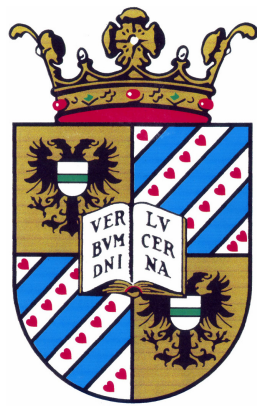


Multiferroic and magnetoelectric materials



Maxim Mostovoy

University of Groningen

**Zernike Institute
for Advanced Materials**

ICMR Summer School
on Multiferroics

Santa Barbara 2008

Outline

- Ferroelectric properties of magnetic defects
- Toroidal moment and magnetoelectric effect
- Electromagnons

Electrostatics of magnetic defects

Easy plane spins: $\mathbf{M} = M [\mathbf{e}_1 \cos \varphi + \mathbf{e}_2 \sin \varphi]$

Polarization: $P_a = -\gamma\chi_e M^2 \varepsilon_{3ab} \partial_b \varphi$

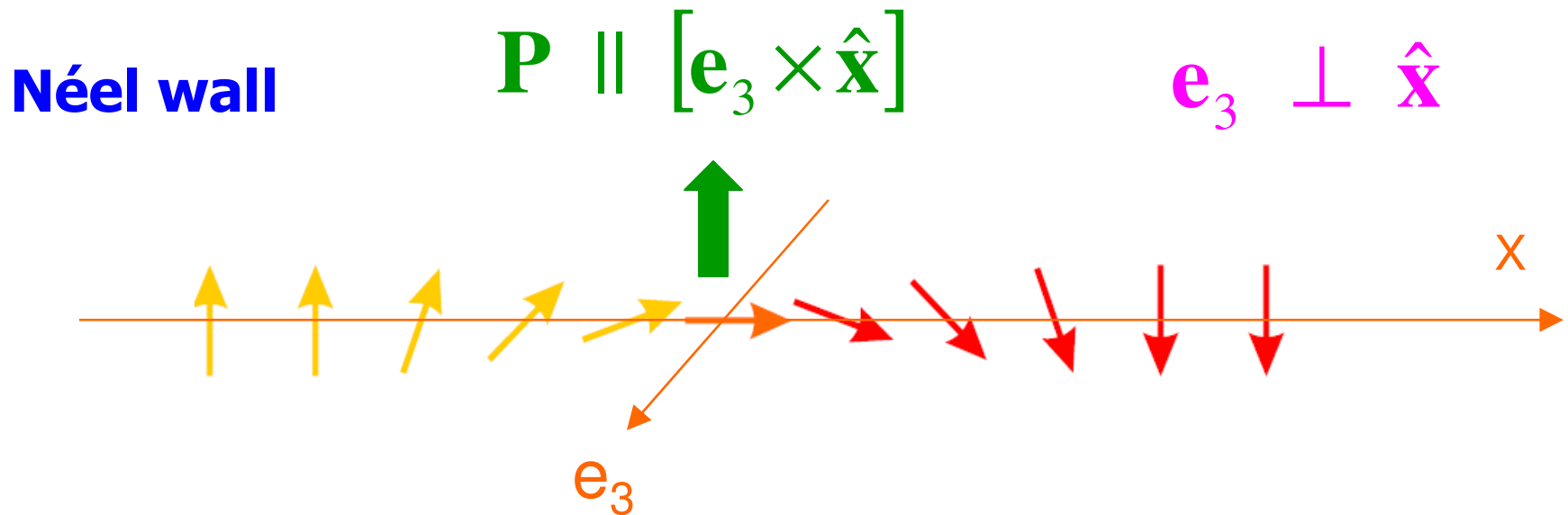
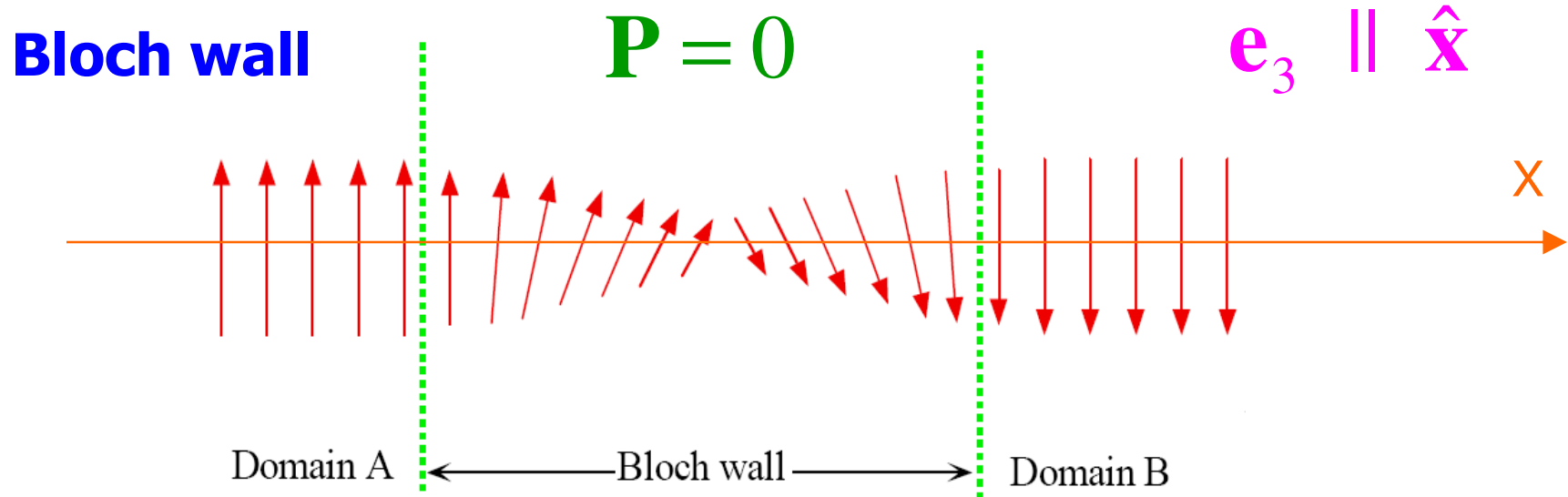
Total polarization of domain wall:

$$\int dx P_y = \gamma\chi_e M^2 [\varphi(+\infty) - \varphi(-\infty)]$$

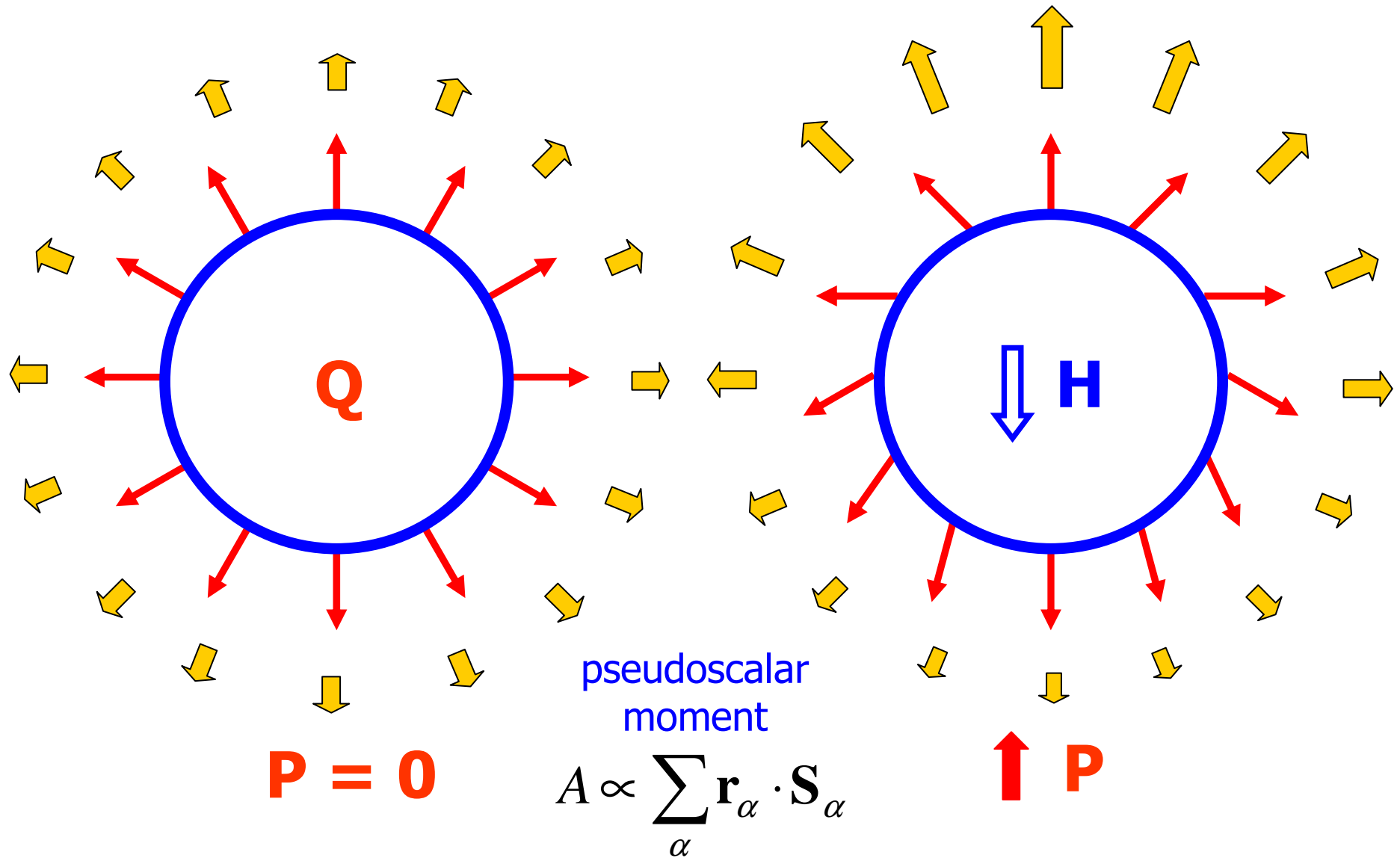
Charge density: $\rho = -\text{div}\mathbf{P} = 2\pi\gamma\chi_e M^2 \Gamma \delta^{(2)}(\mathbf{x}_\perp)$

