Coupled electricity and magnetism: magnetoelectrics, multiferroics and all that

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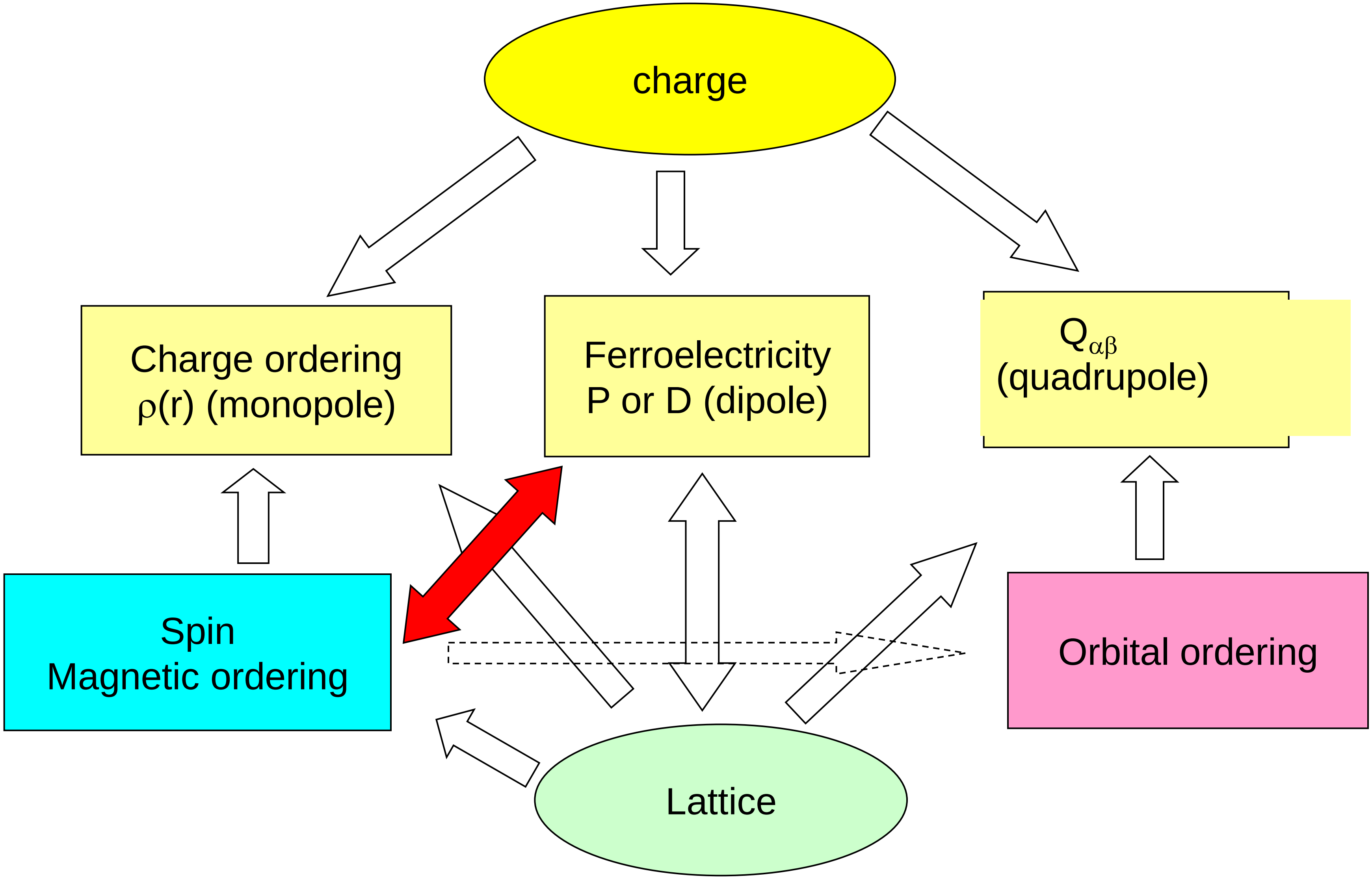
- Introduction
- Magnetoelectrics
- Multiferroics; microscopic mechanisms
- Currents, dipoles and monopoles in frustrated systems
- Magnetic textures: domain walls, vortices, skyrmions
- Dynamics; multiferroics as metamaterials
- Conclusions

Degrees of freedom



Maxwell's equations

$$\nabla \cdot \mathbf{E} = 4\pi\rho_{\mathrm{e}}$$

$$\nabla \cdot \mathbf{B} = 0$$

$$-\nabla \times \mathbf{E} = \frac{1}{c}\frac{\partial \mathbf{B}}{\partial t}$$

$$\nabla \times \mathbf{B} = \frac{1}{c}\frac{\partial \mathbf{E}}{\partial t} + \frac{4\pi}{c}\mathbf{j}_{\mathrm{e}}$$

Magnetoelectric effect

$$M_i = \sum \alpha_{ij} E_j + \sum \beta_{ijk} E_j E_k + \ldots$$

$$P_i = \sum \alpha_{ij} H_j + \sum \beta_{ijk} H_j H_k + \ldots$$

Coupling of electric polarization to magnetism

Time reversal symmetry

$$\mathbf{P} \rightarrow +\mathbf{P}$$

$$\mathbf{M} \rightarrow -\mathbf{M}$$

$$t \rightarrow -t$$

Inversion symmetry

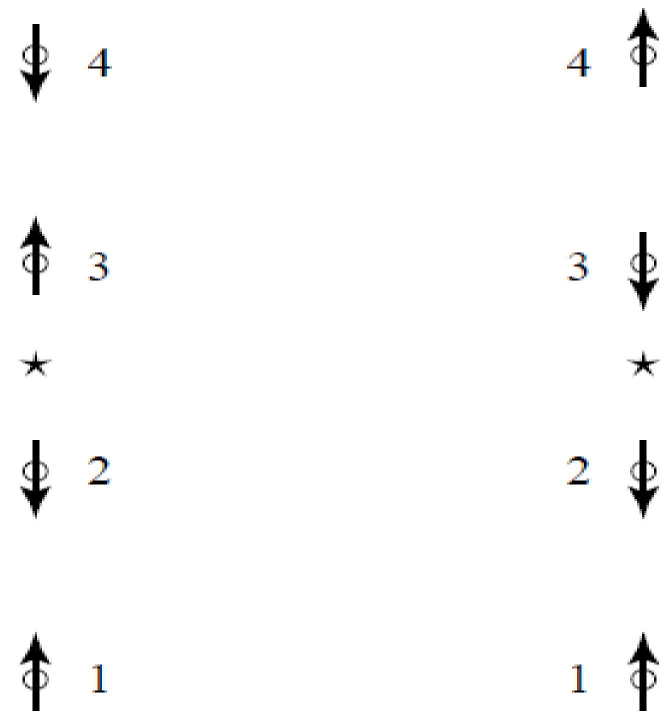
$$\mathbf{P} \rightarrow -\mathbf{P}$$

$$\mathbf{M} \rightarrow +\mathbf{M}$$

$$\mathbf{r} \rightarrow -\mathbf{r}$$

$$F \propto \alpha H E$$

For linear ME effect to exist, both inversion symmetry and time reversal invariance has to be broken



(a) Cr_2O_3 (b) Fe_2O_3

In Cr2O3 inversion is broken --- it is linear magnetoelectric

In Fe2O3 – inversion is not broken, it is not ME (but it has weak ferromagnetism)

Magnetoelectric coefficient α_{ij} can have both symmetric and antisymmetric parts

Symmetric: $\alpha_{ij} = \begin{pmatrix} \alpha_{xx} & 0 & 0 \\ 0 & \alpha_{yy} & 0 \\ 0 & 0 & \alpha_{zz} \end{pmatrix}$ Then

$\mathbf{P}_i = \alpha_{ij} \mathbf{H}_i$; along main axes $\mathbf{P} \parallel \mathbf{H}$, $\mathbf{M} \parallel \mathbf{E}$

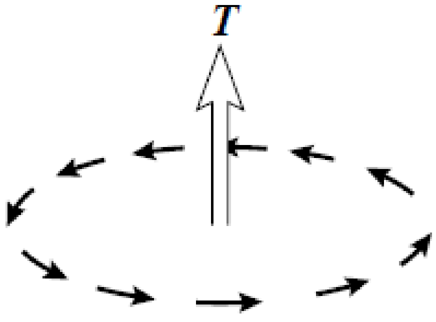
For antisymmetric tensor α_{ij} one can introduce a dual vector $T_i = \varepsilon_{ijk}\alpha_{jk}$

T is the **toroidal moment** (both P and T-odd). Then $\mathbf{P} \perp \mathbf{H}$, $\mathbf{M} \perp \mathbf{E}$,

$\mathbf{P} = [\mathbf{T} \times \mathbf{H}]$, $\mathbf{M} = -[\mathbf{T} \times \mathbf{E}]$

For localized spins $\boldsymbol{T} = \sum_i \boldsymbol{r}_i \times \boldsymbol{S}_i$

For example, toroidal moment exists in a magnetic vortex



MULTIFERROICS

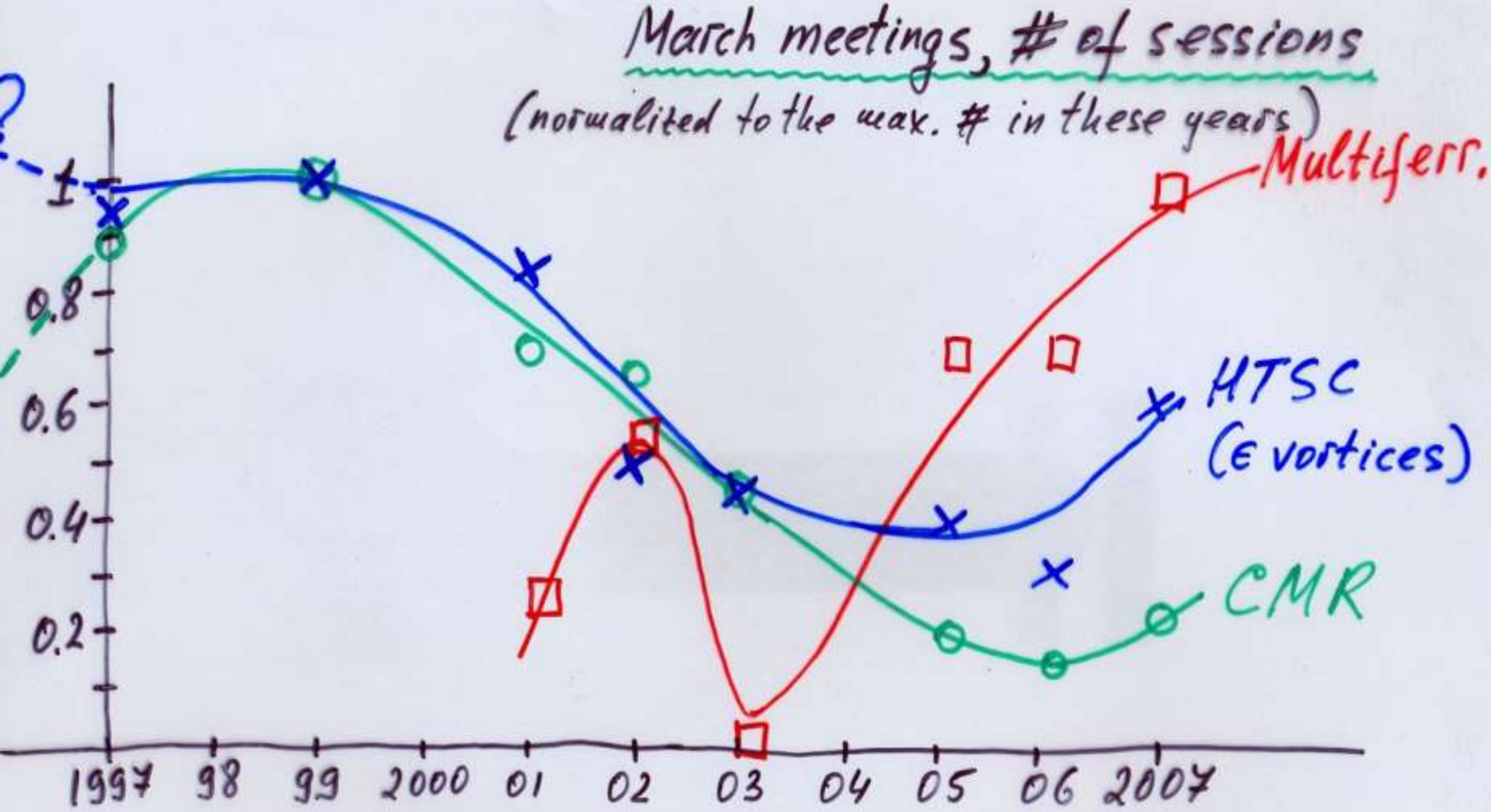
Materials combining ferroelectricity, (ferro)magnetism and (ferro)elasticity

If successful – a lot of possible applications (e.g. electrically controlling magnetic memory, etc)

Field active in 60-th – 70-th, mostly in the Soviet Union

Revival of the interest starting from ~2000

D.Kh. JMMM **306**, 1 (2006);
Physics (Trends) **2**, 20 (2009)



March meetings, # of sessions
(normalized to the max. # in these years)
Multiferr.
HTSC
(∈ vortices)
CMR
1
0.8
0.6
0.4
0.2
1997
98
99
2000
01
02
03
04
05
06
2007

Magnetism: In principle clear: spins; exchange interaction; partially filled d-shells

Ferroelectricity: Microscopic origin much less clear. Many different types, mechanisms ⇨ several different mechanism, types of multiferroics

Type-I multiferroics: Independent FE and magnetic subsystems

1) Perovskites: either magnetic, or ferroelectric; why?
2) “Geometric” multiferroics ($YMnO_3$)
3) Lone pairs (Bi; Pb, ….)
4) FE due to charge ordering

Type-II multiferroics: FE due to magnetic ordering

1) MF due to exchange striction
2) Spiral MF
3) Electronic mechanism

material	T_{FE} (K)	T_M (K)	P(μC cm^{-2})
$BiFeO_3$	1103	643	60 - 90
$YMnO_3$	914	76	5.5
$HoMnO_3$	875	72	5.6
$TbMnO_3$	28	41	0.06
$TbMn_2O_5$	38	43	0.04
$Ni_3V_2O_8$	6.3	9.1	0.01

Type-I multiferroics: Independent ferroelectricity and magnetism

- **Perovskites:** d^0 vs d^n

Empirical rule:FE for perovskites with empty d-shell
($BaTiO_3$, $PbZrO_3$; $KNbO_3$)
contain Ti^{4+}, Zr^{4+}; Nb^{5+}, Ta^{5+}; Mo^{6+}, W^{6+}, etc.

Magnetism – partially filled d-shells, d^n, n>o

Why such mutual exclusion?

Not quite clear. Important what is the mechanism of FE in perovskites like $BaTiO_3$

Classically: polarization catastrophy; Clausius-Mossotti relations, etc.

Real microscopic reason: **chemical bonds**

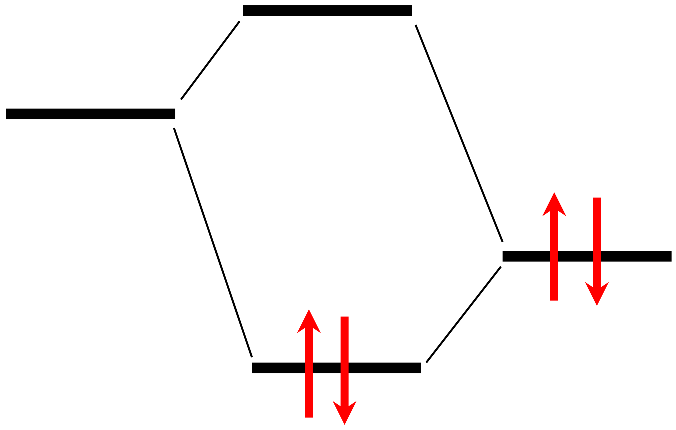
Ti^{4+}: establishes *covalent bond* with oxygens (which “donate” back the electrons), using empty d-levels

O------Ti-----O   O--------Ti--O

Better to have one strong bond with one oxygen that two weak ones with oxygens on the left and on the right

Two possible reasons:

d^0 configurations: only bonding orbitals are occupied



Other localized d-electrons break *singlet* chemical bond by Hund’s rule pair-breaking (a la pair-breaking of Cooper pairs by magnetic impurities)

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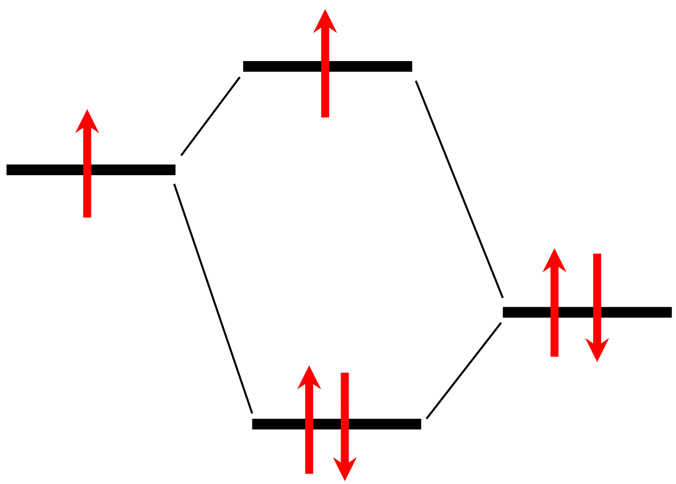

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"Geometric" multiferroics: hexagonal manganites $RMnO_3$

$YMnO_3$: T_{FE}~900 K; T_N~70 K

The origin (T.Palstra, N. Spaldin): tilting of MnO_5 trigonal bipiramids – a la tilting of MO_6 octahedra in the usual perovskites leading to orthorombic distortion.

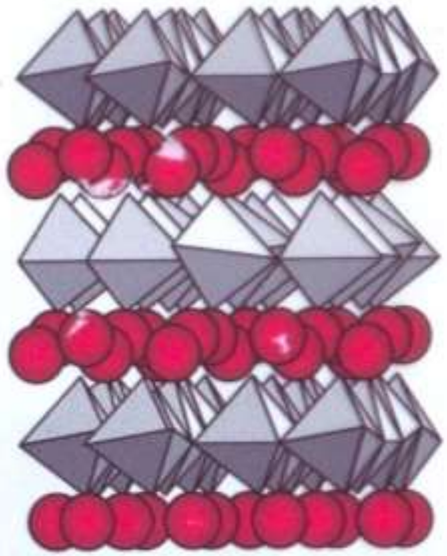
In perovskites one AMO_3 one A-O distance becomes short, but no total dipole moment – dipole moments of neighbouring cells compensate.

In $YMnO_3$ – total dipole moment, between Y and O; Mn plays no role!

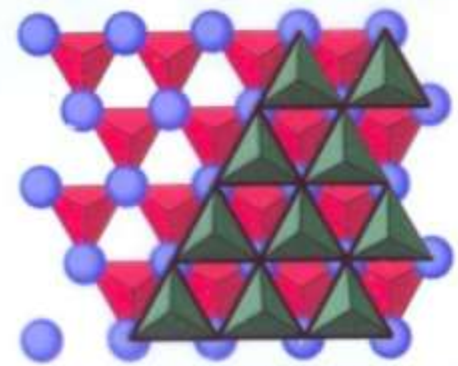
Crystal structure of $YMnO_3$

Ferroelectric distortion

Displacements from:
centrosymmetric high temp
to ferroelectric room temp

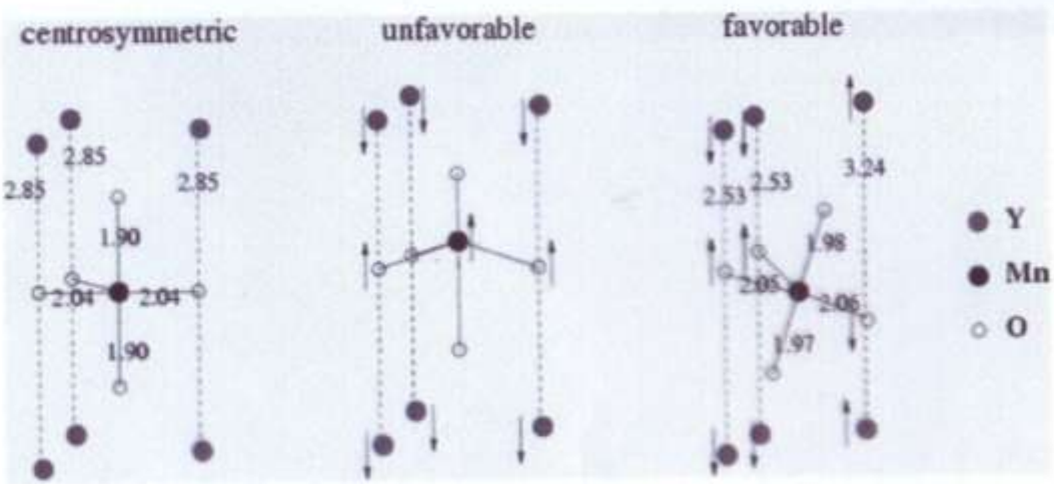


Perpendicular hexagonal axis



Parallel hexagonal axis

B. Van Aken, A. Meetsma, T. Palstra
Acta Cryst. C and several E (2001)



Van Aken, Palstra, Filipetti, Spaldin, Nature Materials 2004

Lone pairs and ferroelectricity

Bi^{3+}; Pb^{2+}. Classically – large polarizability. Microscopically – easy orientation of the lone pairs

Many nonmagnetic ferroelectrics with Bi^{3+}; Pb^{2+} . – e.g. PZT [Pb(ZrTi)O3]

Some magnetic:

Aurivillius phases: good ferroelectrics, layered systems with perovskite slabs/Bi_2O_2 layers ($SrBi_2Nb_2O_9$; $SrBi_4Ti_4O_{15}$, etc). Exist with magnetic ions, but not really studied.

PbVO3 – a novel compound. Distortion so strong that probably impossible to reverse polarization – i.e. it is probably not ferroelectric, but rather pyroelectric

Ferroelectricity due to charge ordering

Bond- versus site-centred ordering and possible ferroelectricity in manganites

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Transition metal oxides with a perovskite-type structure constitute a large group of compounds with interesting properties. Among them are materials such as the prototypical ferroelectric system $BaTiO_3$, colossal magnetoresistance manganites and the high-T_c superconductors. Hundreds of these compounds are magnetic[1], and hundreds of others are ferroelectric[2], but these properties very seldom coexist. Compounds with an interdependence of magnetism and ferroelectricity could be very useful: they would open up a plethora of new applications, such as switching of magnetic memory elements by electric fields. Here, we report on a possible way to avoid this incompatibility, and show that in charge-ordered and orbitally ordered perovskites it is possible to make use of the coupling between magnetic and charge ordering to obtain ferroelectric magnets. In particular, in manganites that are less than half doped there is a type of charge ordering that is intermediate between site-centred and bond-centred. Such a state breaks inversion symmetry and is predicted to be magnetic and ferroelectric.

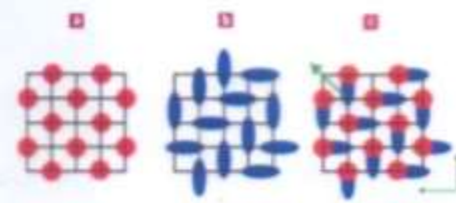


Figure 1 Three types of charge ordering. a, Site-centred charge order; **b,** bond-centred charge order (the Zener polaron state); and **c,** a ferroelectric intermediate state. The charge-ordered structure in **c** lacks inversion symmetry. Thin green arrows indicate the dipole moments of horizontal and vertical dimers, and the diagonal arrow is the total ferroelectric moment.

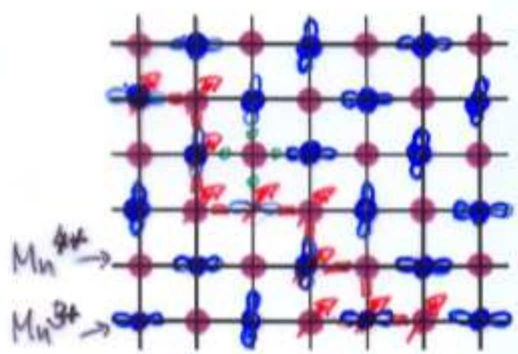
Perovskites consist of corner-sharing O_6 octahedra with a transition metal ion in the centre. Almost all the ferroelectric perovskites contain non-magnetic transition metal ions with an empty d-shell (d^0 configuration), for example Ti^{4+}, Nb^{5+} and W^{6+}. Apparently the presence of the d^0 plays an important role in formation of a ferroelectric state[3,4]. In all of these systems ferroelectricity originates from a shift of the transition metal ion from the centre of the O_6 octahedron. In this way a stronger covalent bond with one (or three) instead of six weaker bonds with neighbouring oxygen atoms is formed[5].

