Coupled electric and magnetic effects in frustrated Mott insulators: currents, dipoles and monopoles

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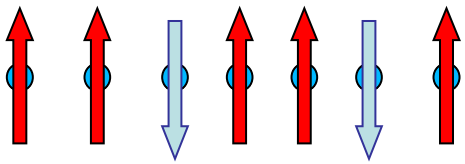
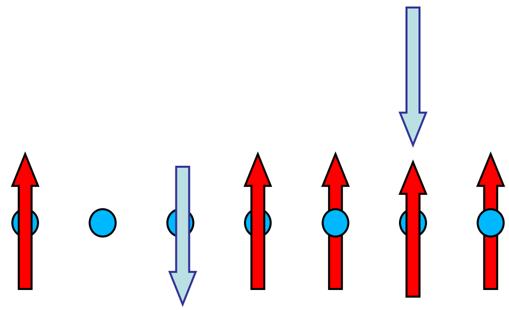
- Introduction
- Spontaneous currents and dipoles in Mott insulators
- Dipoles on monopoles in spin ice
- Conclusions

$$\mathcal{H} = -t \sum_{\langle ij \rangle, \sigma} c_{i\sigma}^{\dagger} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

The Hubbard model

$$E_{\mathrm{g}} \sim U - 2zt$$

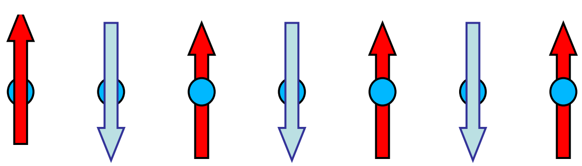




n=1, U>t: **Mott insulator**

Localized electrons/localized magnetic moments

$$H_{eff} = \frac{2t^2}{U} \sum_{\langle ij \rangle} \mathbf{S}_i \mathbf{S}_j .$$



$\Delta E = 0$ $\quad\quad$ $\Delta E = -\frac{2t^2}{U}$

Fig. 1. Two possible configurations and corresponding energy gain for non-degenerate orbitals.

Electronic Orbital Currents and Polarization in 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

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

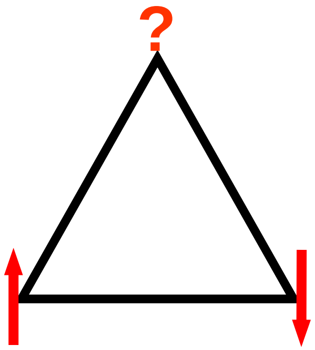
Not the full truth!

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

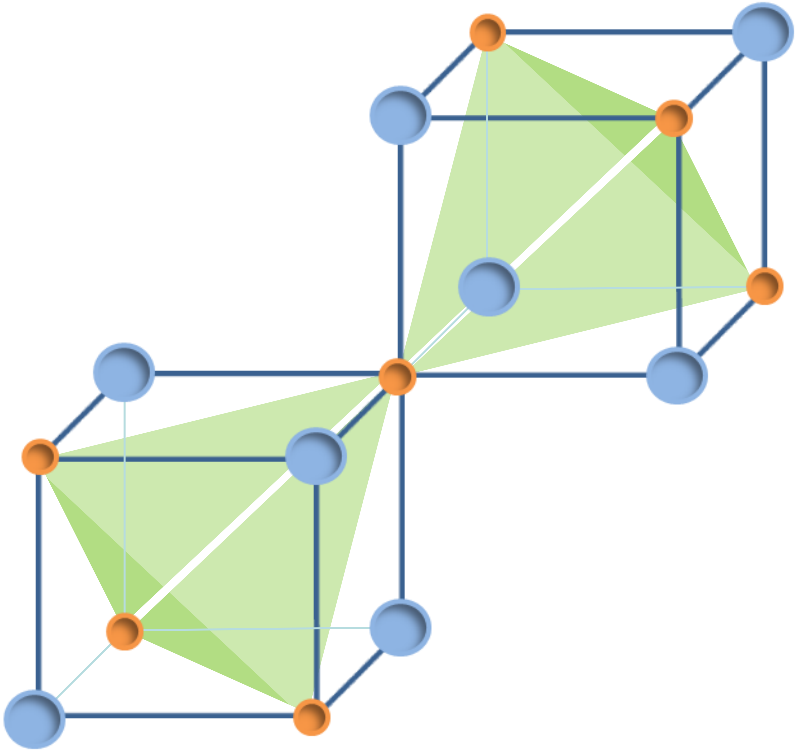


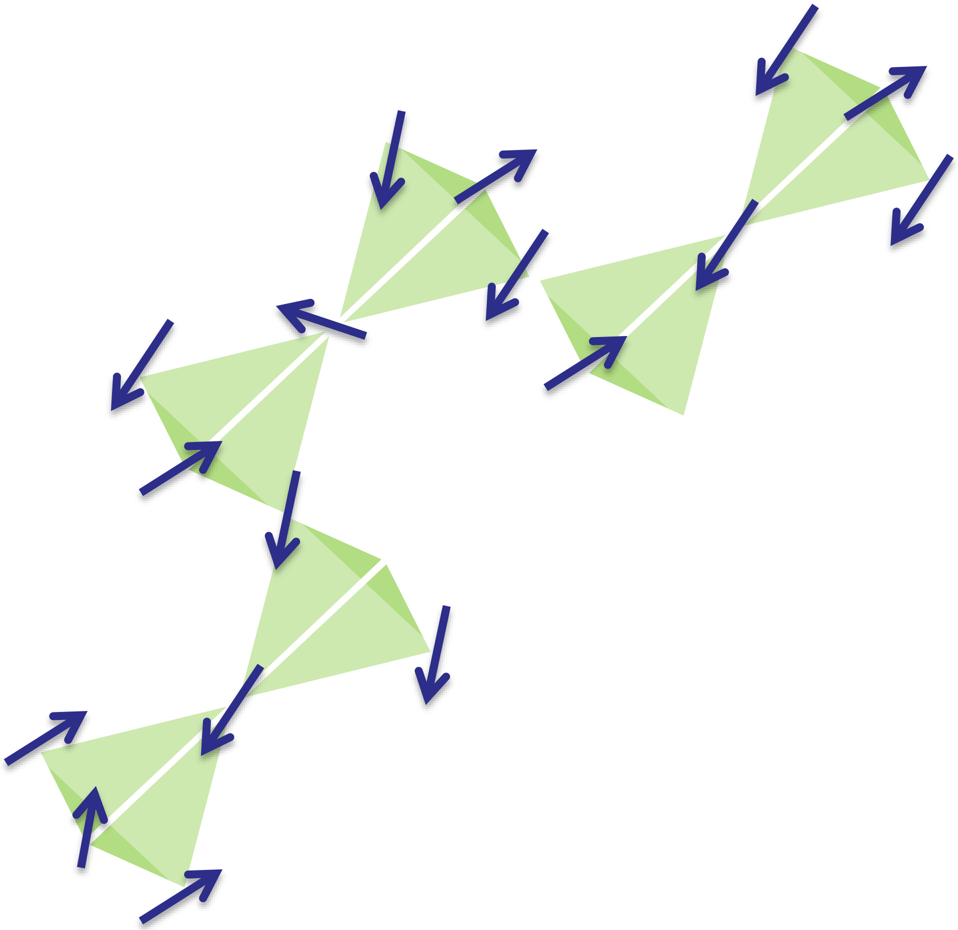
- Isolated triangles (trinuclear clusters) - e.g. in some magnetic molecules (V15, …)
- Solids with isolated triangles ($La_4Cu_3MoO_{12}$)
- Triangular lattices
- Kagome
- Pyrochlore

The Cathedral San Giusto, Trieste, 6-14 century

Spinels:

The B-site pyrochlore lattice: geometrically frustrated for AF





Antiperovskites (octahedra of transition metals);

1d-systems of this type

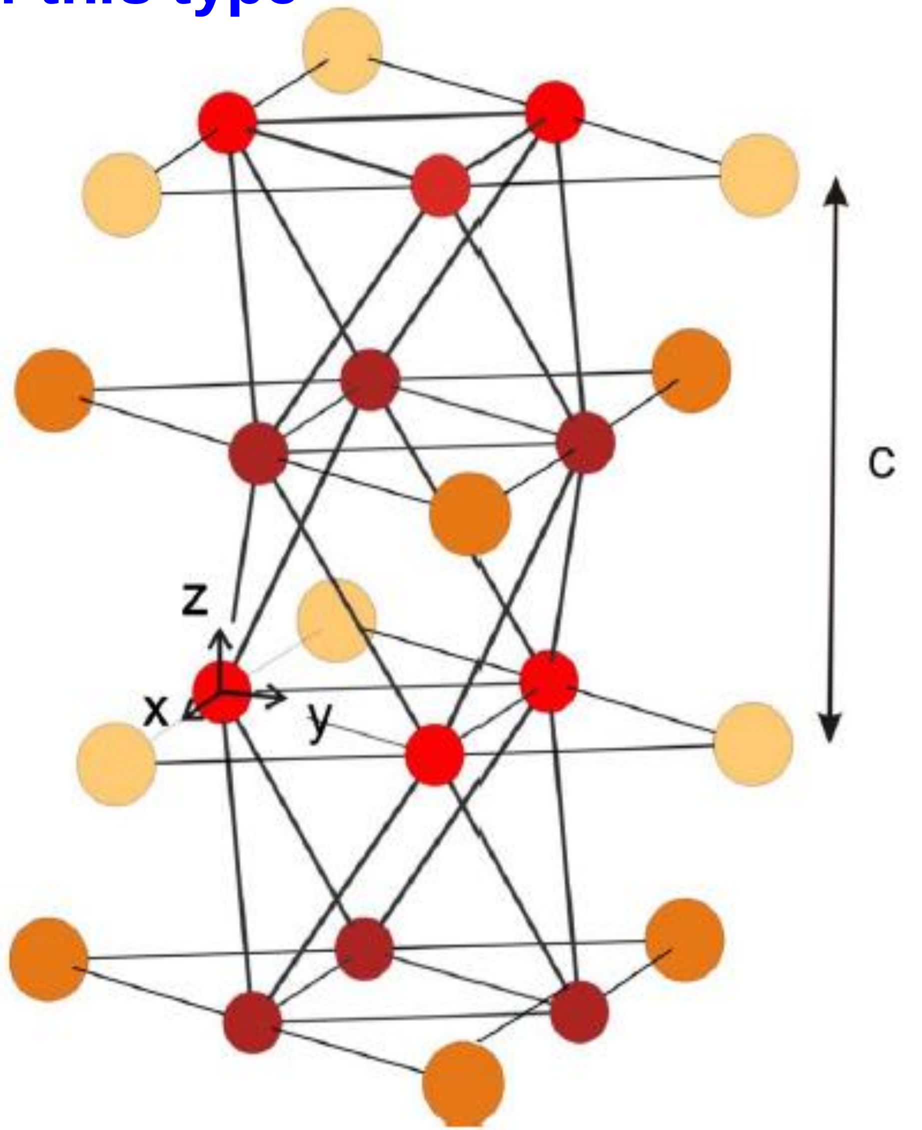


FIG. 1. (Color online) Crystal structure of $M_2Mo_6Se_6$ com-

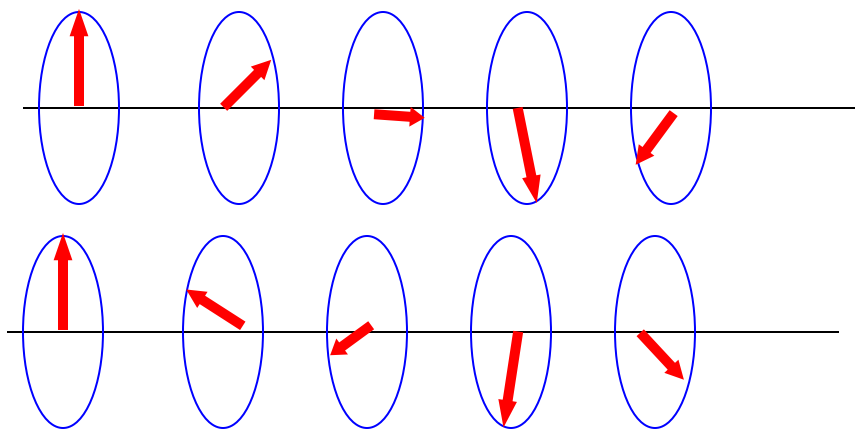
Often complicated ground states; sometimes $\langle \vec{S}_i \rangle = 0$

spin liquids

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

Vector chirality

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

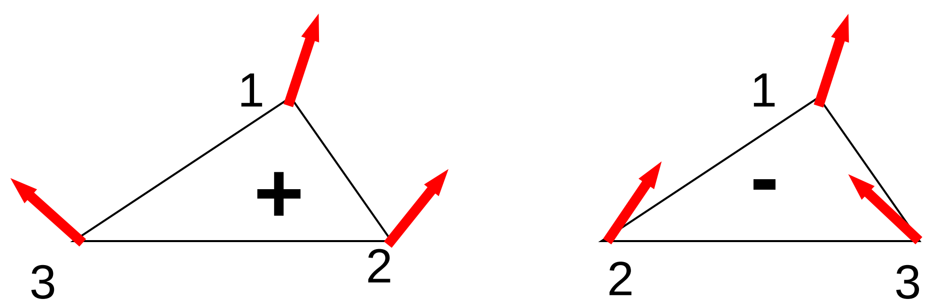


Scalar chirality

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

- solid angle

χ may be + or - :

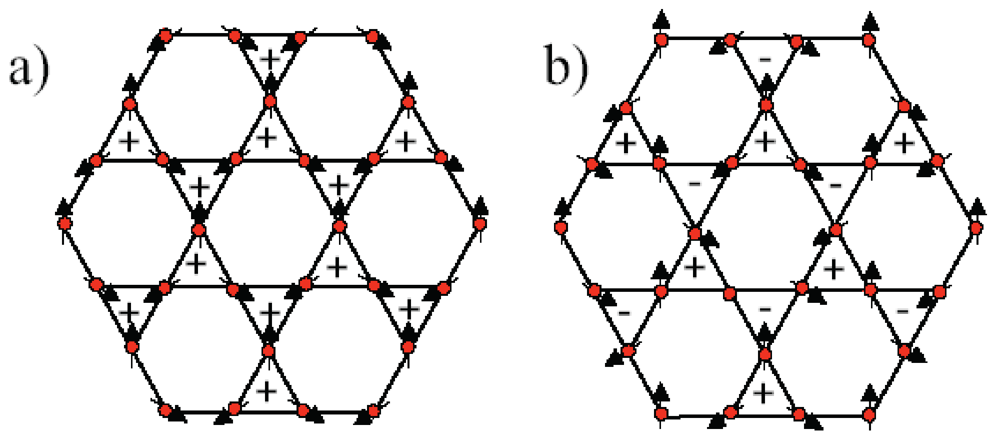


Scalar chirality χ is often invoked in different situations:

- 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$\sqrt{3}$)



But what is the scalar chirality physically?

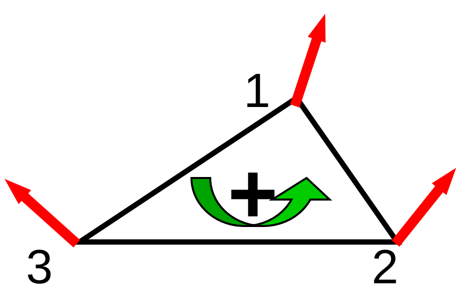
What does it couple to?

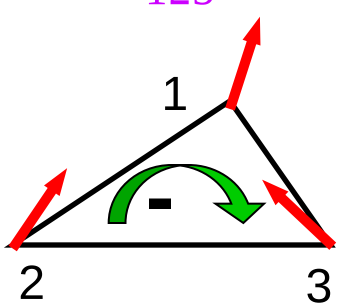
How to measure it?

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

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

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





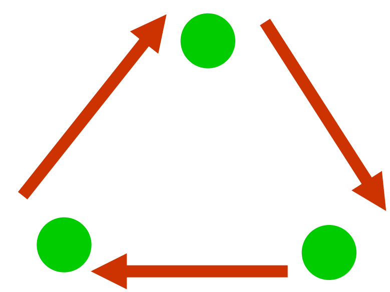
Couples to magnetic field:

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

Difference between Mott and band insulators

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

- Only in the limit $U \to \infty$ electrons are localized on sites.
- At $t/U \neq 0$ electrons can hop between sites.