Vortex charge: $Q \propto \Gamma = \frac{1}{2\pi} \oint_C d\mathbf{x} \cdot \nabla \varphi$ **winding number**

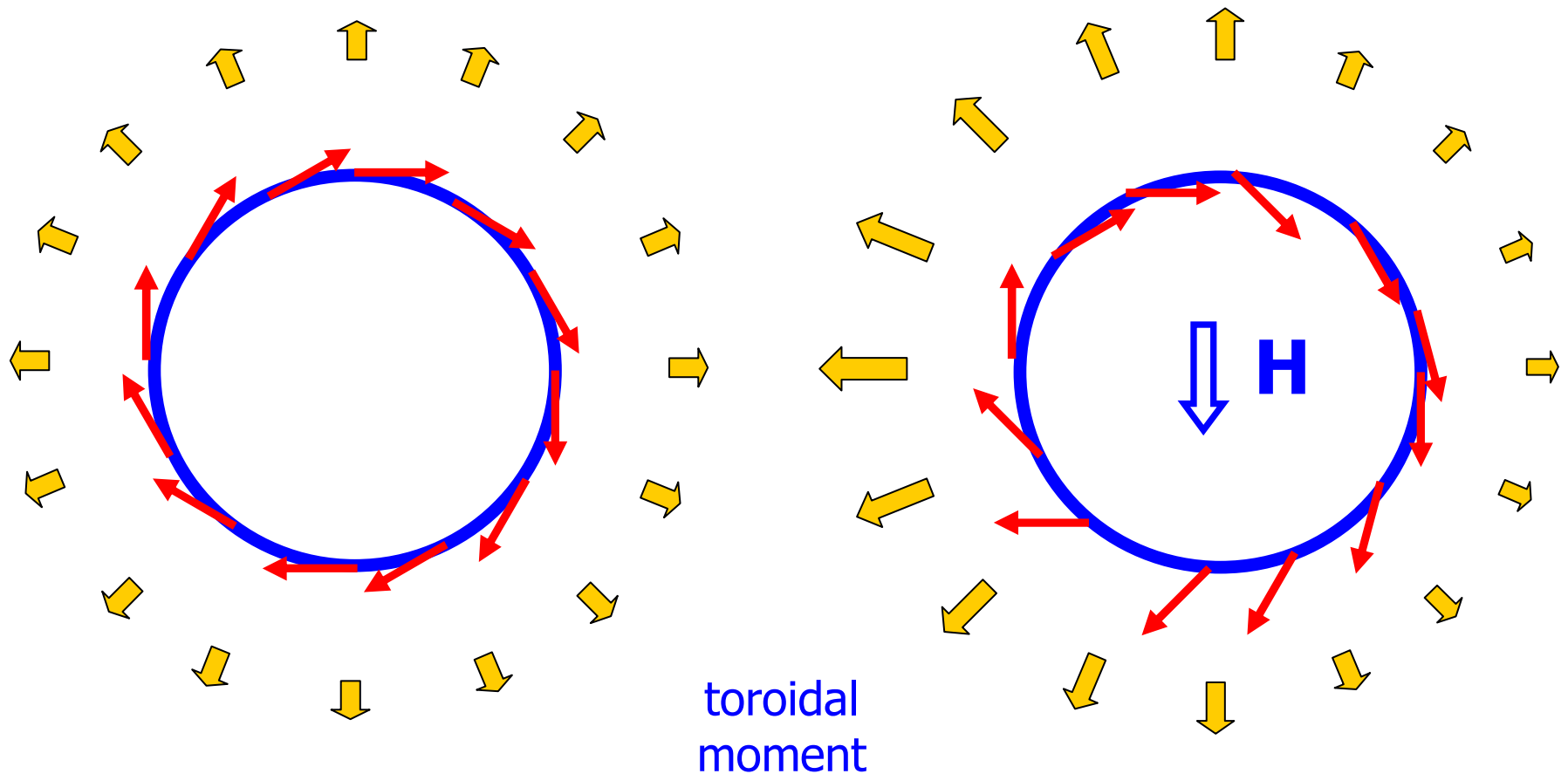
Polarization of domain walls



Magnetic vortex in magnetic field



Magnetic vortex in magnetic field



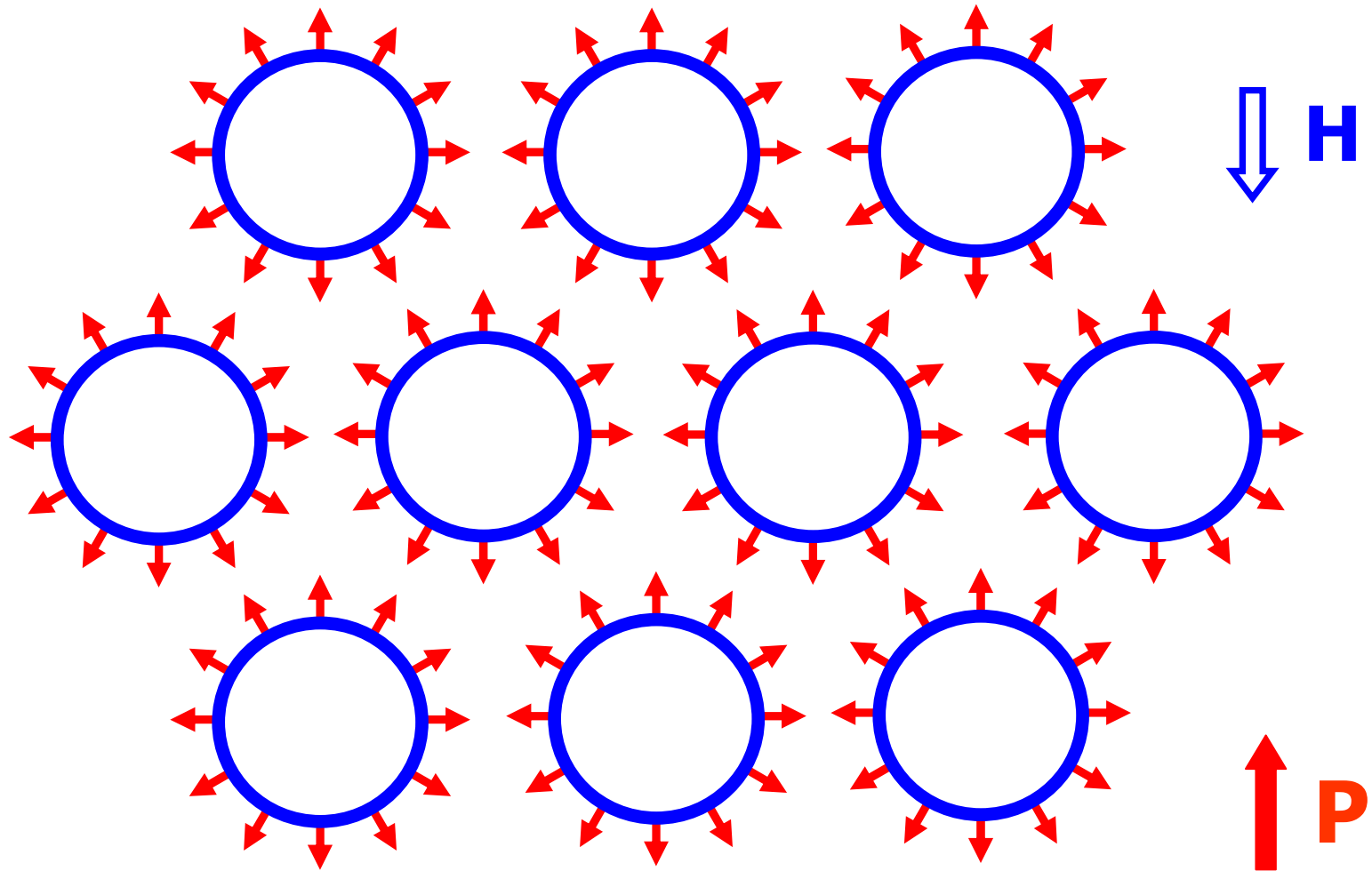
$$\mathbf{P} = \mathbf{0}$$

toroidal
moment

$$\mathbf{T} \propto \sum_{\alpha} \mathbf{r}_{\alpha} \times \mathbf{S}_{\alpha}$$

$$\leftarrow \mathbf{P}$$

Array of magnetic vortices is magnetoelectric



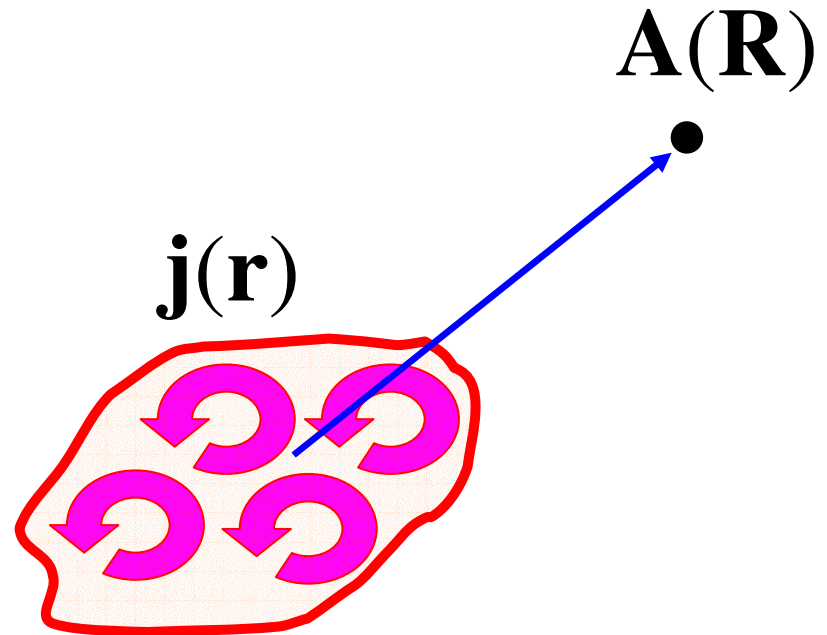
Outline

- Ferroelectric properties of magnetic defects
- Toroidal moment and magnetoelectric effect
- Electromagnons