The problem of why magnetism and ferroelectricity seem

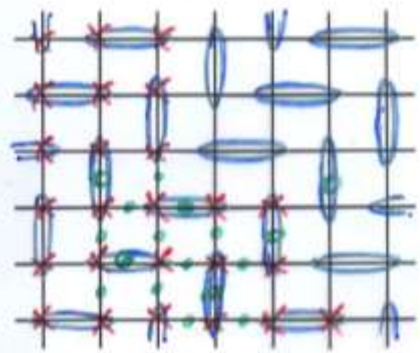
$BiFeO_3$, $BiMnO_3$ and $RMnO_3$ ($R = Y$ or another small rare-earth ion). But actually even these compounds are not exceptions to the general rule, as the mechanism of ferroelectricity here is different from the conventional one. In $BiFeO_3$ and in $BiMnO_3$ ferroelectricity is due to the lone pairs of non-magnetic[6] Bi, and in $YMnO_3$ it is due to tilting of almost rigid MnO_5 trigonal bipyramids[7]. This last example shows that the shift of the transition metal ion from the centre of the O_6 octahedron is not the only feasible mechanism of ferroelectricity. Recently ferroelectricity was observed in $TbMnO_3$,

- Charge density waves:
  - manganites
  - magnetite Fe_3O_4 (Vervey transition at 119K)

- Bond-centered charge density waves vs site centered charge density waves

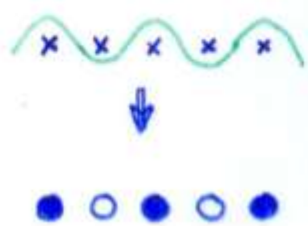


SCDW

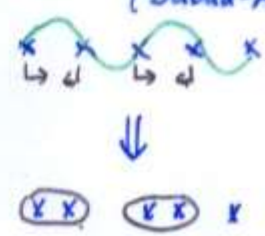


BCDW (Zener polarons)
(Daoud-Aladine et al., PRL 2002)

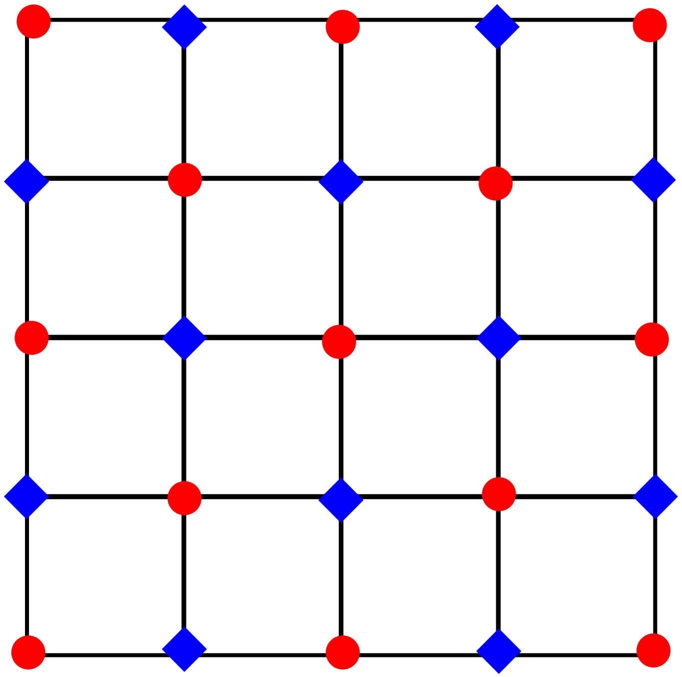
1d:

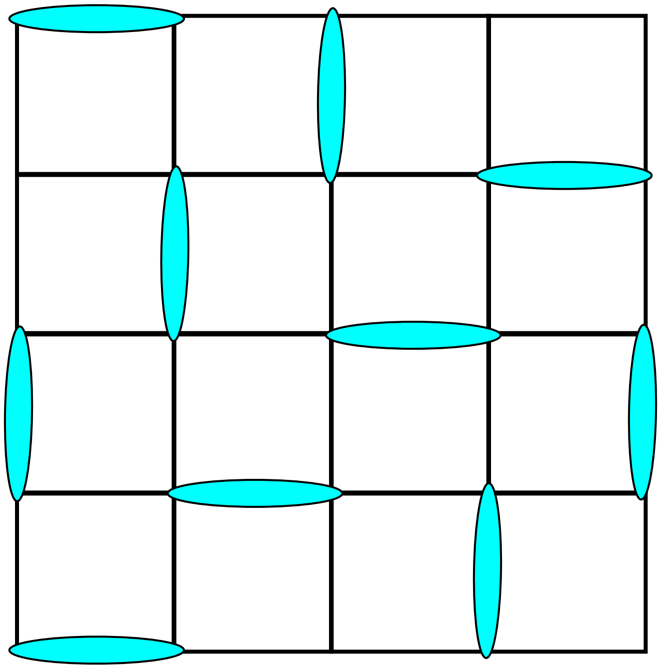


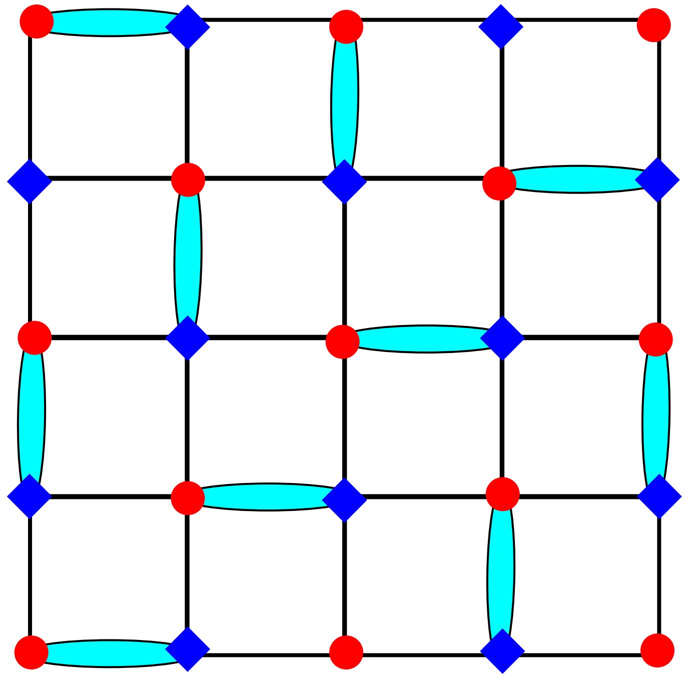
CO

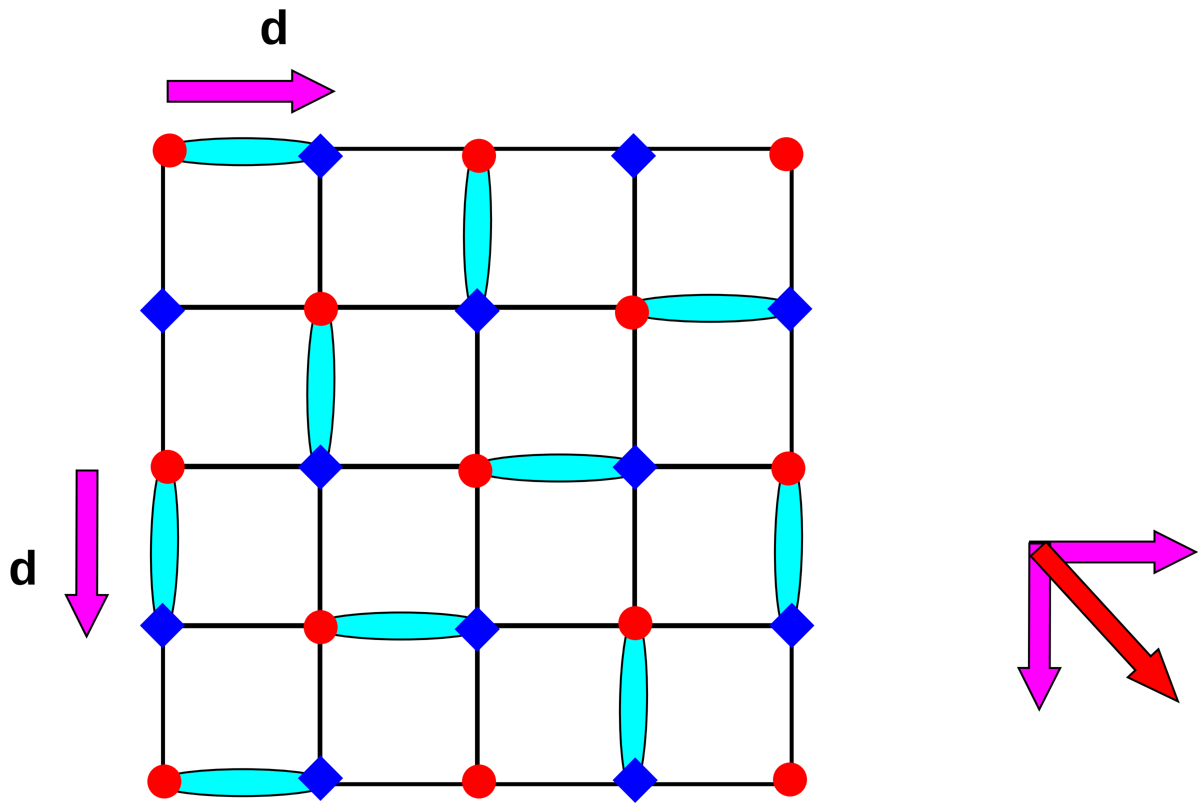


Peierls state





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Systems with ferroelectricity due to charge ordering

Some quasi-one-dimensional organic materials (Nad’, Brazovskii & Monceau; Tokura)

Fe3O4: ferroelectric below Verwey transition at 119 K ! Also ferrimagnetic with large magnetization and high Tc

LuFe2O4 ?

RNiO3 ?

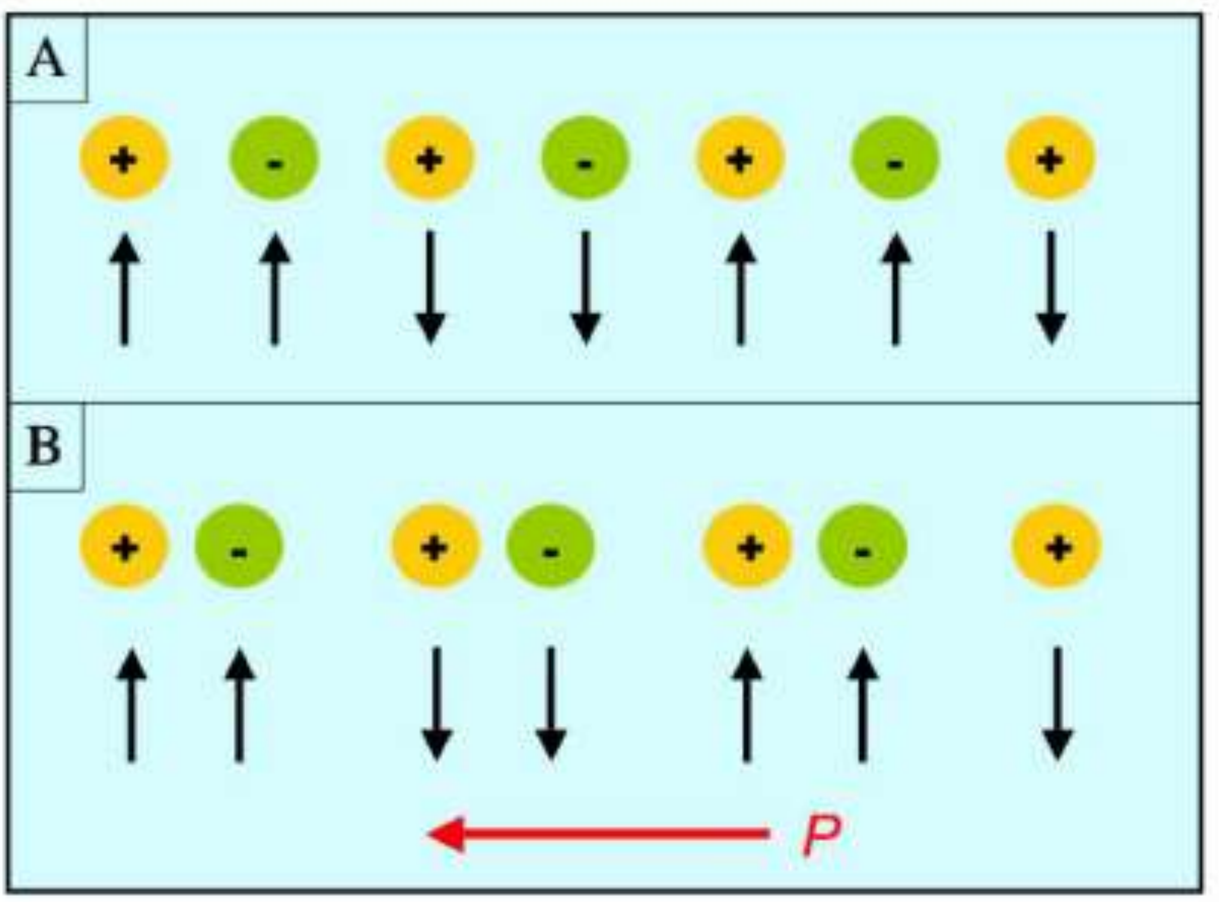
PRL **103**, 156401 (2009) PHYSICAL REVIEW LETTERS week ending 9 OCTOBER 2009

Multiferroicity in Rare-Earth Nickelates $R\mathrm{NiO}_3$

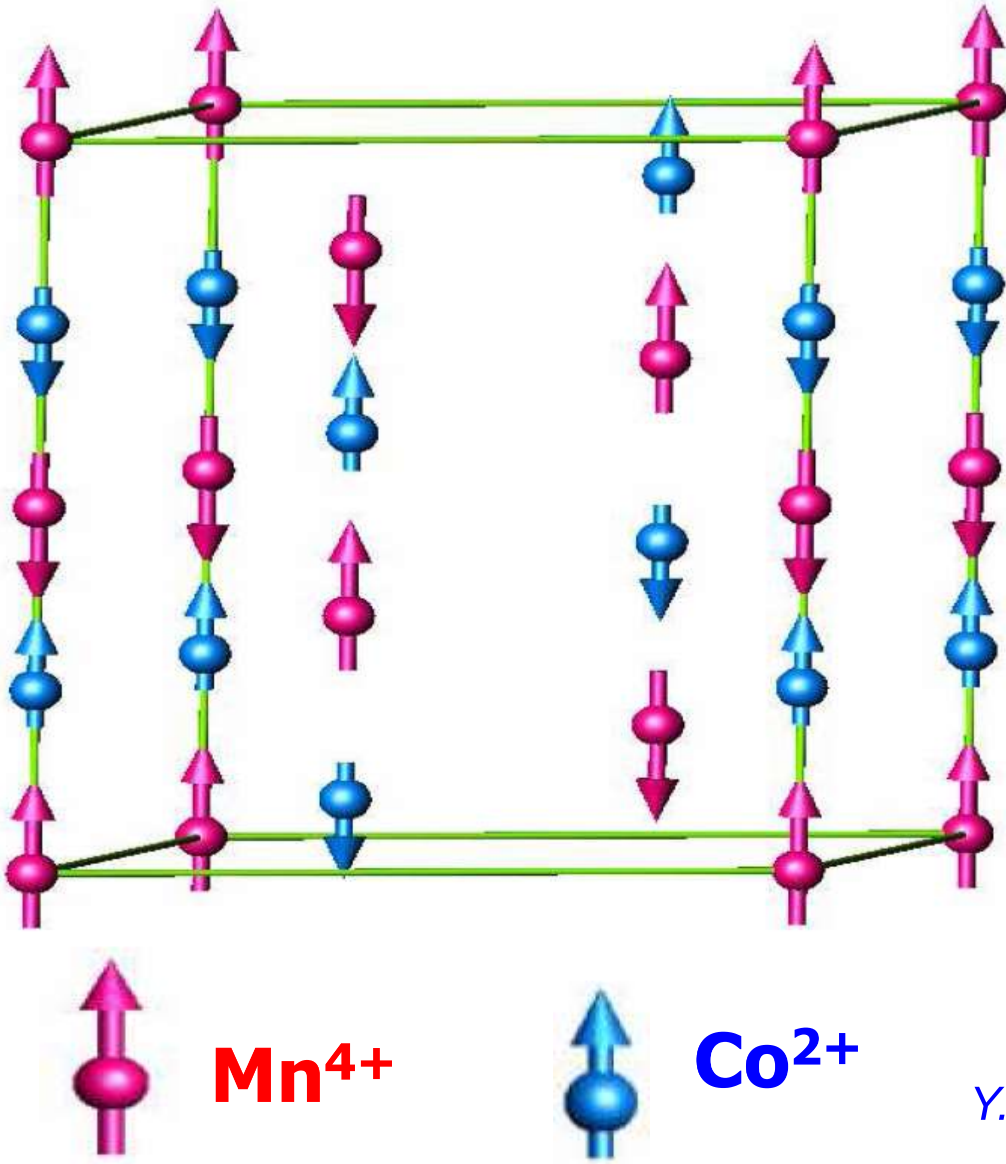
Gianluca Giovannetti,[1,2,3] Sanjeev Kumar,[1,2] Daniel Khomskii,[4] Silvia Picozzi,[3] and Jeroen van den Brink[1,5,6,7]

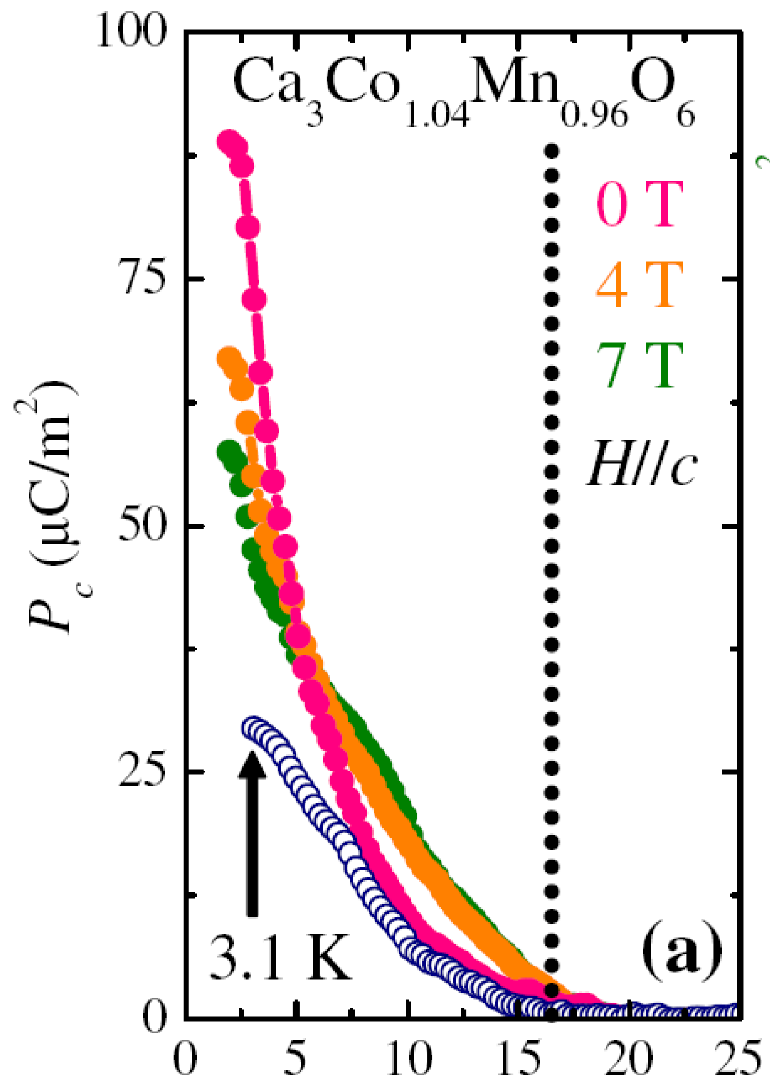
Type-II multiferroics: Ferroelectricity due to magnetic ordering

- Magnetostriction mechanism



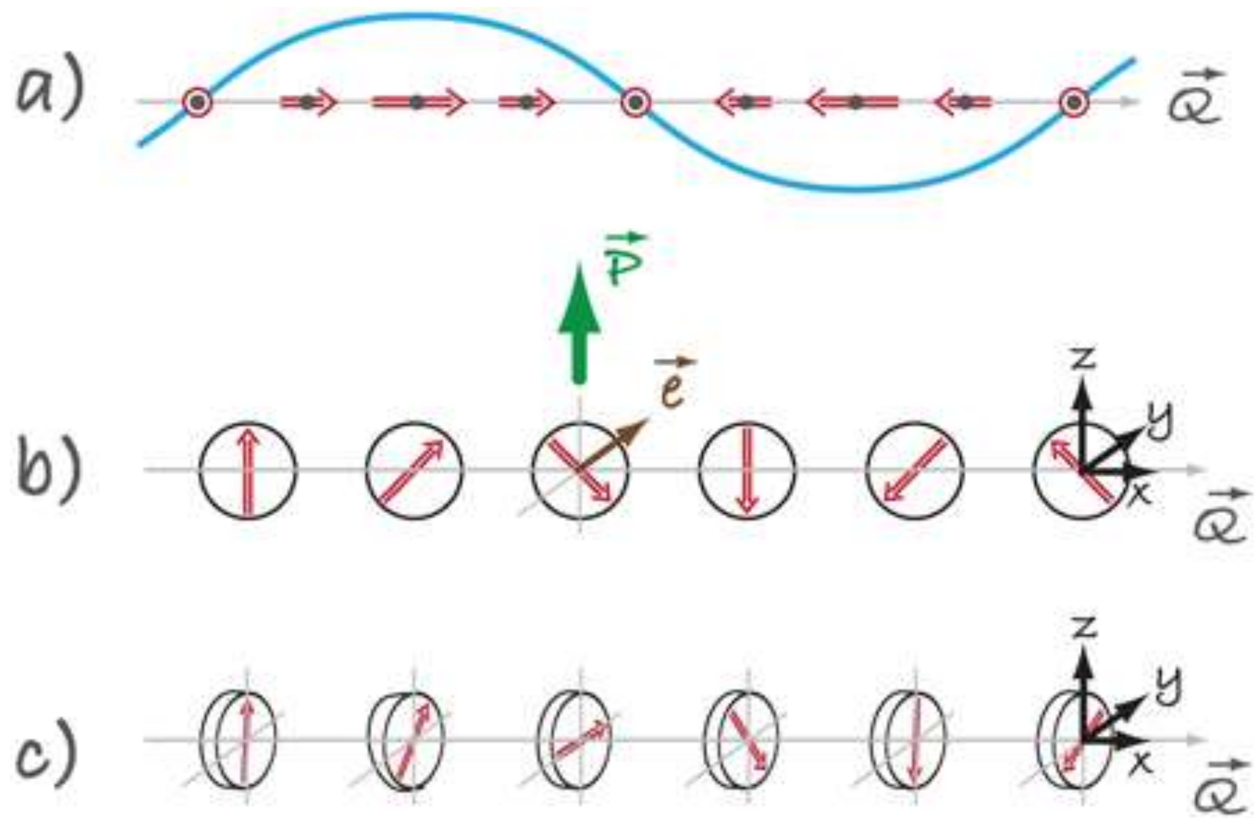
$Ca_3Co_{2-x}Mn_xO_6$





*Y.J. Choi et al PRL **100** 047601 (2008)*

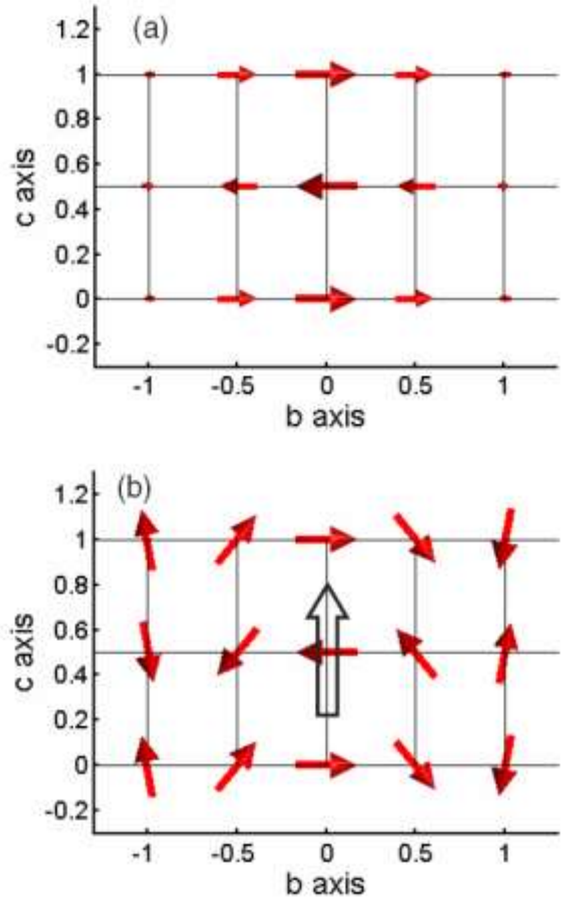
Spiral mechanism (cycloidal spiral)



$$\boldsymbol{P} = c(\boldsymbol{Q} \times \boldsymbol{e}),\ \boldsymbol{e} \sim \boldsymbol{S}_1 \times \boldsymbol{S}_2$$ **(Mostovoy)**

$$\boldsymbol{P}_{ij} = c' \boldsymbol{r}_{ij} \times (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$ **(Katsura, Nagaosa and Balatsky)**

Magnetic ordering in $TbMnO_3$



M. Kenzelmann et al (2005)

28K < T < 41K

Sinusoidal SDW spins along b axis

T < 28K

Helicoidal SDW spins rotating in bc plane

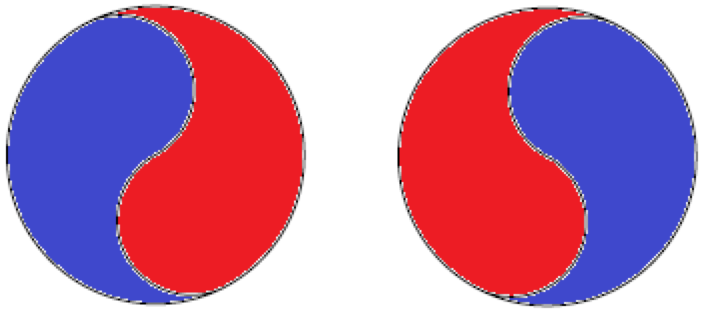
Ferroelectricity in a proper screw

Sometimes also proper screw structures can give ferroelectricity

They should not have 2-fold rotation axis perpendicular to the helix

Special class of systems: ferroaxial crystals (L.Chapon, P.Radaelli)

crystals with inversion symmetry but existing in two inequivalent modifications, which are mirror image of one another



Characterised by pseudovector (axial vector) **A**

Proper screw may be characterised by chirality $\kappa = \mathbf{r}_{12}\,[\mathbf{S}_1 \times \mathbf{S}_2]$

Then one can have polarization $\mathbf{P} = \kappa\,\mathbf{A}$ (or have invariant $(\kappa\,\mathbf{A}\,\mathbf{P})$)

Examples: $AgCrO_2$, $CaMn_7O_{12}$, $RbFe(MoO_4)_2$

Electronic Orbital Currents and Polarization in frustrated Mott Insulators

L.N. Bulaevskii, C.D. Batista, M. Mostovoy and D. Khomskii

PRB **78**, 024402 (2008)

D. Khomskii J.Phys.-Cond. Mat. **22**, 164209 (2010)

Mott insulators

$$H = -\sum_{ij\sigma} t_{ij} (c_{i\sigma}^{+} c_{j\sigma} + c_{j\sigma}^{+} c_{i\sigma}) + \frac{U}{2} \sum_{i} (n_i - 1)^2,$$

Standard paradigm: for U>>t and one electron per site electrons are localized on sites. All charge degrees of freedom are frozen out; only spin degrees of freedom remain in the ground and lowest excited states

$$H_S = \frac{4t^2}{U} (\vec{S}_1 \cdot \vec{S}_2 - 1/4).$$

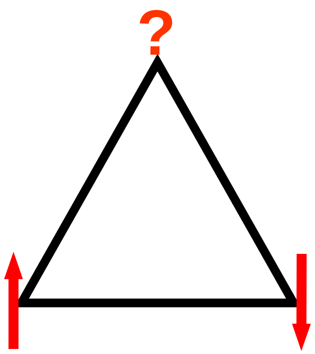
Not the full truth!

