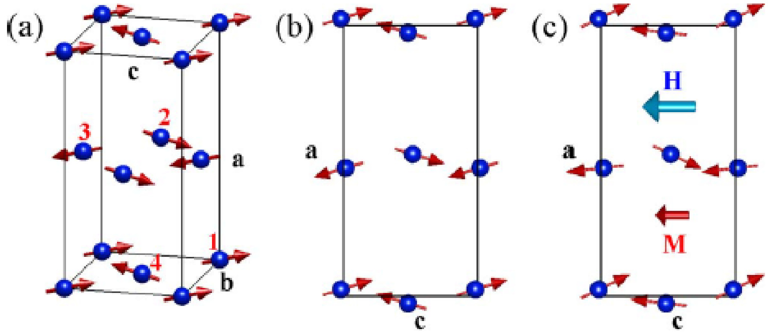
Pnma - grupa przestrzenna oliwinów





C

- Quasi – two-dimensional Isin
- Antiferromagnetic order deve
  - 2^{nd} order transition: param antiferromagnetic) T_{N1}=21
  - 1^{st} order transition: incomm
- **Specific heat** studies *(M. Gu*)



A more complicated structure without inversion, presence of the polarization allowed in B ≠ 0

Vaknin et al. Phys. Rev. Lett. 92, 207201 (2004).

Jensen et al. Phys. Rev. B 79, 092412 (2009).

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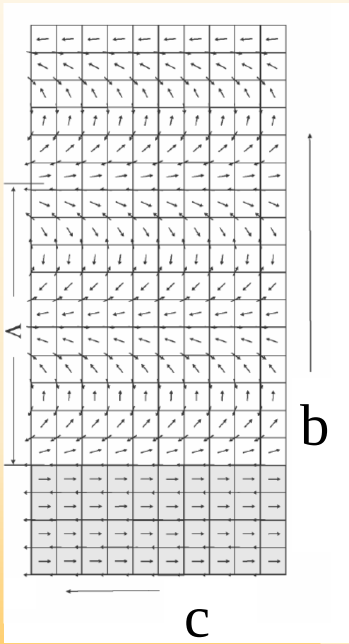
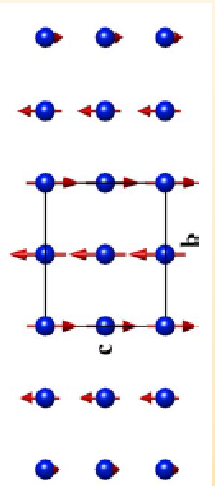
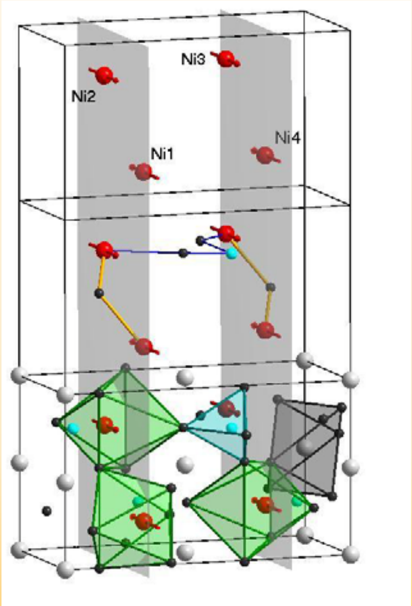
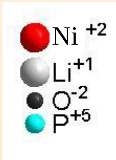
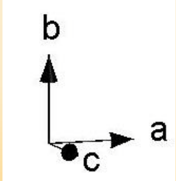