Multipole expansion

Average vector potential

$$\mathbf{A}(\mathbf{R}) = \frac{1}{c} \int d^3r \frac{\mathbf{j}(\mathbf{r})}{|\mathbf{r} - \mathbf{R}|}$$



Magnetic dipole moment

$$\mathbf{A}^{(1)} = -\mathbf{M} \times \nabla \frac{1}{R}$$

$$\mathbf{M} = \frac{1}{2c} \int d^3r \mathbf{r} \times \mathbf{j}$$

Quadrupole and toroidal moments

Quadrupole moment

$$A_i^{(2)} = -\varepsilon_{ijk} Q_{kl} \partial_j \partial_l \frac{1}{R}$$

$$Q_{ij} = \frac{1}{6c} \int d^3 r \left([\mathbf{r} \times \mathbf{j}]_i r_j + [\mathbf{r} \times \mathbf{j}]_j r_i \right)$$

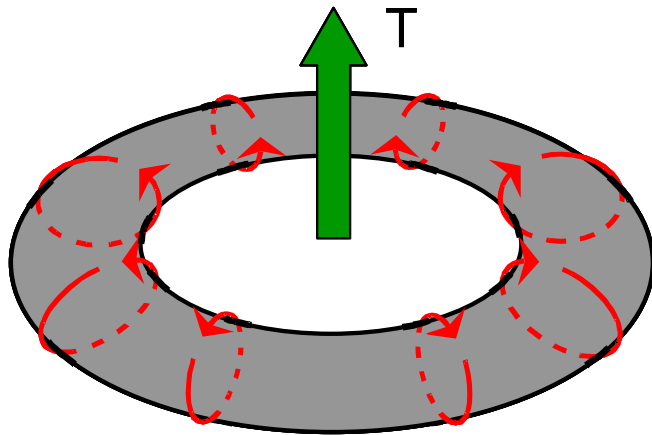
Toroidal moment (not in textbooks)

$$\mathbf{A}^{(2)} = \nabla(\mathbf{T} \cdot \nabla) \frac{1}{R} + 4\pi \mathbf{T} \delta(\mathbf{R})$$

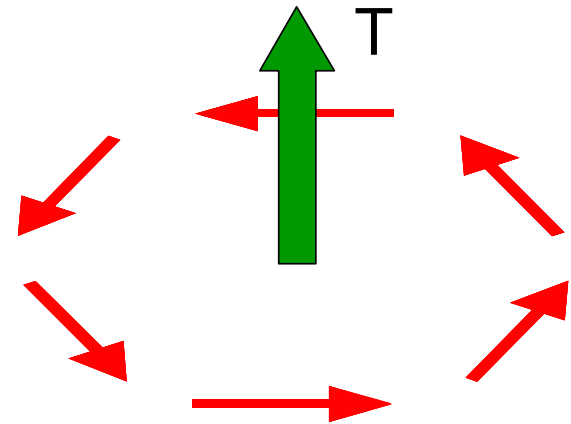
$$\mathbf{T} = \frac{1}{6c} \int d^3 r \mathbf{r} \times [\mathbf{r} \times \mathbf{j}]$$

Meaning of toroidal moment

$$\mathbf{T} = \frac{1}{6c} \int d^3r \mathbf{r} \times [\mathbf{r} \times \mathbf{j}]$$

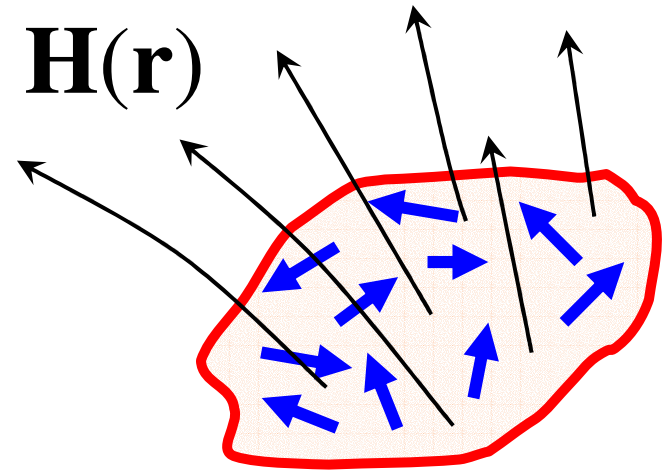


$$\mathbf{T} = \mu_B \sum_{\alpha} \mathbf{r}_{\alpha} \times \mathbf{S}_{\alpha}$$



Spins interacting with inhomogeneous magnetic field

$$E = -2\mu_B \sum_{\alpha} \mathbf{S}_{\alpha} \cdot \mathbf{H}(\mathbf{r}_{\alpha})$$



Gradient expansion

$$E^{(0)} = -\mathbf{M} \cdot \mathbf{H}(0)$$

~~$$E^{(1)} = -A \nabla \cdot \mathbf{H} - \mathbf{T} \cdot \nabla \times \mathbf{H} - Q_{ij} (\partial_i H_j + \partial_j H_i)$$~~

$$A = -\frac{2}{3} \mu_B \sum_{\alpha} \mathbf{r}_{\alpha} \cdot \mathbf{S}_{\alpha}$$

Magnetolectric properties of magnetic multipoles

$$\Phi_{\text{me}} = -a \mathbf{E} \cdot \mathbf{H} - \mathbf{t} \cdot \mathbf{E} \times \mathbf{H} - q_{ij} (E_i H_j + E_j H_i)$$

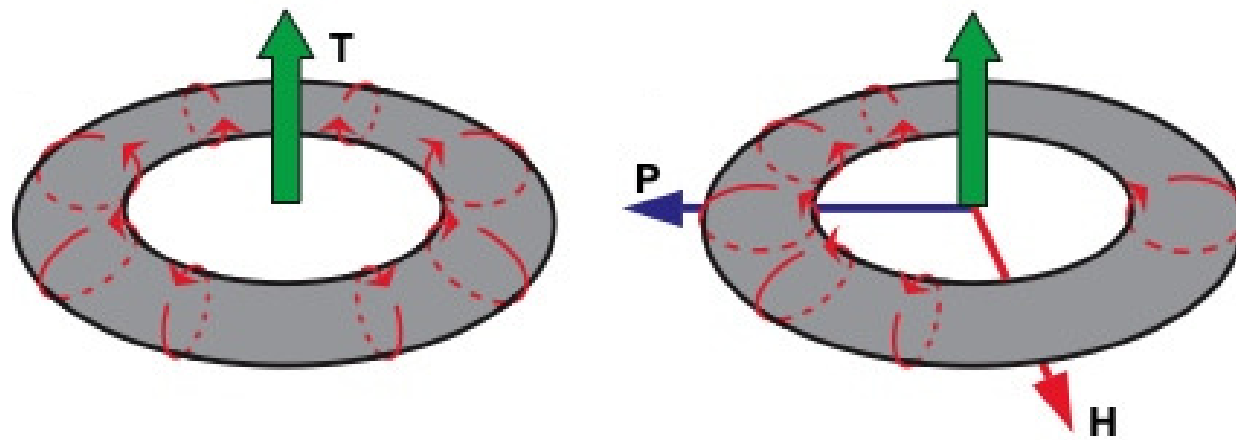
$$\Phi_H = -A \nabla \cdot \mathbf{H} - \mathbf{T} \cdot \nabla \times \mathbf{H} - Q_{ij} (\partial_i H_j + \partial_j H_i)$$

The magnetic multipoles have the same symmetry as magnetolectric tensor

$$a \propto A \quad \mathbf{t} \propto \mathbf{T} \quad q_{ij} \propto Q_{ij}$$

Magnetoelectric effect

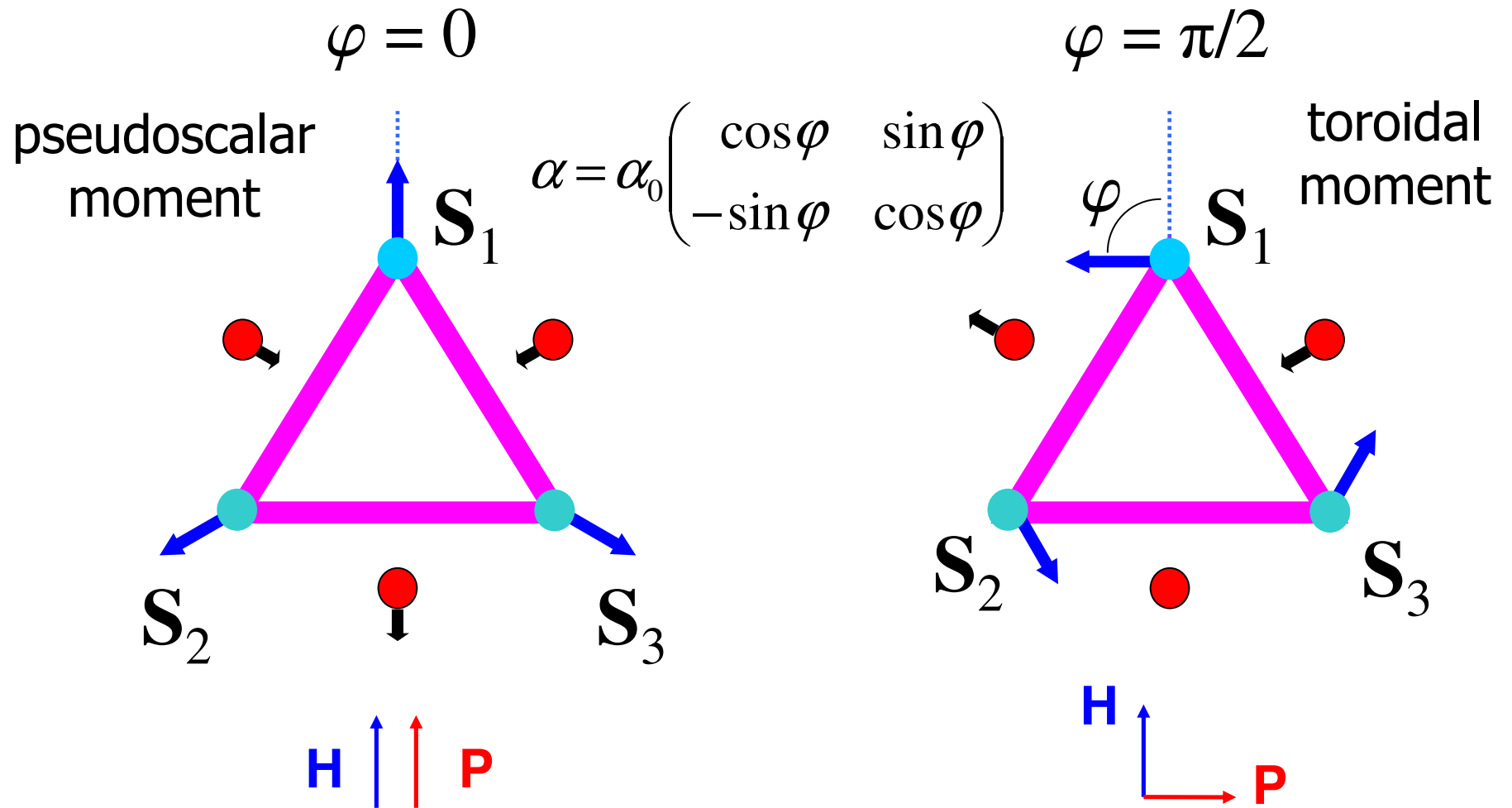
$$\Phi_{me} = -\lambda \mathbf{T} \cdot \mathbf{E} \times \mathbf{H}$$