For certain spin configurations there exist in the ground state of strong Mott insulators **spontaneous electric currents** (and corresponding orbital moments)!

For some other spin textures there may exist a **spontaneous charge redistribution**, so that $\langle n_i \rangle$ is not 1! This, in particular, can lead to the appearance of a spontaneous **electric polarization** (a purely ***electronic mechanism of multiferroic behaviour***)

These phenomena, in particular, appear in frustrated systems, with **scalar chirality** playing important role

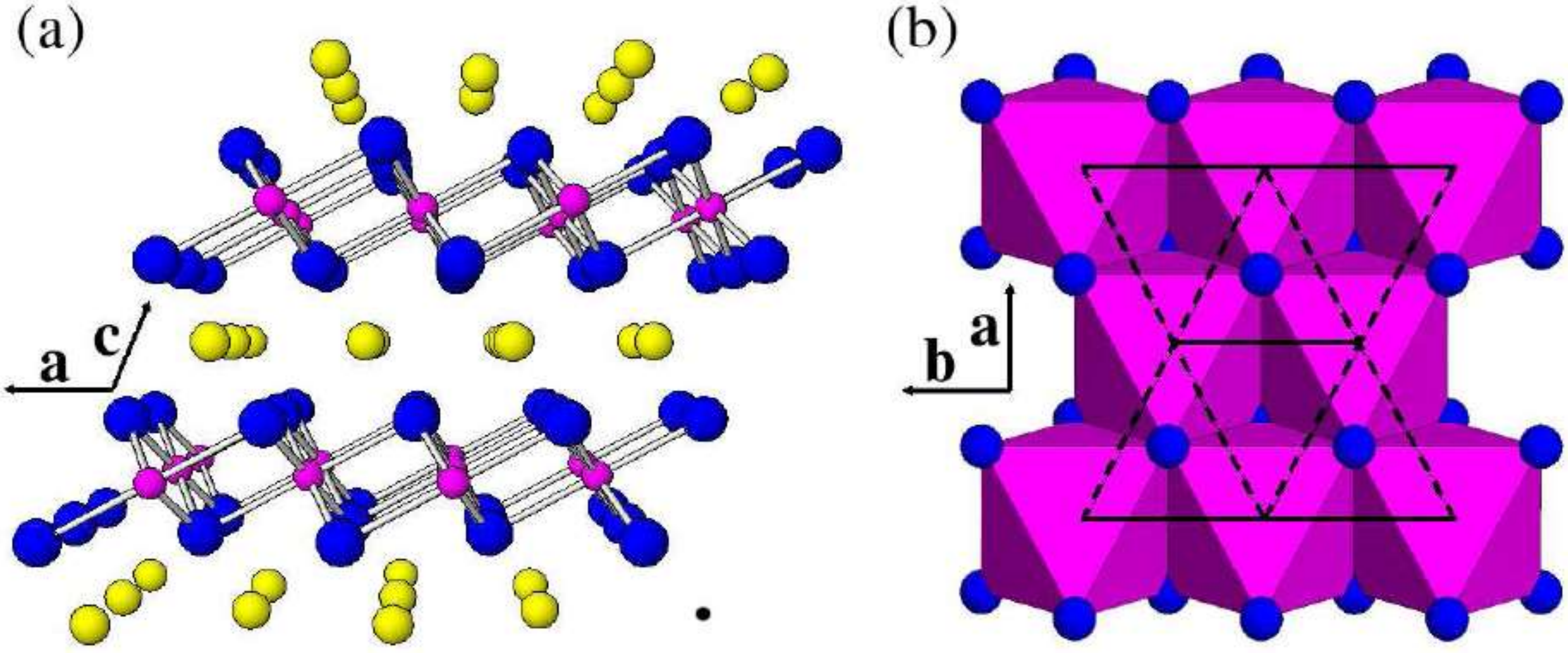
Spin systems: often complicated spin structures, especially in frustrated systems – e.g. those containing triangles as building blocks



- Isolated triangles (trinuclear clusters) - e.g. in some magnetic molecules (V15, …)
- Solids with isolated triangles ($La_4Cu_3MoO_{12}$)
- Triangular lattices
- Kagome
- Pyrochlore

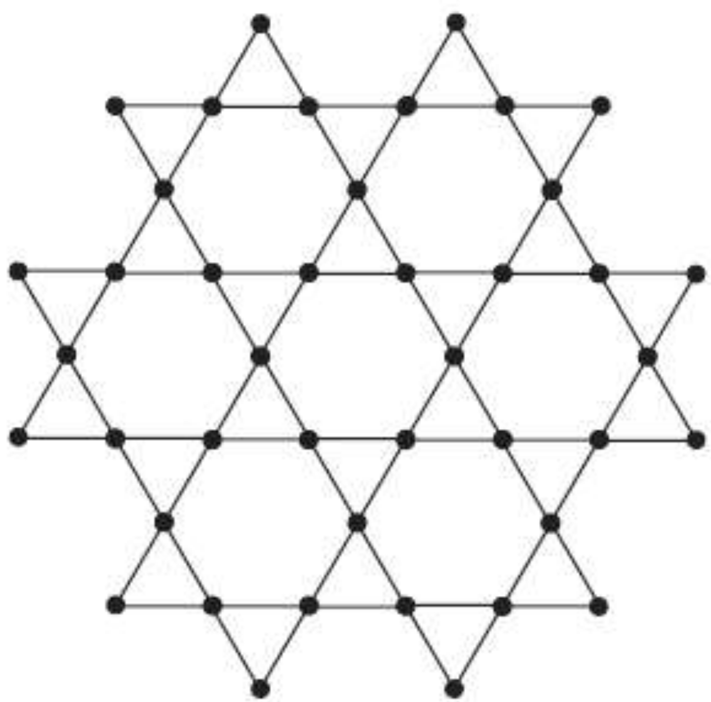
Triangular lattices:

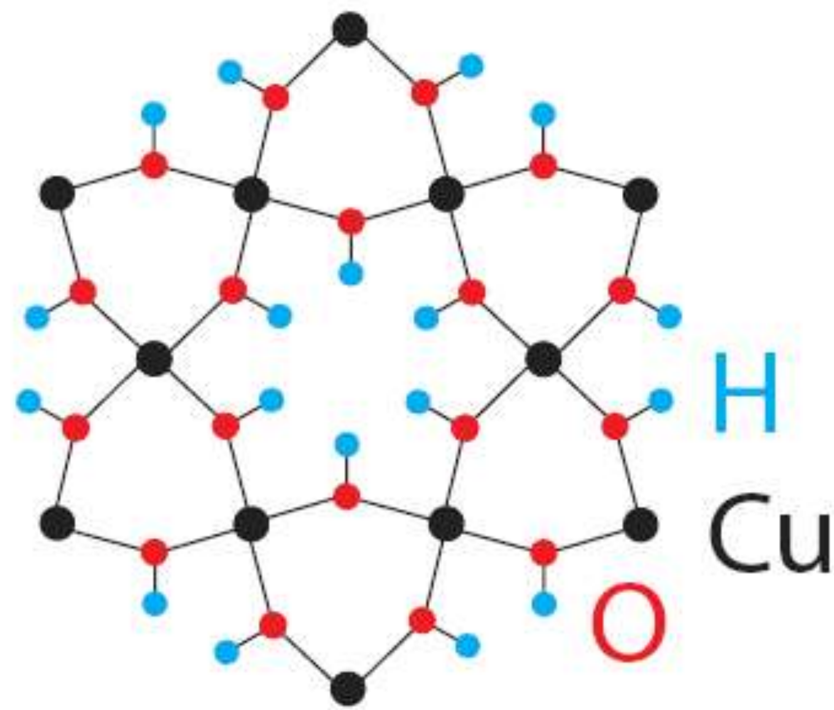
Na_xCoO_2, $LiVO_2$, $CuFeO_2$, $LiNiO_2$, $NiGa_2S_4$, …



Kagome:

$ZnCu_3(OH)_6Cl_2$ (herbertsmithite)

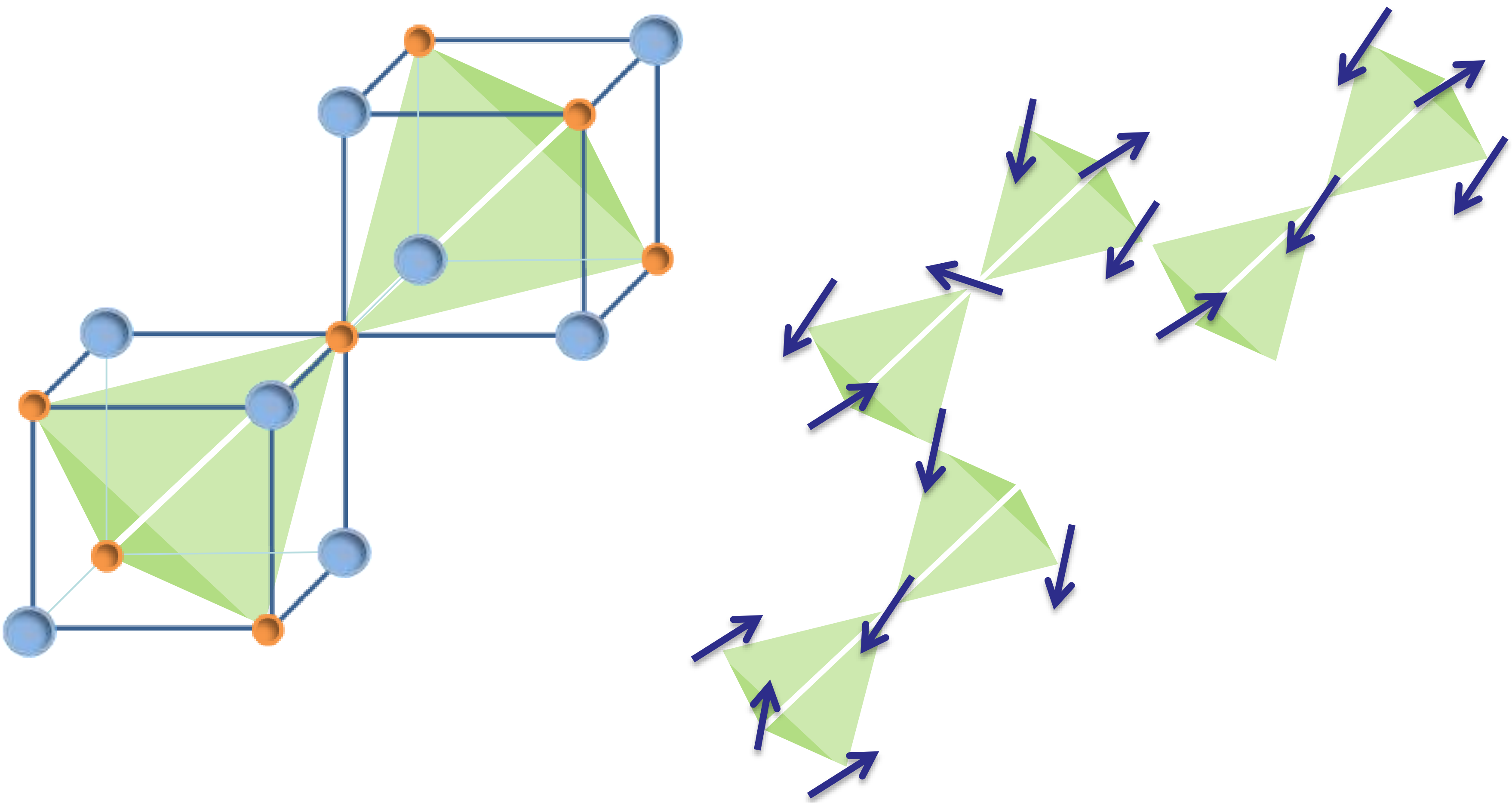




The Cathedral San Giusto, Trieste, 6-14 century

Spinels, pyrochlores:

The B-site pyrochlore lattice: geometrically frustrated for AF



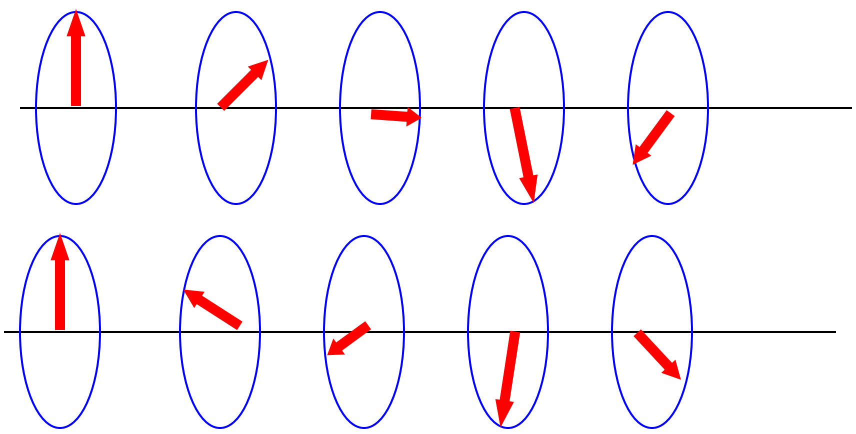
Often complicated ground states; sometimes $\langle \vec{S}_i \rangle = 0$ ⟹

⟹ spin liquids

Some structures, besides $\langle \vec{S}_i \rangle$, are characterized by:

Vector chirality

$$\left[\vec{S}_i \times \vec{S}_j \right]$$

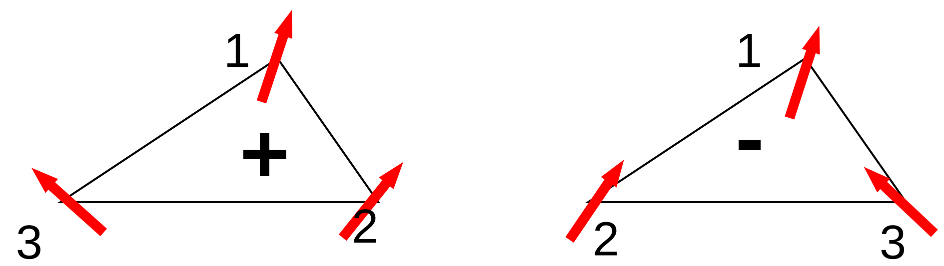


Scalar chirality

$$\chi_{123} = \vec{S}_1 \left[\vec{S}_2 \times \vec{S}_3 \right]$$

- solid angle

χ may be + or - :



But what is the scalar chirality physically?

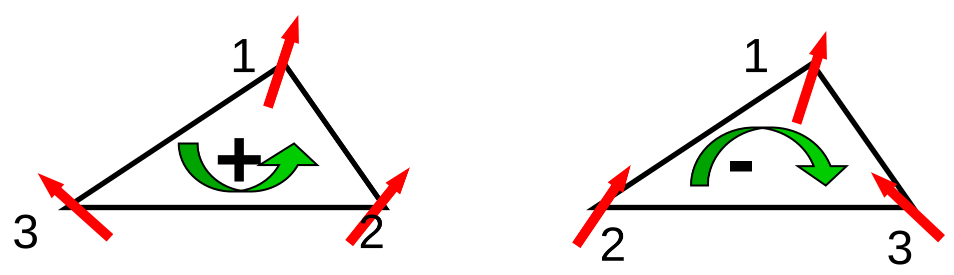
What does it couple to?

How to measure it?

Breaks time-reversal-invariance T and inversion P - like currents!

$\Longrightarrow$ $\chi_{123} \neq 0$ means spontaneous circular electric current $j_{123} \neq 0$ and orbital moment $L_{123} \neq 0$

$$L_{123} \propto j_{123} \propto \chi_{123}$$



Couples to magnetic field:

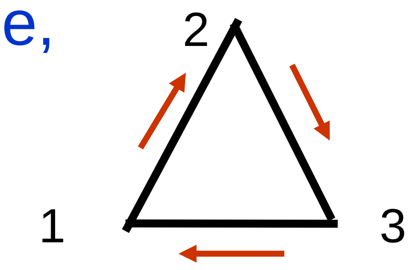
$$-\vec{L}\vec{H} \sim -\chi H$$

Spin current operator and scalar spin chirality

- Current operator for Hubbard Hamiltonian on bond ij:

$$\vec{I}_{ij} = \frac{iet_{ij}\vec{r}_{ij}}{\hbar r_{ij}} \sum_{\sigma} (c^{+}_{i\sigma} c_{j\sigma} - c^{+}_{j\sigma} c_{i\sigma}).$$

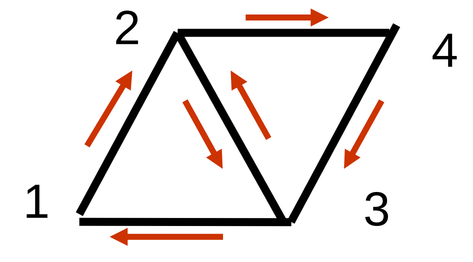
- Projected current operator: odd # of spin operators, scalar in spin space. For smallest loop, triangle,



$$\vec{I}_{S,12}(3) = \frac{\vec{r}_{ij}}{r_{ij}} \frac{24et_{12}t_{23}t_{31}}{\hbar U^2} [\vec{S}_1 \times \vec{S}_2] \cdot \vec{S}_3.$$

- Current via bond 23

$$I_{S,23} = I_{S,23}(1) + I_{S,23}(4).$$



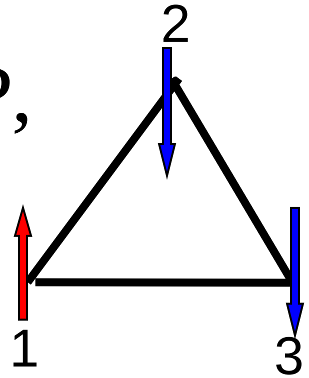
- On bipartite nn lattice I_S is absent.

Spin-dependent electronic polarization

Charge operator on site i: $$Q_i = e\sum_{\sigma} c_{i\sigma}^{+} c_{i\sigma}.$$

Projected charge operator $n_{S,i} = P e^{S} n_i e^{-S} P,$

$$\langle n_1 \rangle = 1 + \delta n_1 = 1 - 8\left(\frac{t}{U}\right)^3 \left[\mathbf{S}_1(\mathbf{S}_2 + \mathbf{S}_3) - 2\mathbf{S}_2\mathbf{S}_3\right]$$

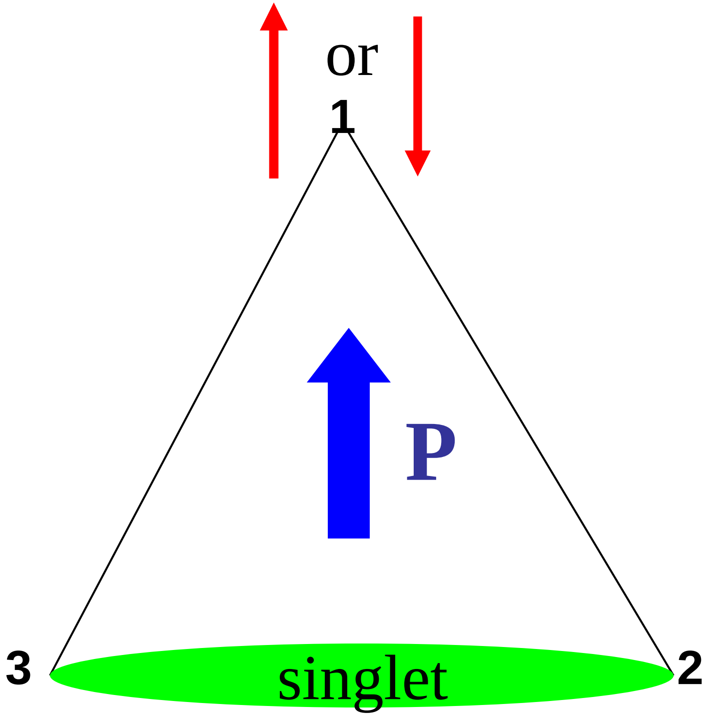


Polarization on triangle $$\vec{P}_{123} = e\sum_{i=1,2,3} n_{S,i}\vec{r}_i, \qquad \sum_i n_{S,i} = 3.$$

Charge on site *i* is sum over triangles at site *i*.

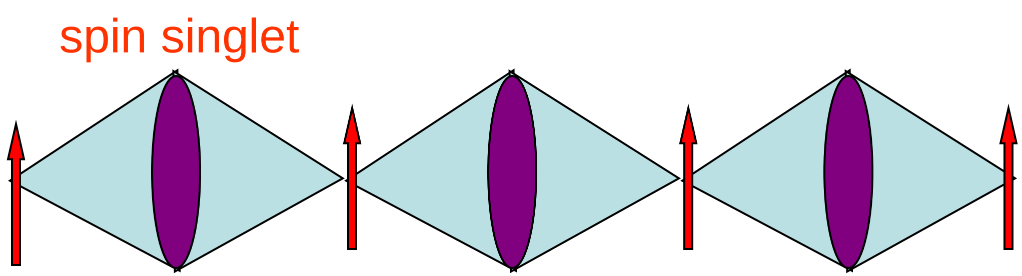
Electronic polarization on triangle

$$\langle n_1 \rangle = 1 + \delta n_1 = 1 - 8\left(\frac{t}{U}\right)^3 \left[\mathbf{S}_1(\mathbf{S}_2 + \mathbf{S}_3) - 2\mathbf{S}_2\mathbf{S}_3\right]$$



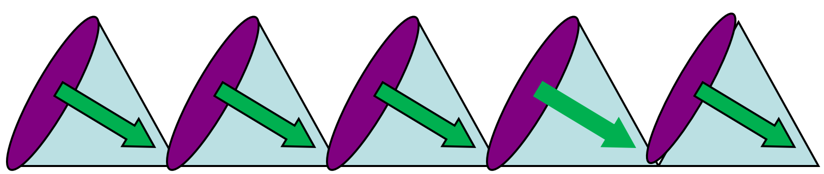
Purely electronic mechanism of multiferroic behavior!

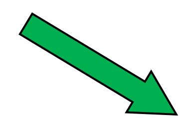
- Diamond chain (azurite $Cu_3(CO_3)_2(OH)_2$)

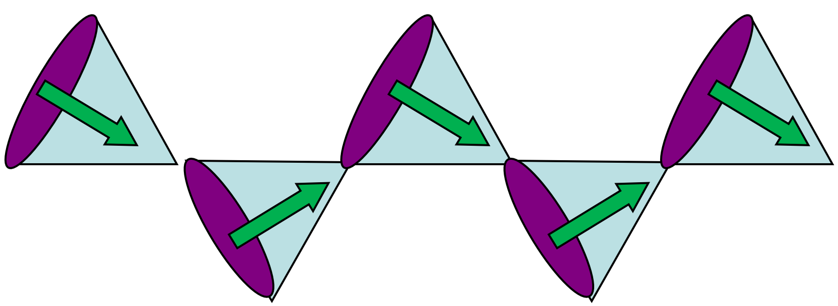


-will develop S-CDW

- Saw-tooth (or delta-) chain



Net polarization 



Net polarization 

$Cu_2(AsO_4)(OH)\cdot 3H_2O$ (euchroite)

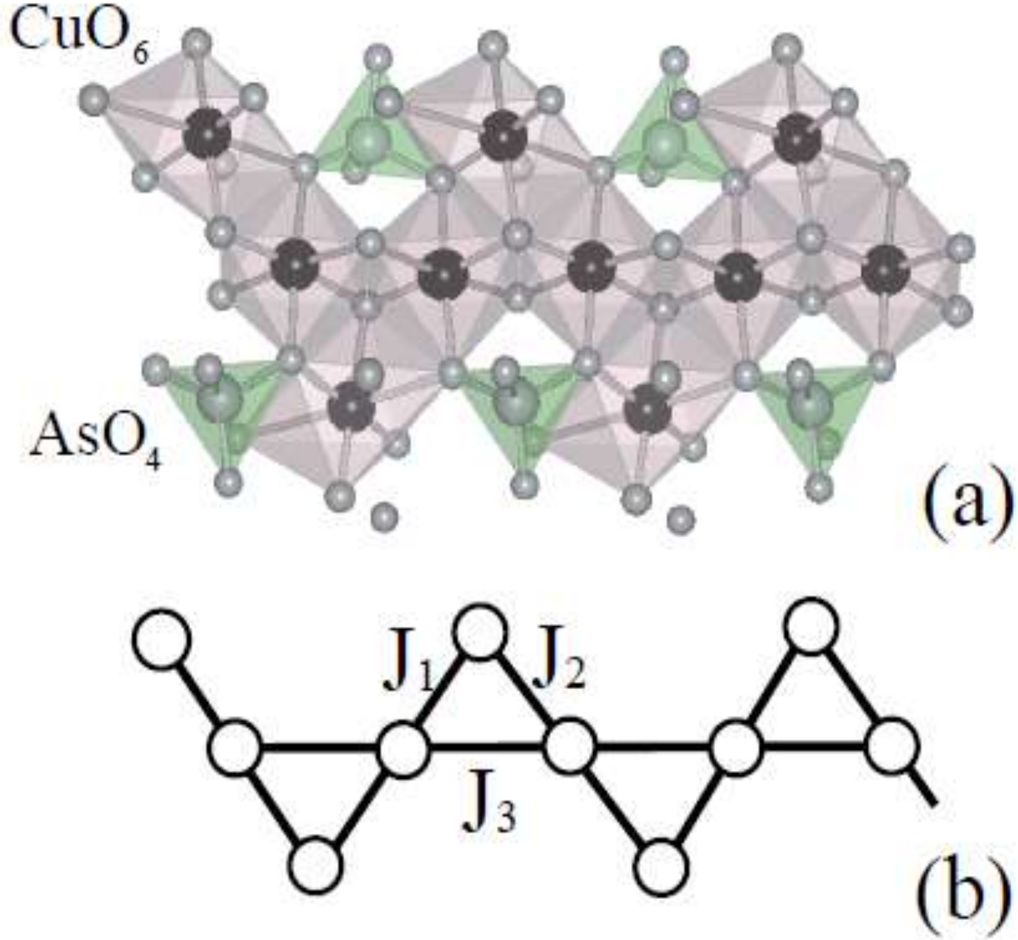
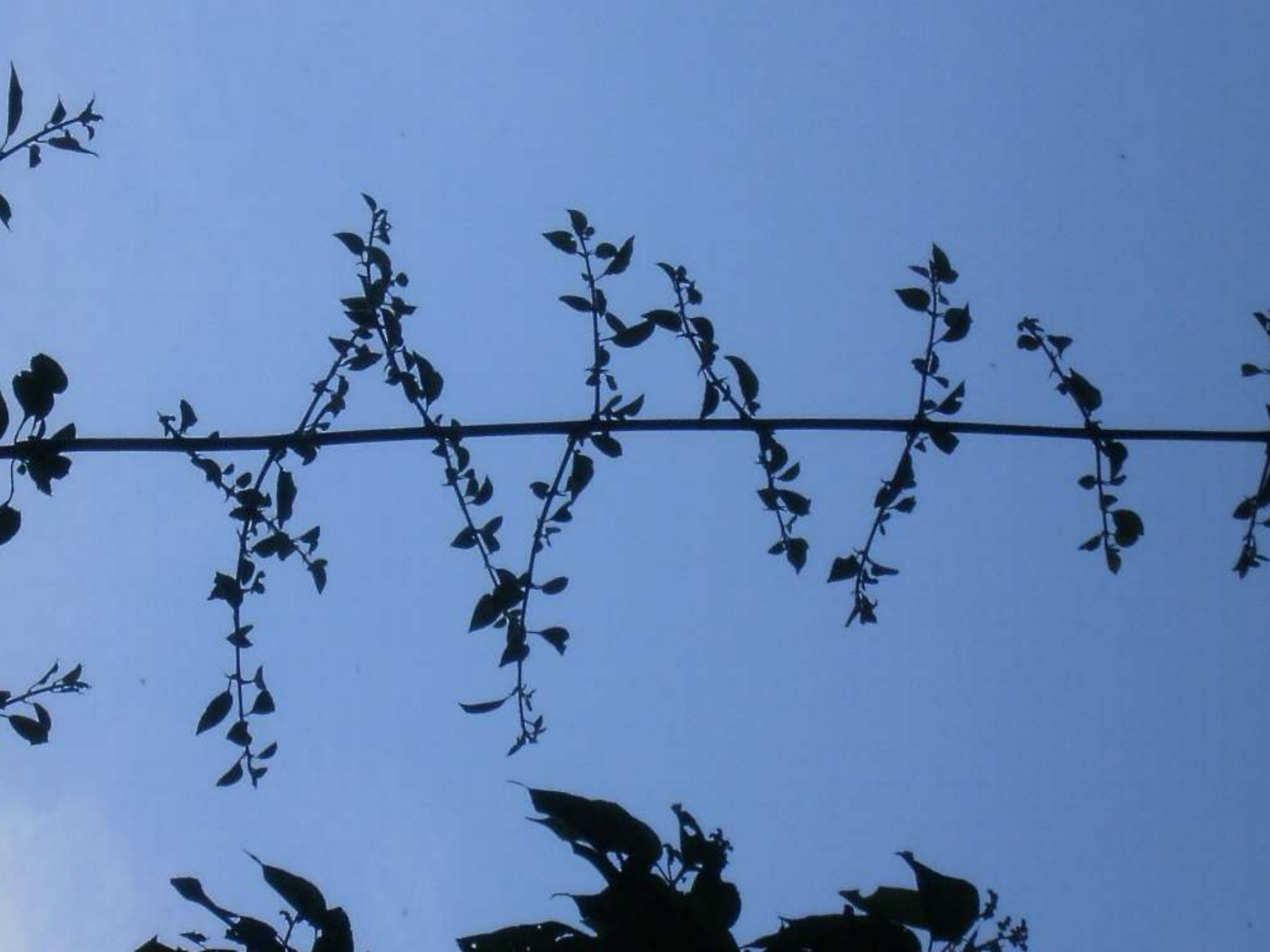


Figure 1. (a) Structure of euchroite, (b) schematic view of the delta chain.