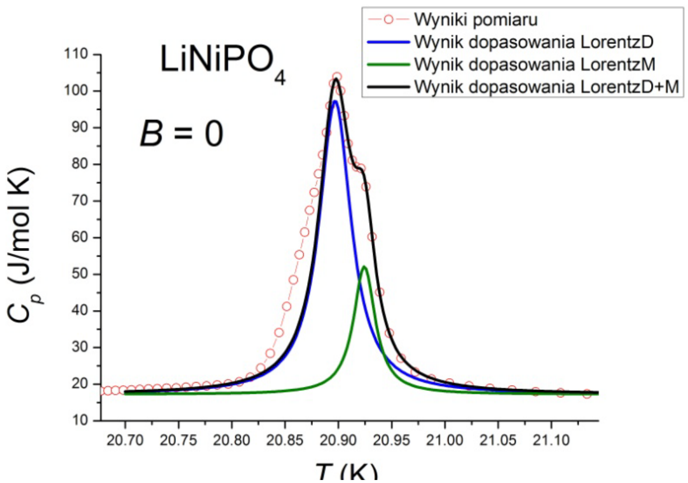


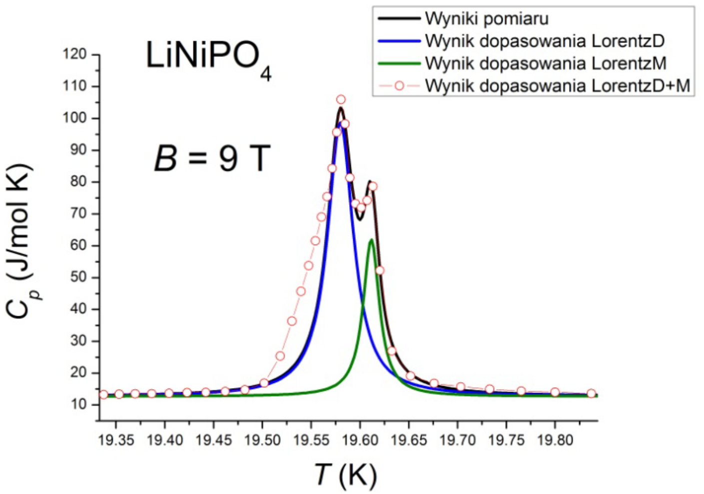


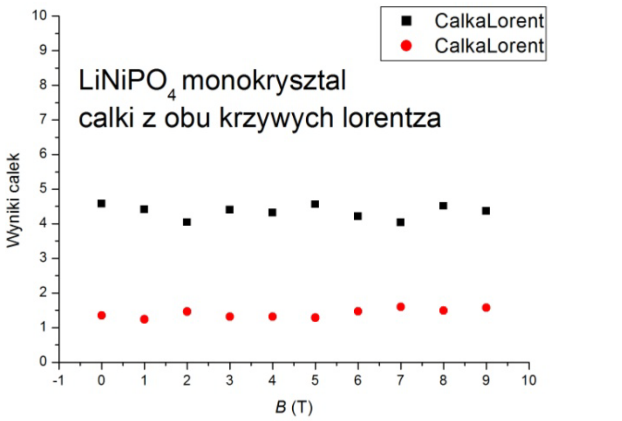


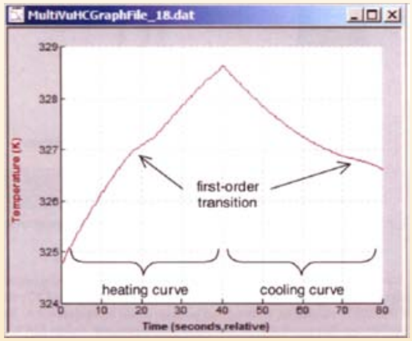


Olivine $LiNiPO_4$ – slope analysis









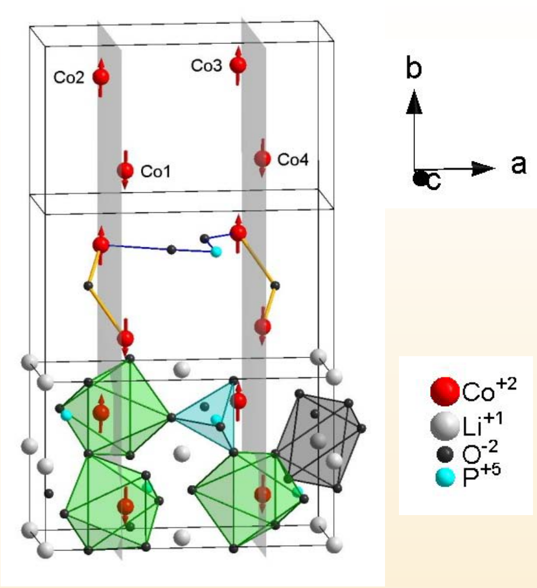
Slope analysis allows to put forward a hypothesis that the 1st order phase transition to the commensurate antiferromagnetic phase is coupled to a transition to a ferroelectric phase. Thus, $LiNiPO_4$ is **a multiferroic (?)**

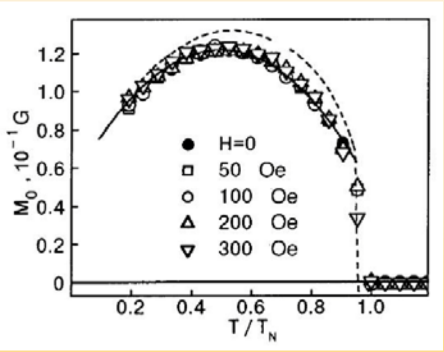
Toroidal ordering - olivine $LiCoPO_4$

- Co^{2+} in „corrugated” (100) planes
- Strong coupling between Co^{2+} ions in (100) planes
- Strong magnetic anisotropy (with *b* easy axis)