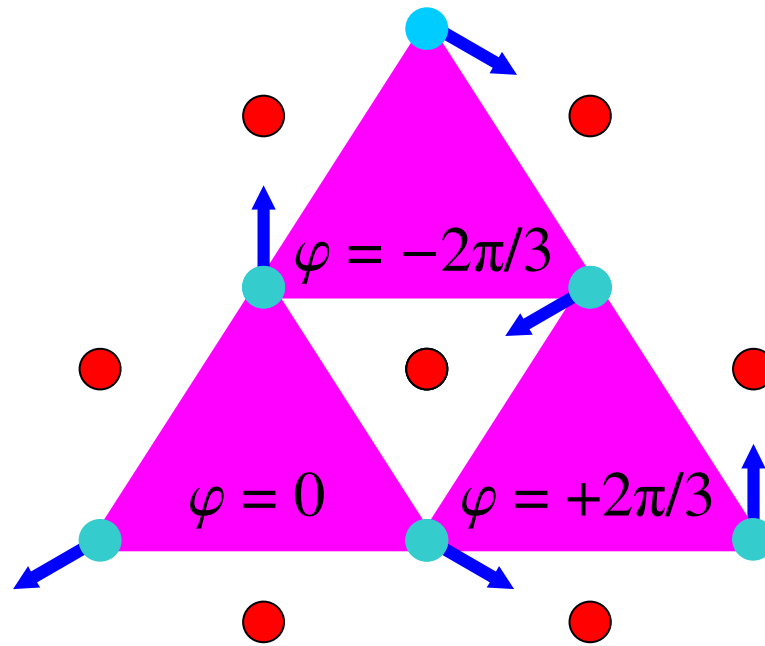
$$\mathbf{P} = -\frac{\partial \Phi_{me}}{\partial \mathbf{E}} = -\mathbf{T} \times \mathbf{H}$$

$$\mathbf{M} = -\frac{\partial \Phi_{me}}{\partial \mathbf{H}} = +\mathbf{T} \times \mathbf{E}$$