- ESR : magnetic field (**-HM**) causes transitions

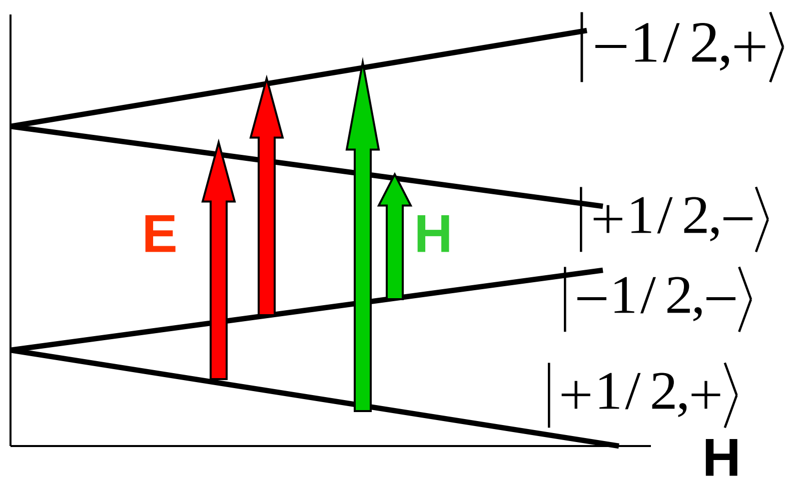
$$|1/2,\chi\rangle \to |-1/2,\chi\rangle,\ or\ |-1/2,\chi\rangle \to |1/2,\chi\rangle$$

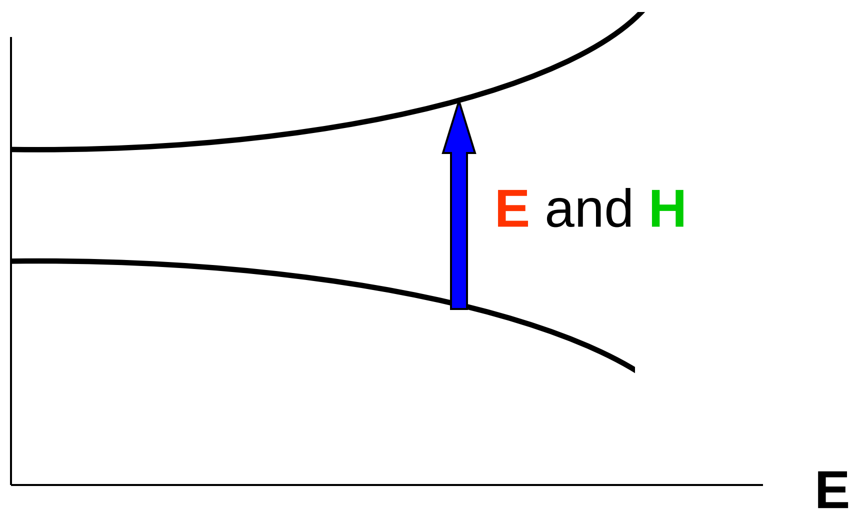
Here: electric field (**-Ed**) has nondiagonal matrix elements in χ:

$$\langle \chi = + | \mathbf{d} | \chi = - \rangle \neq 0$$ → electric field will cause

dipole-active transitions $$|S^z,+\rangle \Leftrightarrow |S^z,-\rangle$$

-- ESR caused by electric field E !





Chirality as a qubit?

Triangle: S=1/2, chirality (or pseudosin T) = ½

Can one use chirality instead of spin for quantum computation etc, as a qubit instead of spin?

We can control it by magnetic field (chirality = current = orbital moment) and by electric field

Georgeot, Mila, Phys. Rev. Lett. **104**, 200502 (2008)

Magnetoelectrics as methamaterials

(systems with negative refraction index)

Multiferroics as metamaterials

LHM: negative $\varepsilon(\omega) < 0$ and $\mu(\omega) < 0$

Maxwell Eqs: $\nabla \times \vec{E} = \frac{i\omega}{c}\vec{B}$ $\nabla \times \vec{H} = -\frac{i\omega}{c}\vec{D}$

$\nabla \cdot \vec{D} = 0$ $\nabla \cdot \vec{B} = 0$ $\vec{D} = \varepsilon(\omega)\vec{E},$ $\vec{B} = \mu(\omega)\vec{H},$

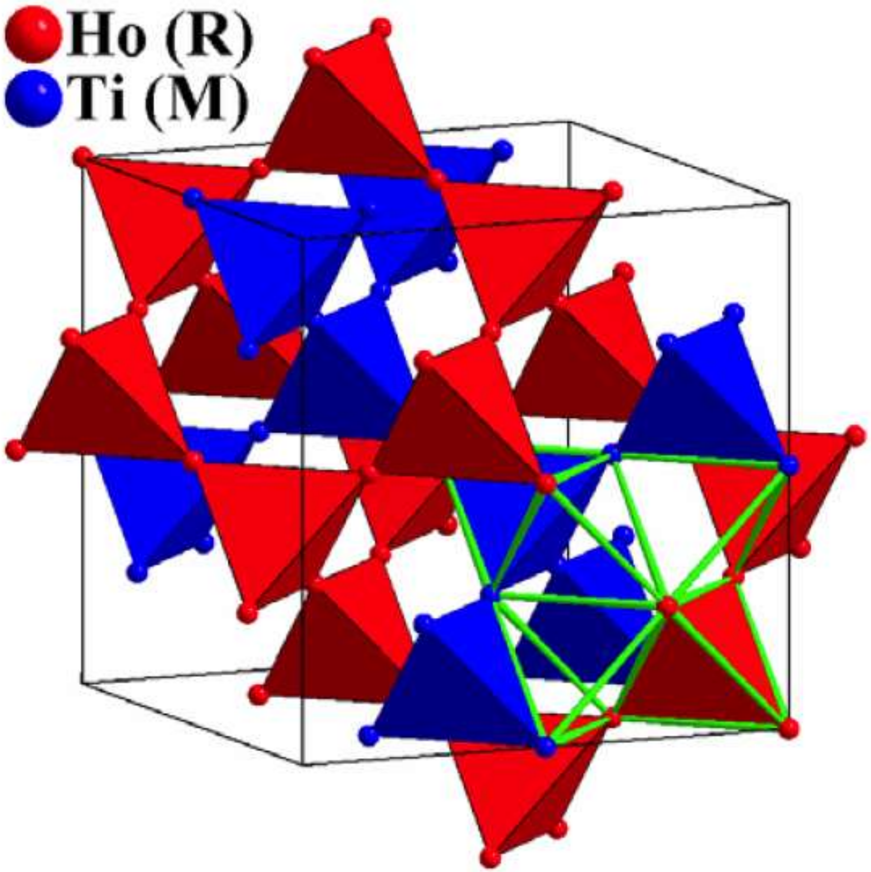
$$n^2 = \varepsilon\mu > 0$$

Vectors $\vec{k},\ \vec{E},\ \vec{H}$

form a left-handed orthogonal set (LHM) (V. Veselago, 1967).

Monopoles and dipoles in spin ice

Pyrochlore: Two interpenetrating metal sublattices



$Ho_2Ti_2O_7$


nature COMMUNICATIONS

ARTICLE

Received 23 Feb 2012 | Accepted 14 May 2012 | Published 19 Jun 2012
DOI: 10.1038/ncomms1904

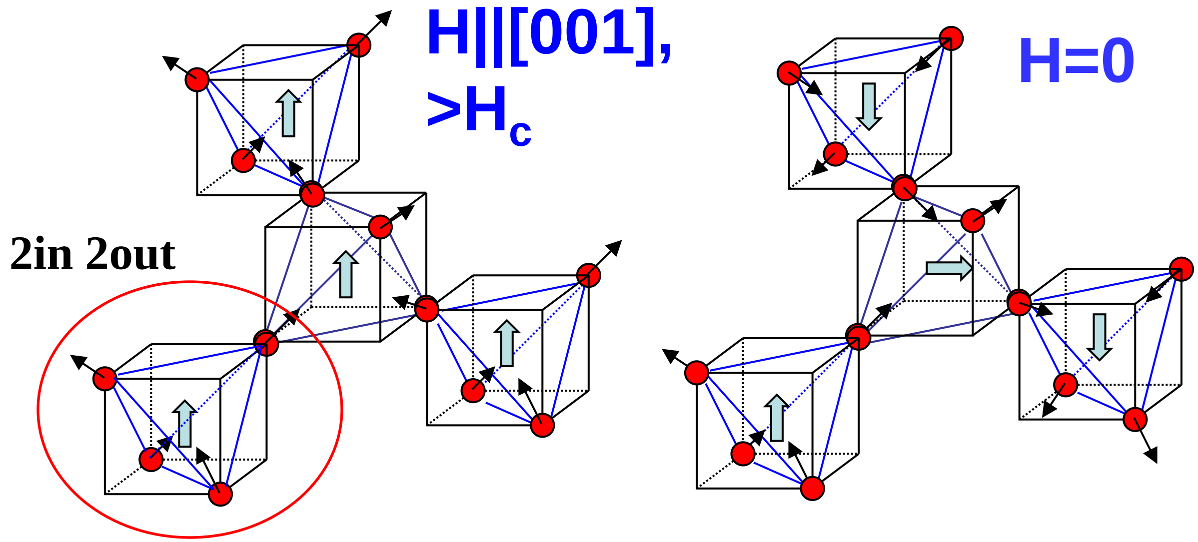
Electric dipoles on magnetic monopoles in spin ice

D.I. Khomskii

pyrochlore $R_2Ti_2O_7$ ･･geometrical spin frustration

***R*=Ho**

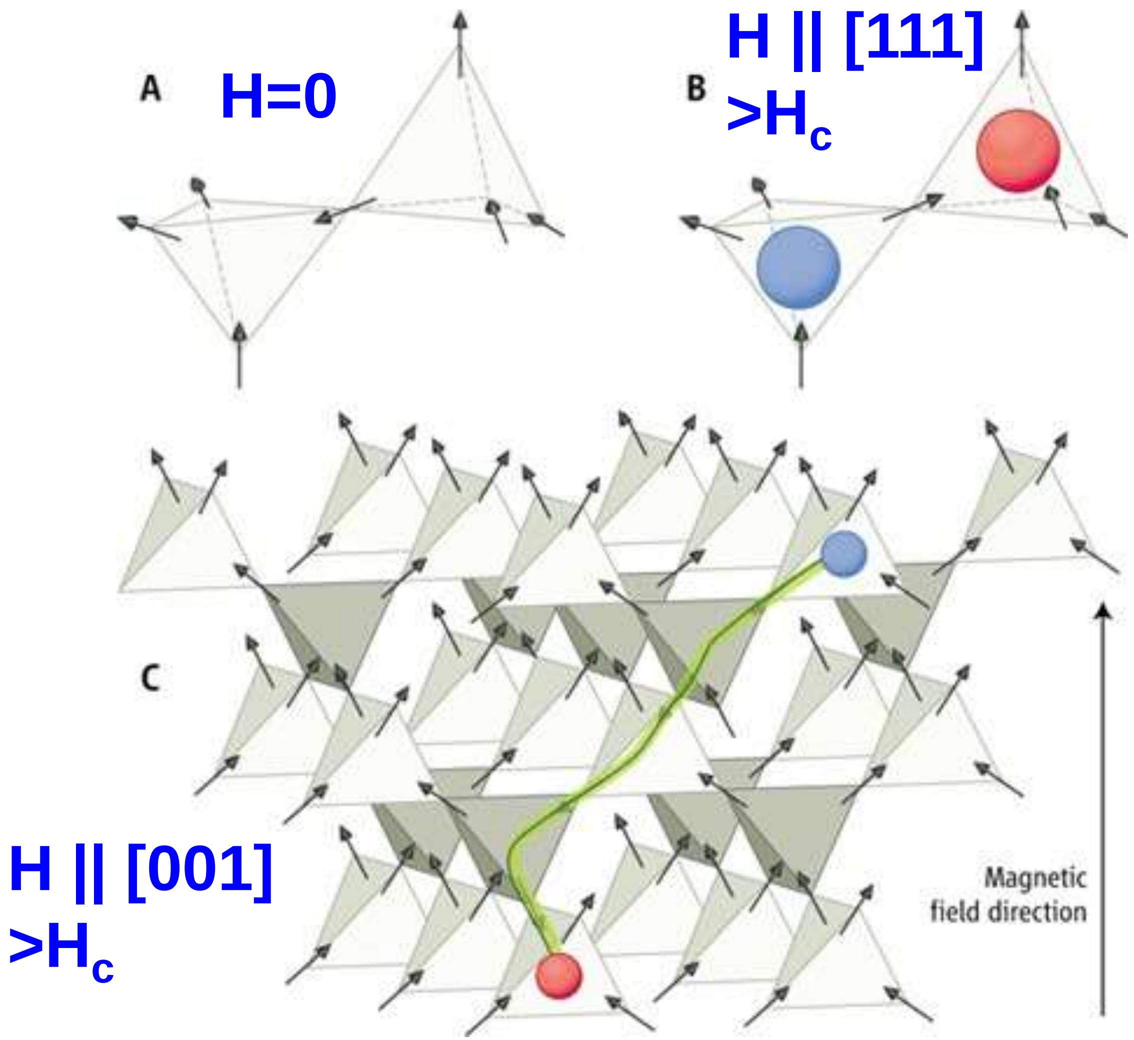
Ferromagnetic interaction, Ising spin (spin ice)



***R*=Gd**

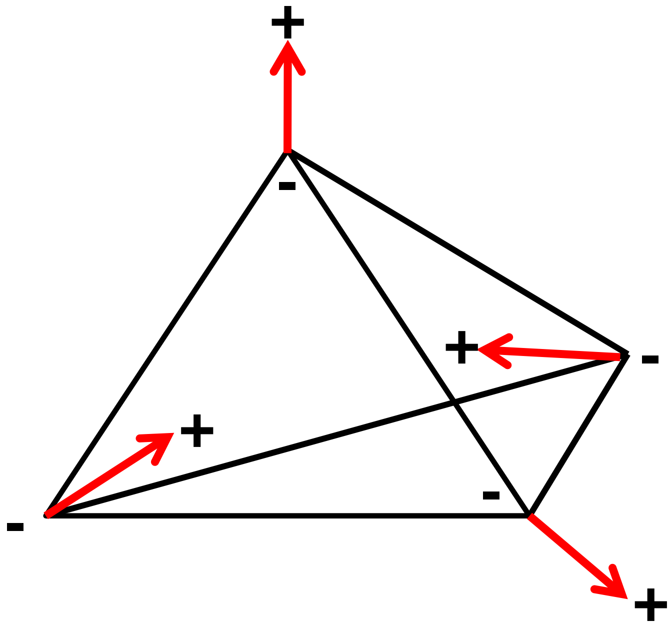
Antiferromagnetic interaction, Heisenberg spin

Excitations creating magnetic monopole (Castelnovo, Moessner and Sondhi)

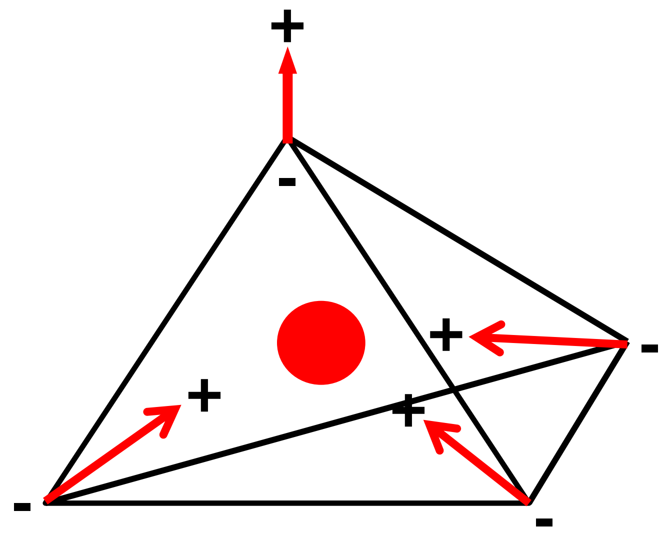


M J P Gingras Science 2009;326:375-376

Science AAAS



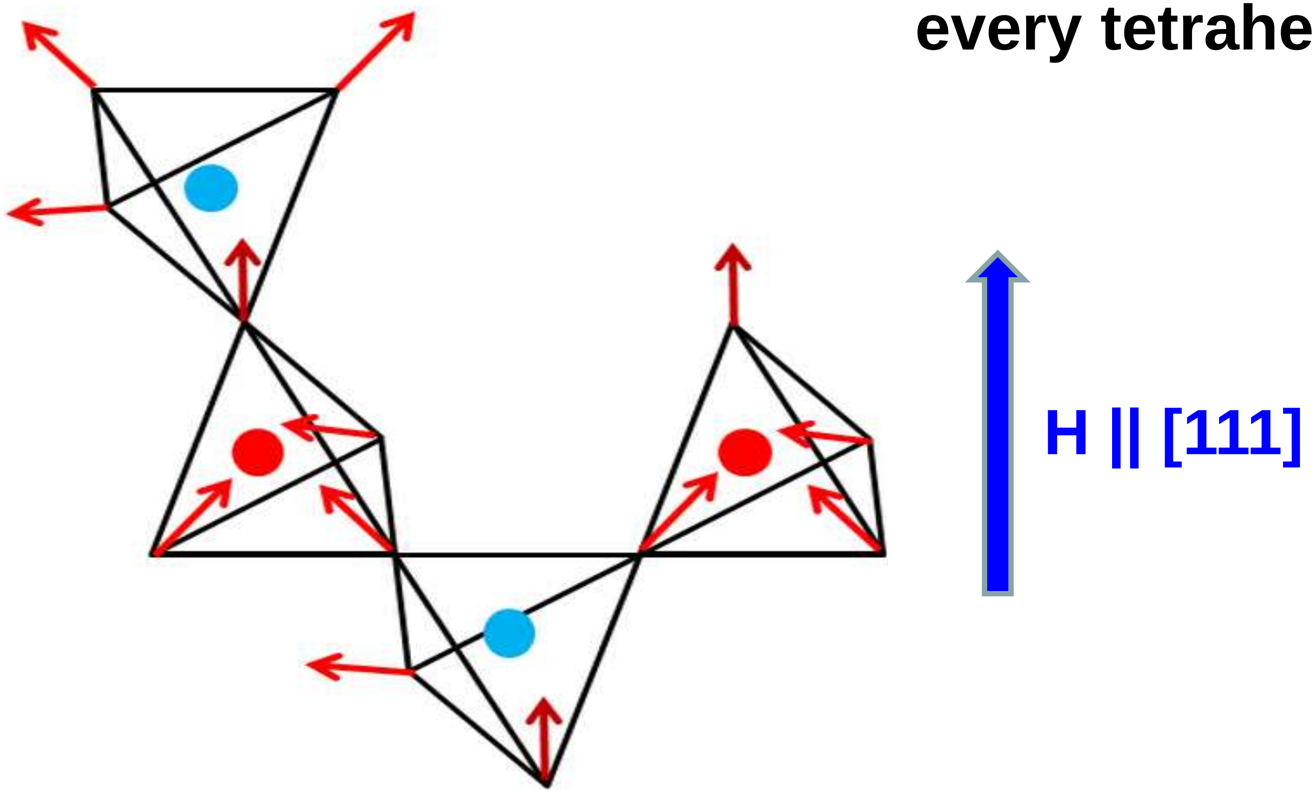
2-in/2-out: net magnetic charge inside tetrahedron zero



3-in/1-out: net magnetic charge inside tetrahedron ≠ 0 – monopole or antimonopole

$H \parallel [111]$, $>H_c$

Monopoles/antimonopoles at every tetraheder, staggered



H Aoki et al., JPSJ 73, 2851 (2004)

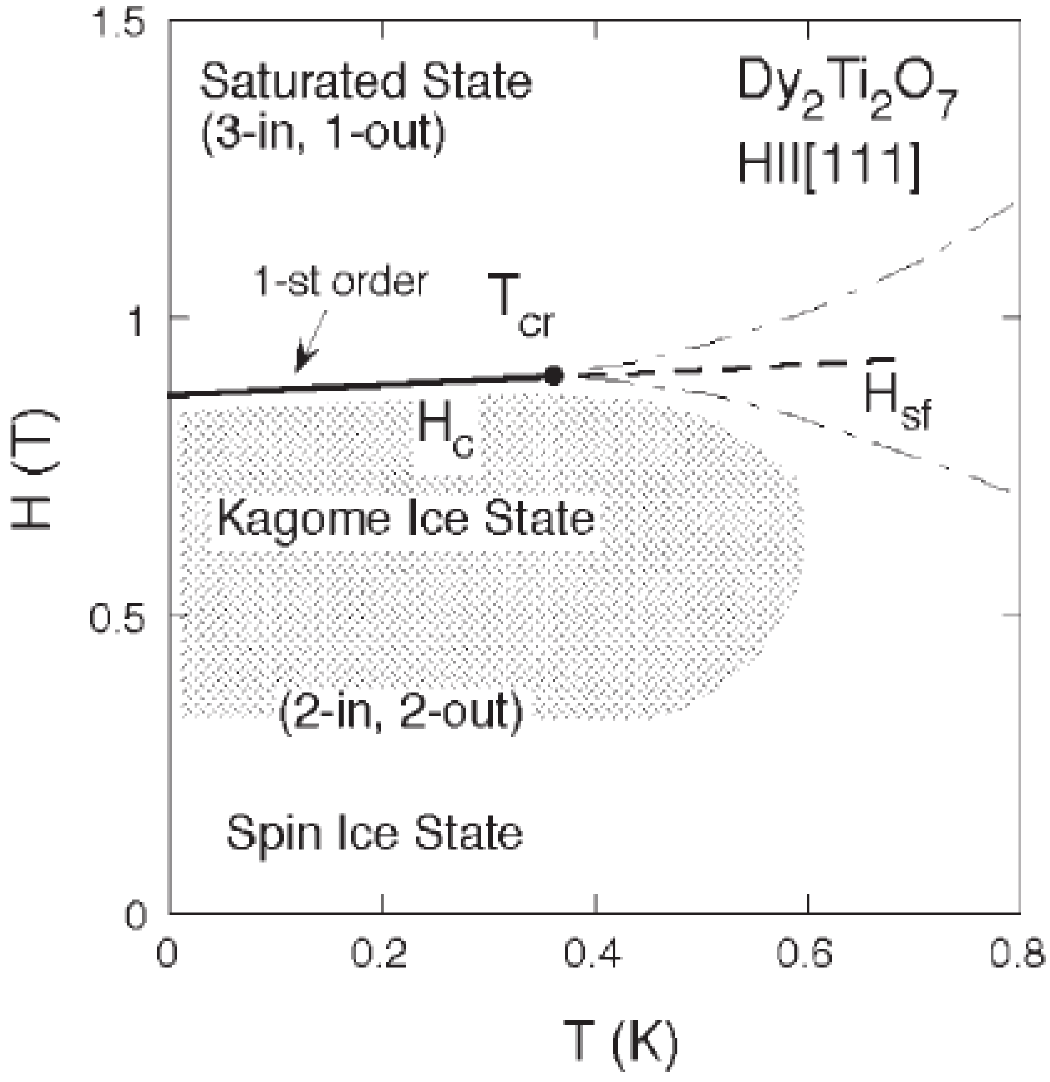
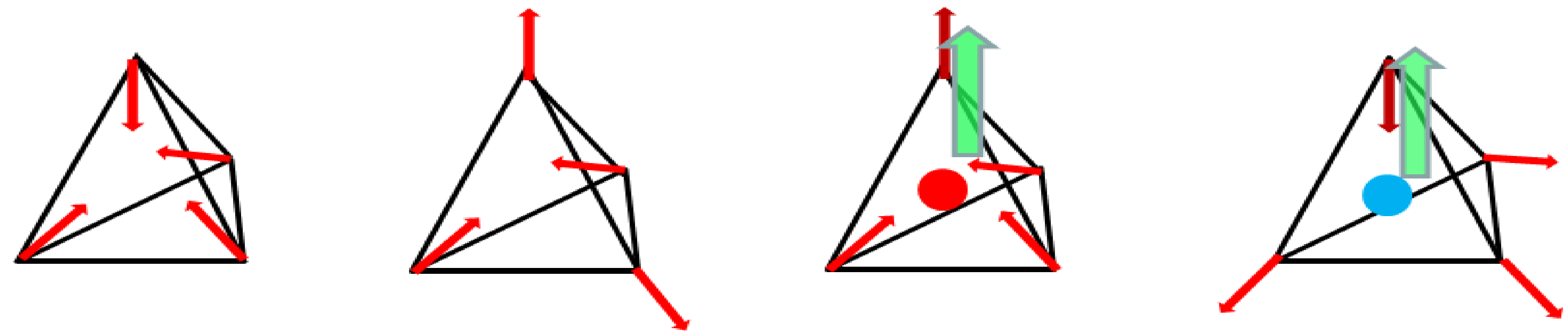