  [Vaknin et al., Phys. Rev. B 65, 224414 (2002)]
- **Quasi – two-dimensional Ising system**
- Antiferromagnetic order at T_N= 21.6 K
- Co^{2+} magnetic moments **deflected from the b axis** by 4.6° (within the b-c plane)
  [Vaknin et al., Phys. Rev. B 65, 224414 (2002)]
- **A small spontaneous net magntization exists** (directed along the b axis !!!)
  [Kharchenko et al. (among them R. Szymczak), Low Temp.Phys. 28, 646 (2002)]

conclusion: magnetic symmetry is not orthorhombic but monoclinic - $P12_1'1$

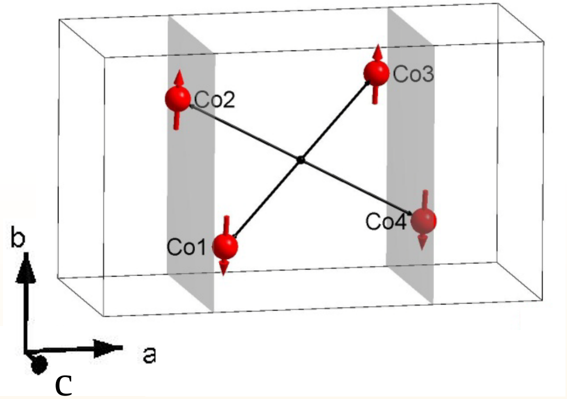
Pnma – space group of olivines





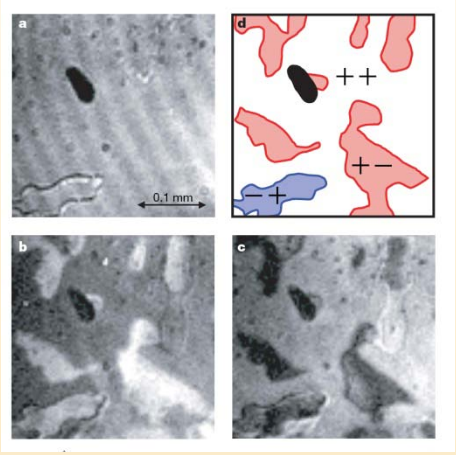
Uporządkowanie toroidalne - oliwin $LiCoPO_4$