Orbital current

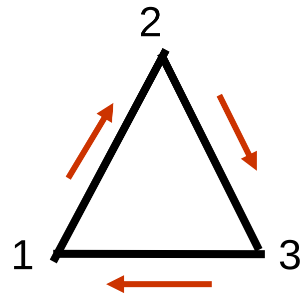
$$H_S = \frac{4t^2}{U}(\vec{S}_1 \cdot \vec{S}_2 - 1/4).$$

Spin current operator and scalar spin chirality

- Current operator for Hubbard Hamiltonian on bond ij:

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

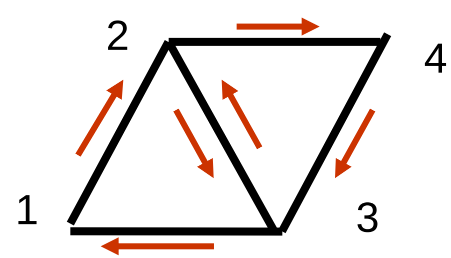
- Projected current operator: odd # of spin operators, scalar in spin space. For smallest loop, triangle,



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

- Current via bond 23

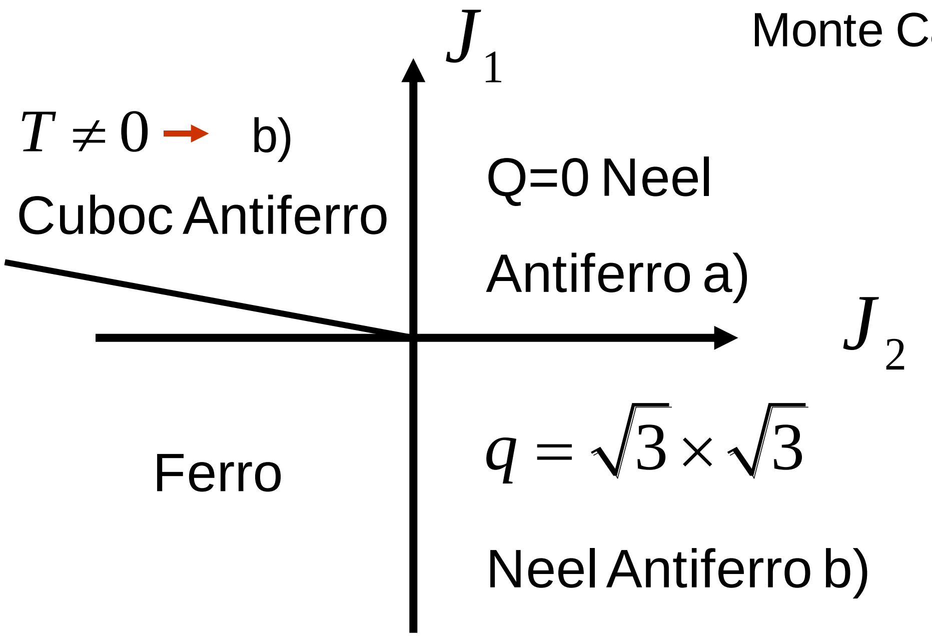
$$I_{S,23} = I_{S,23}(1) + I_{S,23}(4).$$



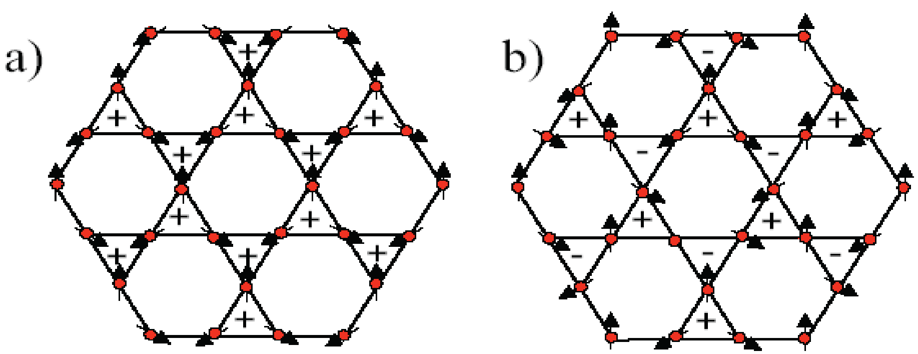
- On bipartite nn lattice I_S is absent.

Ordering in $J_1 - J_2$ model on kagome lattice at T=0

Monte Carlo results by Domenge et al., 2007.

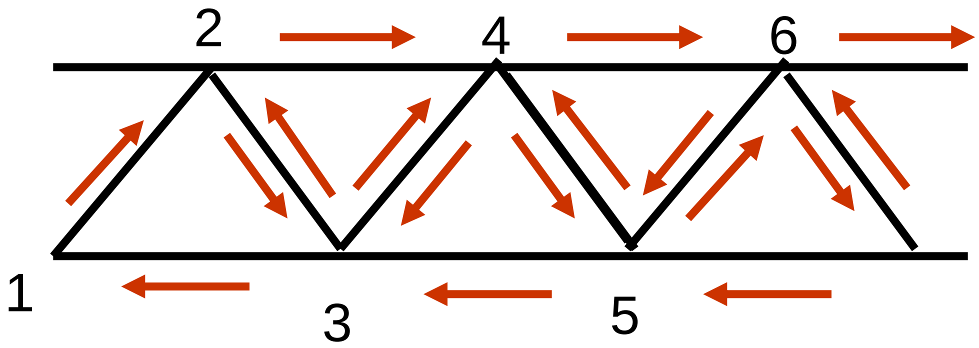


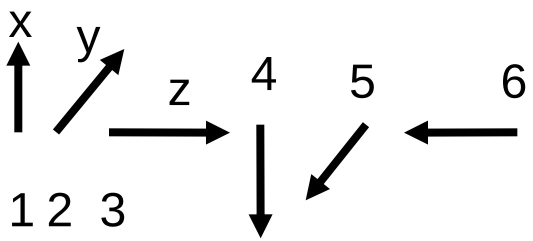
Phase diagram for classical order parameter



For S=1/2 for low T cub'oc is chiral with orbital currents and without spin ordering.

Boundary and persistent current





$$\chi = const$$

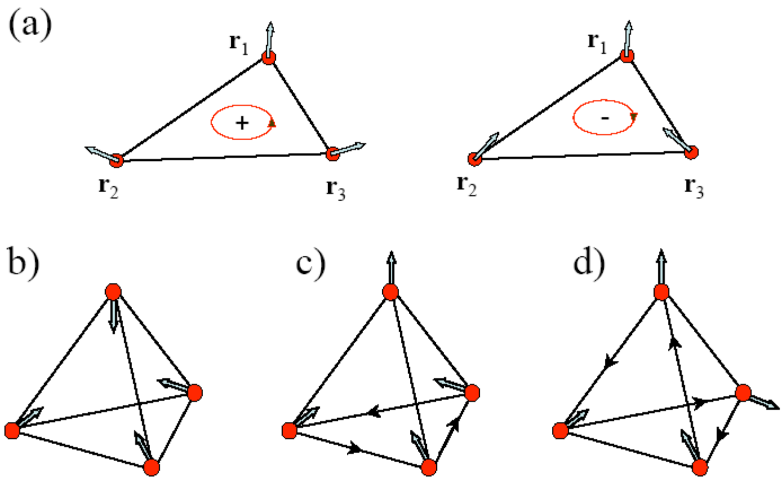
Boundary current in
gaped 2d insulator

Orbital currents in the spin ordered ground state $\langle \vec{S}_i \rangle \neq 0$

- Necessary condition for orbital currents is nonzero average chirality

$$\chi_{12,3} = [\vec{S}_1 \times \vec{S}_2] \cdot \vec{S}_3, \qquad \langle \chi_{ij,k} \rangle \neq 0.$$

- It may be inherent to spin ordering or induced by magnetic field

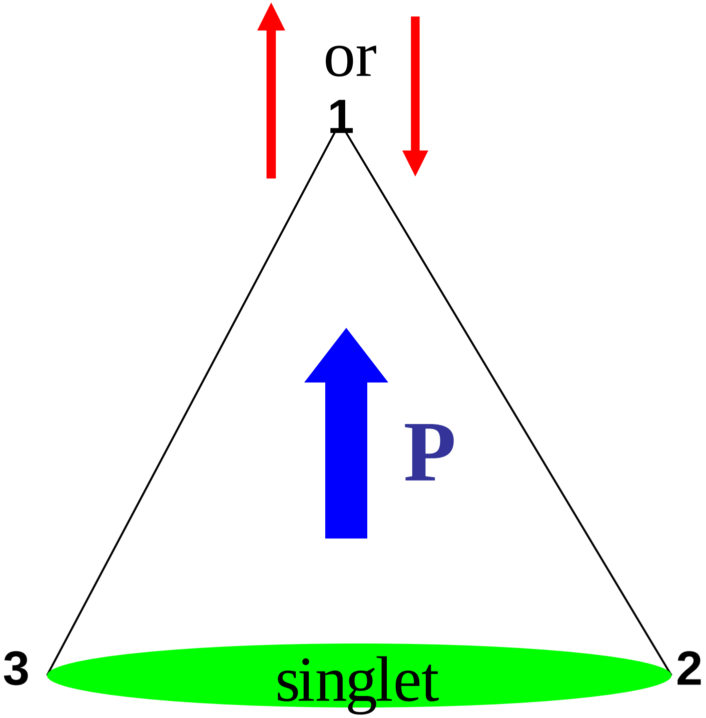


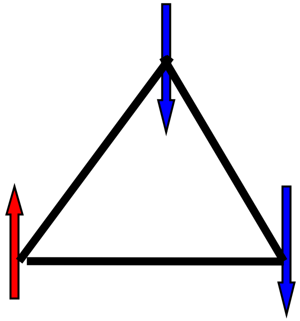
Triangles with $\pm$ chirality

Tetrahedra with [111] anisotropy

Electronic polarization on triangle

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

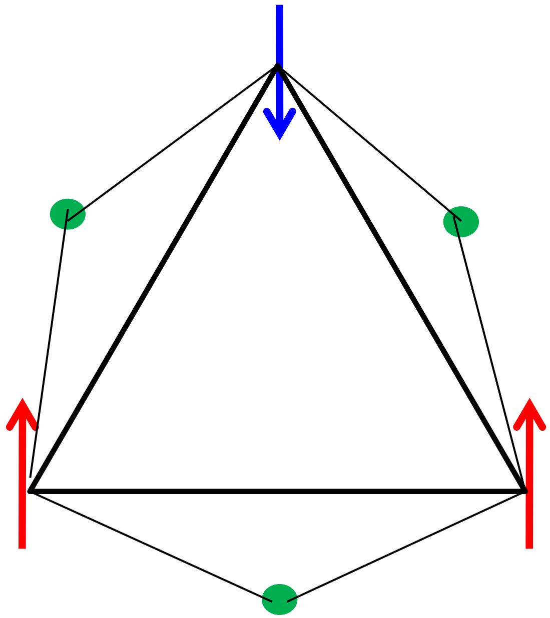




Purely electronic mechanism of multiferroic behavior!

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)



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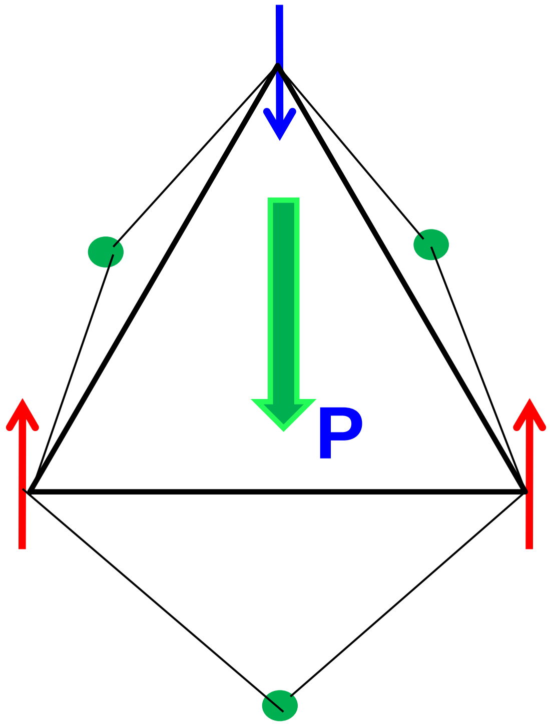
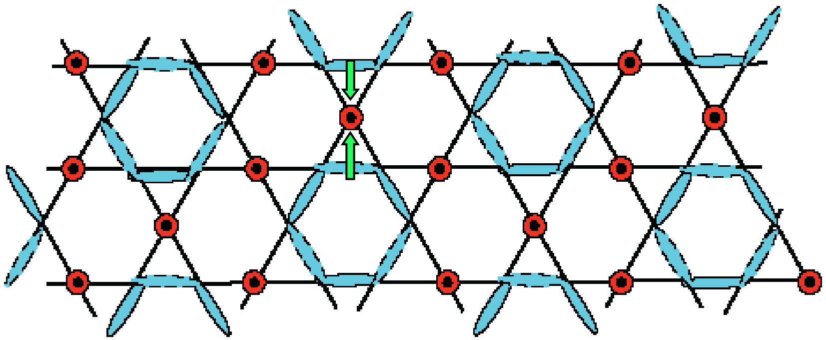


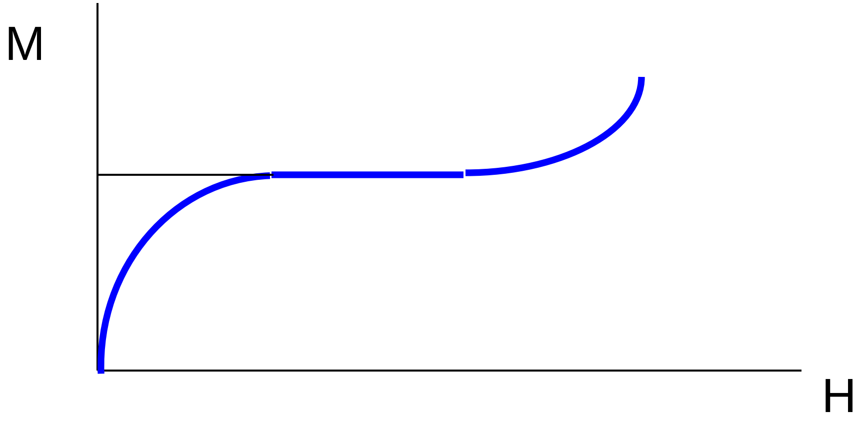
Charges on kagome lattice



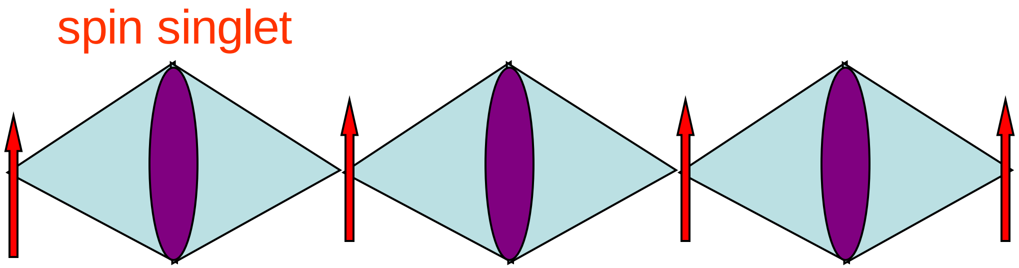
1/3 magnetization plateau:

Charge ordering for spins 1/3 in magnetic field: spin-driven CDW

•Typical situation at the magnetization plateaux!