Magnetoelectric effect in spin triangle

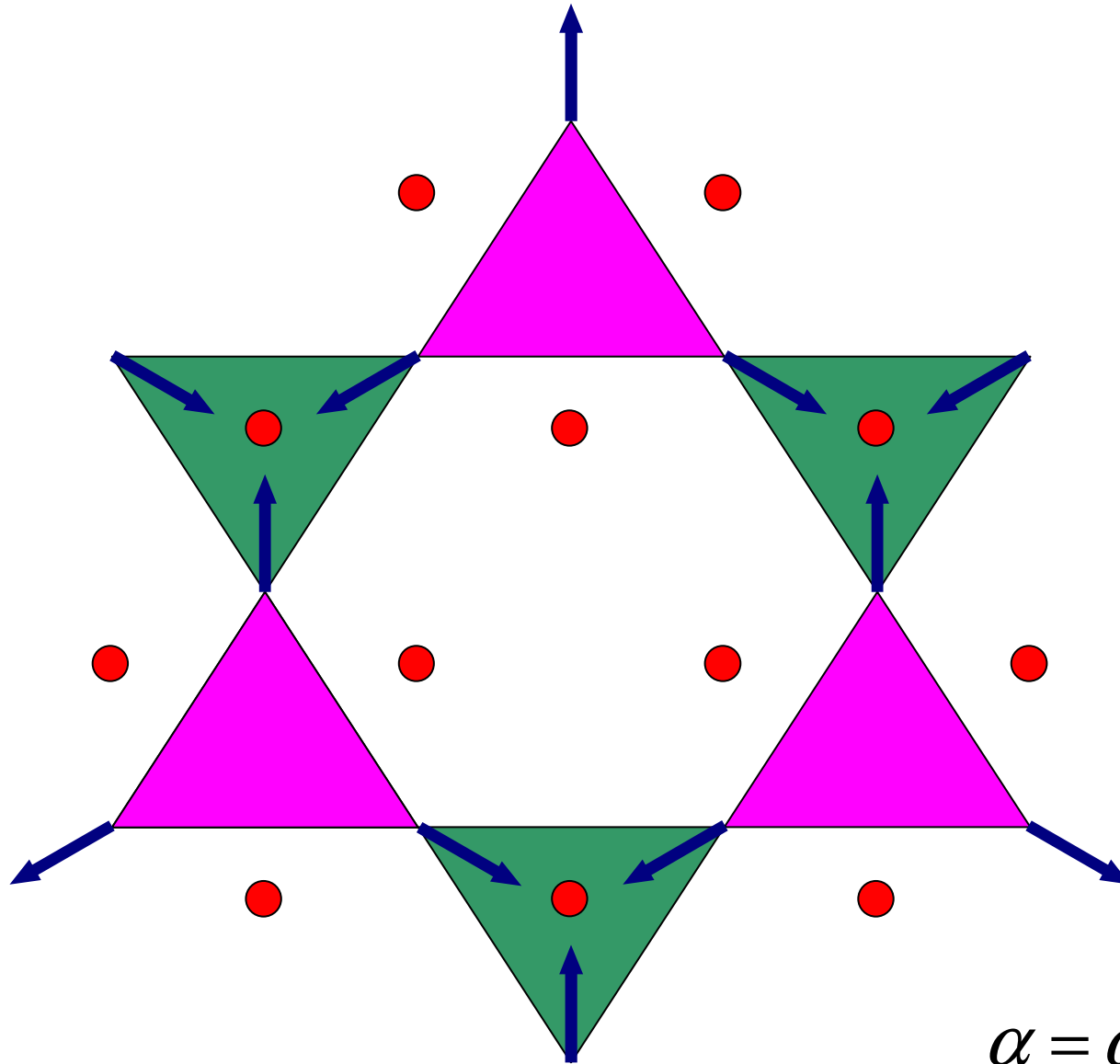


Triangular lattice



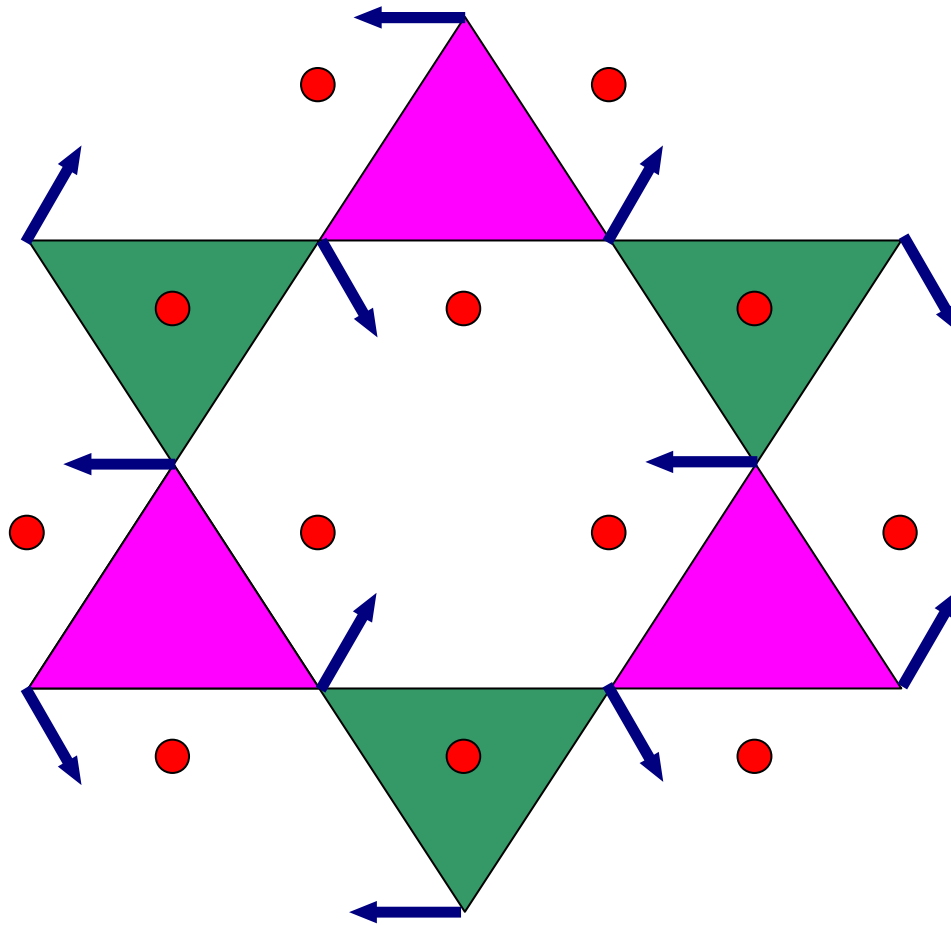
Cancellation of
magnetoelectric
effect

Pseudoscalar Kagome



$$\alpha = \alpha_0 \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$$

Toroidal Kagome

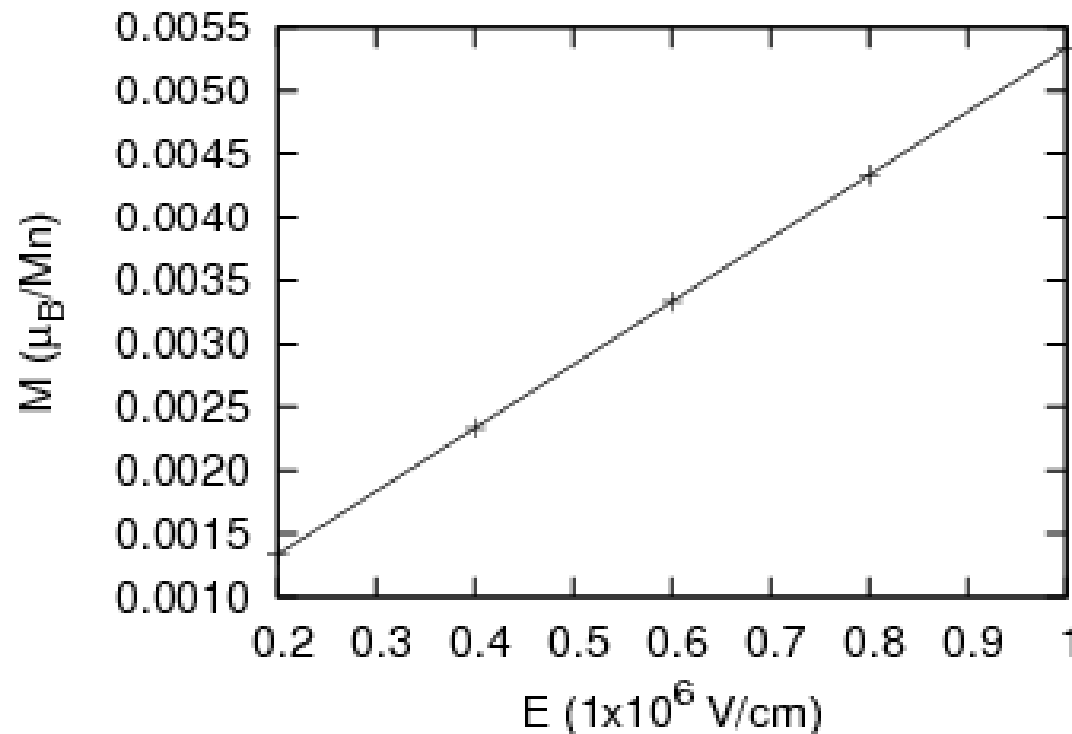


layered
Kagomé lattice

$$\alpha = \alpha_0 \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}$$

Magnetolectric Coupling

- KITPITE: $\alpha = 3 \cdot 10^{-3}$ Gaussian units)



LDA+U
Cris Delaney &
Nicola Spaldin

- Cr₂O₃ $\alpha = 1 \cdot 10^{-4}$

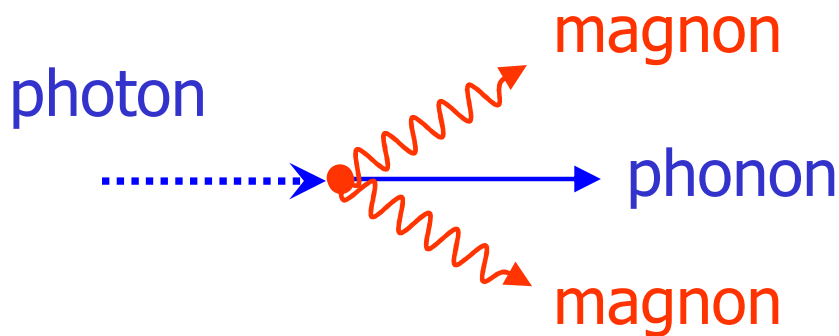
Outline

- Ferroelectric properties of magnetic defects
- Toroidal moment and magnetoelectric effect
- Electromagnons

Phonon-magnon continuum

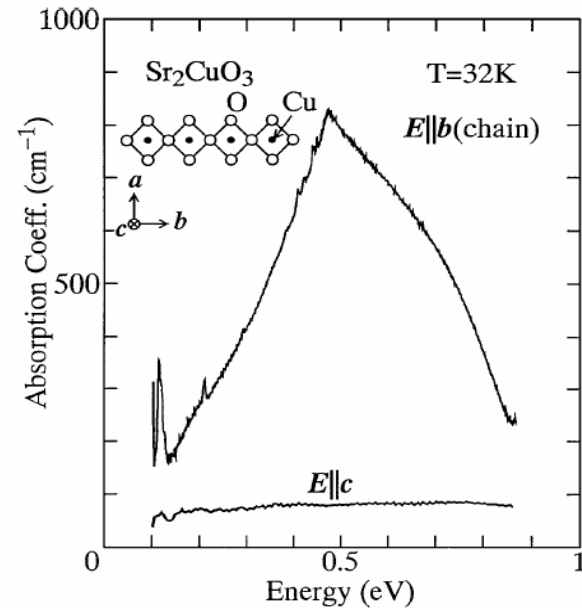
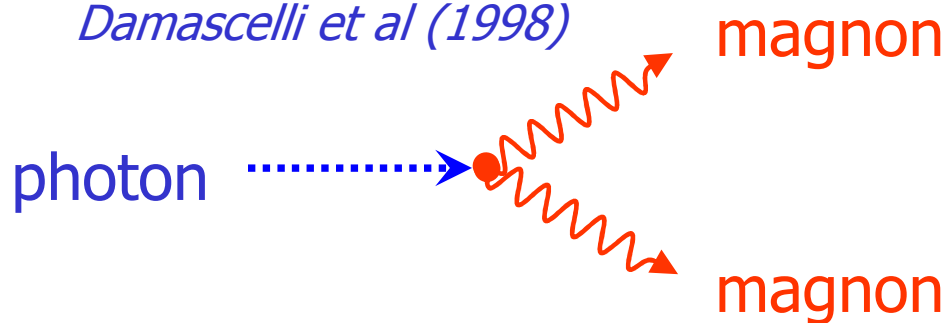
$$H_{me} = -\mathbf{P} \sum_{ijk} \lambda_{ijk} \mathbf{u}_i (\mathbf{S}_j \cdot \mathbf{S}_k)$$

Lorenzana & Sawatzky (1995)

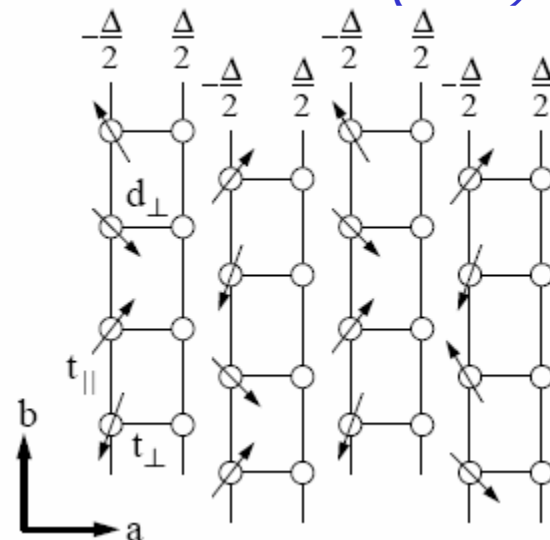


'Charged magnons' in NaV_2O_5

Damascelli et al (1998)

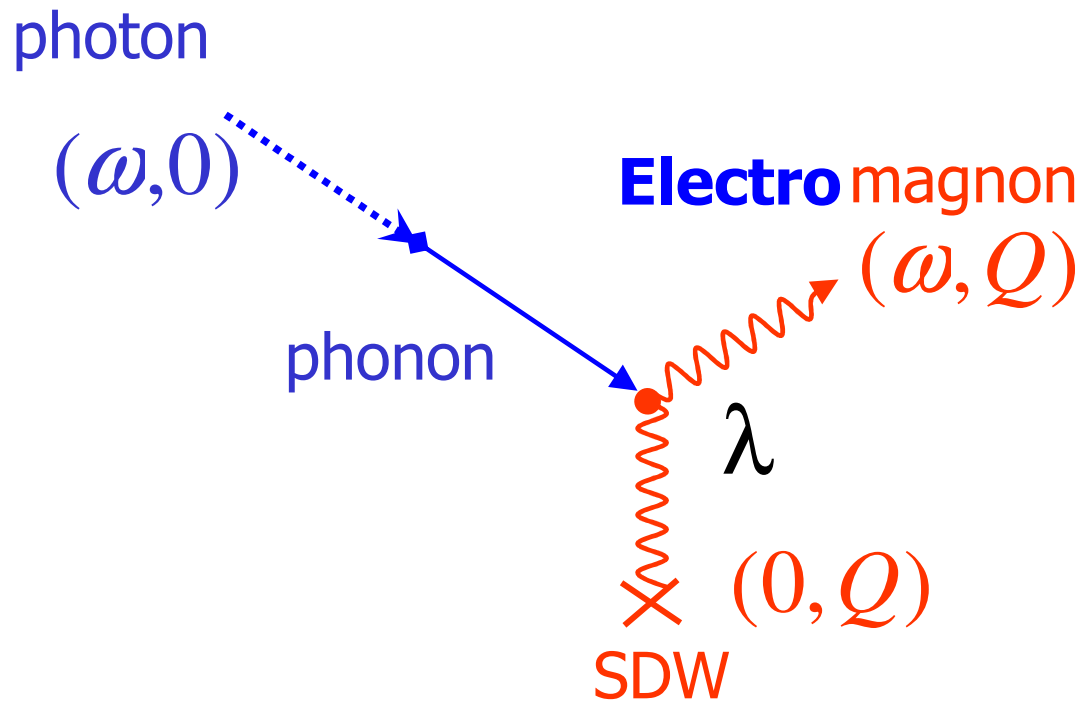


Suzuura et al (1996)