- In the $P2'_11$ symmetry **there can exist:**
  - net magnetization along the *b* axis
  - modulated structures ($\perp$ *b* $\mathbf{M}_n$ components)
  - nonzero **toroidal moment** $\mathbf{T} = \frac{1}{2}\sum_n \mathbf{r}_n \times \mathbf{M}_n$
  - nonzero dielectric polarization



- The nonzero toroidal moment was found based on neutron studies) *[Van Aken et al., Nature **449**, 702 (2007); Vaknin et al., Phys. Rev. B 65, 224414 (2002)]*

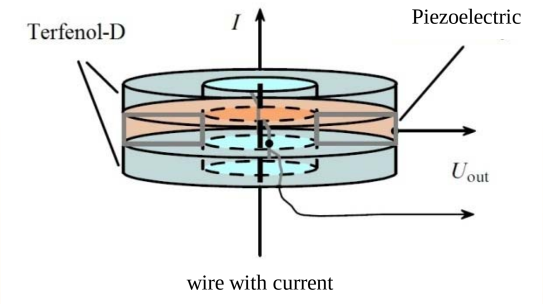
- Based on the optical studies (second harmonic generation), **4 different domain states were found** and interpreted as 2 weakly ferromagnetic domains, each of which is divided into 2 torroidic domains, differing in sign of **T**. This was claimed to be the **first observation of toroidic domains** *[Van Aken et al., Nature **449**, 702 (2007) **120 citations**].* Kharchenko, and Schmid (*J.Phys. Condens. Matter 20, 434201 (2008)*) proved **this interpretation to be erroneous!**



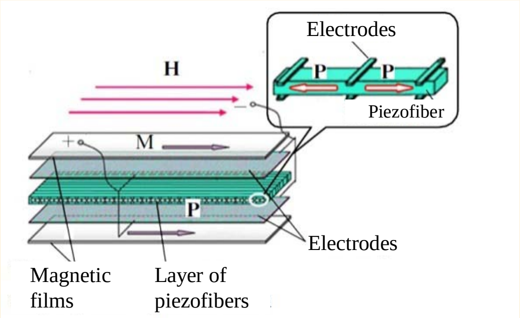
Ideas of application of magnetoelectrics and multiferroics

(*review paper: A.V. Pyatakov, A.K. Zvezdin, Physics-Uspekhi Fiz. Nauk 182, 593 (2012)).*

- Magnetic field sensors (the most advanced idea)



Terfenol-D = alloy $Tb_xDy_{1-x}Fe_2$ (x ~ 0.3)



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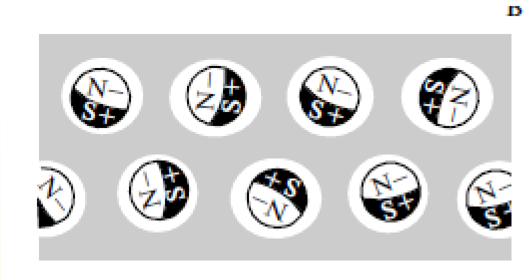