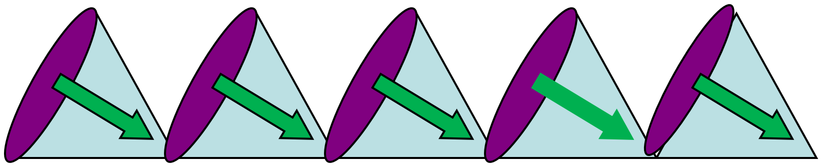


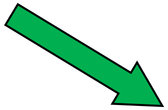
- Diamond chain (azurite $Cu_3(CO_3)_2(OH)_2$)

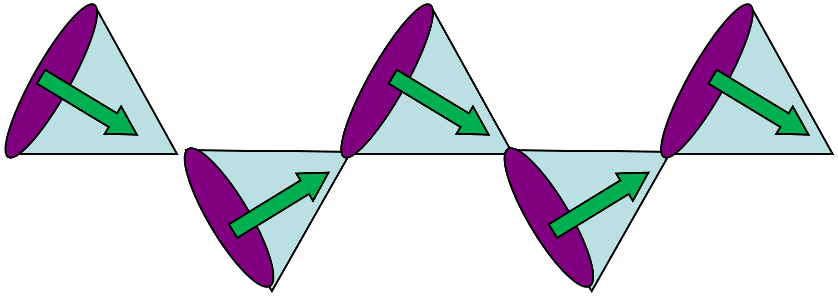


-will develop S-CDW

- Saw-tooth (or delta-) chain

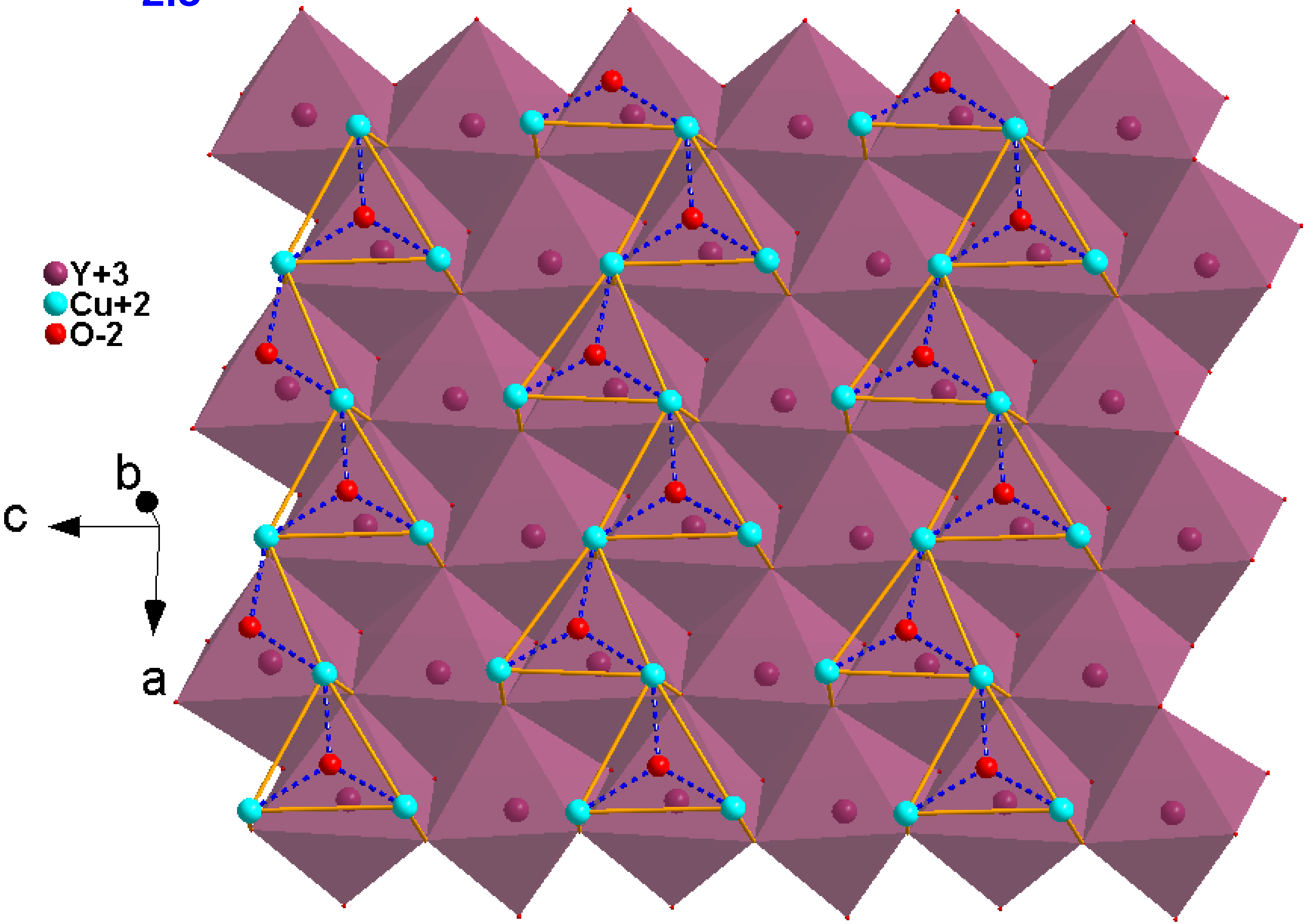


Net polarization 



Net polarization 

$CuYO_{2.5}$



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

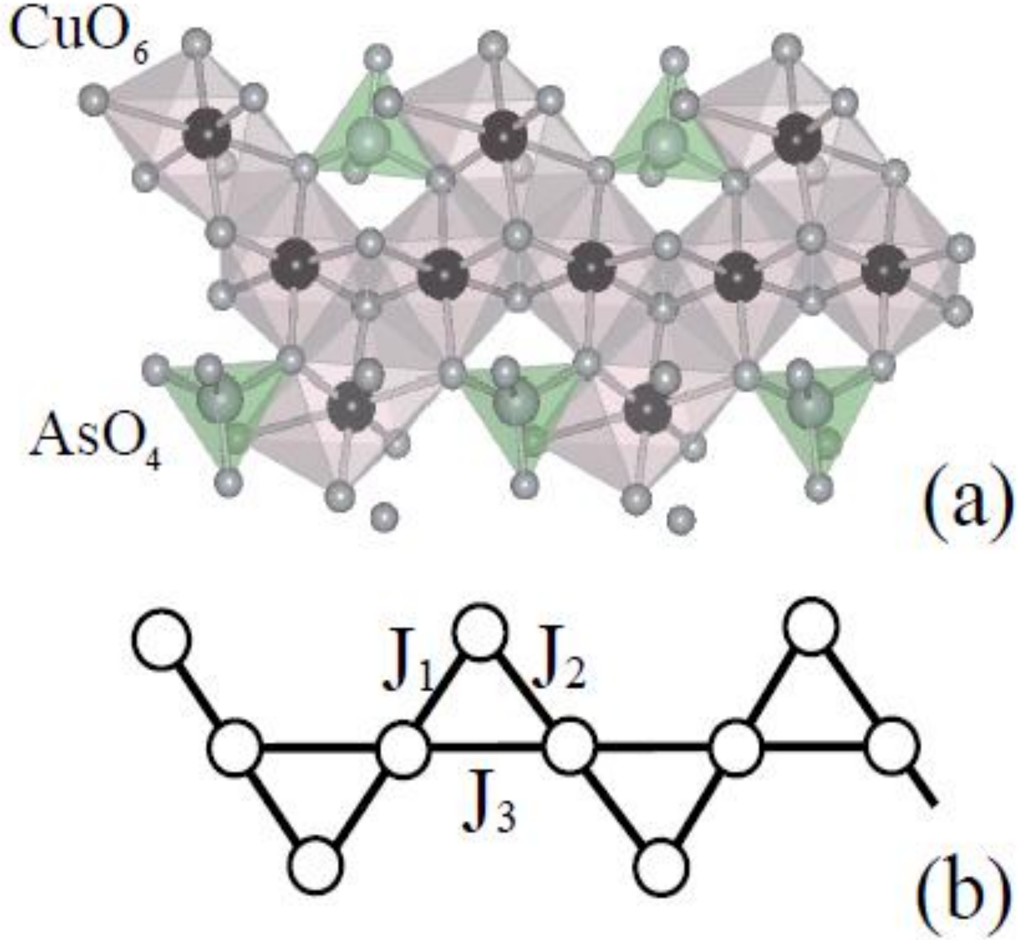


Figure 1. (a) Structure of euchroite, (b) schematic view of the delta chain.

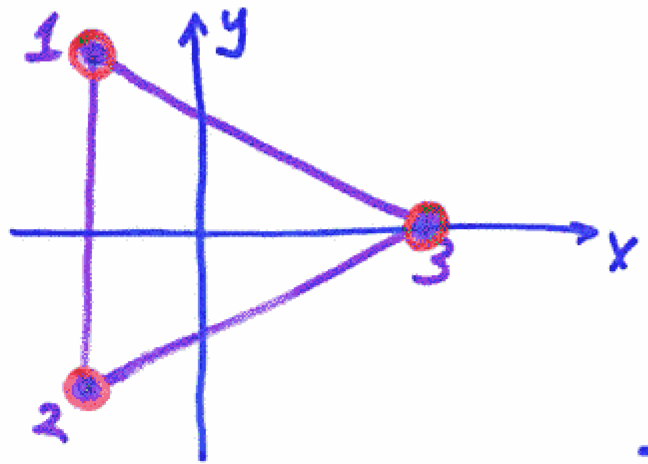
Isolated triangle: accounting for DM interaction

- DM coupling: $H_{DM} = \sum_{ij} D_{ij} \vec{S}_i \times \vec{S}_j.$

- For V15 $H_{DM} \approx D_z L_z S_z.$
- Splits lowest quartet into 2 doublets $|+\uparrow\rangle, |-\downarrow\rangle$ and $|+\downarrow\rangle, |-\uparrow\rangle$ separated by energy $\Delta = D_z.$
- Ac electric field induces transitions between $\chi = \pm 1.$
- Ac magnetic field induces transitions between

  $S_z = \pm 1/2.$

Polarization of a triangle:



$$P_x = 4\sqrt{3}\,ea\left(\frac{t}{U}\right)^3\left[\vec{S}_1\cdot(\vec{S}_2+\vec{S}_3)-2\vec{S}_2\cdot\vec{S}_3\right]$$

$$P_y = 12\,ea\left(\frac{t}{U}\right)^3\vec{S}_1\cdot(\vec{S}_2-\vec{S}_3)$$

Dipole moments, or polarization, and current on a triangle, can be combined in one „isospin"-$\frac{1}{2}$:

$$\left\{\begin{array}{l} P_x \longrightarrow -c\,T_x \\ P_y \longrightarrow c\,T_y \end{array}\right\} \text{ real (T-even)}$$

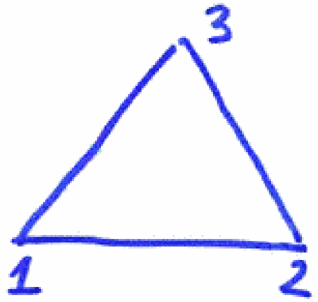
$$\frac{ta}{U} I \longrightarrow c\,T_z \;\} \text{ imaginary (T-odd)}$$

– somewhat similar to pseudospin $\vec{\tau}$ for e_g-orbitals

$$\left(\underbrace{\tau_z \rightarrow |z^2\rangle,\ \tau_x \rightarrow |x^2-y^2\rangle}_{\text{real (T-even)}},\ \underbrace{\tau_y \rightarrow \frac{1}{\sqrt{2}}\left(|z^2\rangle \pm i|x^2-y^2\rangle\right)}_{\text{Im. (T-odd)}}\right)$$

Consequenses for dynamic properties:

Equilateral triangle of $S = \frac{1}{2}$ with antiferro. exchang



$2^3 = 8$ states

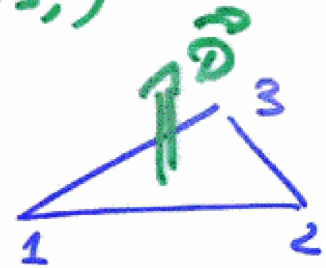
$S_{tot} = 3/2$

$S_{tot} = \frac{1}{2}$, $\chi = \pm$

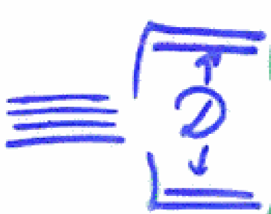
Dzyaloshinskii-Moriya interaction:

$$-\vec{D}_{12}\cdot(\vec{S}_1\times\vec{S}_2) - \vec{D}_{23}(\vec{S}_2\times\vec{S}_3) - \vec{D}_{31}(\vec{S}_3\times\vec{S}_1)$$

E.g. $\vec{D}_{12} = \vec{D}_{23} = \vec{D}_{3} \perp$ (123) plane



Splits ground state quartet



$(S^z = \frac{1}{2}, \chi = -)$, $(S^z = -\frac{1}{2}, \chi = +)$

$(S^z = \frac{1}{2}, \chi = +)$, $(S^z = -\frac{1}{2}, \chi = -)$

(DM interaction plays a role of spin-orbit interaction for ground state quartet)

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

$$\left|1/2,\chi\right\rangle \rightarrow \left|-1/2,\chi\right\rangle,\ or\ \left|-1/2,\chi\right\rangle \rightarrow \left|1/2,\chi\right\rangle$$

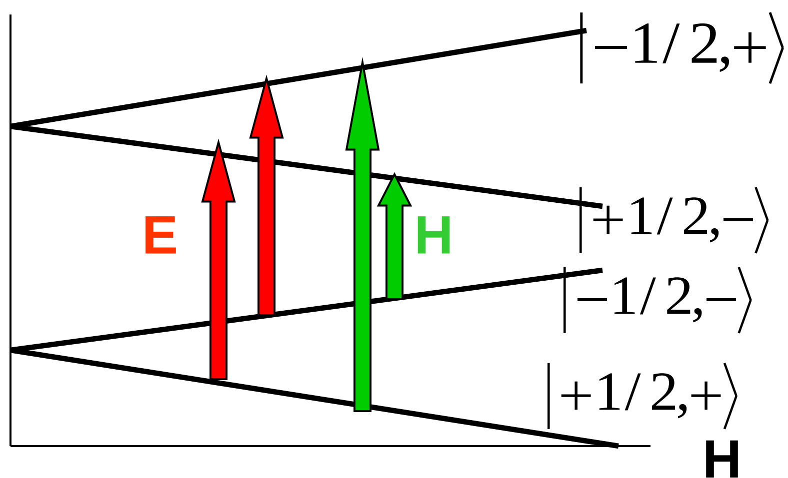
Here: electric field (**-Ed**) has nondiagonal matrix elements in χ:

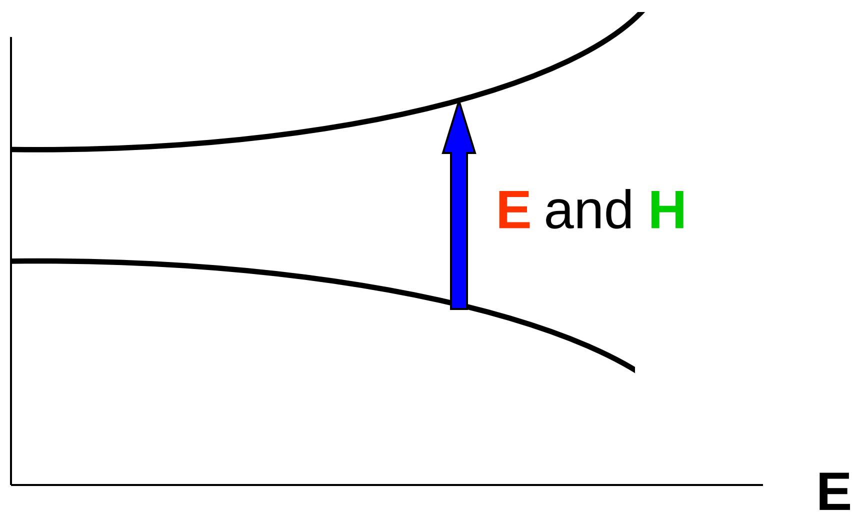
$$\left\langle \chi=+\left|\mathbf{d}\right|\chi=-\right\rangle \neq 0$$

→ electric field will cause

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

-- ESR caused by electric field E !





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?