Fig. 1. Phase diagram of $Dy_2Ti_2O_7$ in a [111] magnetic field, determined by magnetization and specific heat measurements. The dashed line

★ Dipoles on tetrahedra:



4-in or 4-out:
d=0

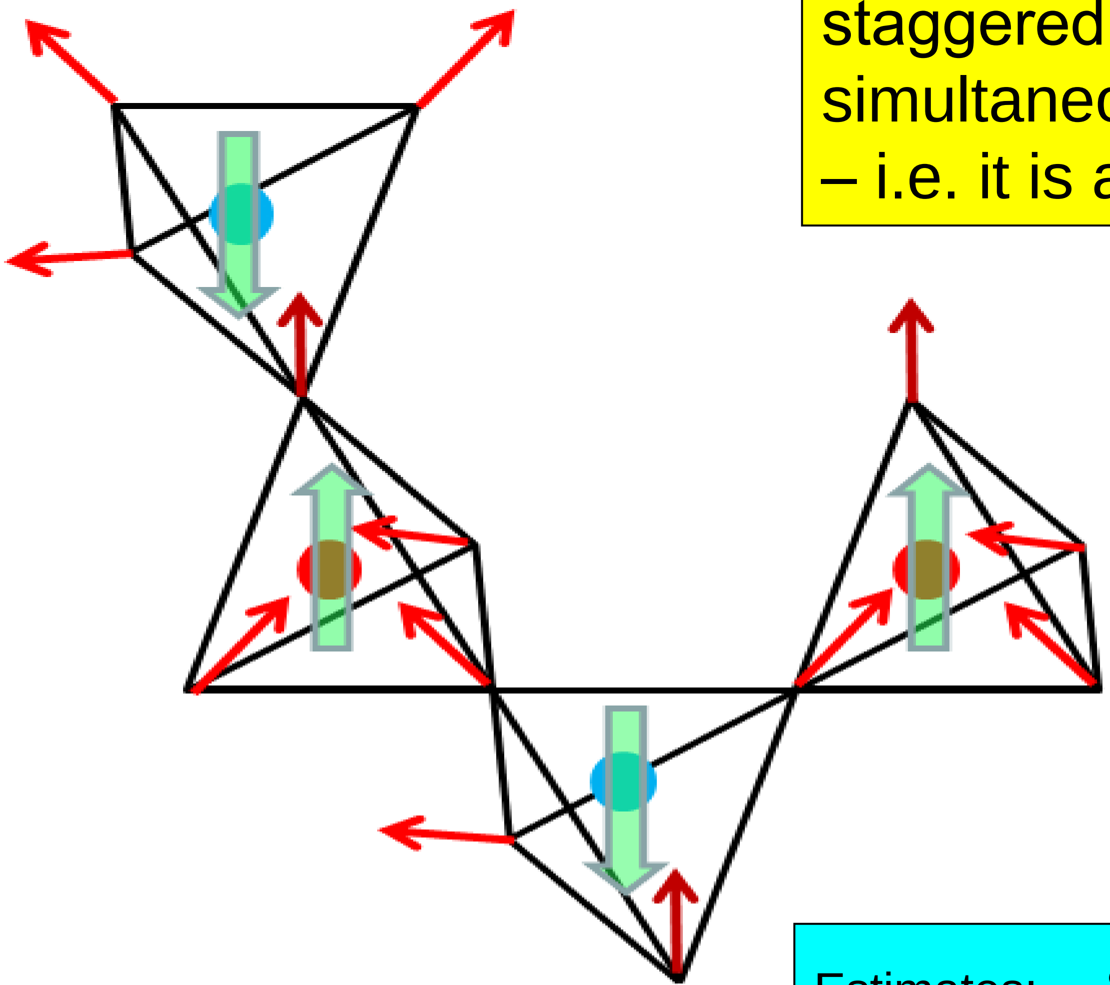
2-in/2-out (spin ice): **d=0**

3-in/1-out or 1-in/3-out (monopoles/antimonopoles): **d** $\neq$ 0

$$\langle n_1 \rangle = 1 + \delta n_1 = 1 - 8\left(\frac{t}{U}\right)^3 \left[\mathbf{S}_1(\mathbf{S}_2 + \mathbf{S}_3) - 2\mathbf{S}_2\mathbf{S}_3\right]$$

For 4-in state: from the condition $\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4 = 0$ $\delta n_1 = 0$. Change of $\mathbf{S}_1 \rightarrow -\mathbf{S}_1$ (3-in/1-out, *monopole*) gives nonzero charge redistribution and **d** $\neq$ 0.

Charge redistribution and dipoles are *even* functions of S; inversion of all spins does not change direction of a dipole: Direction of dipoles on monopoles and antimonopoles is *the same*: e.g. ***from the center of tetrahedron to a "special" spin***



In strong field **H** || [111] there is a staggered μ/ μ̲, and simultaneously staggered dipoles – i.e. it is an **antiferroelectric**

Estimates: ε=dE =eu(Å)E(V/cm)

for u~0.01Å and E ~10^5V/cm ε~10^{-5} eV~0.1K

Dipoles on monopoles, possible consequences:

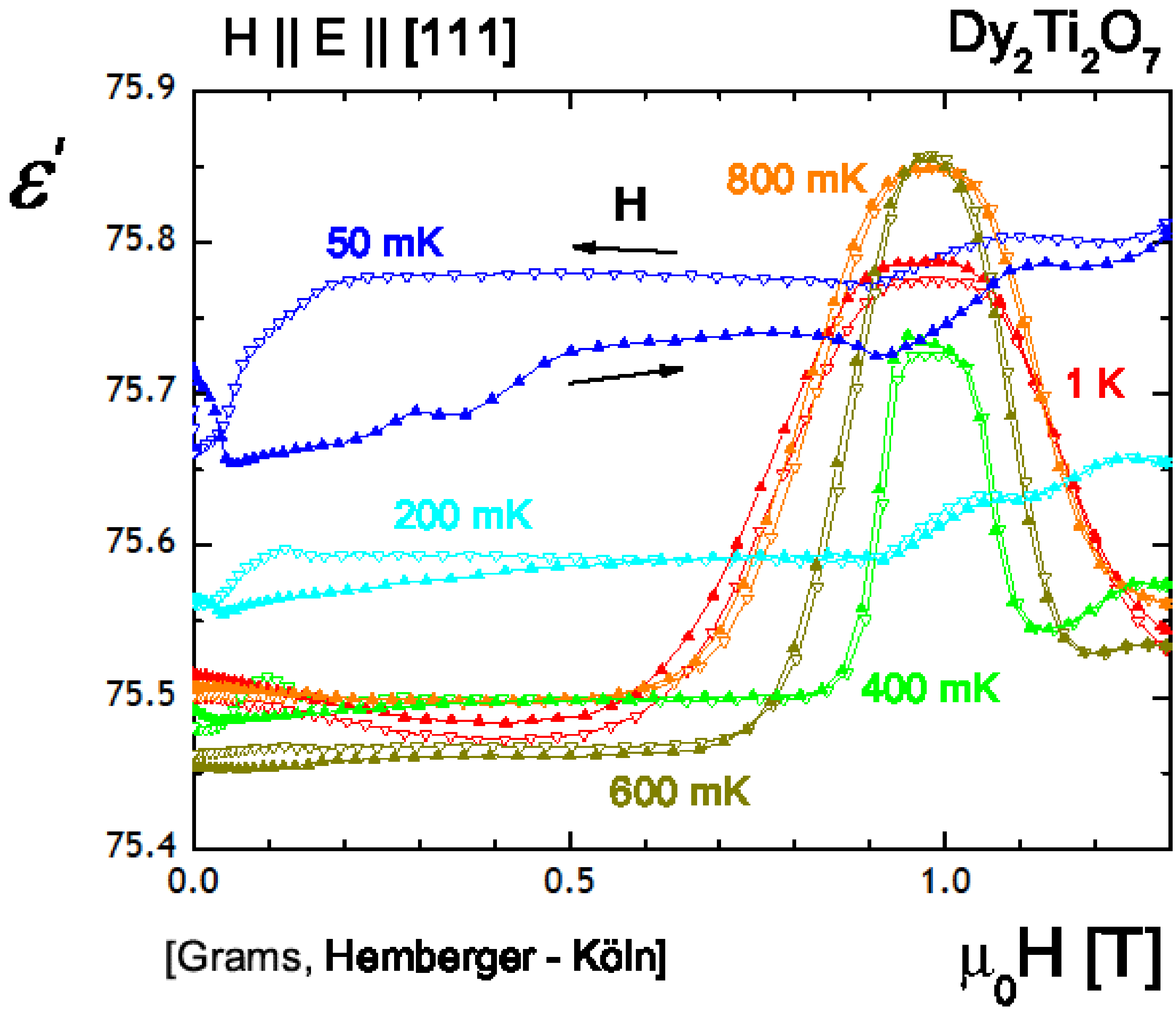
- **"Electric" activity of monopoles**; contribution to dielectric constant $\varepsilon(\omega)$

- **External electric field:**
Decreases excitation energy of certain monopoles
$\omega = \omega_0 - \mathbf{dE}$
Crude estimate: in the field $\mathbf{E} \sim 10^5$ V/cm energy shift ~ 0.1 K

- **Inhomogeneous electric field** (tip): will attract some monopoles/dipoles and repel other

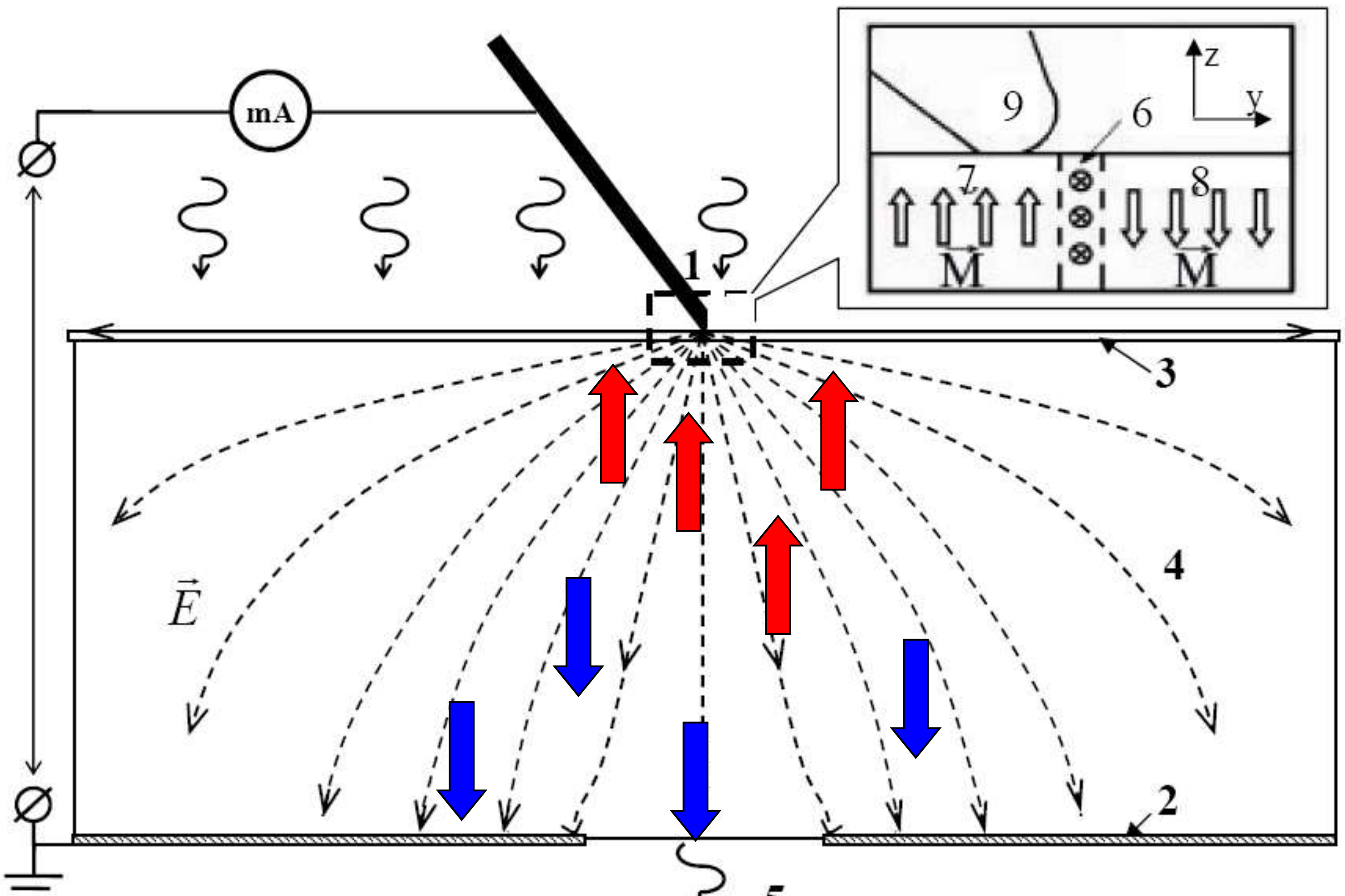
- In the magnetic field H || [001] **E** will promote monopoles, and decrease magnetization **M**, and decrease T_c

- In the field **H** || [111] – staggered Ising-like dipoles; in $\mathbf{E}_\perp$?



H || E || [111]
Dy2Ti2O7
ε'
75.9
75.8
75.7
75.6
75.5
75.4
H
800 mK
50 mK
1 K
200 mK
400 mK
600 mK
0.0
0.5
1.0
[Grams, Hemberger - Köln]
μ0H [T]

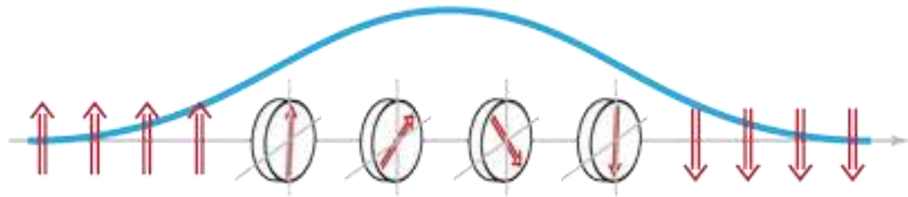
Inhomogeneous electric field



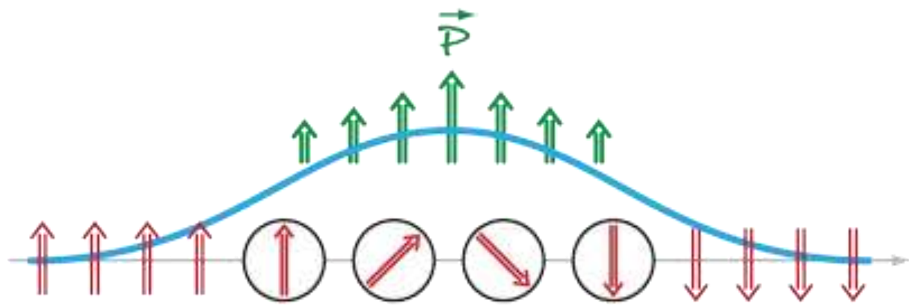
Electric dipoles at domain walls

Was already observed for Neel domain walls in ferromagnets   (cf. spiral multiferroics):

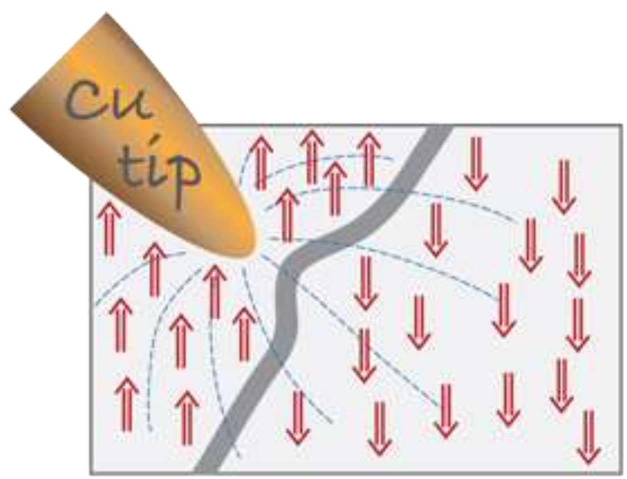
Bloch domain wall: a)



Neel domain wall: b)



c)



Logginov, Pyatakov et al. (Moscow State Univ.)

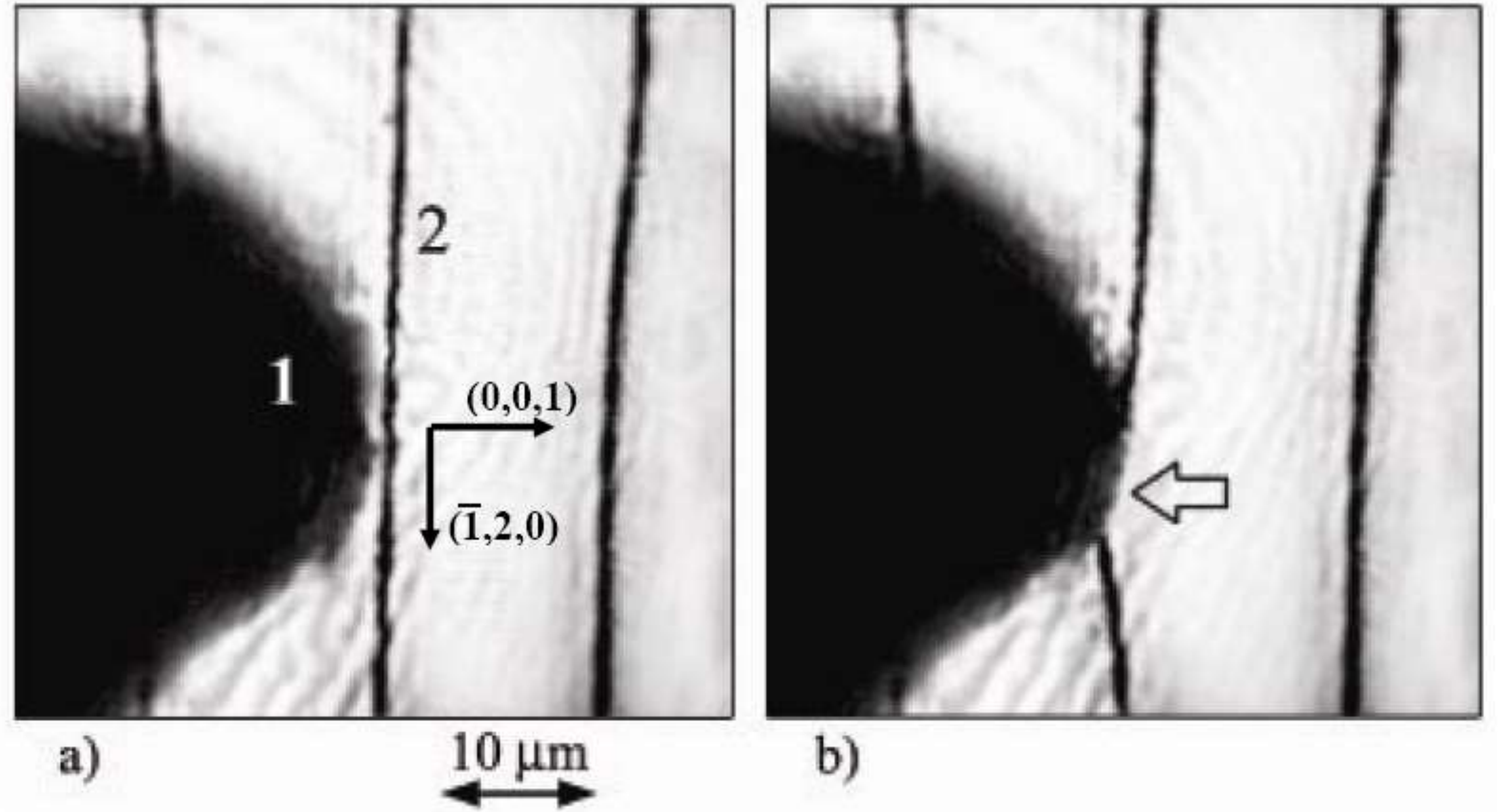


Fig. 2 The effect of electric field in the vicinity of electrode (1) on magnetic domain wall (2) in the films of ferrite garnets: a) initial state b) at the voltage of +1500 V applied

nature Vol 447|10 May 2007|doi:10.1038/nature05802

LETTERS

● Spiral structures in metal monolayers

Chiral magnetic order at surfaces driven by inversion asymmetry

M. Bode[1]†, M. Heide[2], K. von Bergmann[1], P. Ferriani[1], S. Heinze[1], G. Bihlmayer[2], A. Kubetzka[1], O. Pietzsch[1], S. Blügel[2] & R. Wiesendanger[1]

PRL 101, 027201 (2008) PHYSICAL REVIEW LETTERS week ending 11 JULY 2008

Atomic-Scale Spin Spiral with a Unique Rotational Sense: Mn Monolayer on W(001)

P. Ferriani,[1,*] K. von Bergmann,[1] E. Y. Vedmedenko,[1] S. Heinze,[1] M. Bode,[1,†] M. Heide,[2] G. Bihlmayer,[2] S. Blügel,[2] and R. Wiesendanger[1]

[1]*Institute of Applied Physics, University of Hamburg, Jungiusstrasse 11, 20355 Hamburg, Germany*

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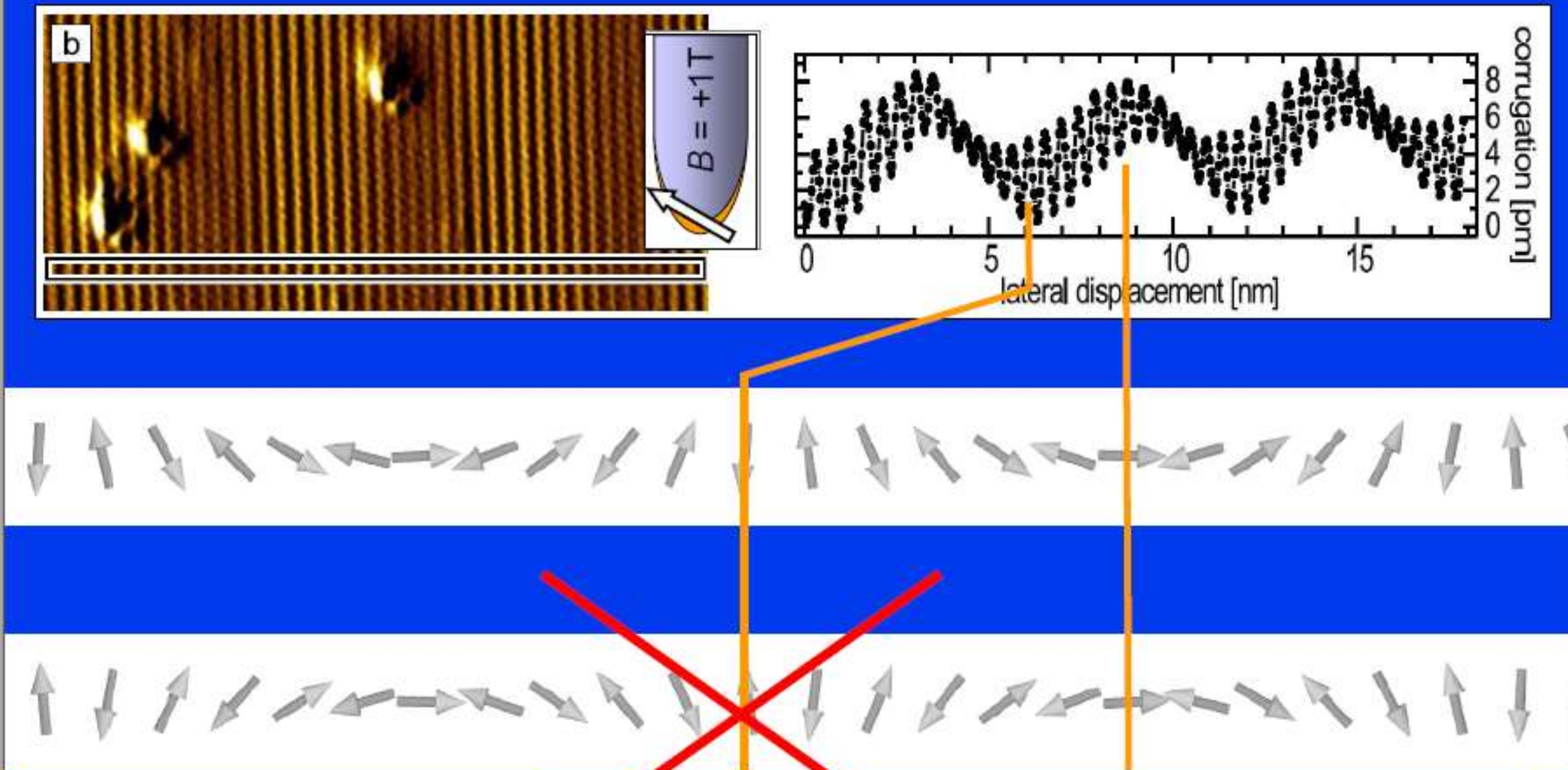
(Received 15 April 2008; published 7 July 2008)