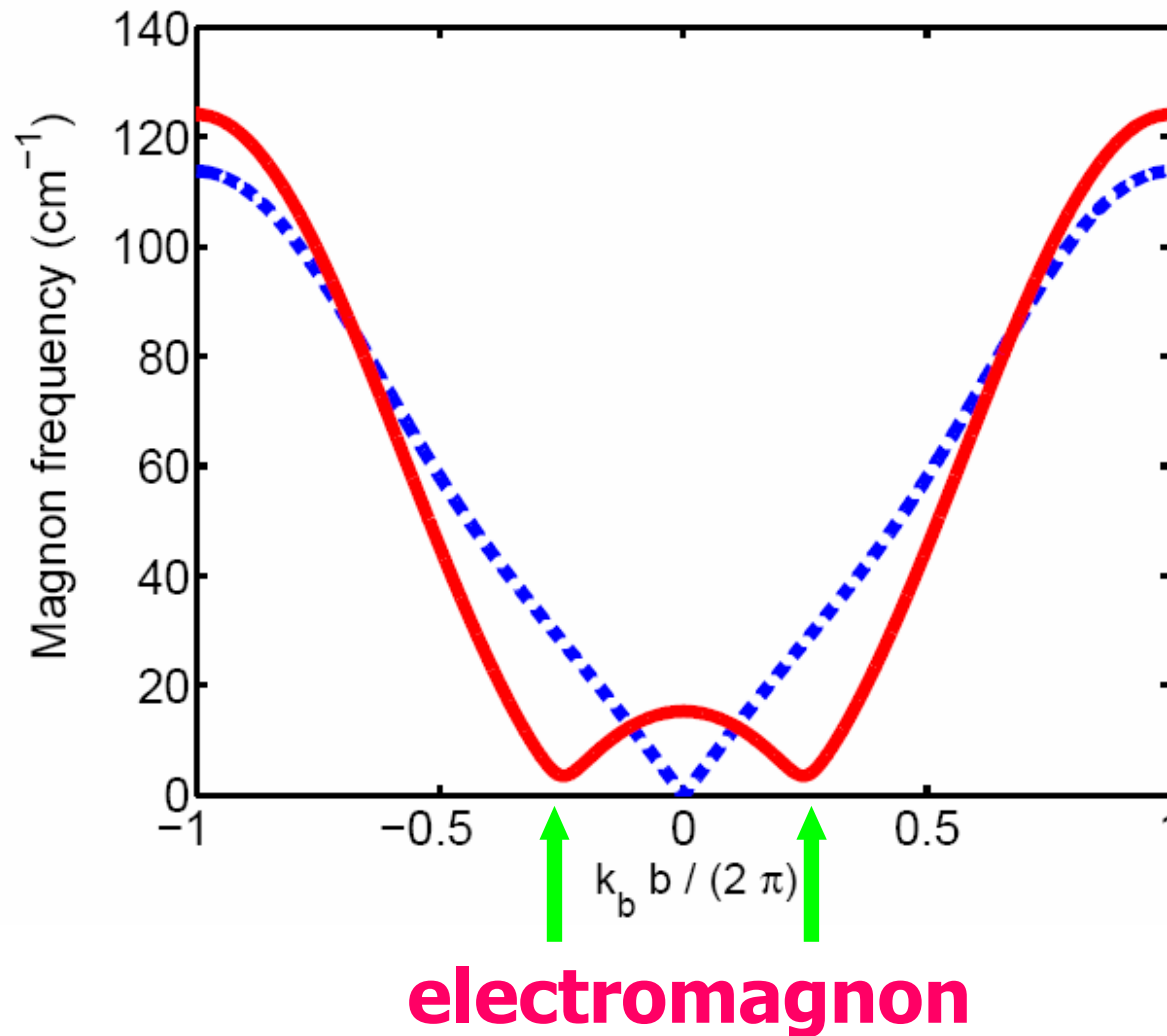
Photoexcitation of magnons

PL ∂ L

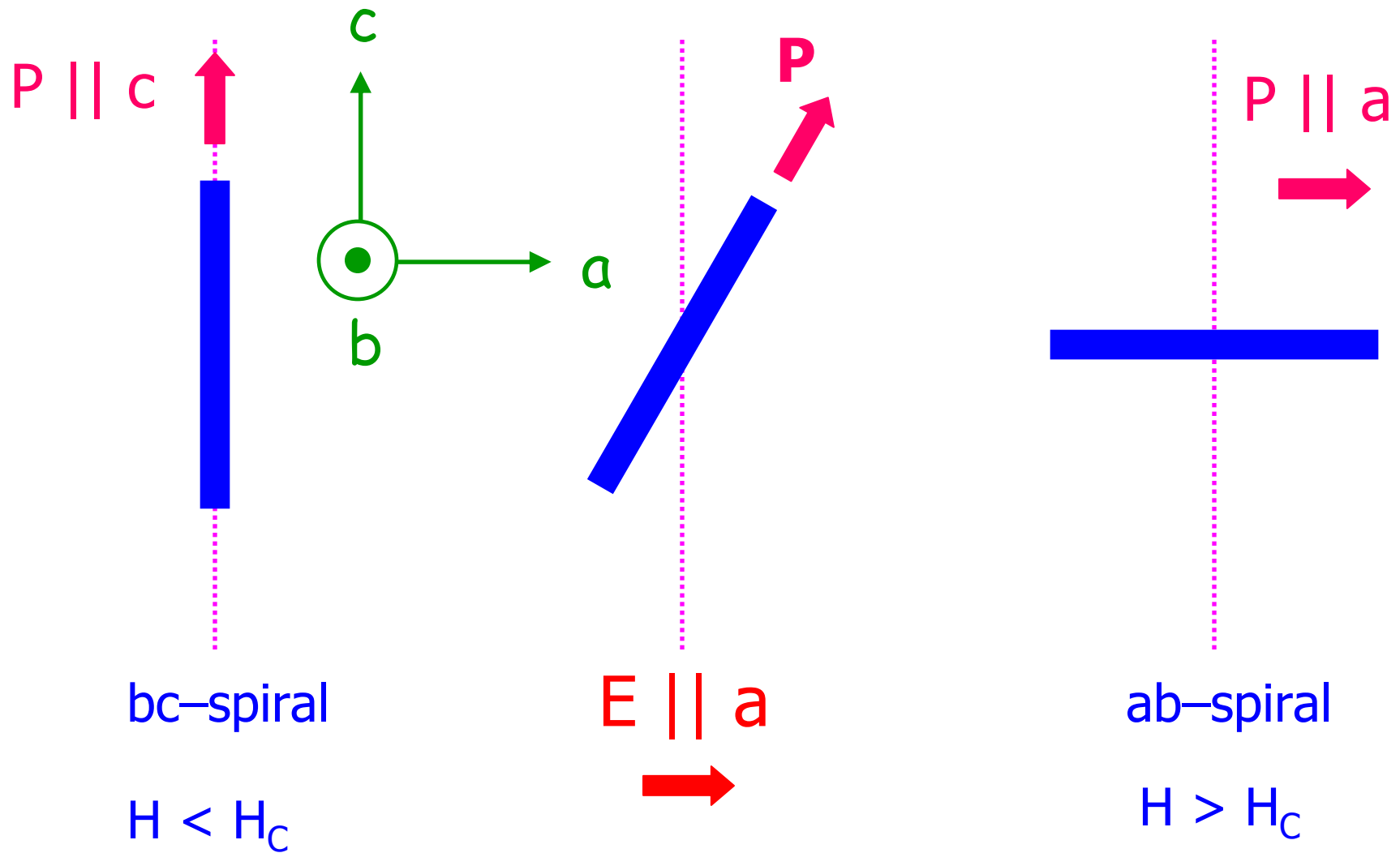


H. Katsura et al. (2006)

Magnons in spiral state of RMnO_3

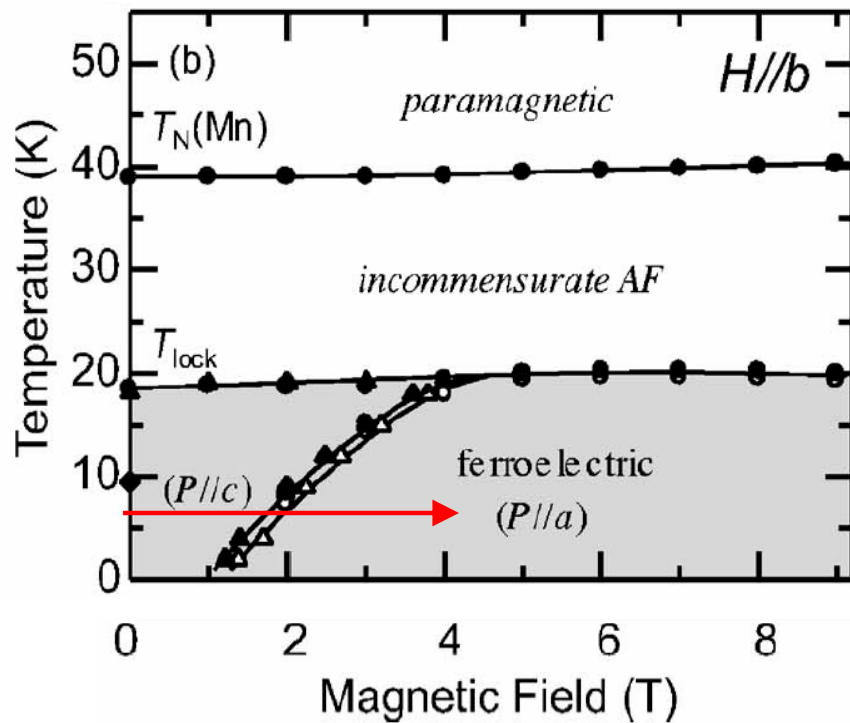


Electromagnon mode

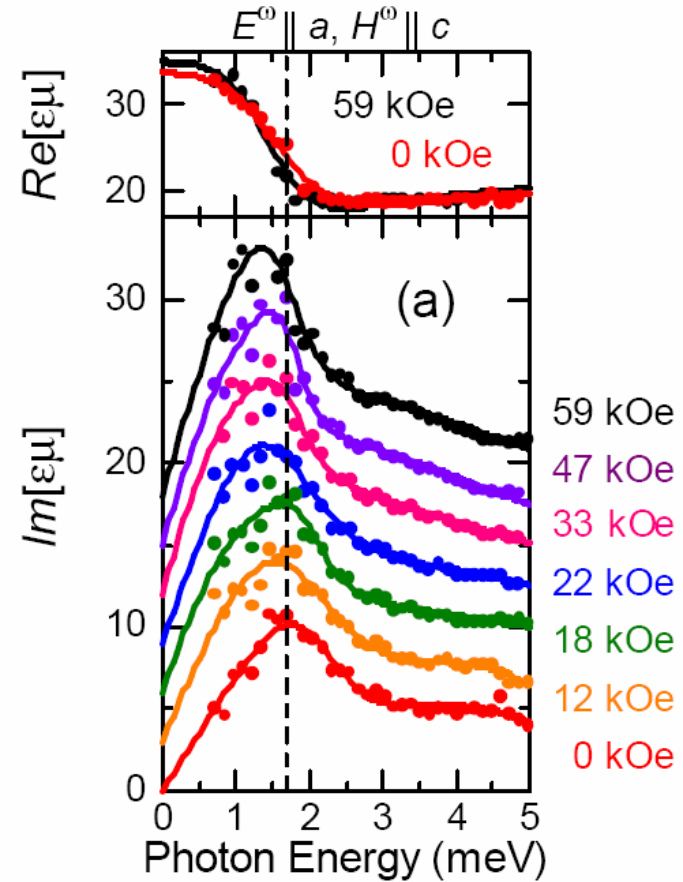