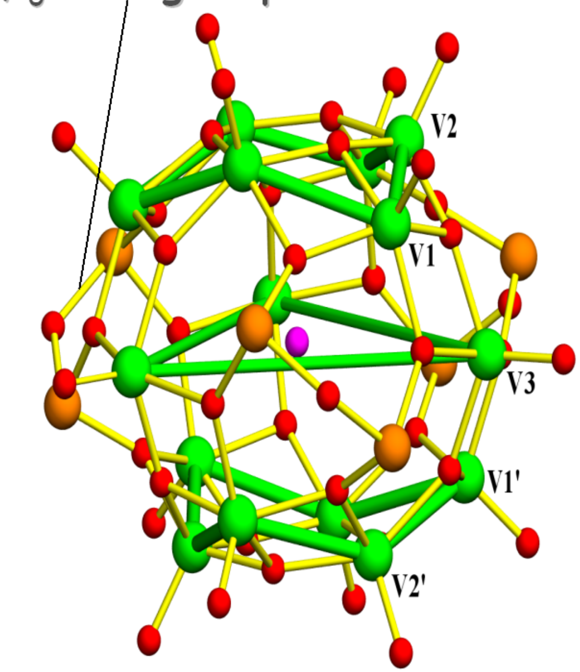
We can control it by magnetic field (chirality = current = orbital moment) and by electric field

Georgeot, Mila, PRL (2010)

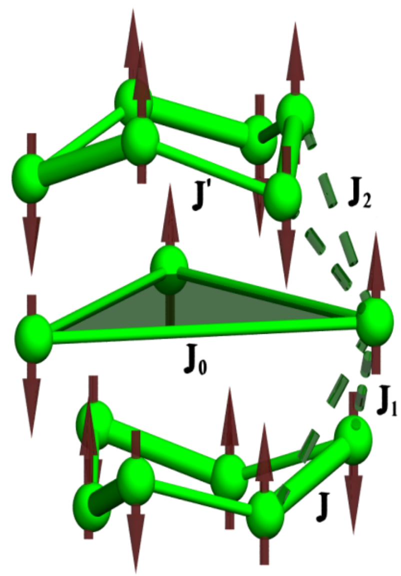
Qusispherical layered molecular structure of $K_6[V^{IV}_{15}As^{III}_6O_{42}(H_2O)]\cdot 8H_2O$ (V_{15} cluster)

First observation of coherent states in which the fifteen cluster spins and the photons are entangled:

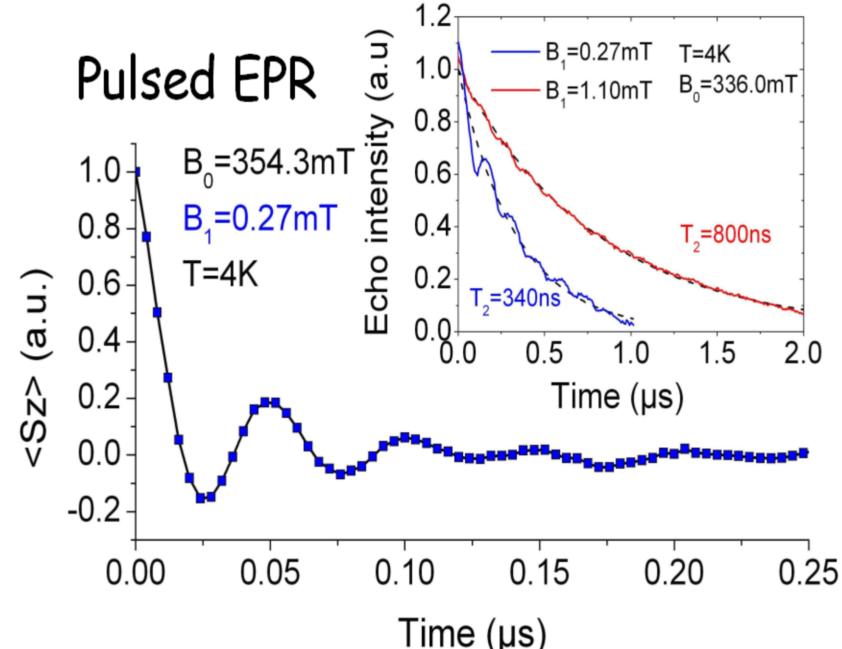
μ_3-oxo groups



Metal network of $\{V_{15}As_6\}$



Spin-echo

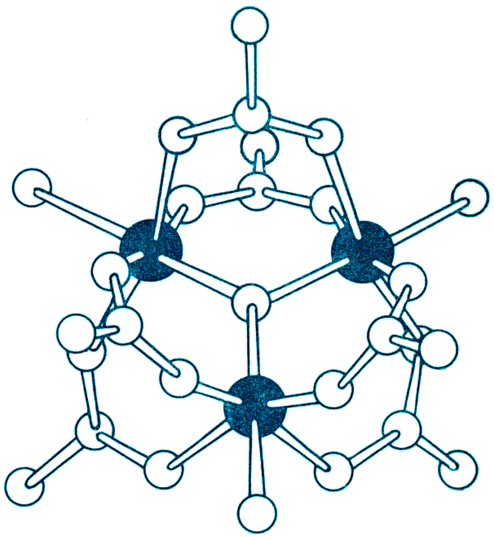


"Quantum oscillations in a molecular magnet"

S. Bertaina, S. Gambarelli, T.Mitra, B. Tsukerblat, A. Müller, B. Barbara, Nature, 2008.

From a triangle to extended frustrated systems

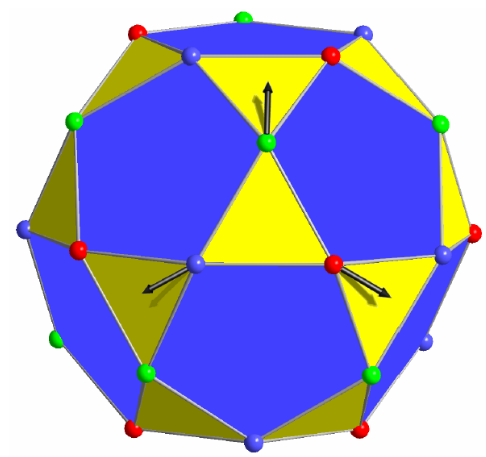
Frustrated trimeric carboxilates



Triads of the metal ions

$$(Cr_3),(Fe_3),$$
$$(FeCr_2),(Fe_2Cr)$$

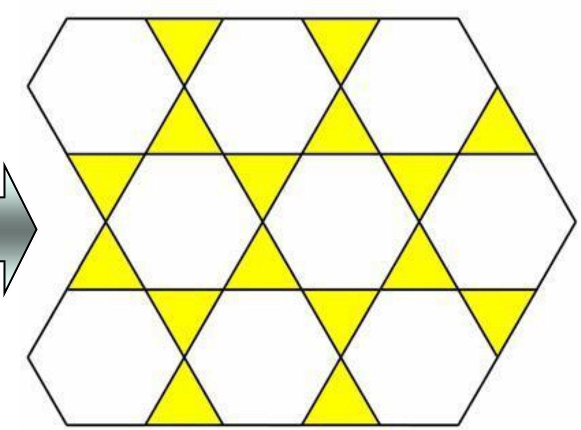
Frustrated nanoscopic systems



The M centers (colored spheres) of the $\{(Mo)Mo_5\}M_{30}$ type Keplerates ($M = V^{IV}, Fe^{III}$)

Achim Müller & coworkers

Crystal lattice

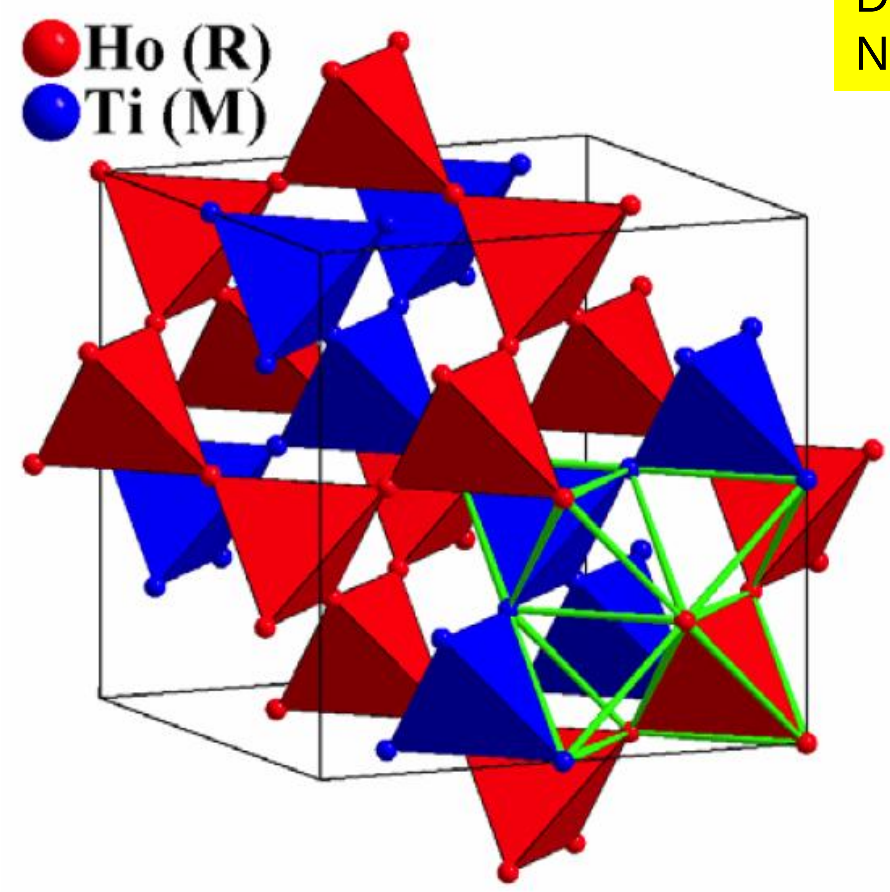


Kagomé lattice in which equilateral triangles are arranged around regular hexagons in a two dimensional plane.

● Monopoles and dipoles in spin ice

Pyrochlore: Two interpenetrating metal sublattices

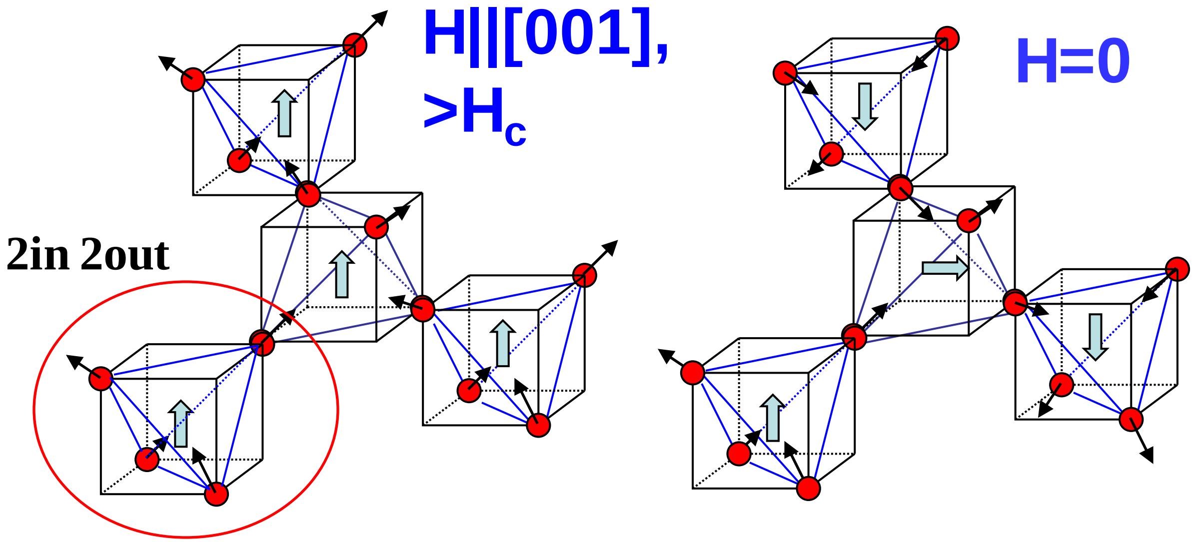
D.Khomskii
Nature Comm. 3, 904 (2012)



$Ho_2Ti_2O_7$

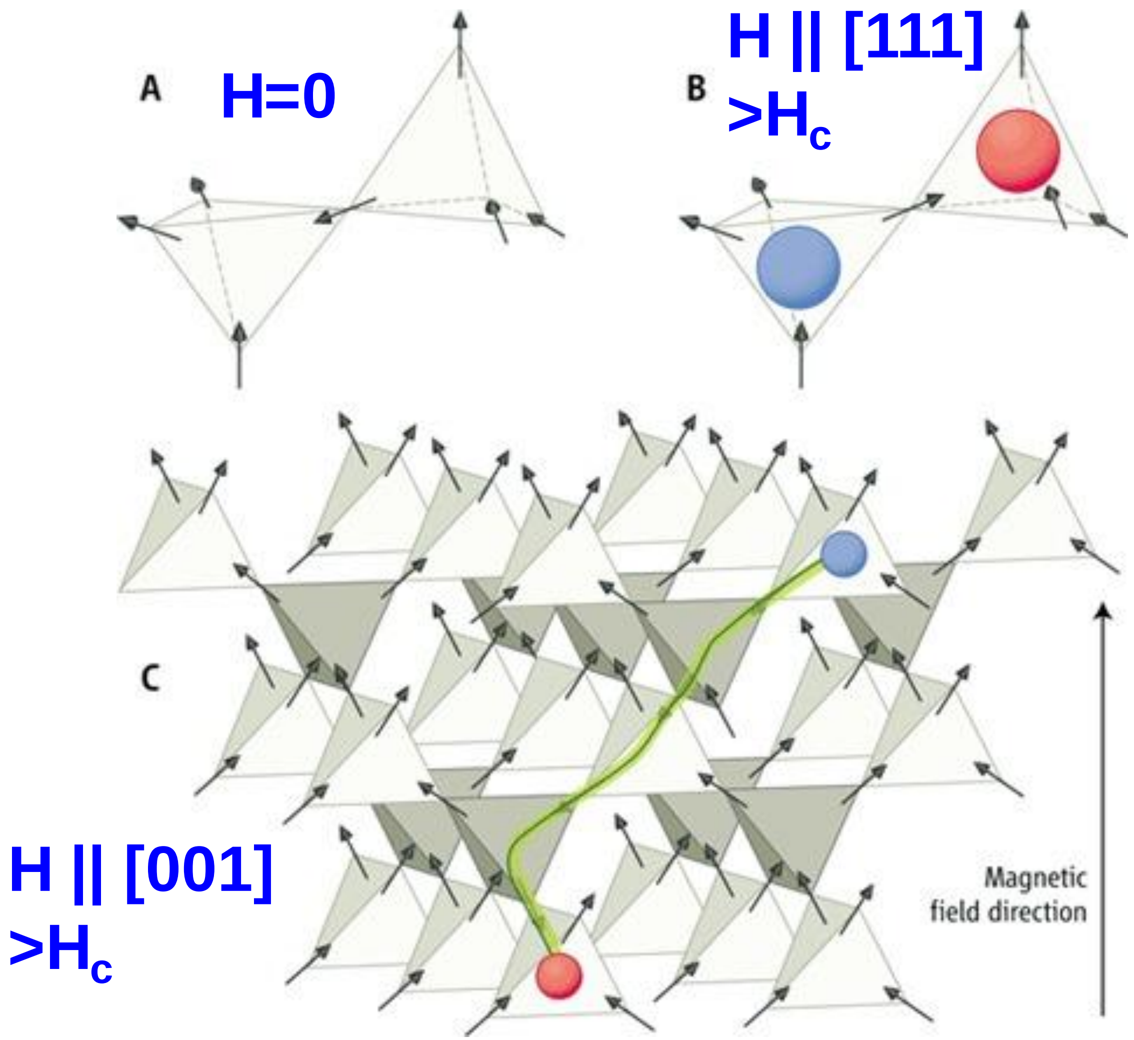
pyrochlore $R_2Ti_2O_7$・・ geometrical spin frustration

***R*=Ho**
Ferromagnetic interaction, Ising spin (spin ice)



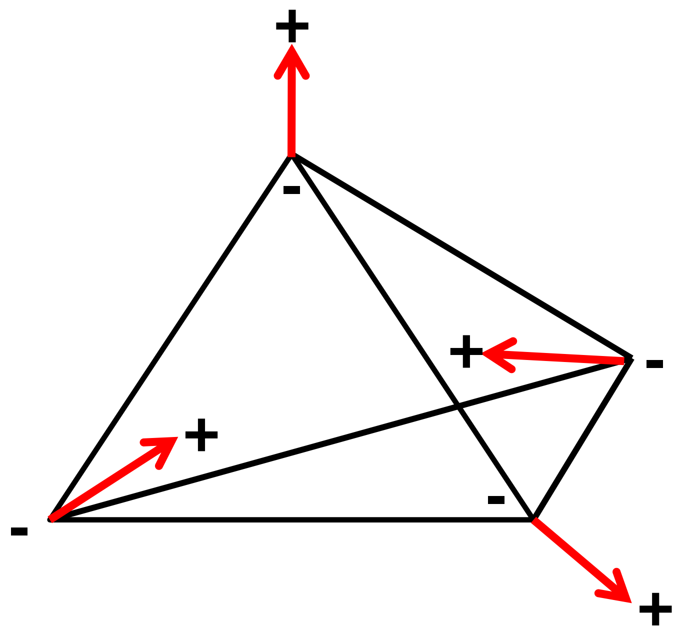
***R*=Gd**
Antiferromagnetic interaction, Heisenberg spin