Using spin-polarized scanning tunneling microscopy we show that the magnetic order of 1 monolayer Mn on W(001) is a spin spiral propagating along ⟨110⟩ crystallographic directions. The spiral arises on the atomic scale with a period of about 2.2 nm, equivalent to only 10 atomic rows. *Ab initio* calculations identify the spin spiral as a left-handed cycloid stabilized by the Dzyaloshinskii-Moriya interaction, imposed by spin-orbit coupling, in the presence of softened ferromagnetic exchange coupling. Monte Carlo simulations explain the formation of a nanoscale labyrinth pattern, originating from the coexistence of the two possible rotational domains, that is intrinsic to the system.

rotation direction of spin spiral

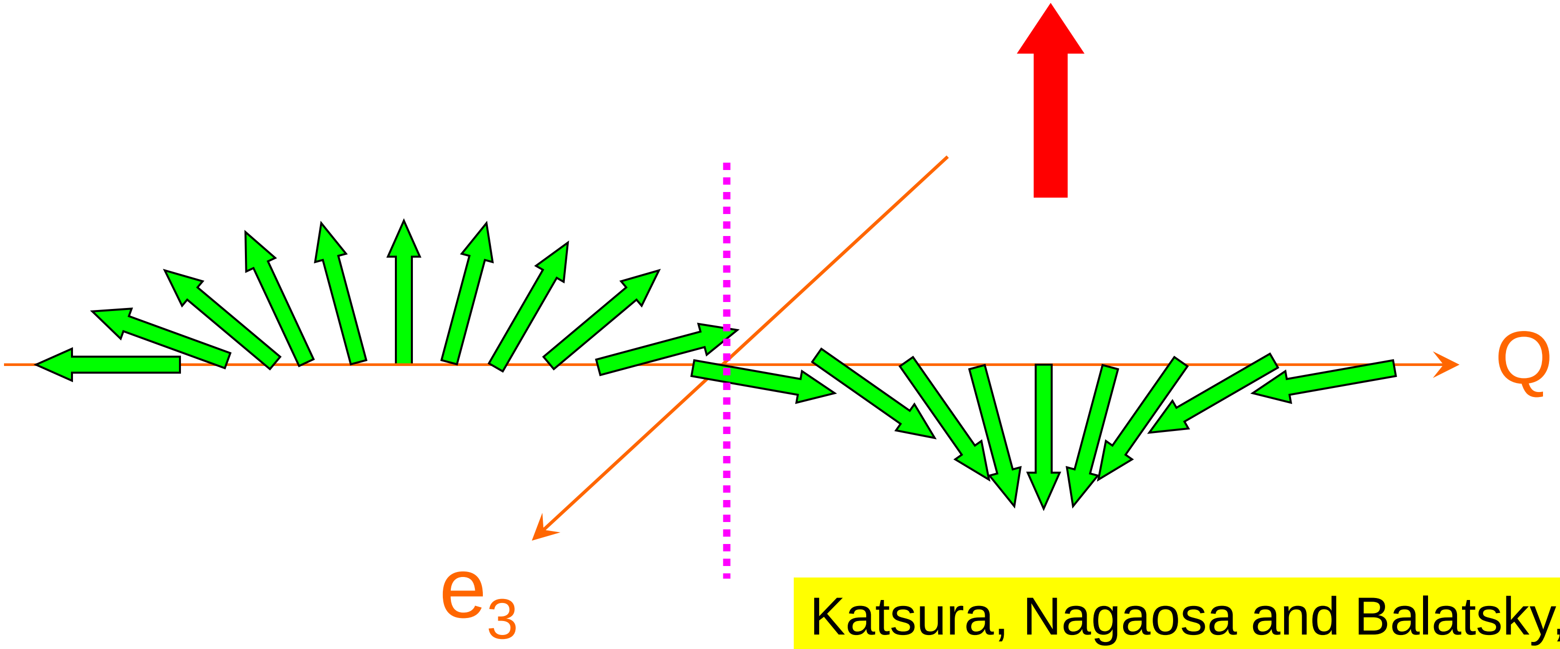


6 out of 6 independent islands: same rotational sense
→ DM-driven spin spiral

Cycloidal SDW

$$\mathbf{M} = A_1\mathbf{e}_1 \cos Qx + A_2\mathbf{e}_2 \sin Qx + A_3\mathbf{e}_3$$

$$\boxed{\overline{\mathbf{P}} \propto [\mathbf{e}_3 \times \mathbf{Q}]}$$



Katsura, Nagaosa and Balatsky, 2005

Mostovoy 2006

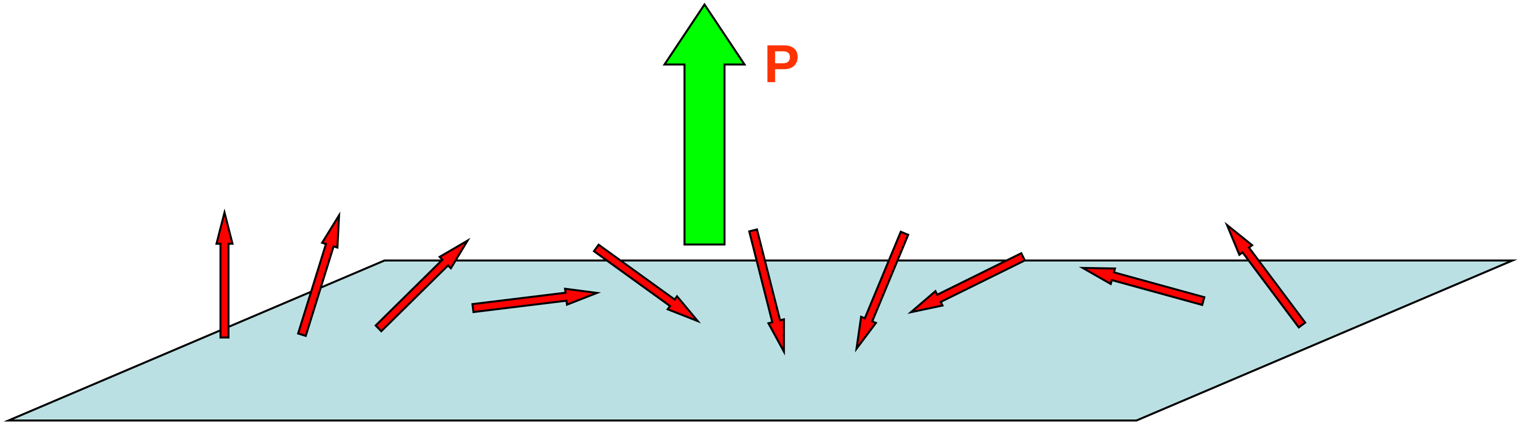
Simple explanation: at the surface there is a drop of a potential (work function, double layer)

I.e. there is an electric field **E**, or polarization **P** perpendicular to the surface

By the relation

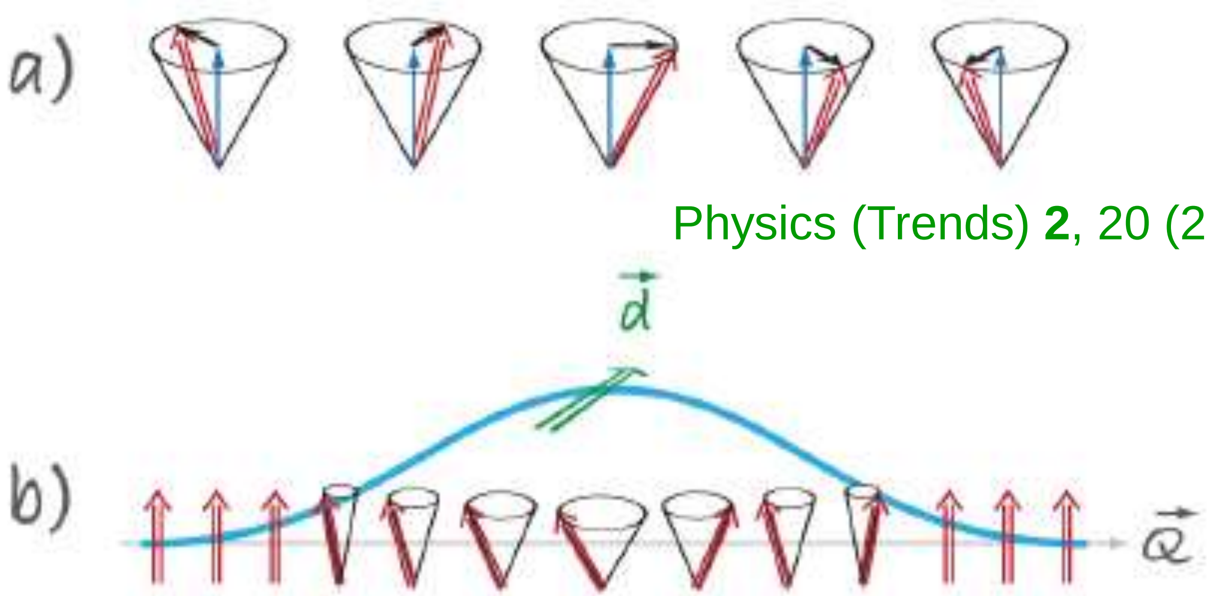
$$\overline{\mathbf{P}} \propto \left[\mathbf{e}_3 \times \mathbf{Q}\right]$$

there will appear magnetic spiral with certain sense of rotation, determined by **P**



Electric dipole carried by the usual spin wave

D.Khomskii, Physics (Trends) **2**, 20 (2009)



Physics (Trends) **2**, 20 (2009)

FIG. 5: How polarization emerges in a spin wave (magnon). (a) The classical picture of a spin wave in a ferromagnet: the spin (red arrow) precesses about a fixed axis (blue). The deviation is measured by the black arrows. (b) According to Eq. (1), as a spin-wave packet propagates along Q, it will also carry an electric dipole moment. (Illustration: Alan Stonebraker)

● Monopoles in magnetoelectrics?

nature COMMUNICATIONS

ARTICLE

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DOI: 10.1038/ncomms5793

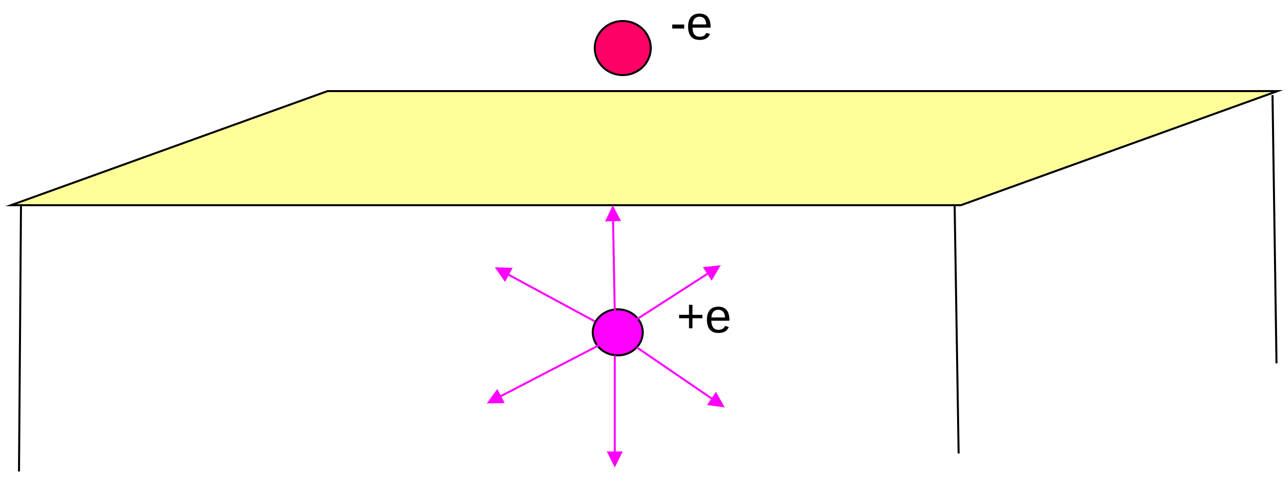
Magnetic monopoles and unusual dynamics of magnetoelectrics

D.I. Khomskii[1]

NATURE COMMUNICATIONS | 5:4793 | DOI: 10.1038/ncomms5793 | www.nature.com/naturecommunications

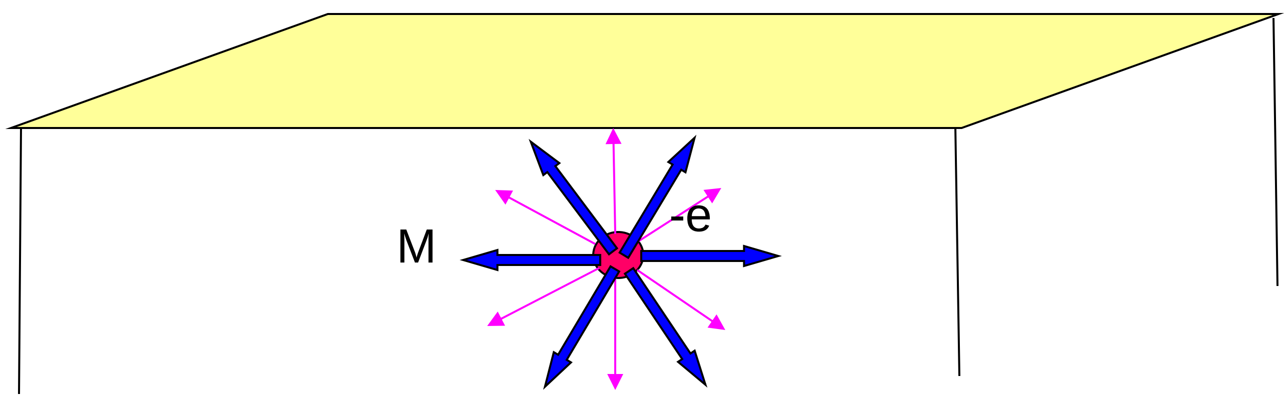
Magnetic monopoles in topological insulators

Charge close to a surface of ME material: $M_i = \alpha_{ij} E_j$



Charge inside of of ME material: $M_i = \alpha_{ij} E_j$, , $H=4\pi M$

Let $\alpha_{ij} = \alpha\delta_{ij}$, diagonal: magnetic field outside of the charge looks like a field of a magnetic monopole **$\mu = 4\pi\alpha e$**



Moving electron $\rightarrow$ moving monopole.

Electron in a magnetic field: force $\mathbf{F} = \mu\mathbf{H} = 4\pi\alpha e\mathbf{H}$

(But one can also consider it as an action of the electric field created in magnetoelectric material on the electric charge: $\mathbf{E} = 4\pi\alpha\mathbf{H}$, $\mathbf{F} = \mathbf{E}e = 4\pi\alpha e\mathbf{H}$)

Other possible effects ? (how to find, to measure such monopoles)

"Electric Hall effect": if electric charge e moving in **H** gives a Hall effect, a monopole moving in electric field will do the same

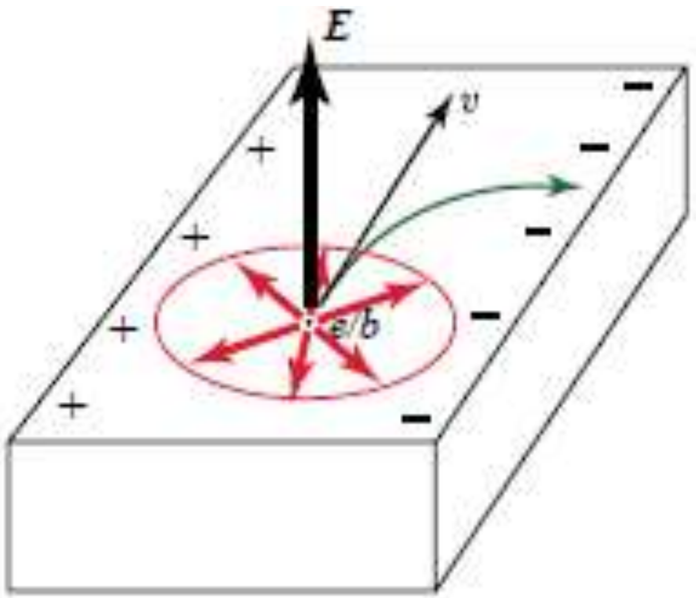


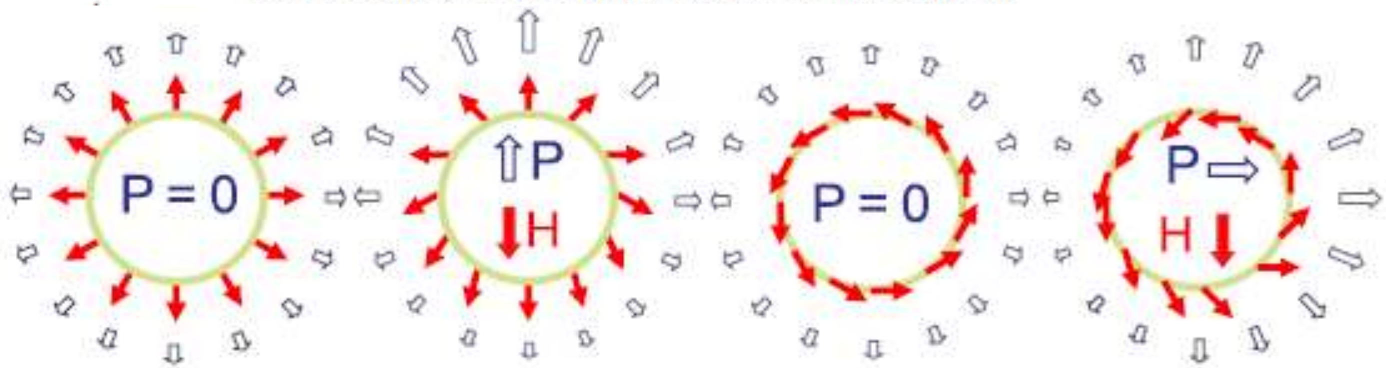
Figure 2: Motion of a magnetic monopole b, accompanying electric charge e, in a perpendicular electric field. The force $b[\boldsymbol{v} \times \boldsymbol{E}]$ will bend the trajectory of this object, leading to an "electric Hall effect".

But one can also explain this effect as the usual Hall effect in an effective magnetic field **B** ~ α**E**

Magnetic vortices as magnetoelectrics

Superexchange-Driven Magnetoelectricity in Magnetic Vortices

Kris T. Delaney,[1] Maxim Mostovoy,[2] and Nicola A. Spaldin[1]



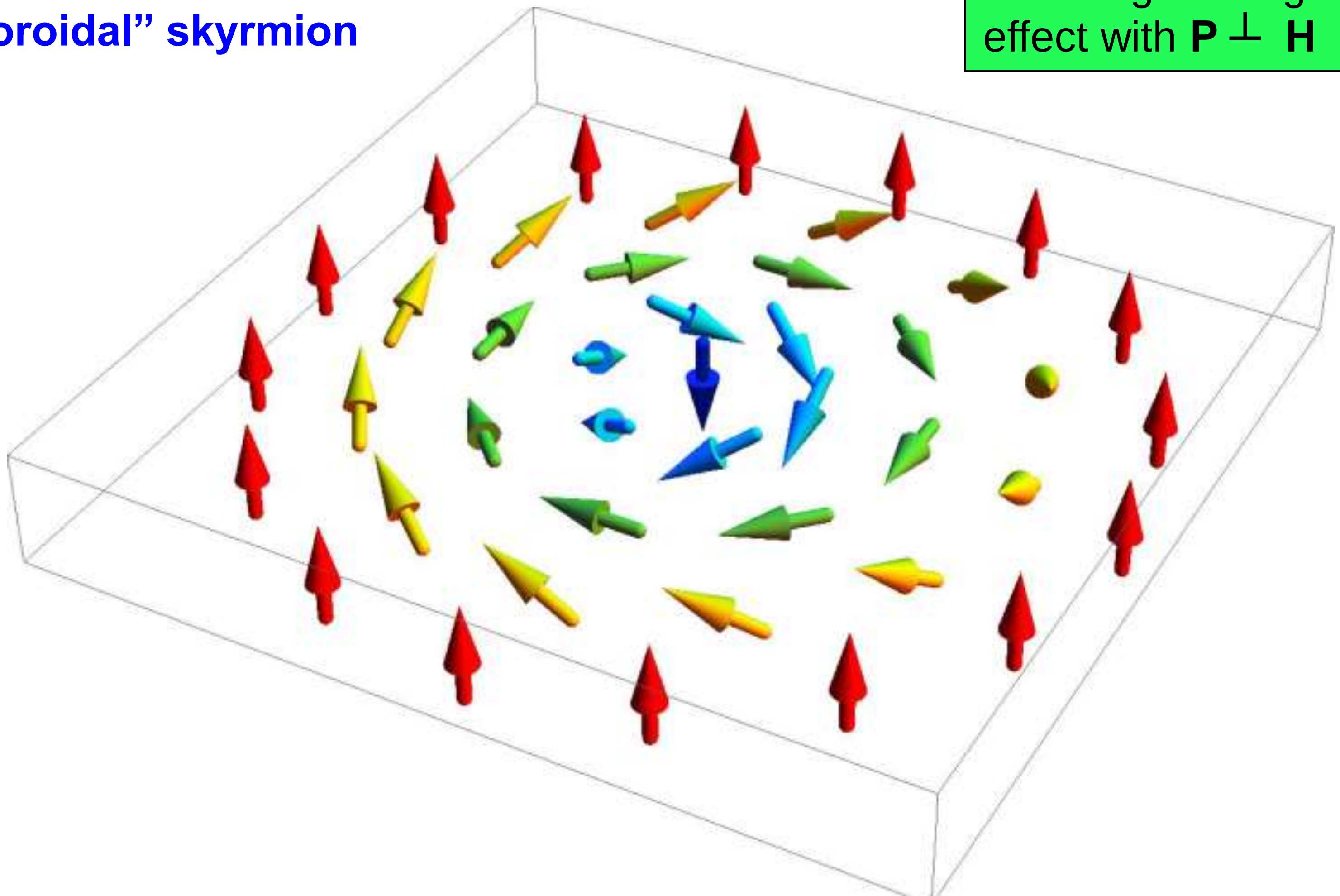
(a) (b) (c) (d)

FIG. 1 (color online). (a) A magnetic vortex carrying a monopole moment. The thin solid arrows indicate the spin orientation, while the thick open arrows show the local polarization vector. (b) A magnetic field applied to the vortex shown in (a) induces a net polarization along the field direction. (c) A magnetic vortex with a toroidal moment. (d) A magnetic field applied to (c) induces an electric polarization perpendicular to the field.