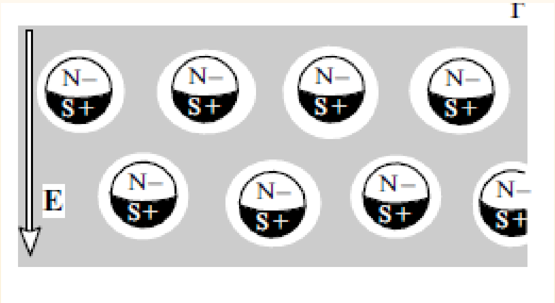

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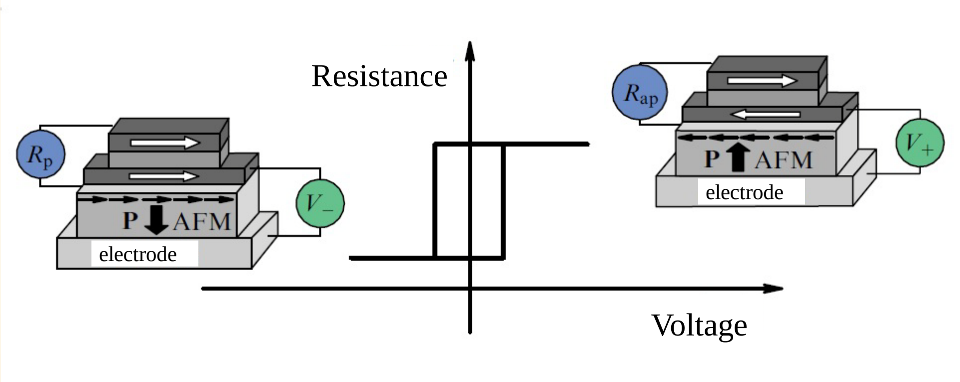

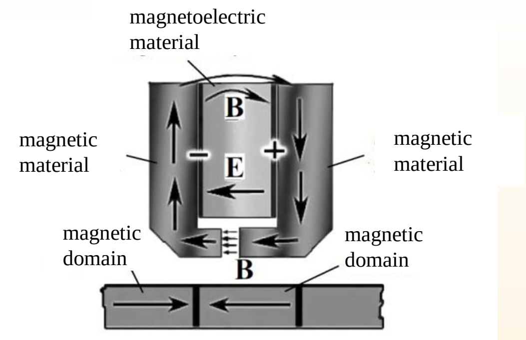

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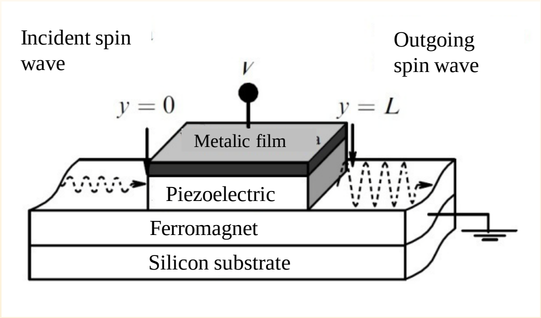

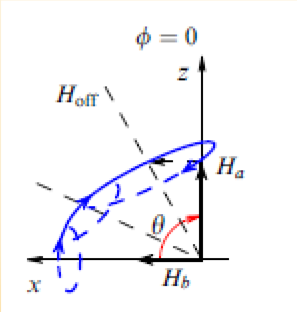

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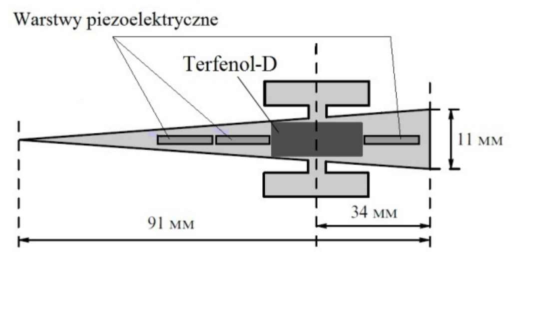
- Suppliers for wireless net of sensors. Idea of an element drawing energy from a variable electromagnetic field and converting this energy into energy of a bank of capacitors.



Conclusions

- Multiferroics are interesting, scarse materials.
- From the classical point of view, conditions necessary for appearance of a ferromagnetic (d^n) and of a ferroelectric order (d^0) exclude each other.
- However, there are several physical mechanisms leading to the coexistence of these orderings in a one material, i.e., leading to the existence of multiferroics (e.g. multiferroic perovskites.
- Multiferroics in which ferrotoroidic order is one of the long-range orderings are particularly interesting.
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Thank you for your attention!