Excitations creating magnetic monopole (Ryzhkin; Castelnovo, Moessner and Sondhi)

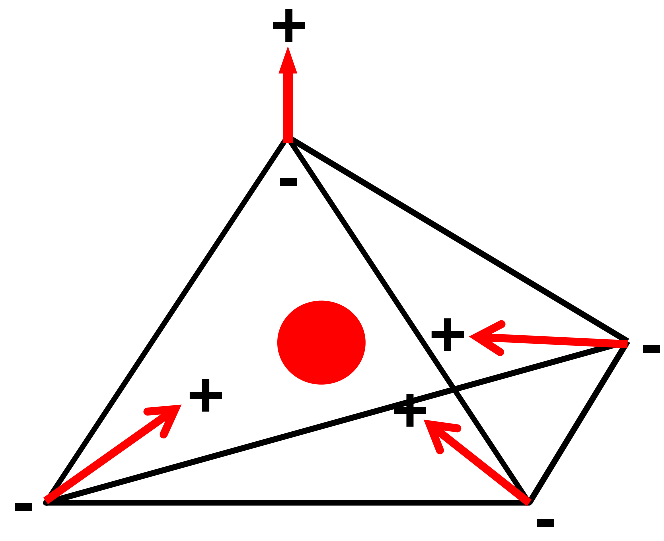


M J P Gingras Science 2009;326:375-376

Published by AAAS



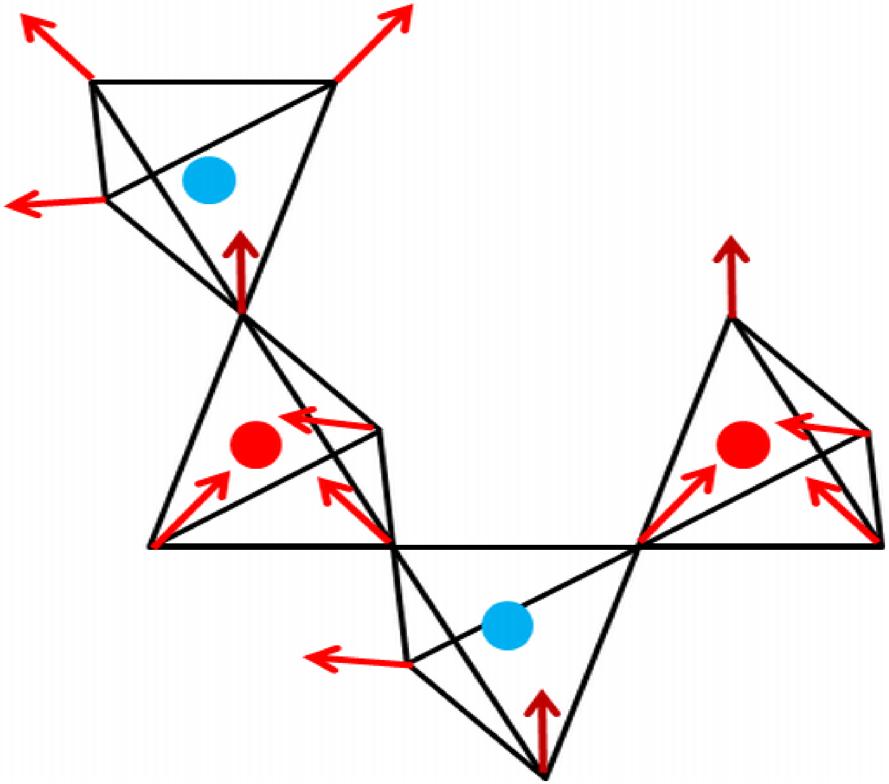
2-in/2-out: net magnetic charge inside tetrahedron zero

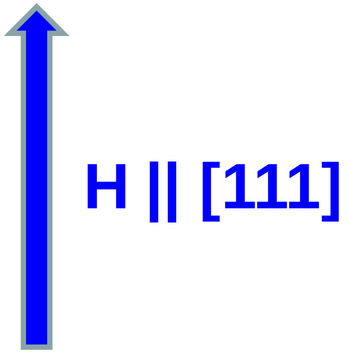


3-in/1-out: net magnetic charge inside tetrahedron ≠ 0 – monopole or antimonopole

H || [111], >H_c

Monopoles/antimonopoles at every tetraheder, staggered





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

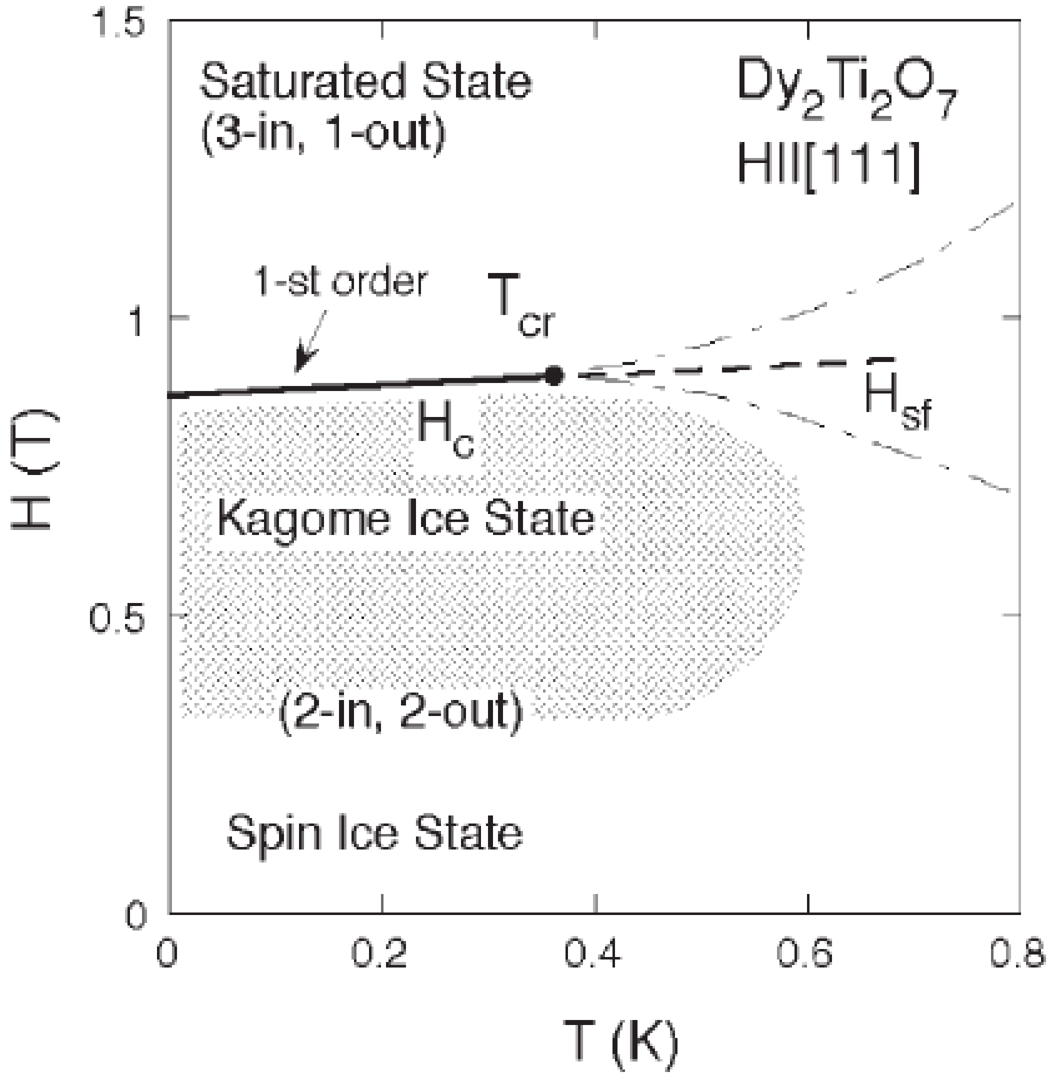
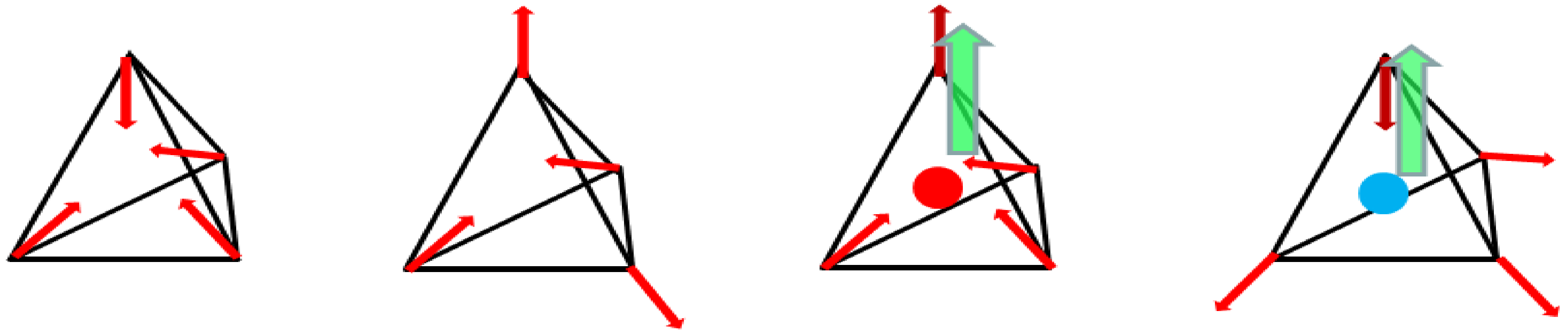


Fig. 1. Phase diagram of $Dy_2Ti_2O_7$ in a [111] magnetic field, determined by magnetization and specific heat measurements. The dashed line

★ Dipoles on tetrahedra:



4-in or 4-out: **d=0**

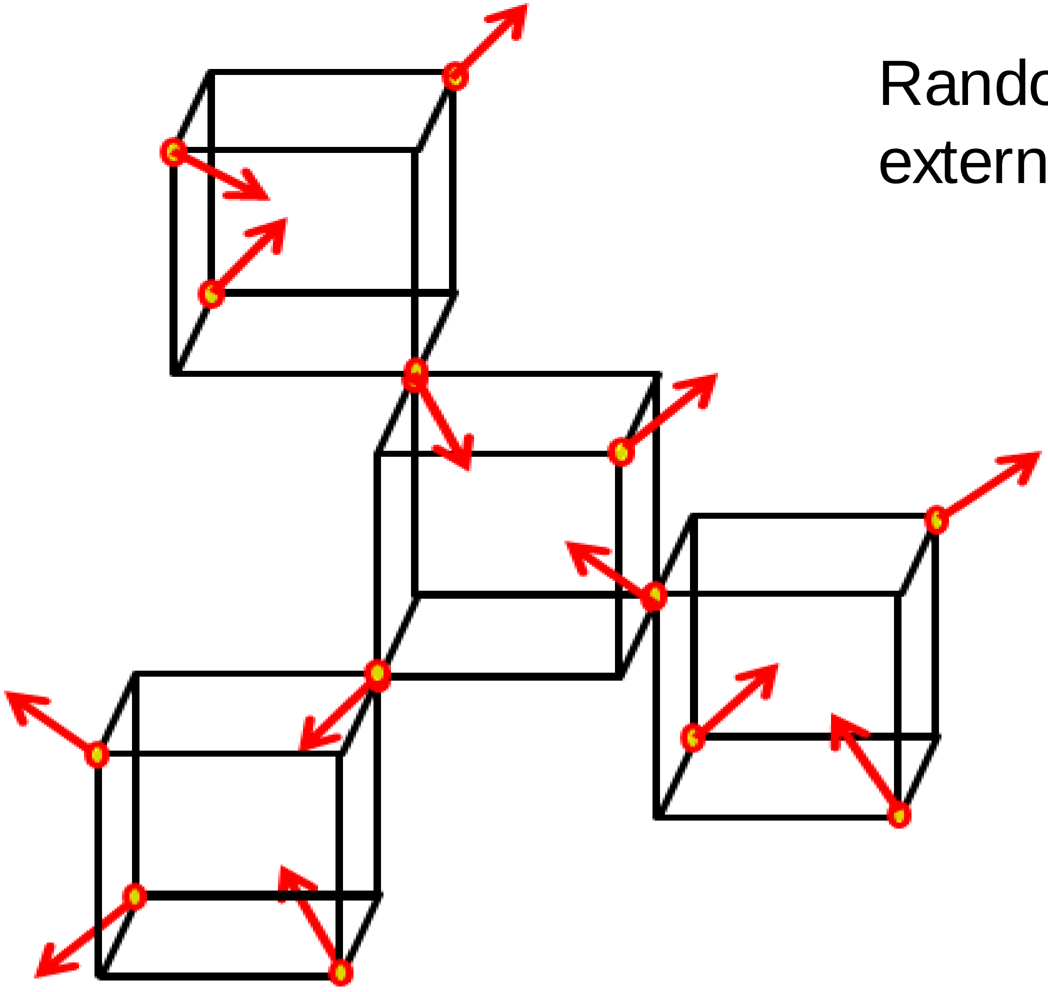
2-in/2-out (spin ice): **d=0**

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** $\neq 0$.

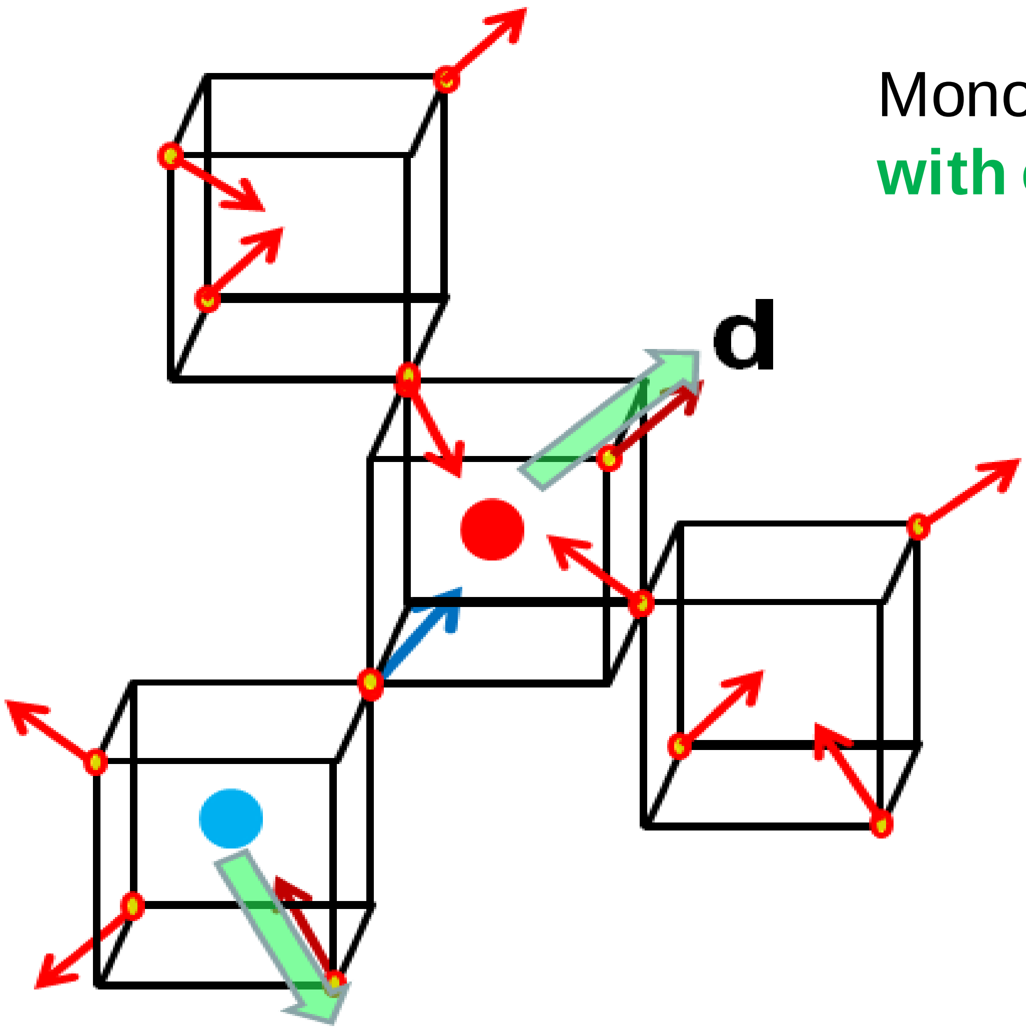
Charge redistribution and dipoles are *even* functions of S; inversion of all spins does not change direction of a dipole: ⇨ Direction of dipoles on monopoles and antimonopoles is *the same*: e.g. ***from the center of tetrahedron to a "special" spin***