● Skyrmions in magnetic crystals

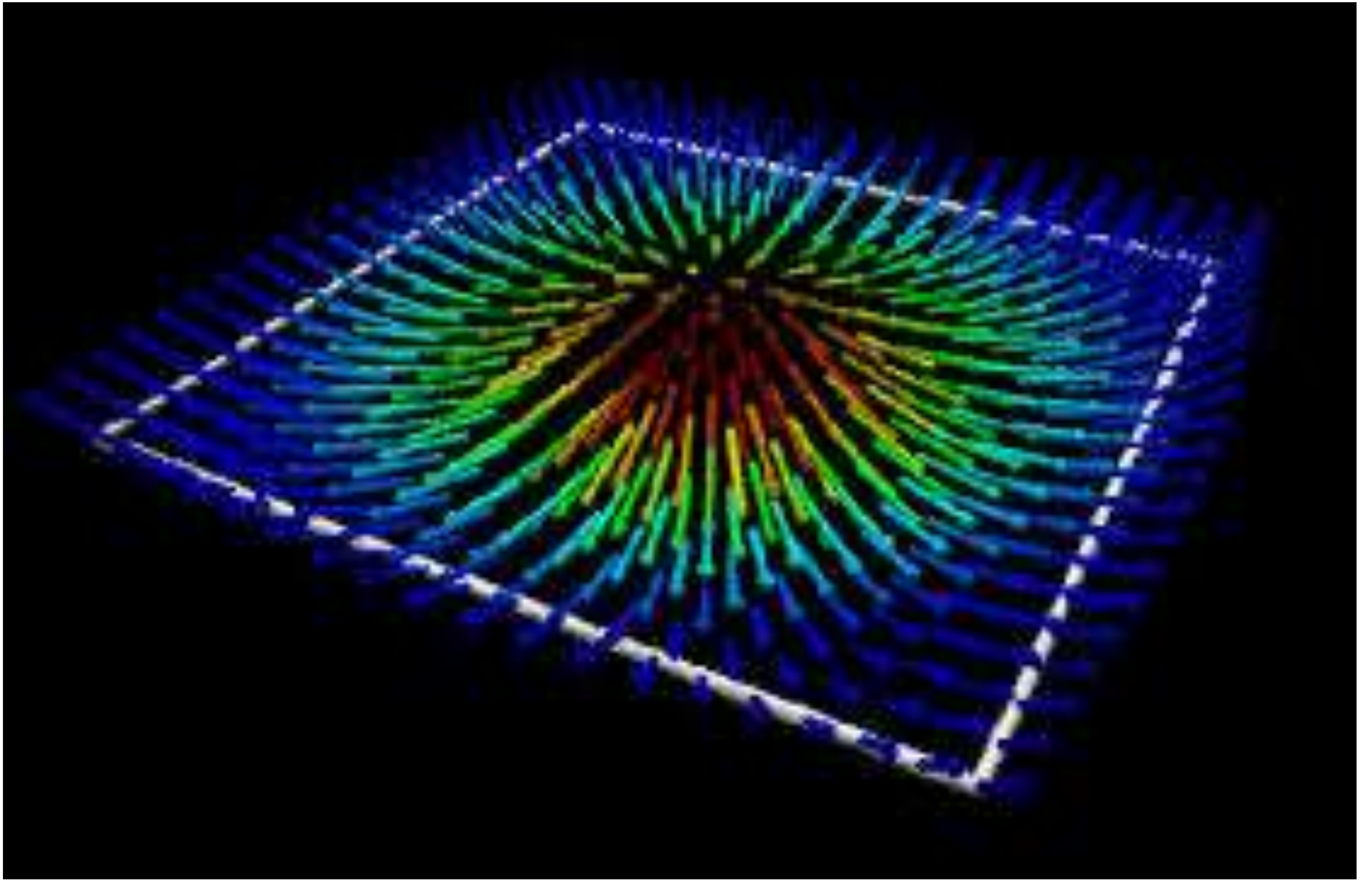
Should give magnetoelectric effect with **P** ⊥ **H**

"Toroidal" skyrmion



Skyrmion lattice (e.g. in MnSi) – C.Pfleiderer, A Rosch

“Radial” skyrmion



Should give magnetoelectric effect with **P** ‖ **H**

Conclusions

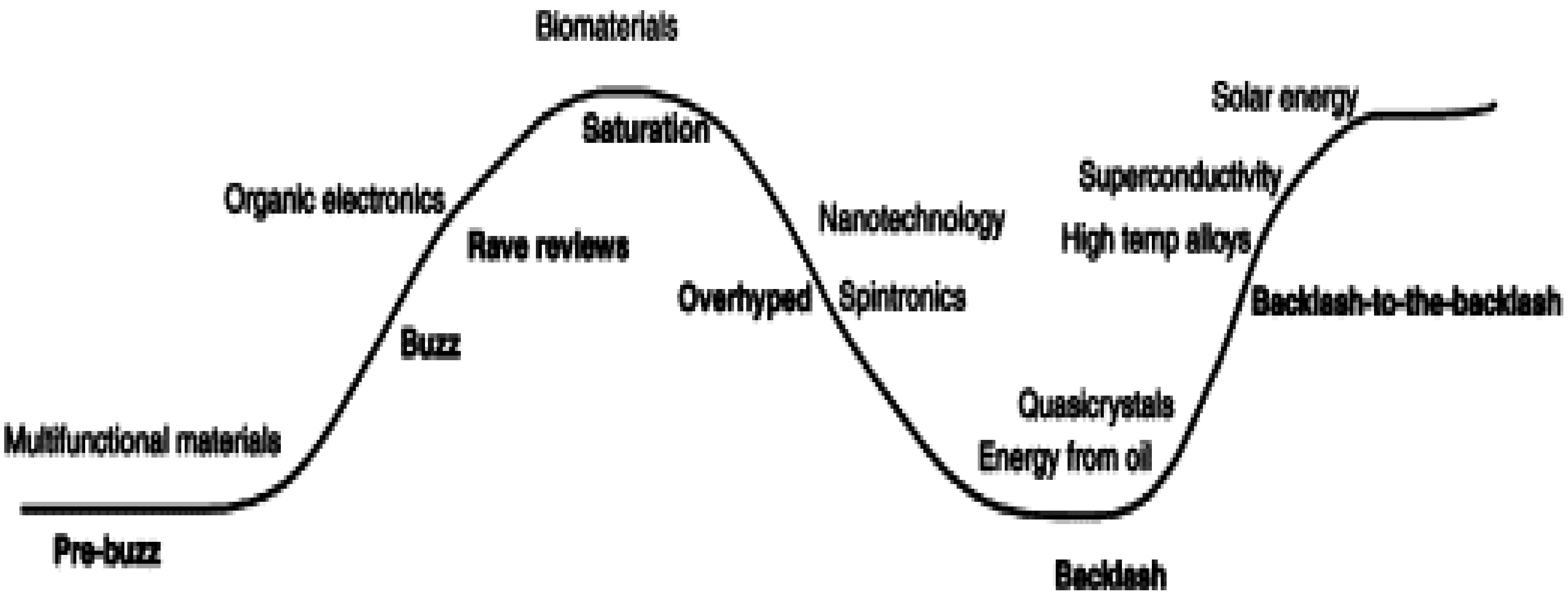
- **There is strong interplay of electric and magnetic properties in solids, having different forms**
- **These are: magnetoelectrics; multiferroics**
- **Multiferroics can be metamaterials at certain frequencies**
- **There should be an electric dipole at each magnetic monopole in spin ice – with different consequences**

  Analogy: electrons have *electric charge* and
  spin/magnetic dipole
  monopoles in spin ice have *magnetic charge*
  and *electric dipole*

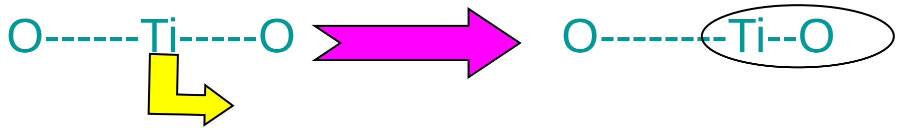
- **Ordinary spin waves in ferromagnets should carry dipole moment**
- **Different magnetic textures (domain walls, magnetic vortices) can either carry dipole moment, or can be magnetoelectric**
- **Electric charges in magnetoelectric should be accompanied by magnetic monopoles**

Steve Pearton, Materials Today **10**, 6 (2007)

"The **F**lorida **L**aw of **O**riginal **P**rognostication maps the shifting tide of expectations in materials science."



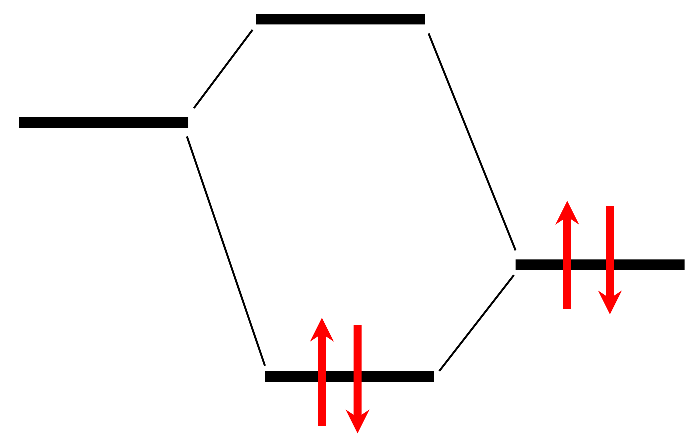
Ti^{4+}: establishes *covalent bond* with oxygens (which “donate” back the electrons), using empty d-levels



Better to have one strong bond with one oxygen that two weak ones with oxygens on the left and on the right

Two possible reasons:

d^0 configurations: only bonding orbitals are occupied



Other localized d-electrons break *singlet* chemical bond by Hund’s rule pair-breaking (a la pair-breaking of Cooper pairs by magnetic impurities)

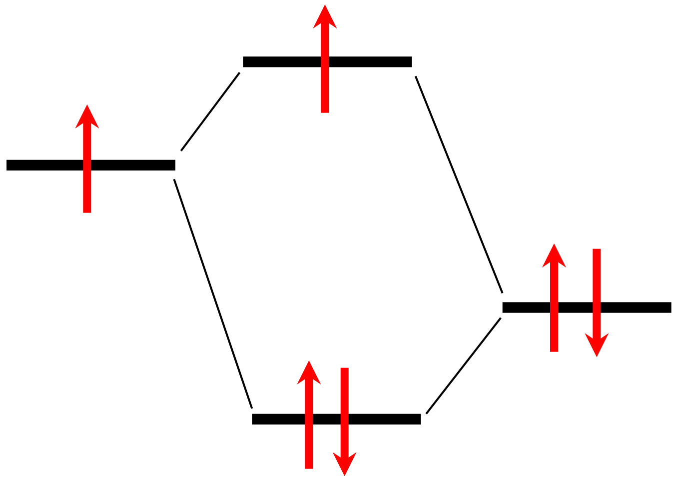
Ti^{4+}: establishes *covalent bond* with oxygens (which “donate” back the electrons), using empty d-levels

O------Ti-----O ⟹ O--------Ti--O

Better to have one strong bond with one oxygen that two weak ones with oxygens on the left and on the right

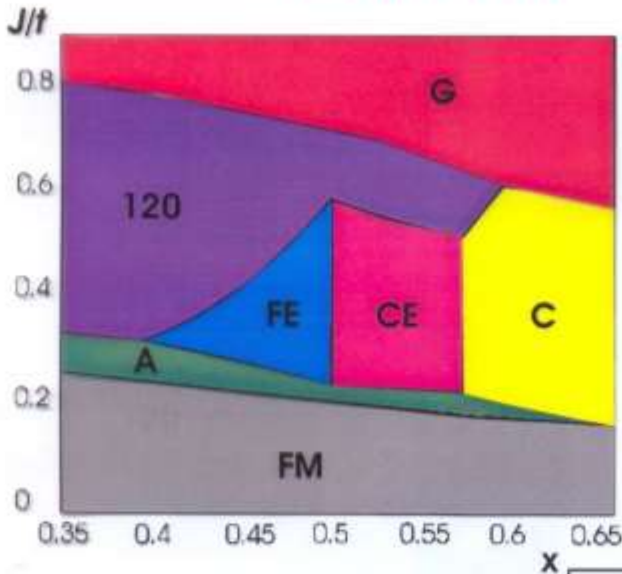
Two possible reasons:

d^0 configurations: only bonding orbitals are occupied

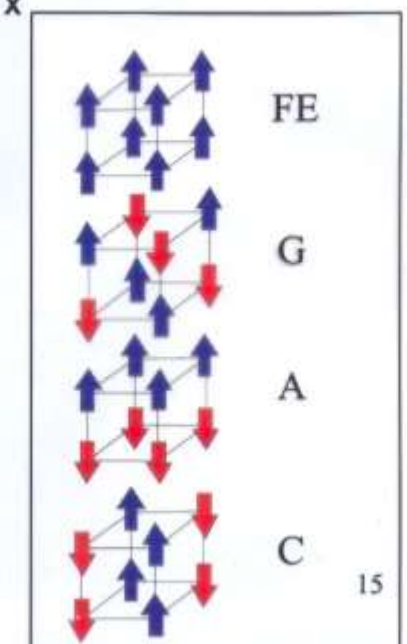


Other localized d-electrons break *singlet* chemical bond by Hund’s rule pair-breaking (a la pair-breaking of Cooper pairs by magnetic impurities)

Phase diagram of manganites near x=0.5



Electron density $n_e = 1\text{-}x$



FM: ferromagnetic phase

G: antiferromagnetic Neel state

A: F planes coupled AFM

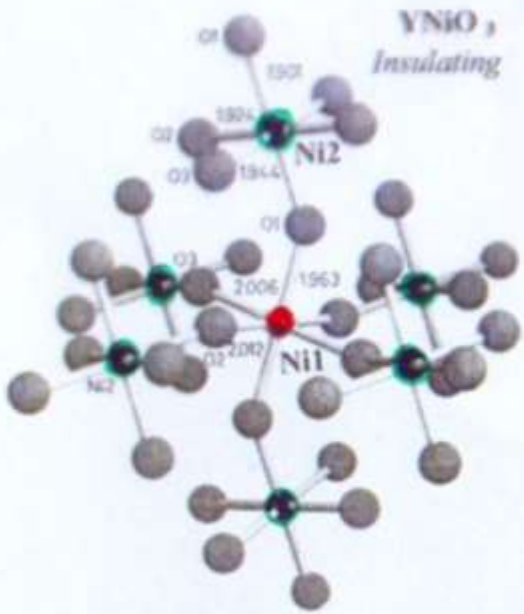
C: F chains coupled AFM

CE: CE-phase

120: Jaffet-Kittel state

FE: Ferroelectric phase

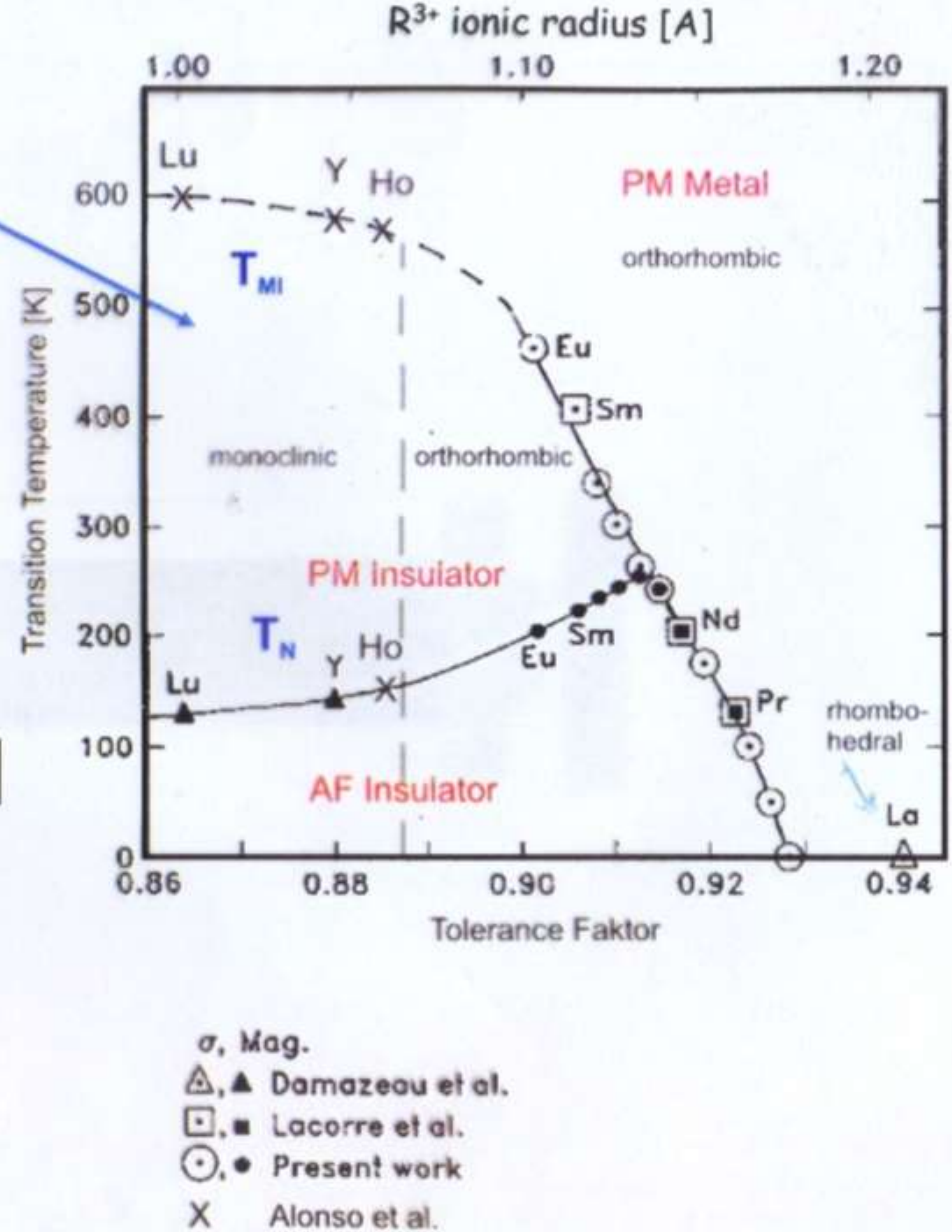
Charge ordering



$2Ni^{3+} \rightarrow Ni^{3+\delta} + Ni^{3-\delta}$ $(\delta \approx 0.35)$

$Ni(1) = 1.4(1)\ \mu_B$
and
$Ni(2) = 0.7(1)\ \mu_B$

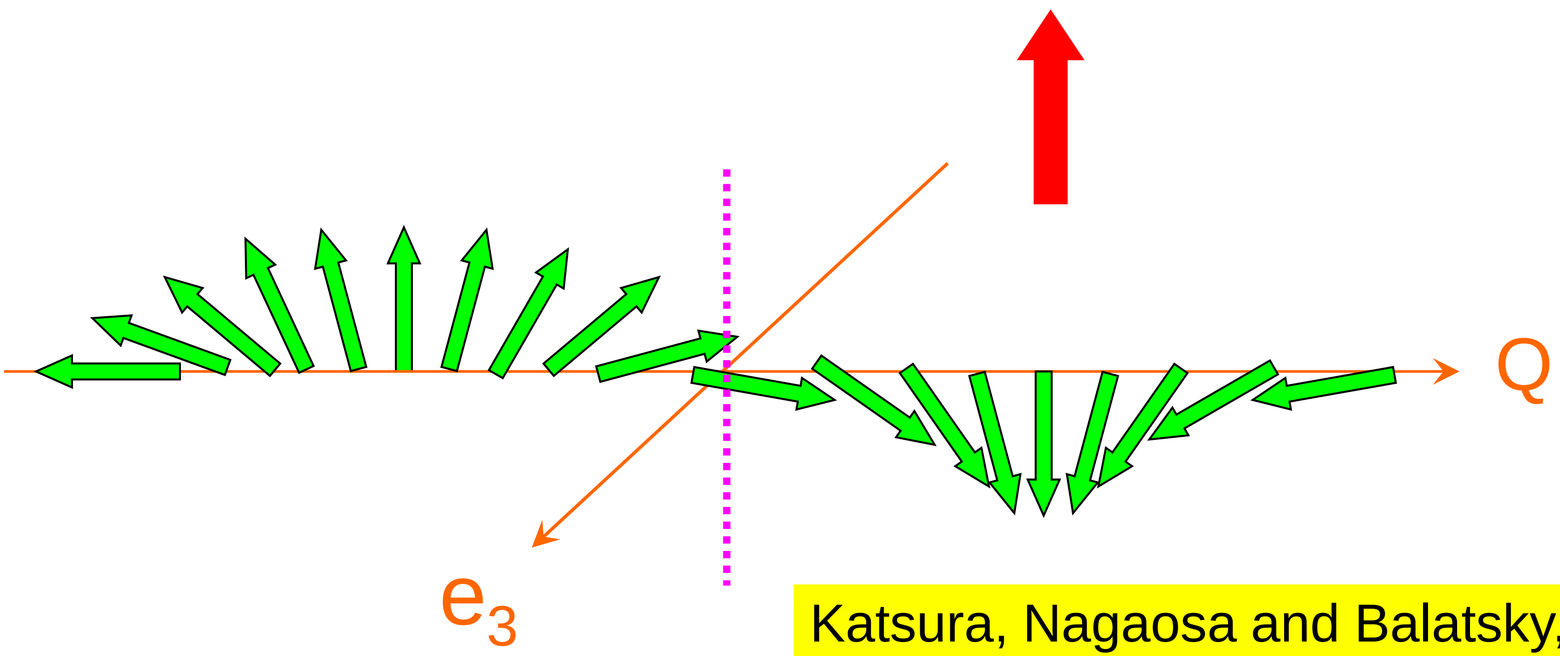
neutron diffraction, J.A. Alonso et al.
PRL 82, 3871 (99)



Cycloidal SDW

$$\mathbf{M} = A_1\mathbf{e}_1 \cos Qx + A_2\mathbf{e}_2 \sin Qx + A_3\mathbf{e}_3$$

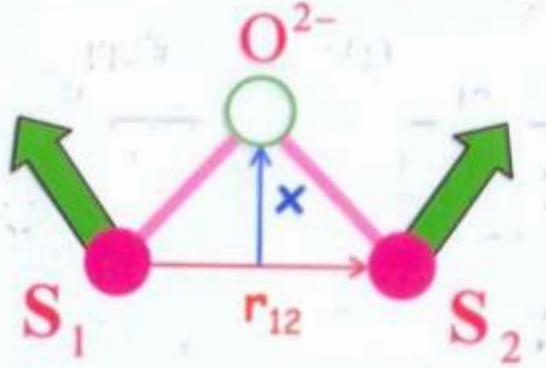
$$\boxed{\overline{\mathbf{P}} \propto [\mathbf{e}_3 \times \mathbf{Q}]}$$



Katsura, Nagaosa and Balatsky, 2005

Mostovoy 2006

Effects of Dzyaloshinskii-Moriya interaction

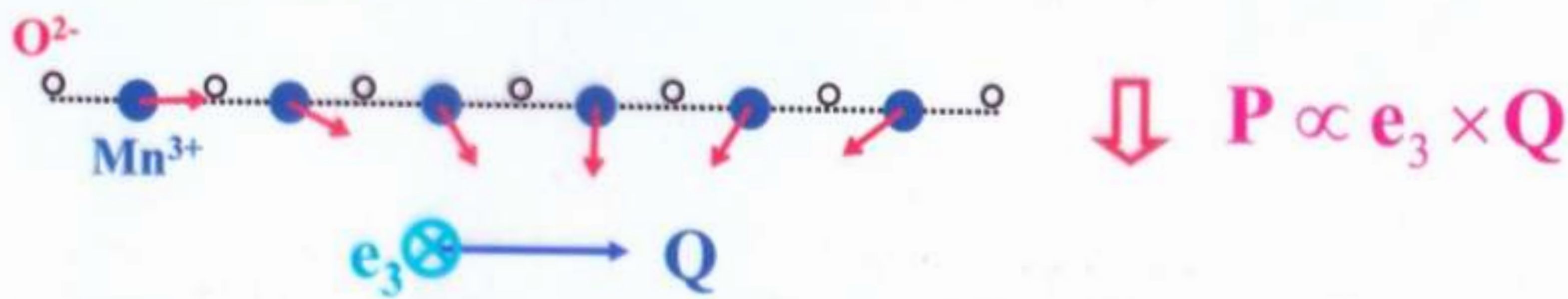


$$E_{DM} = \mathbf{D}_{12} \cdot [\mathbf{S}_1 \times \mathbf{S}_2]$$

$$\mathbf{D}_{12} \propto \lambda \, \mathbf{x} \times \hat{\mathbf{r}}_{12}$$

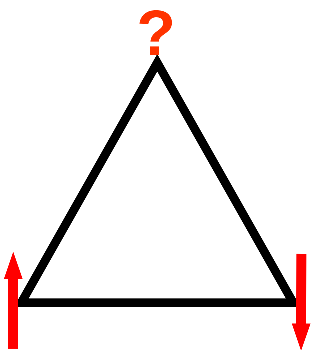
Weak ferromagnetism

Weak ferroelectricity



H. Katsura et al PRL (2005), Sergienko&Dagotto PRB (2006)

Spin systems: often complicated spin structures, especially in frustrated systems – e.g. those containing triangles as building blocks



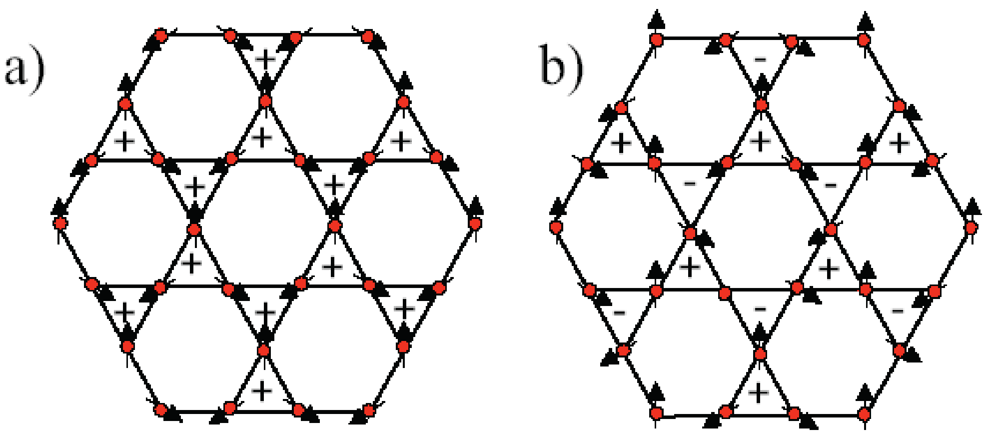
- Isolated triangles (trinuclear clusters) - e.g. in some magnetic molecules (V15, …)
- Solids with isolated triangles ($La_4Cu_3MoO_{12}$)
- Triangular lattices
- Kagome
- Pyrochlore

Scalar chirality χ is often invoked in different situations:

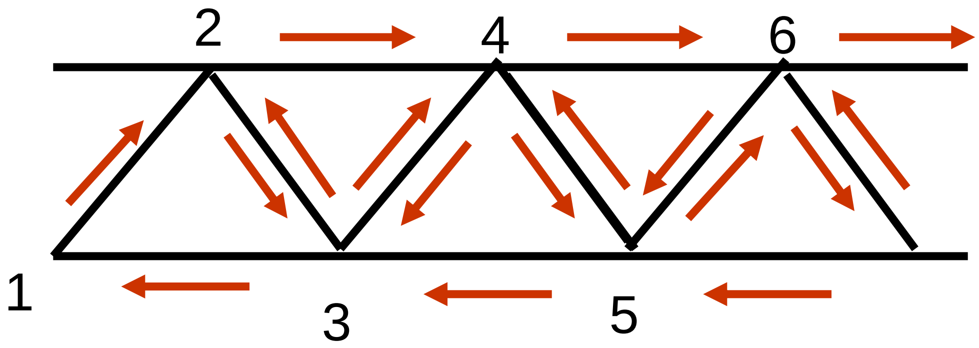
- Anyon superconductivity
- Berry-phase mechanism of anomalous Hall effect
- New universality classes of spin-liquids
- Chiral spin glasses

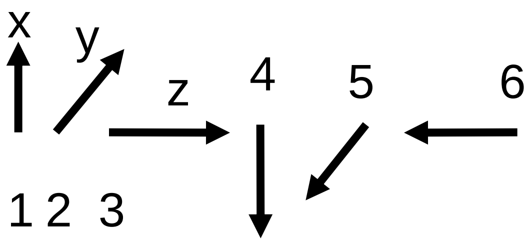
Chirality in frustrated systems: Kagome

a) Uniform chirality (q=0)   b) Staggered chirality ($\sqrt{3}$x$\sqrt{3}$)



Boundary and persistent current





$$\chi = const$$

Boundary current in
gaped 2d insulator

Chirality as a qubit?

Triangle: S=1/2, chirality (or pseudosin T) = ½

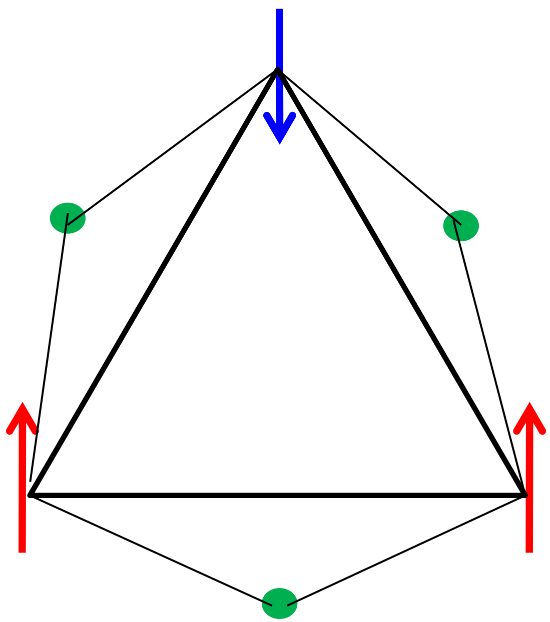
Can one use chirality instead of spin for quantum computation etc,
as a qubit instead of spin?