Softening at $H_c \sim$ divergent ϵ_a

Magnetic field dependence



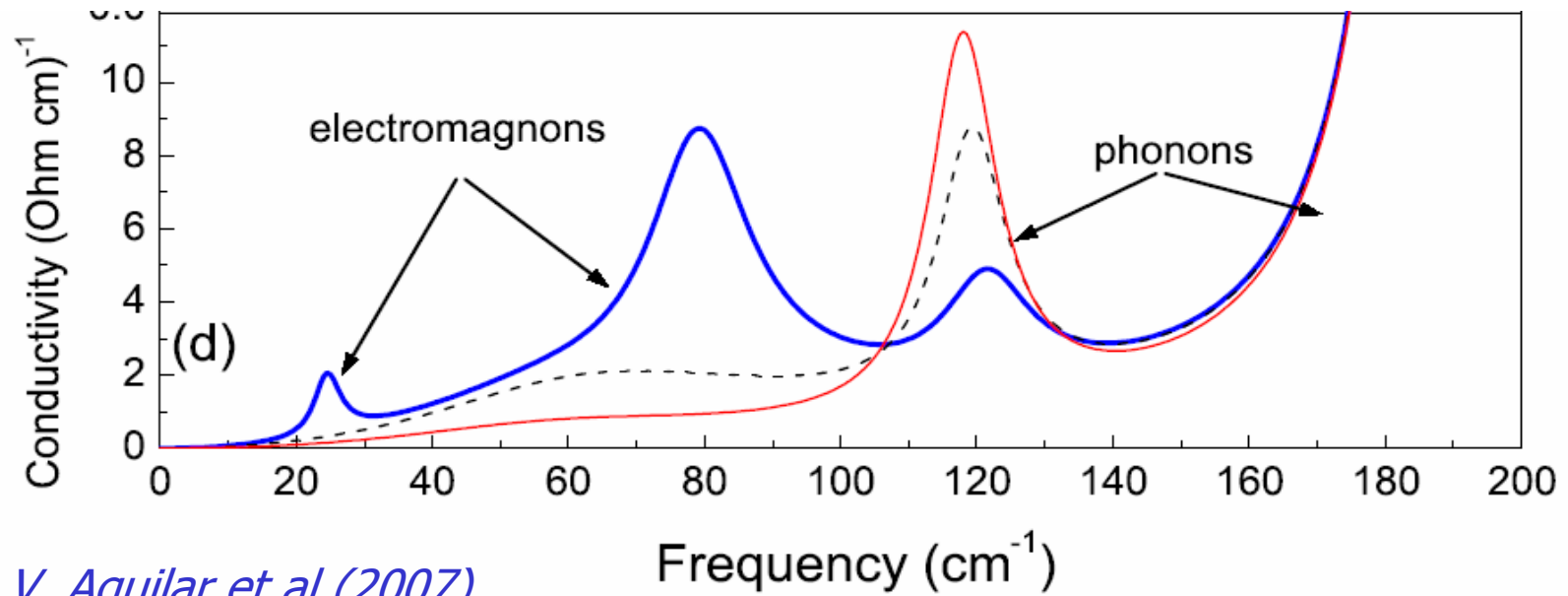
T. Kimura et al (2005)



N. Kida et al (2008)



ab spiral $E \parallel a$



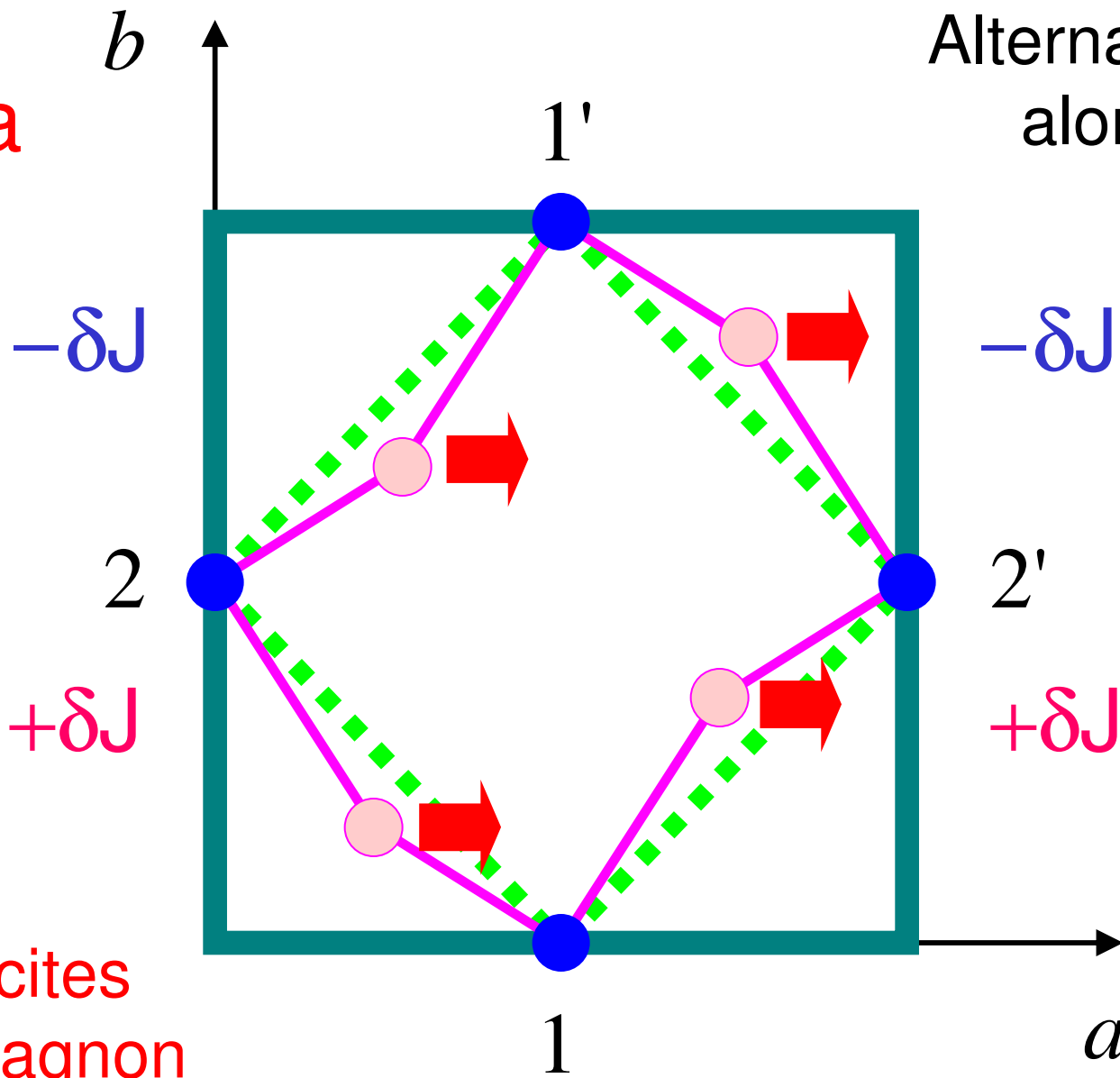
R. V. Aguilar et al (2007)