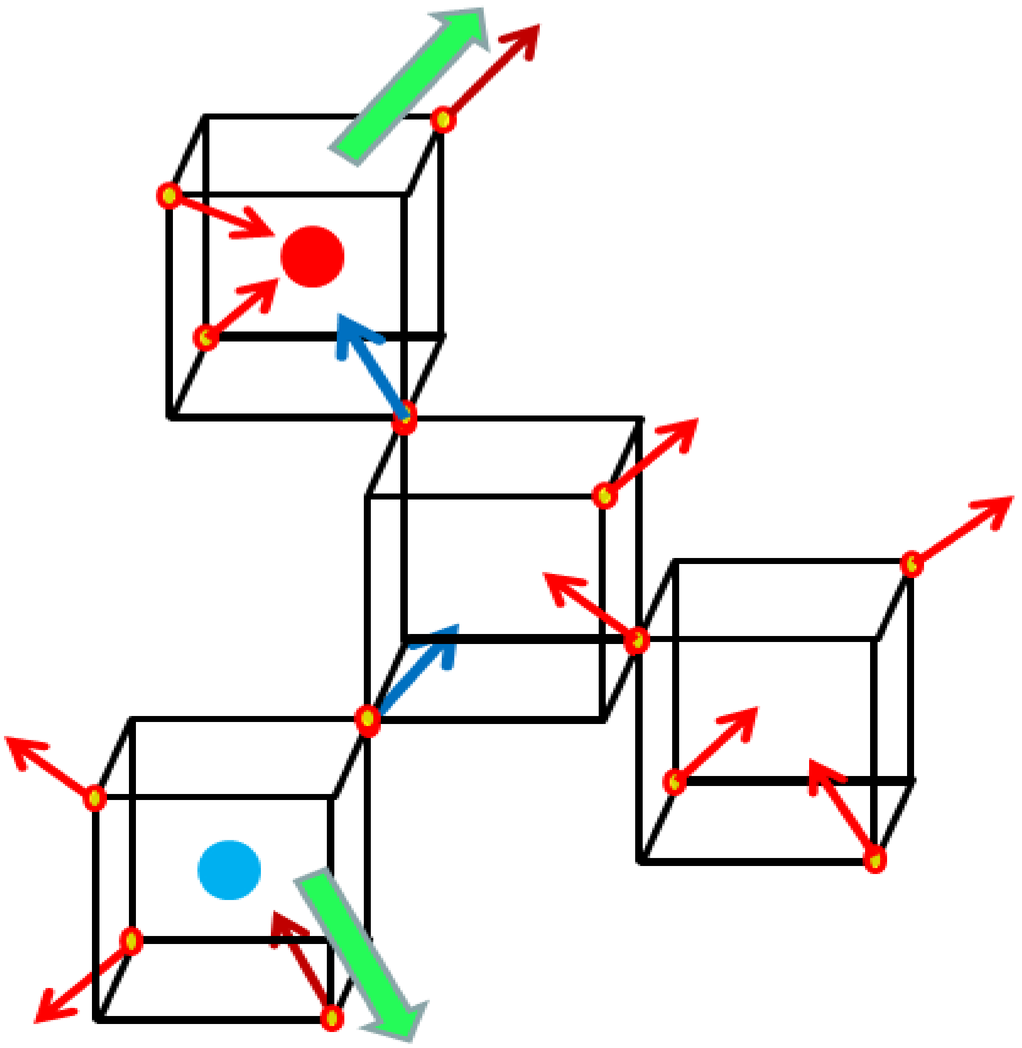


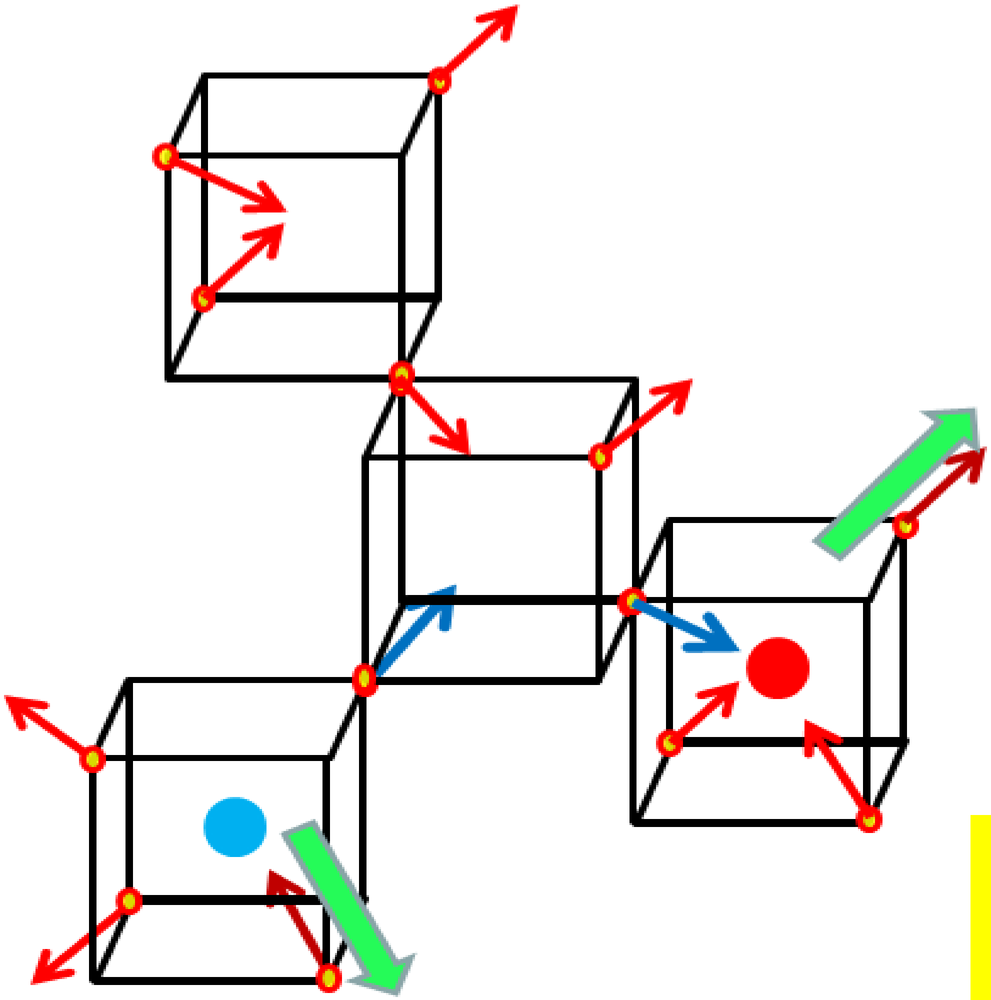
Random ice rule spins (no external magnetic field)

Monopoles/antimonopoles

with electric dipoles

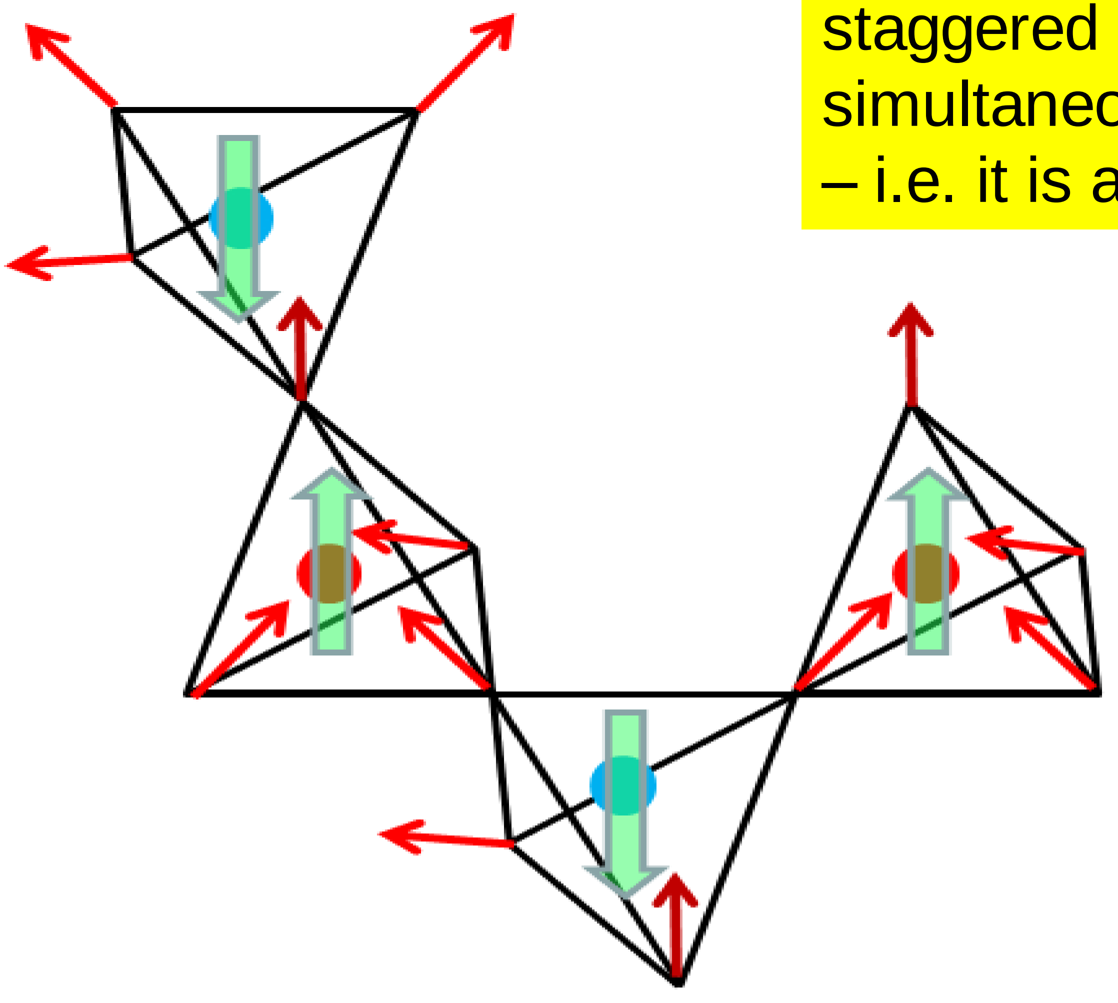






In general directions of electric dipoles are “random” – in any of [111] directions

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



Dipoles on monopoles, possible consequences:

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

- **External electric field:**
Decreases excitation energy of certain monopoles
$\omega = \omega_0 - \mathbf{dE}$

- **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 $\mathbf{E}_\perp$?

● “Electric” activity of monopoles; contribution to dielectric constant $\varepsilon(\omega)$

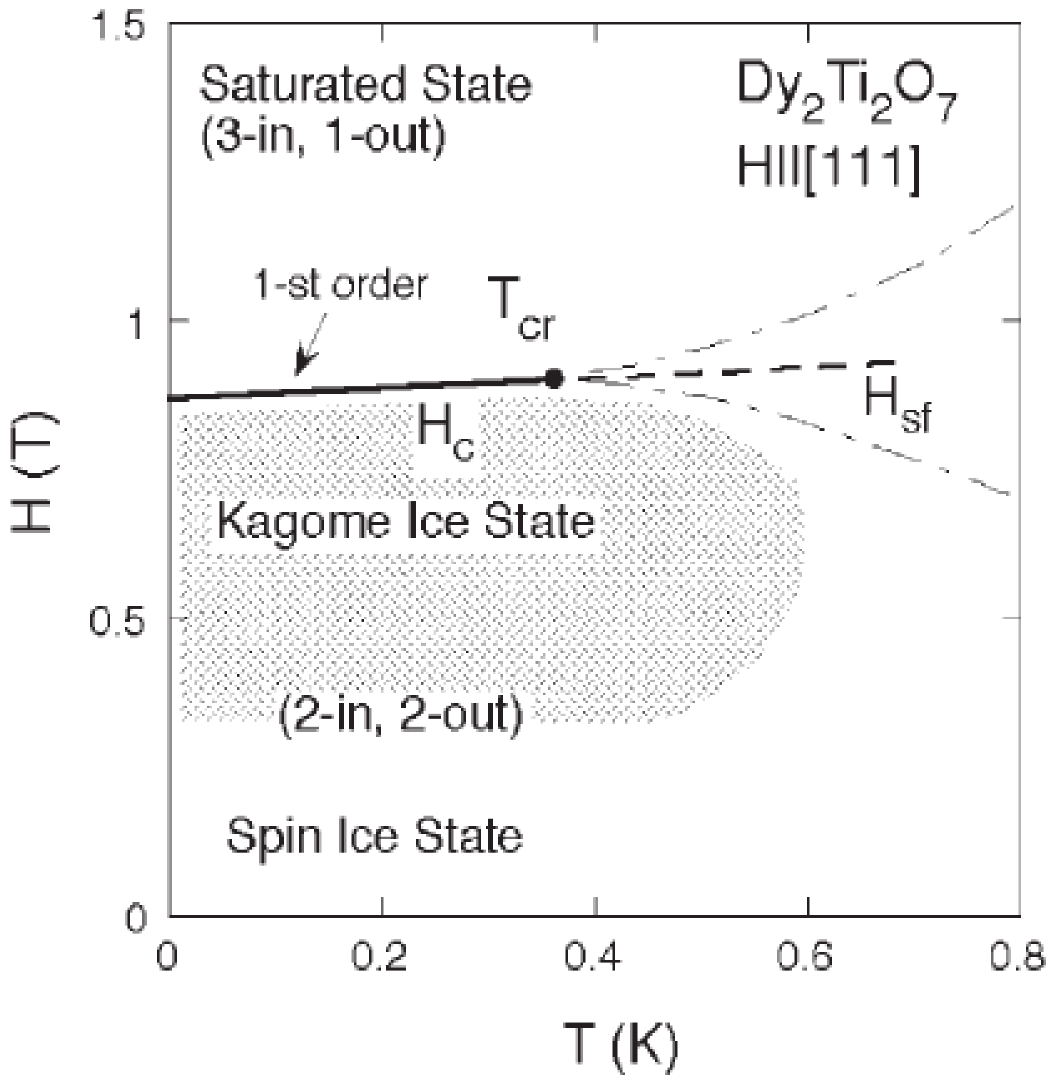


Fig. 1. Phase diagram of $Dy_2Ti_2O_7$ in a [111] magnetic field, determined by magnetization and specific heat measurements. The dashed line

PHYSICAL REVIEW B **72**, 144422 (2005)

Magnetodielectric response of the spin-ice $Dy_2Ti_2O_7$

Masafumi Saito,[1] Ryuji Higashinaka,[1] and Yoshiteru Maeno[1,2,*]