We can control it by magnetic field (chirality = current = orbital moment)
and by electric field

Georgeot, Mila, arXiv 26 February 2009

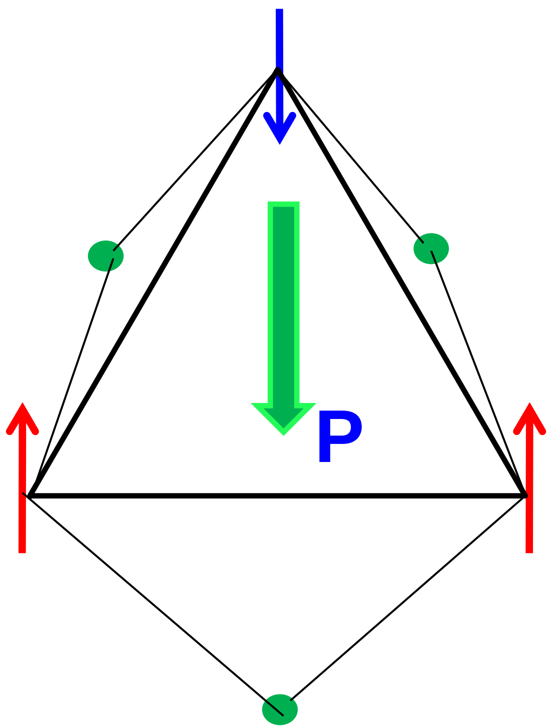
Dipoles are also created by lattice distortions (striction); the expression for

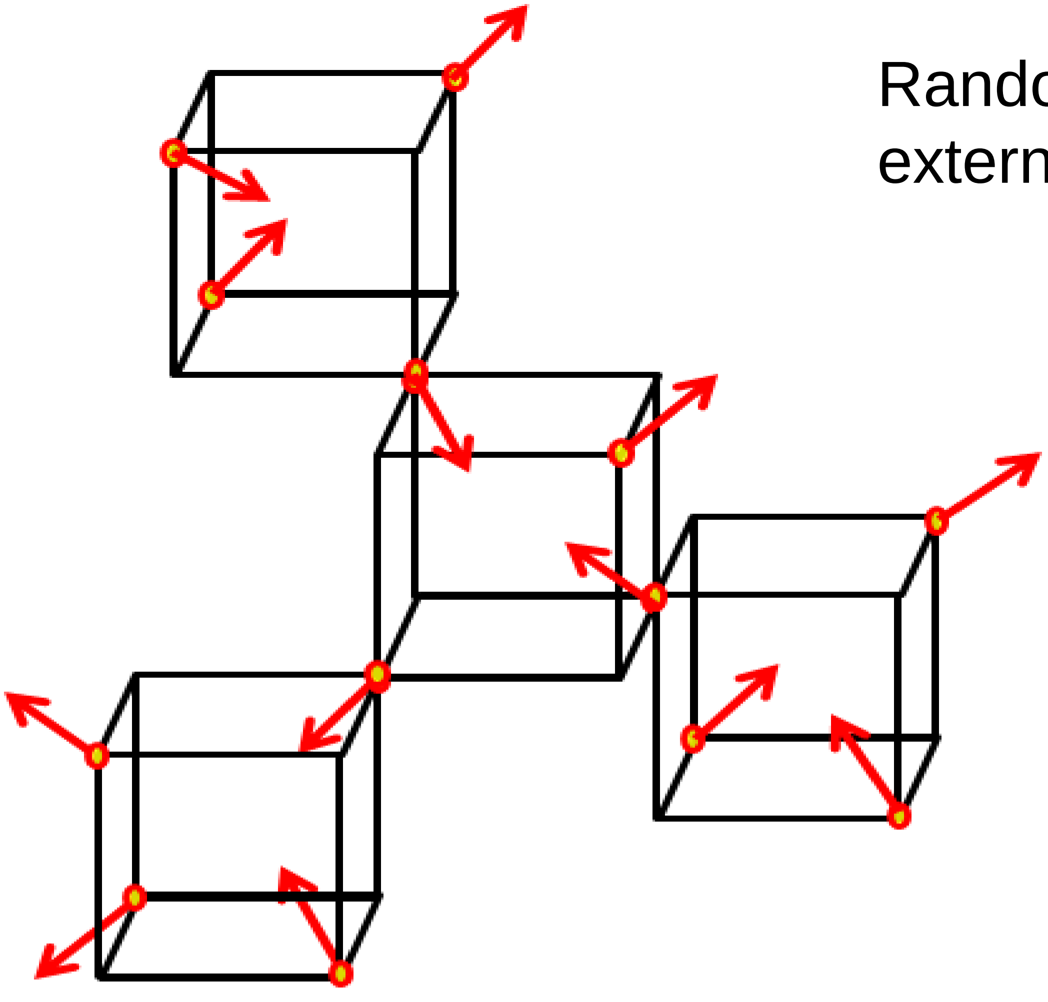
polarization/dipole is the same, $\mathbf{D} \sim \mathbf{P} \sim \mathbf{S}_1(\mathbf{S}_2\text{-}\mathbf{S}_3) - 2\mathbf{S}_2\mathbf{S}_3$ (M.Mostovoy)



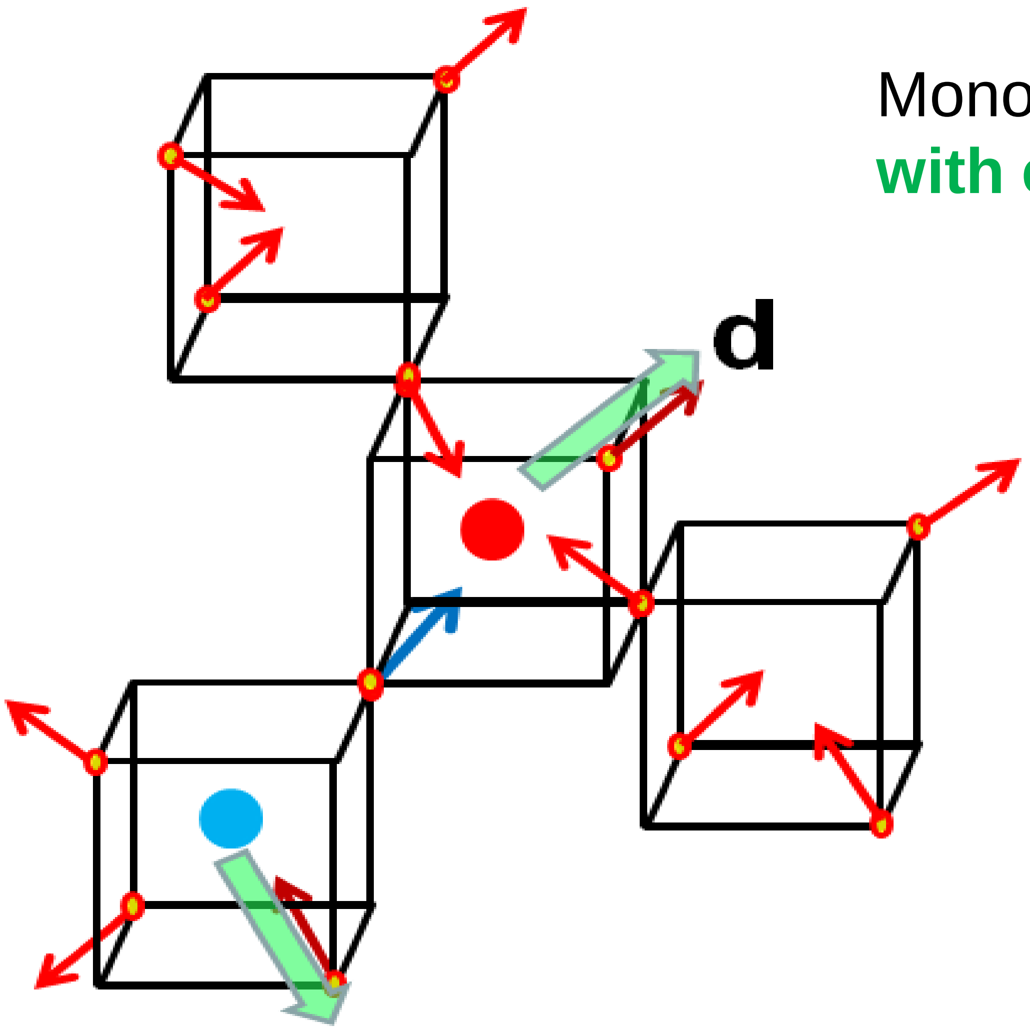
Dipoles are also created by lattice distortions (striction); the expression for

polarization/dipole is the same, $\mathbf{D} \sim \mathbf{P} \sim \mathbf{S}_1(\mathbf{S}_2 - \mathbf{S}_3) - 2\mathbf{S}_2\mathbf{S}_3$ (M.Mostovoy)

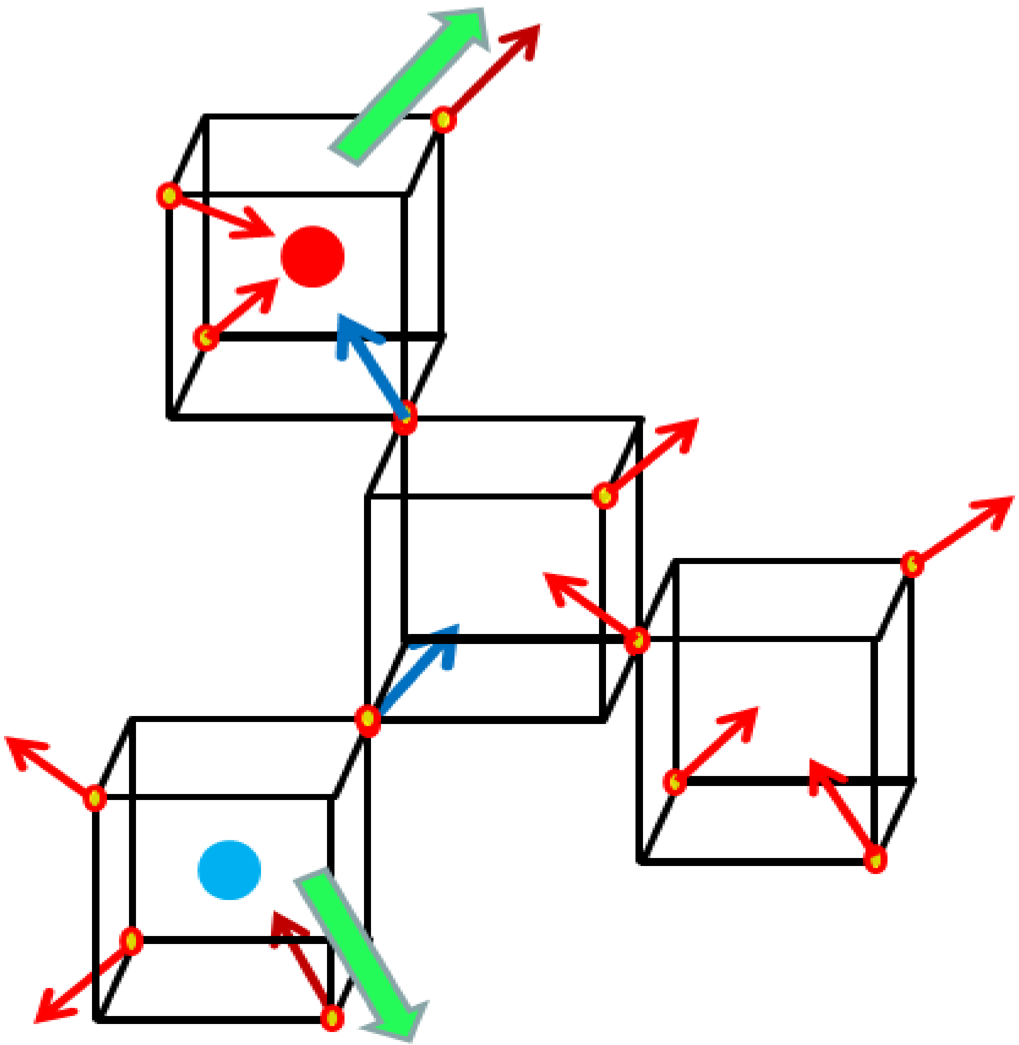


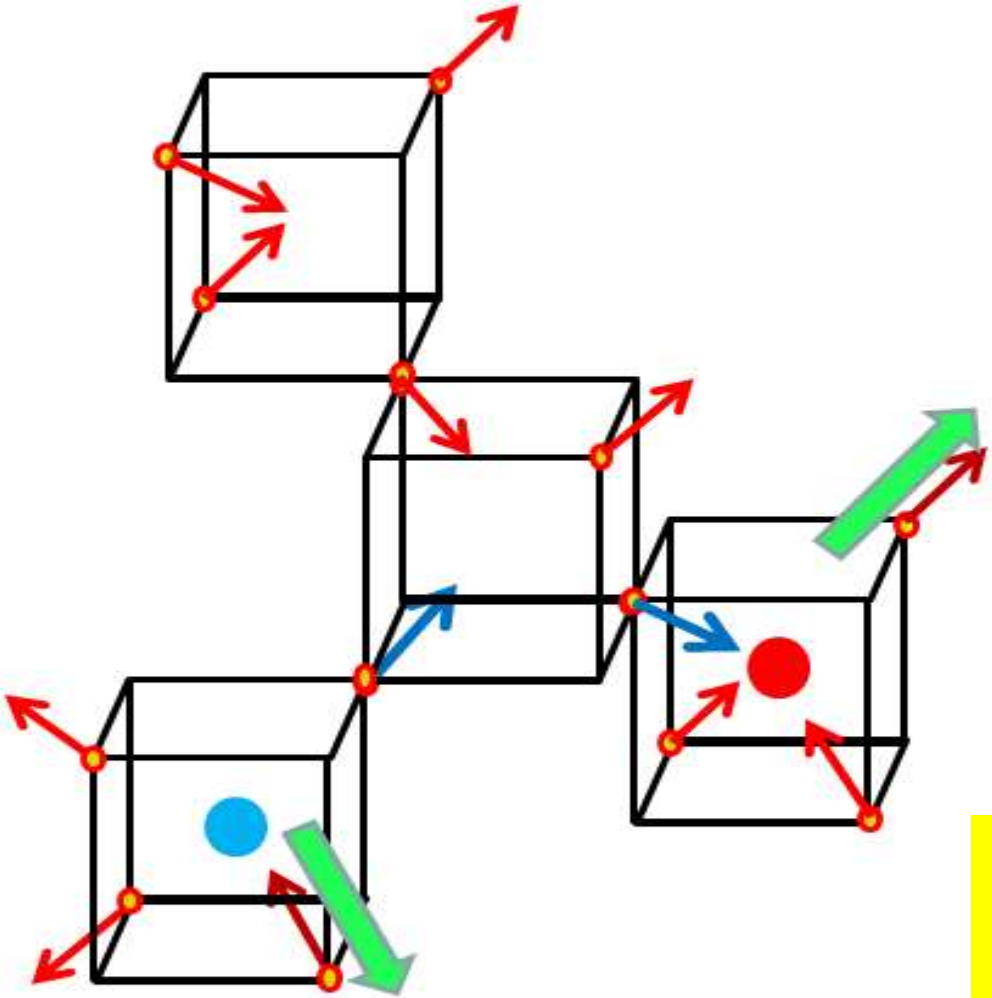


Random ice rule spins (no external magnetic field)



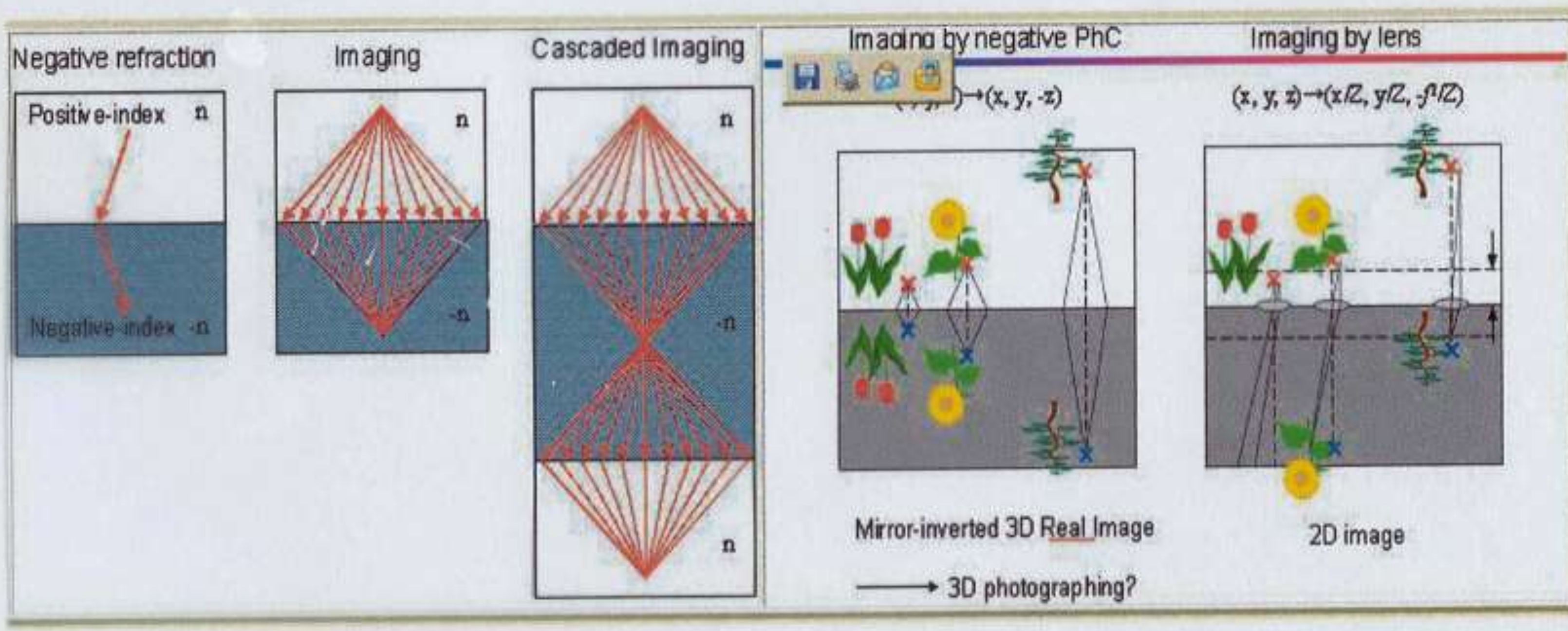
Monopoles/antimonopoles
with electric dipoles





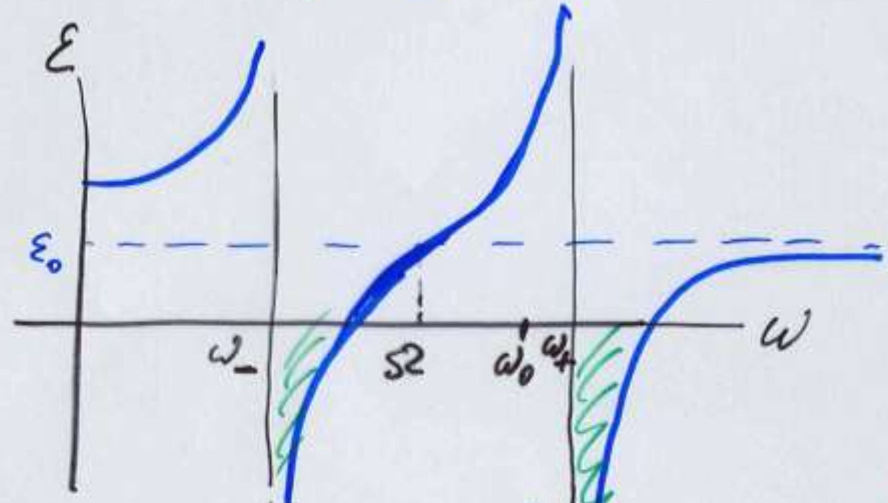
In general directions of electric dipoles are “random” – in any of [111] directions

Why NR is interesting: Focusing by a slab.

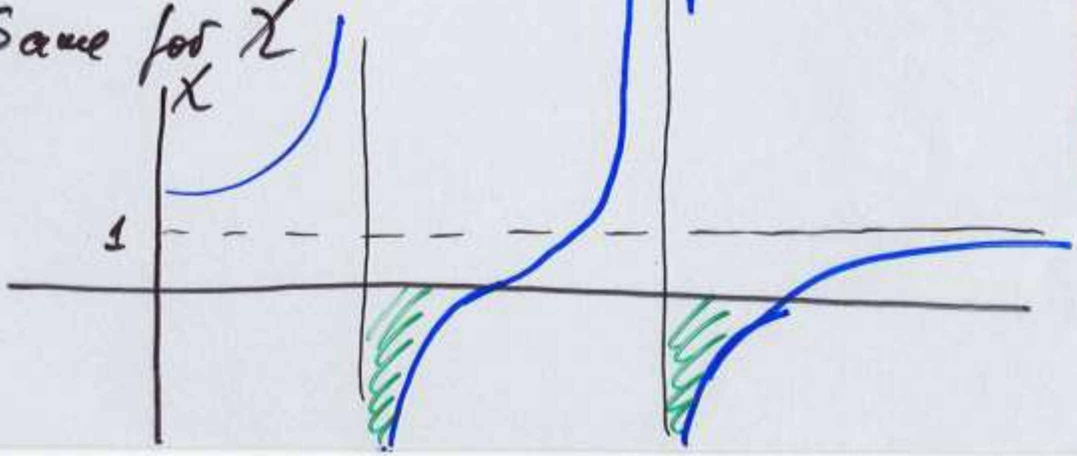


$$\omega_\pm^2 = \frac{\omega_0^2+\Omega^2}{2} \pm \sqrt{\left(\frac{\omega_0^2-\Omega^2}{2}\right)^2+\lambda^2} \approx \begin{cases} \omega_0^2 + \dfrac{\lambda^2}{\omega_0^2-\Omega^2} \\ \Omega^2 - \dfrac{\lambda^2}{\omega_0^2-\Omega^2} \end{cases}$$

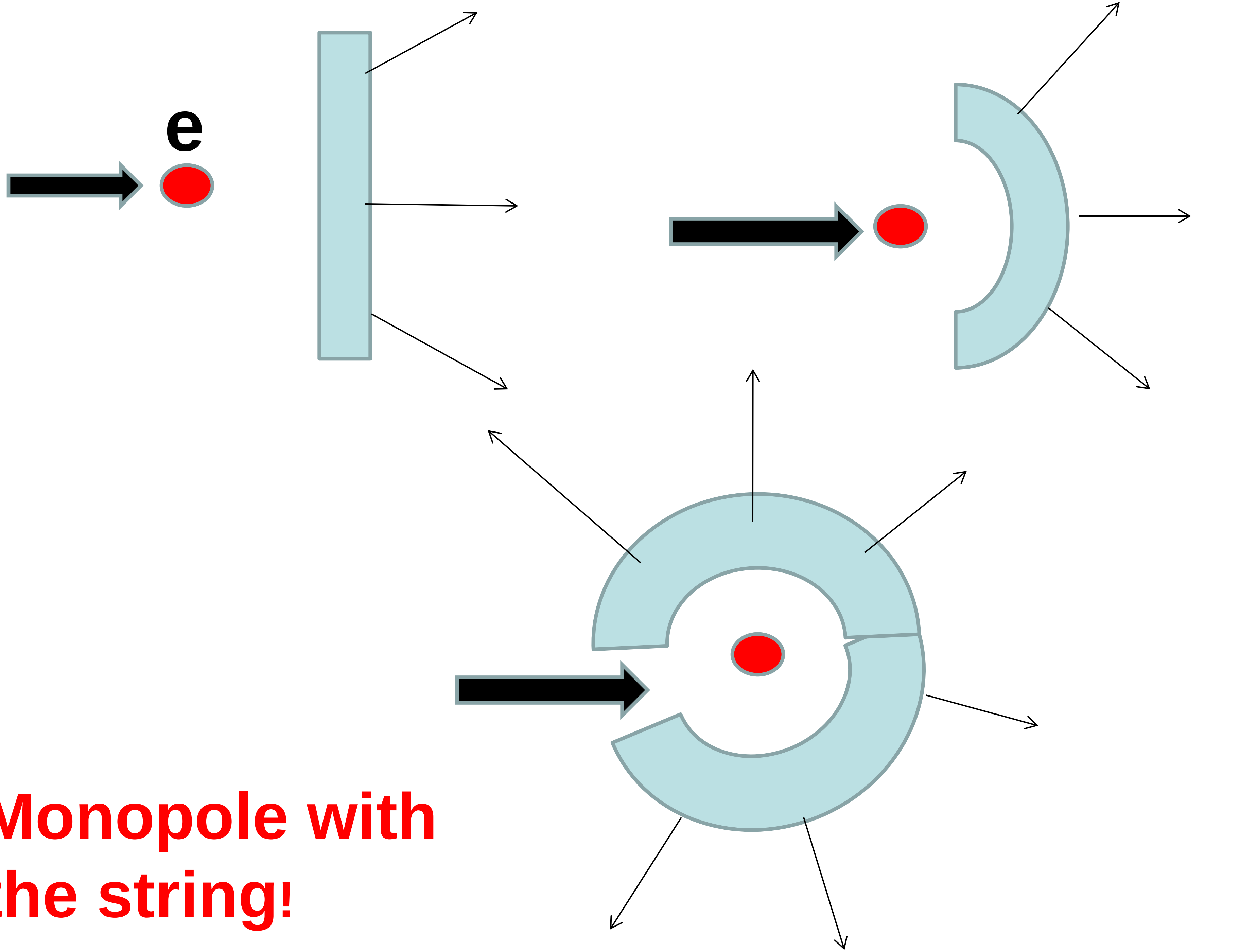
(if $\omega_0^2 > \Omega^2$)



Same for χ



$\Rightarrow$ $\boxed{\text{For } \omega \gtrsim \omega_+,\ \omega \gtrsim \omega_- \text{ - both } \varepsilon \text{ \& } \mu < 0, \text{ and } n < 0}$



Monopole with the string!

Visualization of skyrmion crystal (Y.Tokura et al.)