RMnO₃

$Q \parallel b$

$E \parallel a$

Alternation of J
along Q



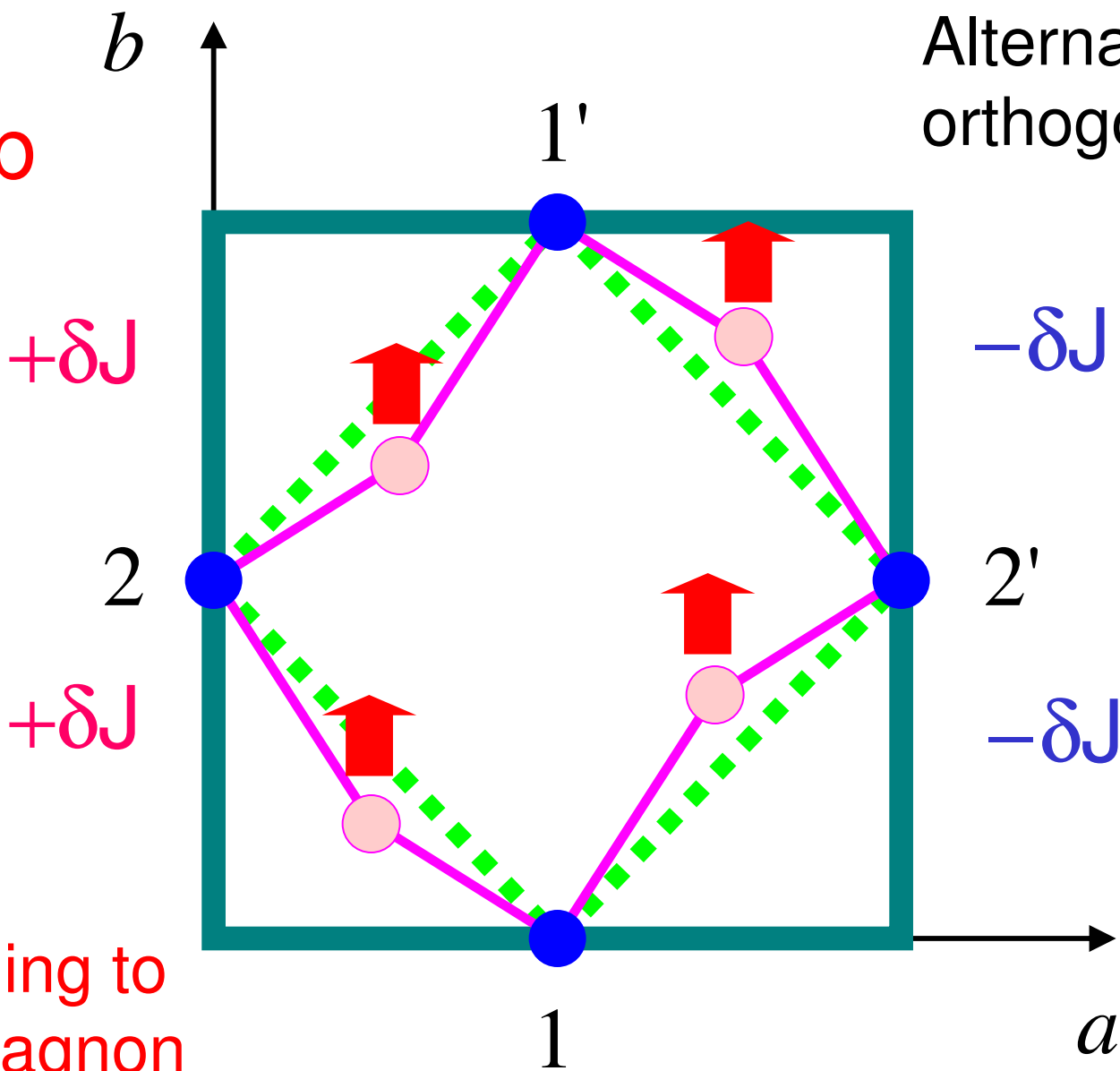
$E \parallel a$ excites
single magnon

RMnO₃

$Q \parallel b$

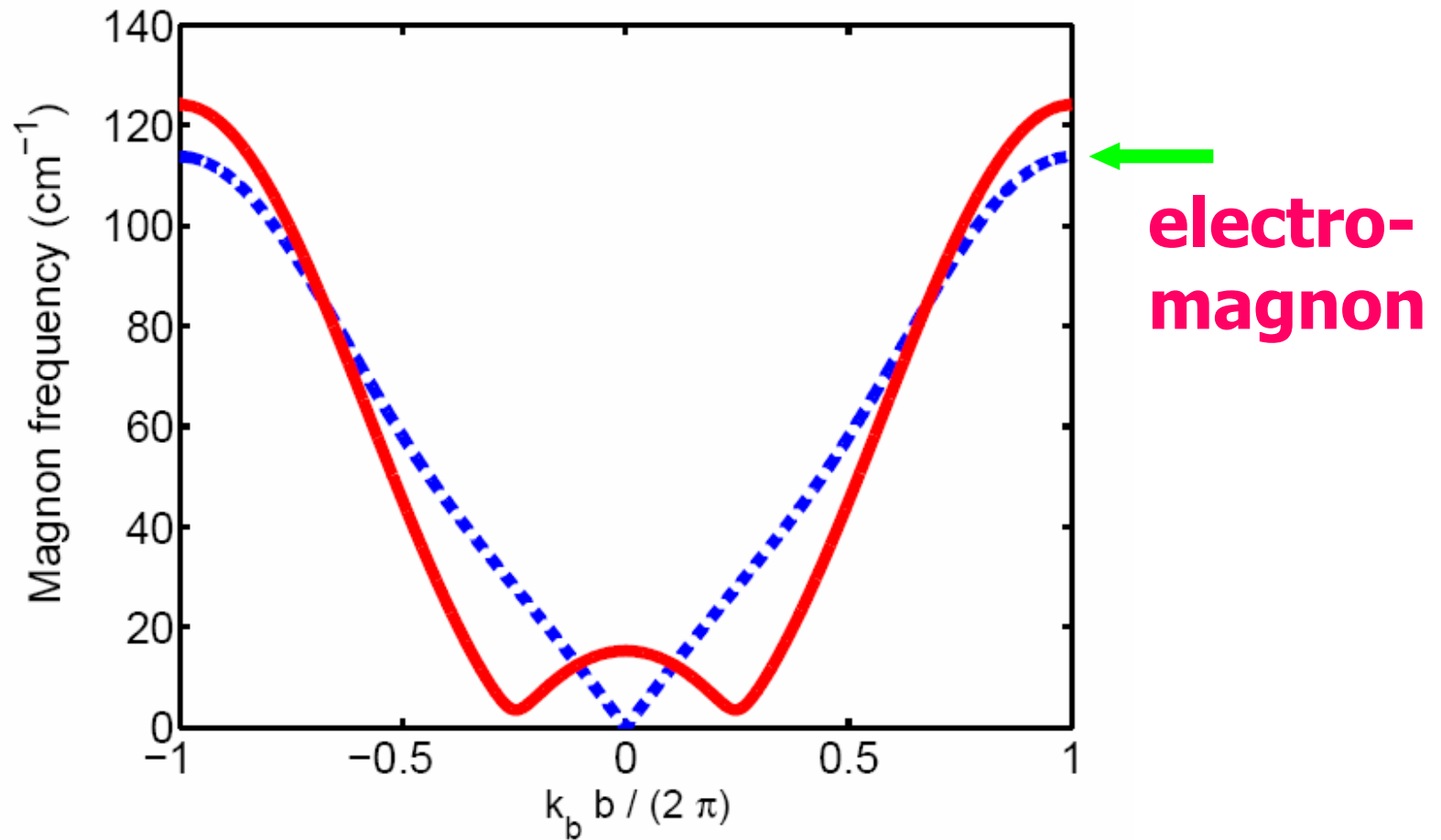
$E \parallel b$

Alternation of J
orthogonal to Q



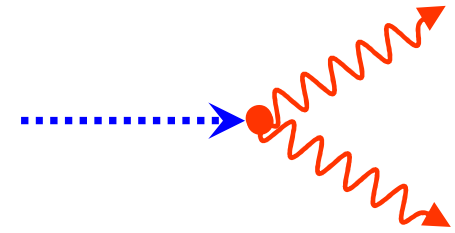
no coupling to
single magnon

Magnons in spiral state of RMnO₃

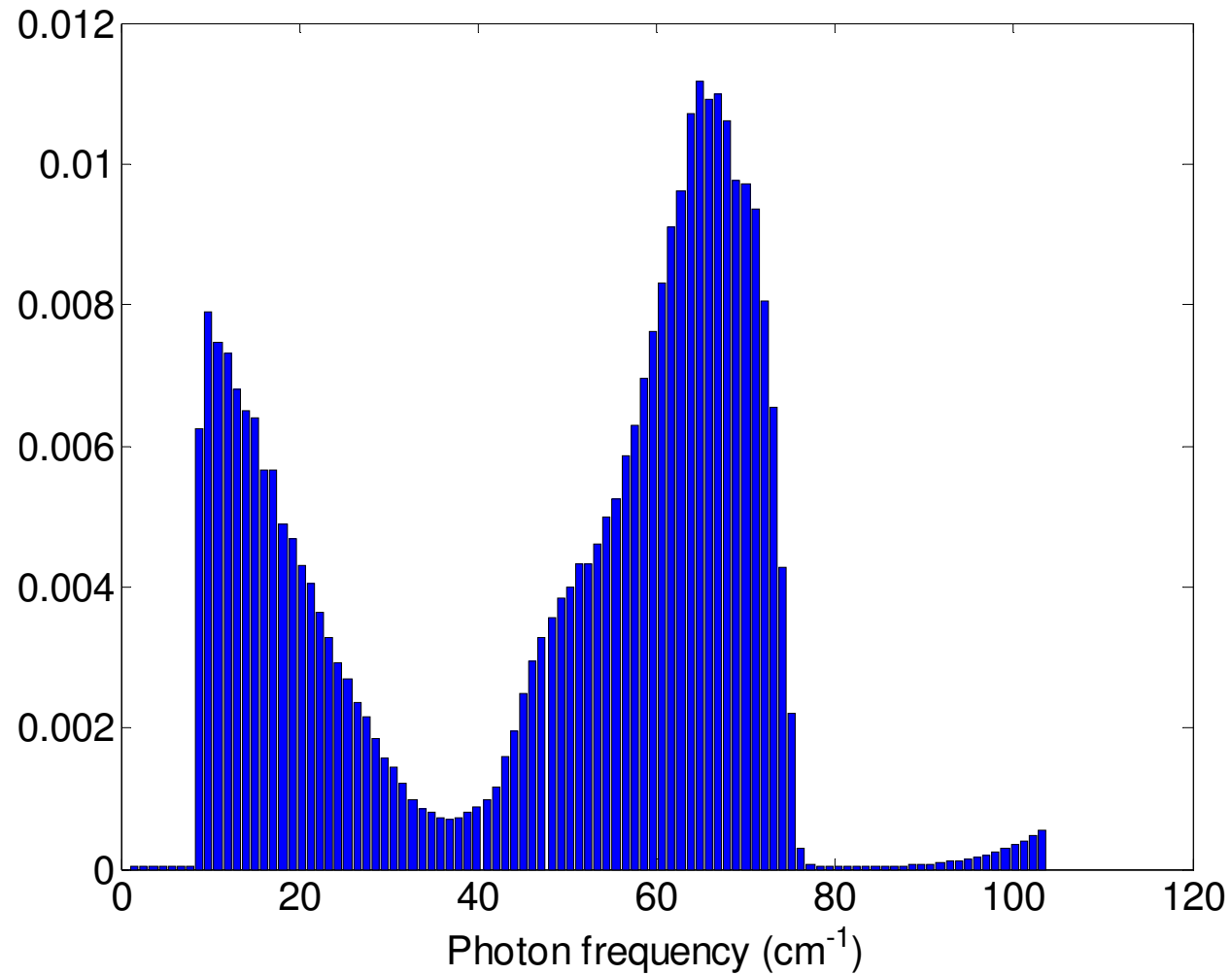


Bi-magnon continuum

e l a



χ''



Conclusions

- Magnetic frustration gives rise to unusual spin orders that break inversion symmetry and give rise to multiferroic behavior and linear magnetoelectric effect