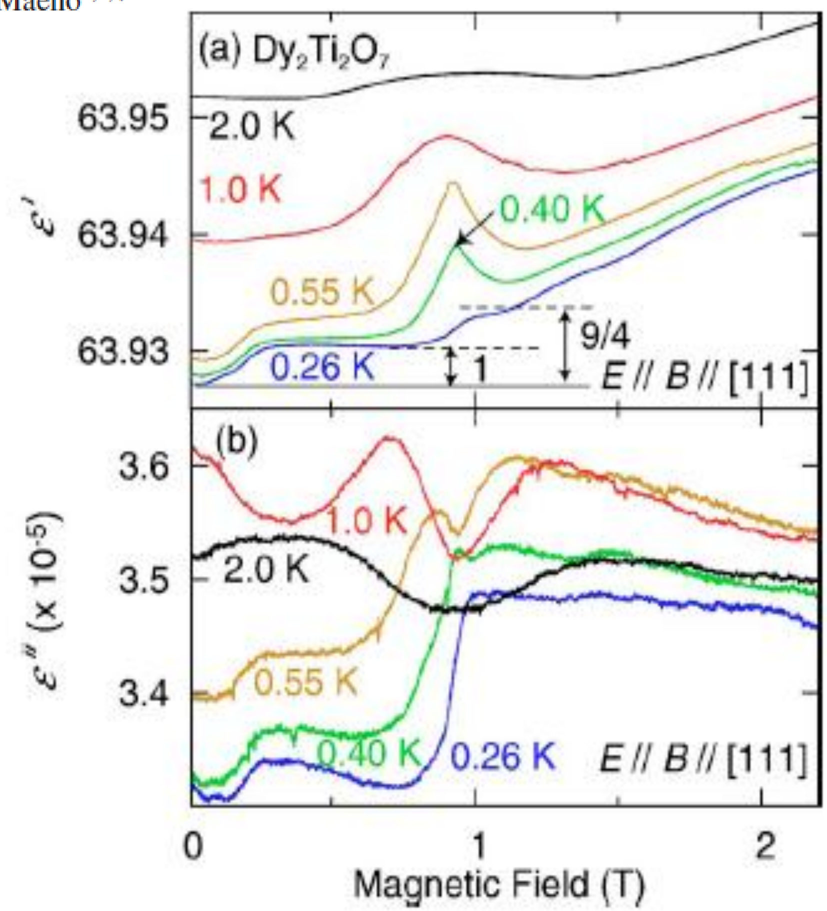
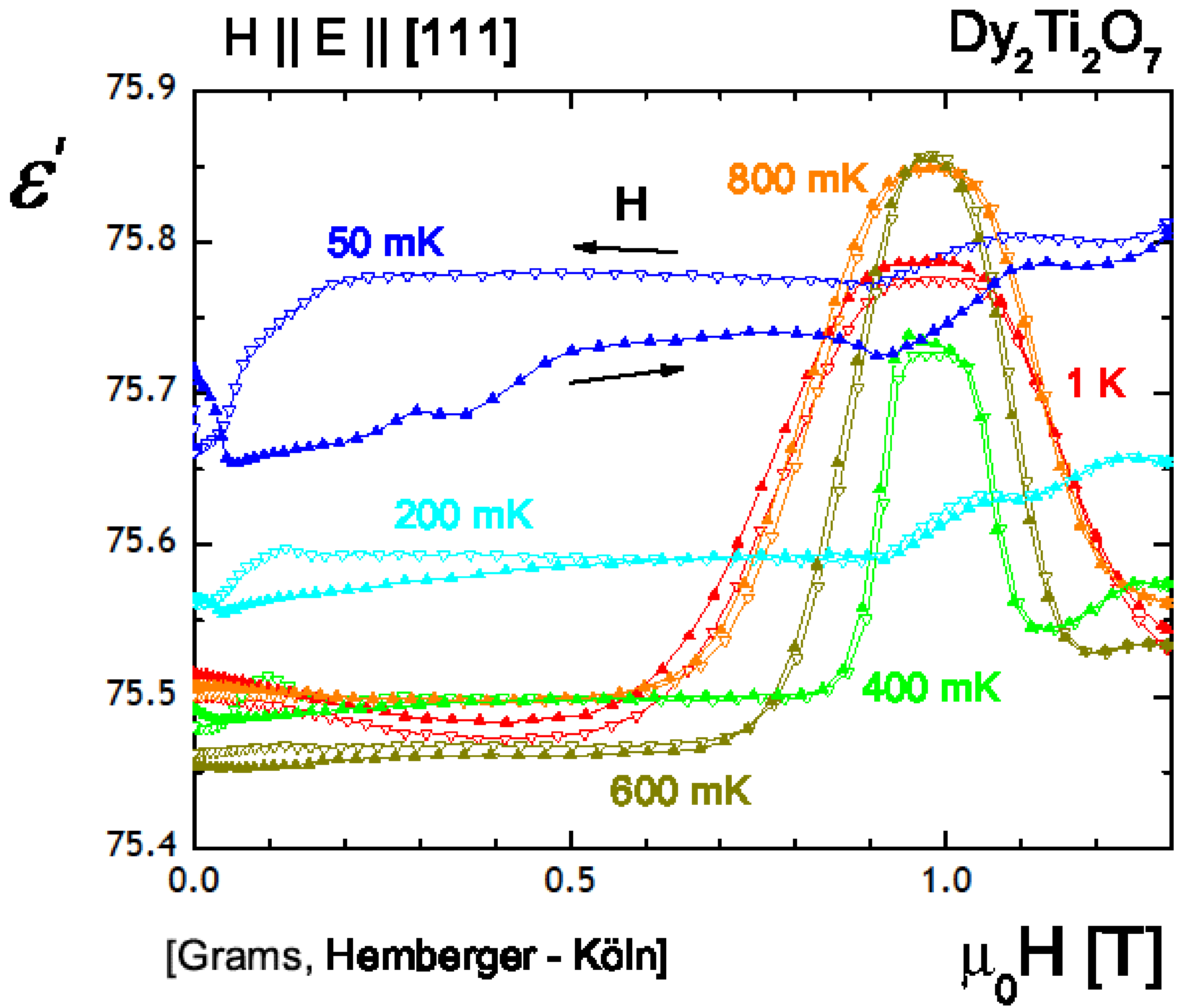


FIG. 6. (Color online) Magnetic field dependence of (a) the real and (b) the imaginary parts of the dielectric constant of $Dy_2Ti_2O_7$



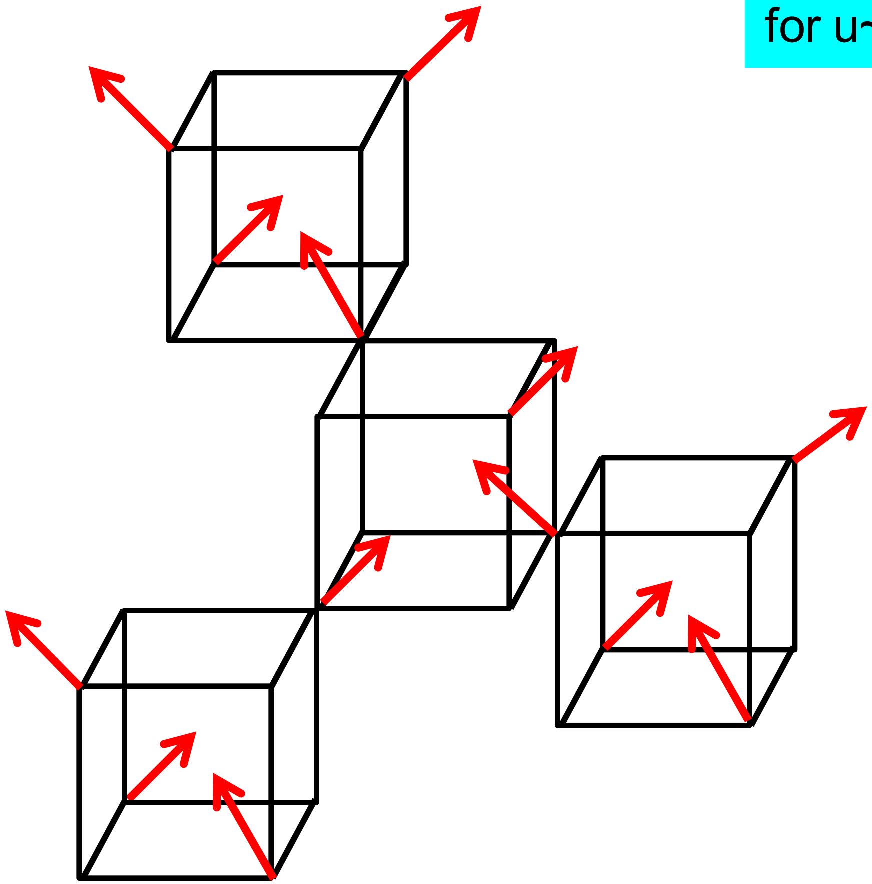
H || E || [111]
Dy2Ti2O7
ε'
75.9
75.8
75.7
75.6
75.5
75.4
0.0
0.5
1.0
H
50 mK
800 mK
1 K
200 mK
400 mK
600 mK
[Grams, Hemberger - Köln]
μ0H [T]

- **External electric field: Decreases excitation energy of certain monopoles**

$\omega = \omega_0 - \mathbf{dE}$

Estimates: ε=dE =eu(Å)E(V/cm)

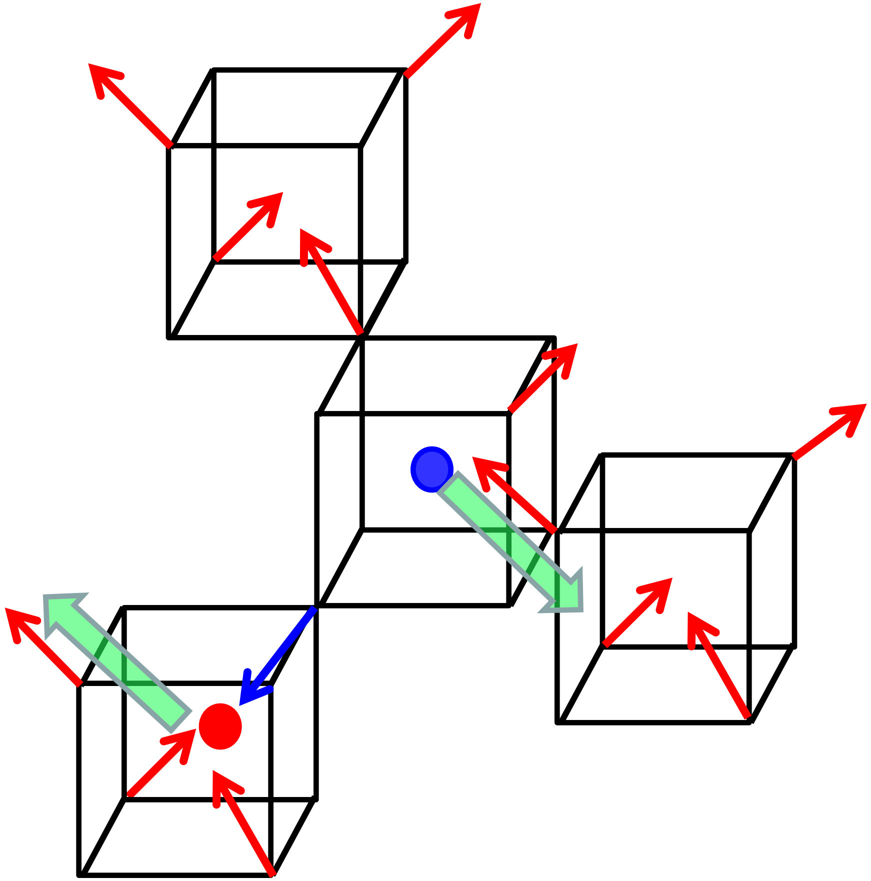
for u~0.01Å and E ~10^5V/cm ε~10^{-5} eV~0.1K



In strong magnetic field **H** || [001]

External electric field: Decreases excitation energy of certain monopoles

$\omega = \omega_0 - \mathbf{dE}$

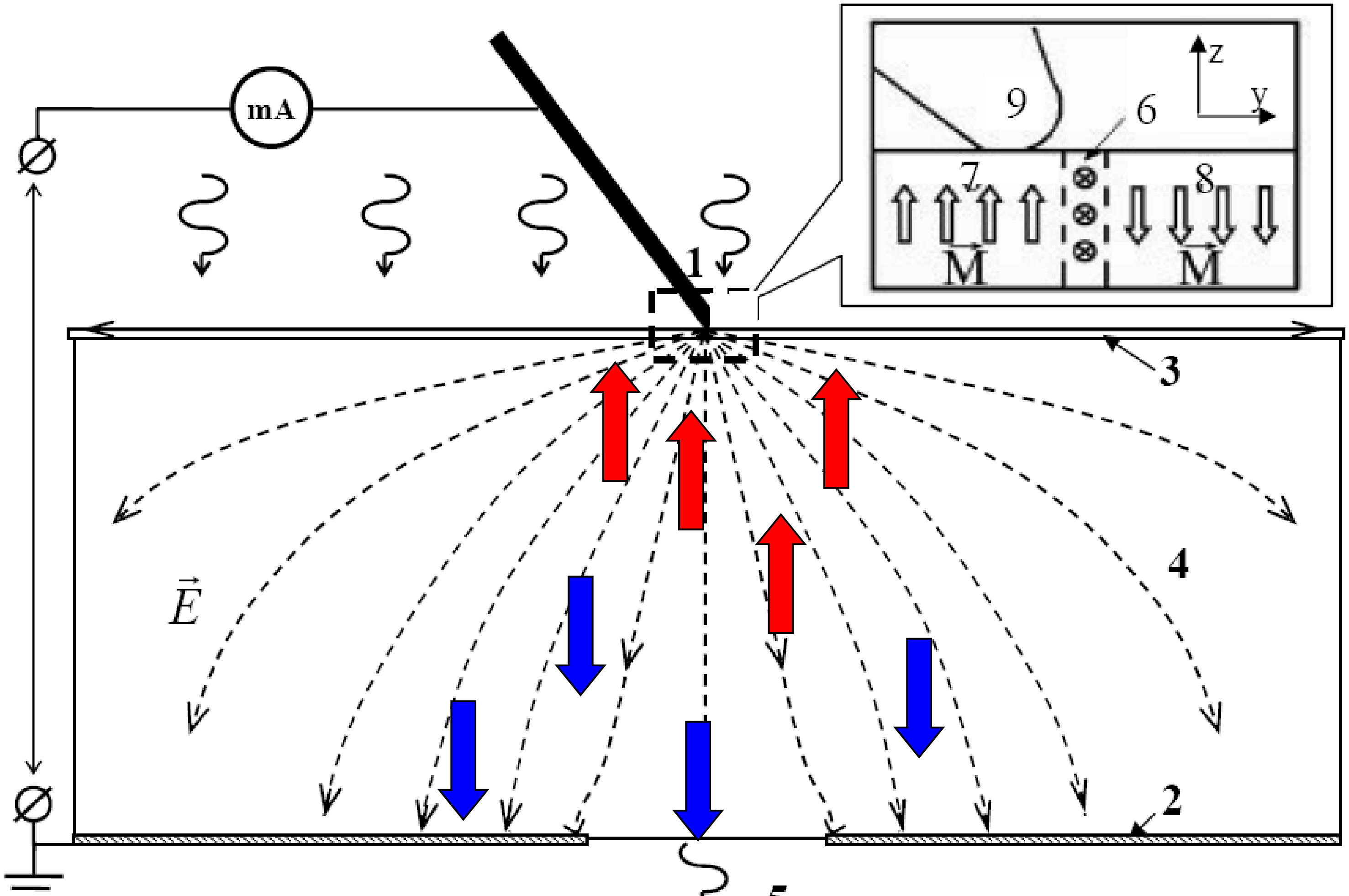


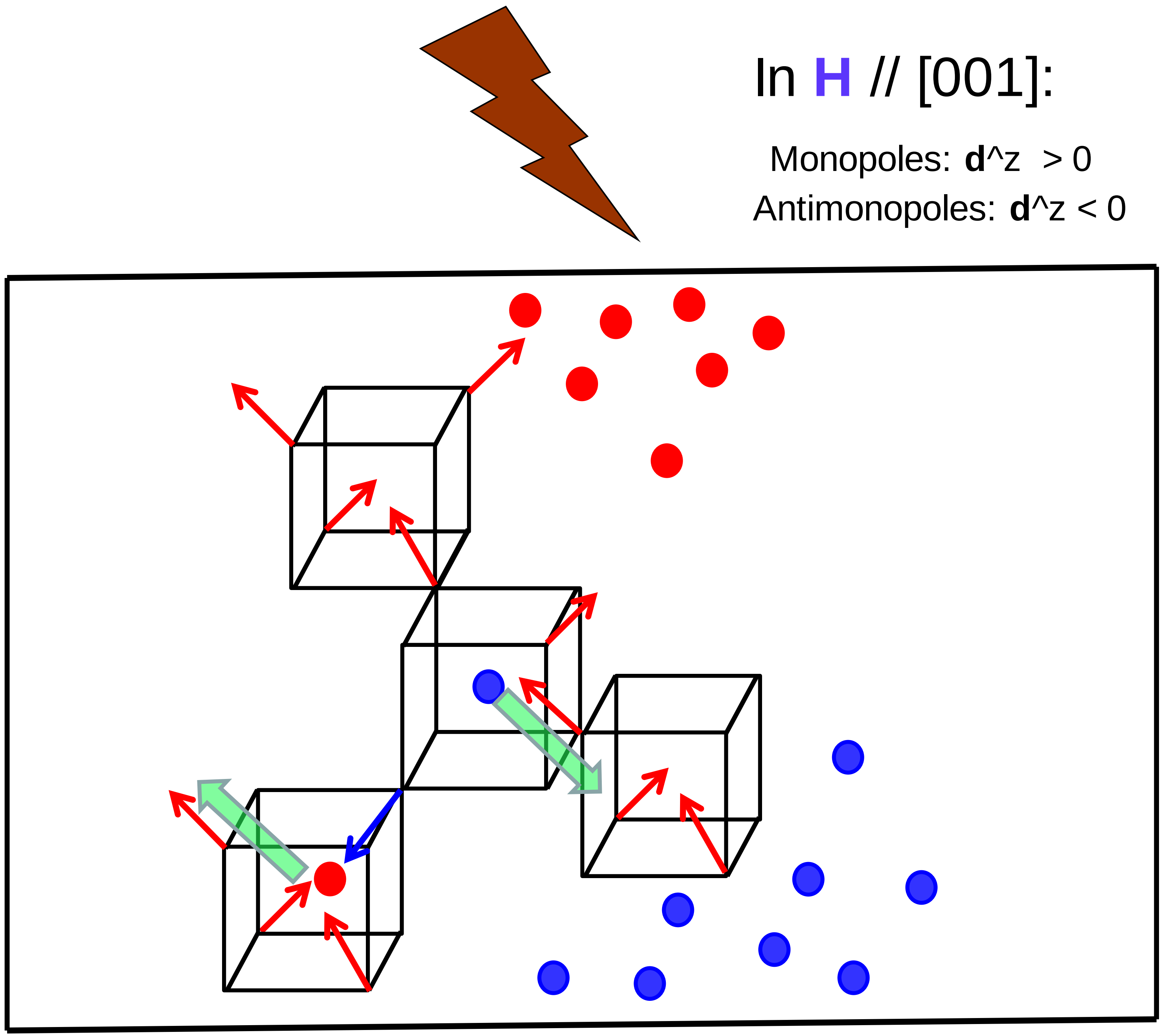
In strong magnetic field **H** || [001]

Monopoles: $\mathbf{d}^z > 0$

Antimonopoles: $\mathbf{d}^z < 0$

Inhomogeneous electric field

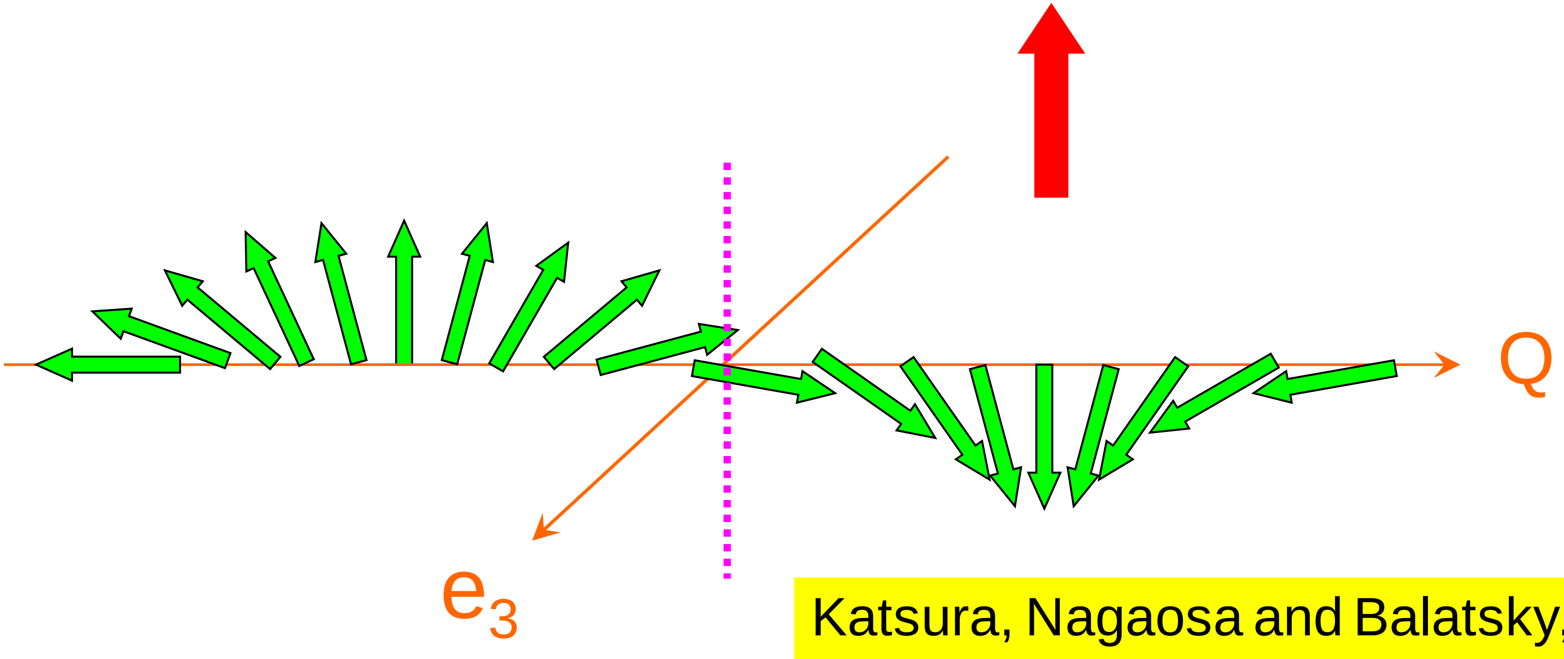



In H // [001]:
Monopoles: d^z > 0
Antimonopoles: d^z < 0

Cycloidal SDW :

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

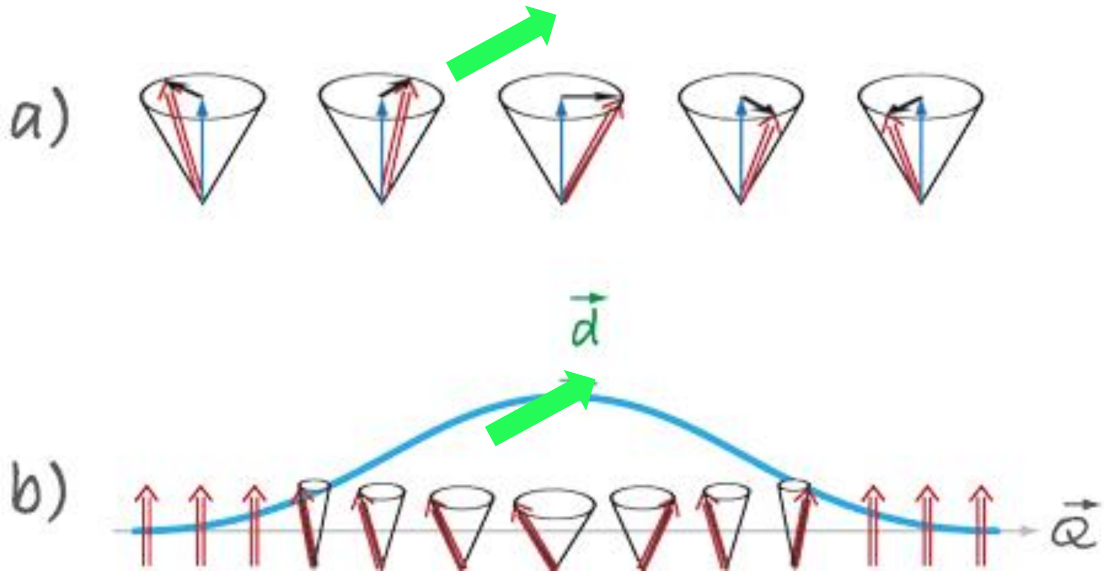
$$\overline{\mathbf{P}} \propto \left[\mathbf{e}_3 \times \mathbf{Q}\right]$$



Katsura, Nagaosa and Balatsky, 2005

Mostovoy 2006

Polarization carried by the usual spin waves



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

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

CONCLUSIONS 1

- Contrary to the common belief, there are real charge effects in strong Mott insulators (with frustrated lattices): spin-driven spontaneous electric currents and orbital moments, and charge redistribution in the ground state

- Spontaneous currents are ~ scalar spin chirality $\chi_{123} = \vec{S}_1 \left[\vec{S}_2 \times \vec{S}_3 \right]$

- Charge redistribution ($\langle n_i \rangle$ is not 1!) may lead to **electric polarization** (purely electronic mechanism of multiferroicity)

- Many consequences:

  - In the ground state: lifting of degeneracy; formation of spin-driven CDW, …….

  - In dynamics: electric field-induced "ESR"; rotation of electric polarization by spins; contribution of spins to low-frequency dielectric function; possibility of negative refraction index; etc

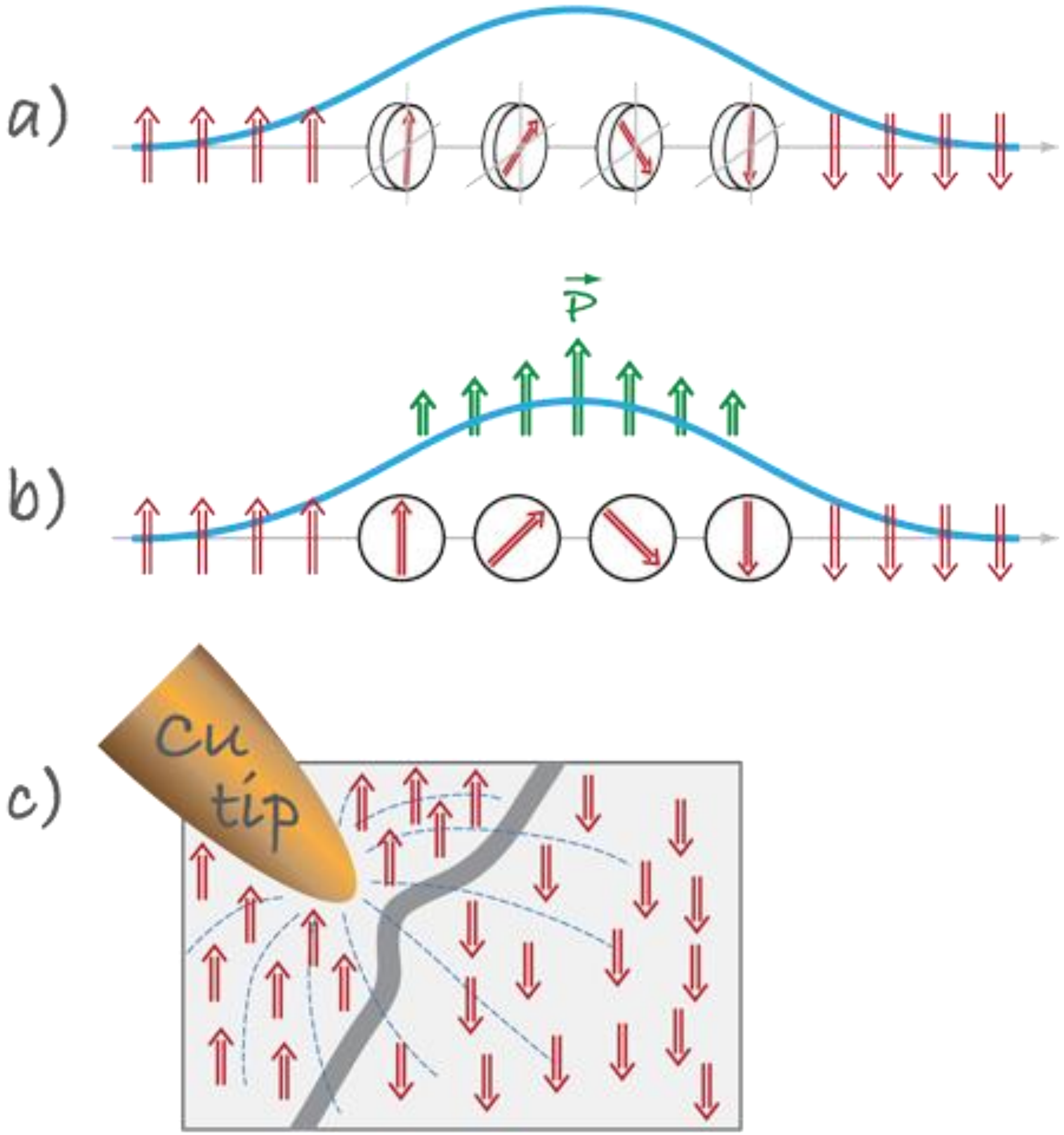
Conclusions 2

★ 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

Such effect was already observed for Neel domain walls in ferromagnets (cf. spiral multiferroics):



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

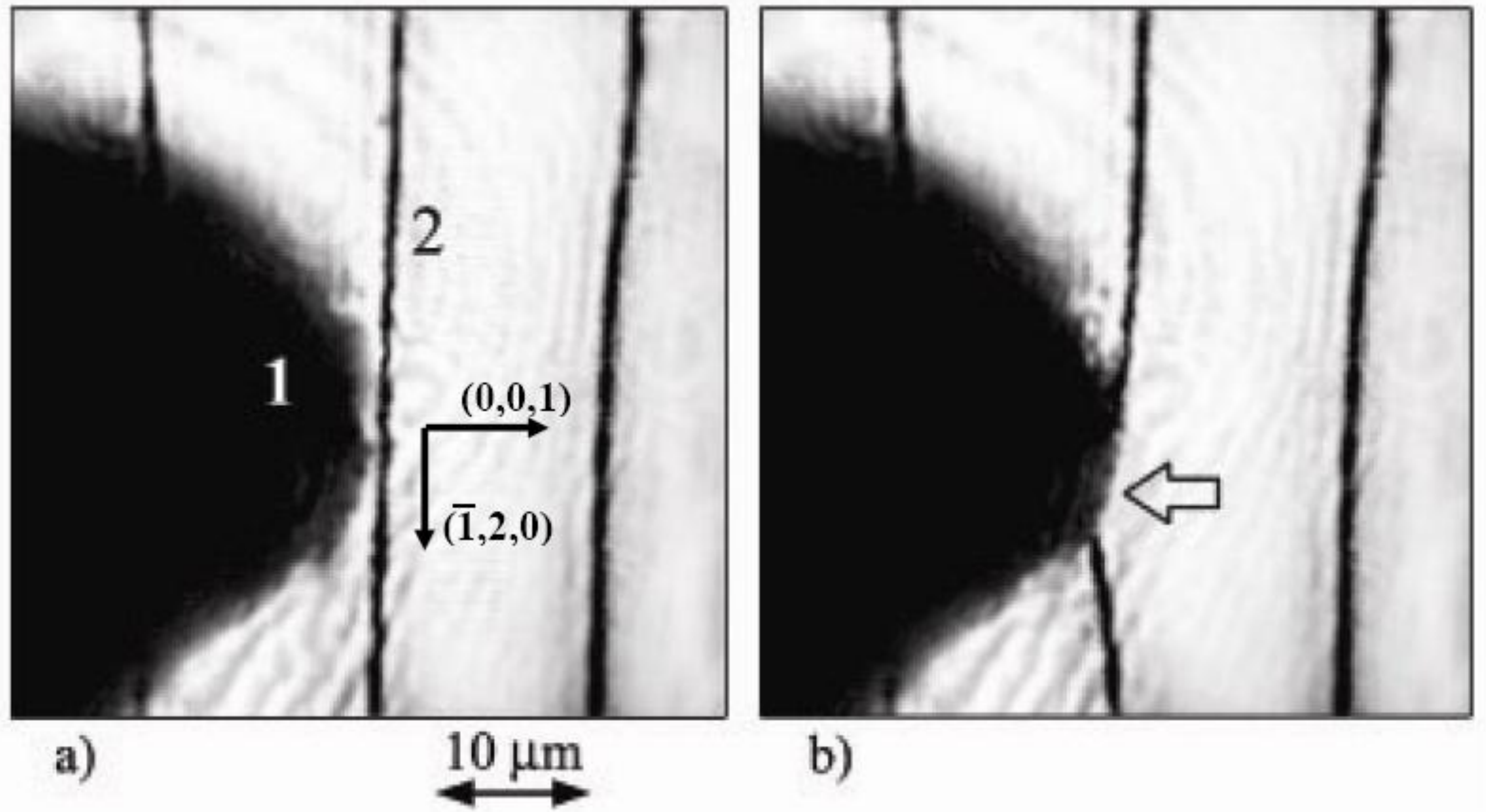


Fig. 2 The effect of electric field in the vicinity of electrode (1) on magnetic domain wall (2) in the films of ferrite garnets: a) initial state b) at the voltage of +1500 V applied