Unidirectional light propagation in multiferroics and multi-antiferroics

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TRR 80

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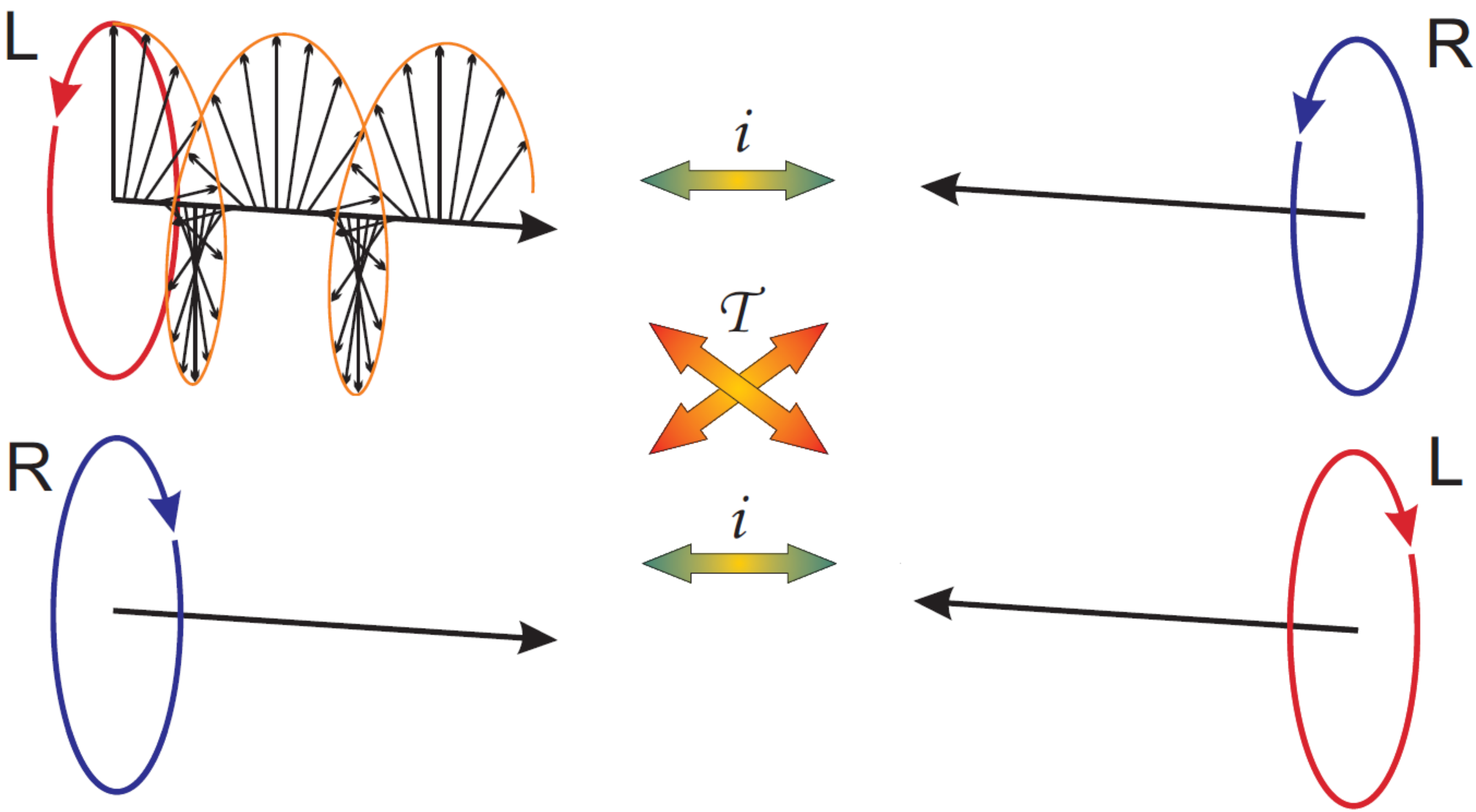
Dr. T. Rõõm, Dr. U. Nagel, NICPB, Estonia

Prof. J. Hemberger, Prof. M. Grüninger, University of Cologne, Germany

Dr. K. Penc, Dr. J. Romhányi, Wigner Res. Inst. for Physics, Hungary

Dr. R. Fishman, Oak Ridge National Lab, USA

Quadrochroism

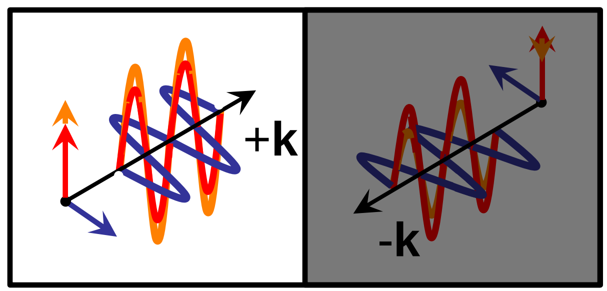


Outline

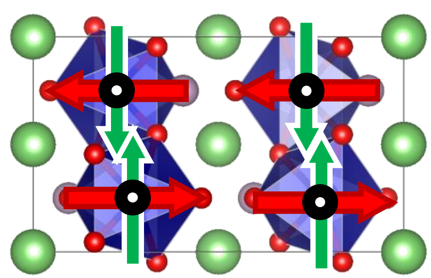
Static & optical magnetoelectric effects in multiferroics

$$\begin{bmatrix} D \\ B \end{bmatrix} = \begin{bmatrix} \hat{\varepsilon} & \hat{\chi}^{em} \\ \hat{\chi}^{me} & \hat{\mu} \end{bmatrix} \begin{bmatrix} E \\ H \end{bmatrix}$$

Quadrochroism & one-way trasparency via the optical magnetoelectric effect

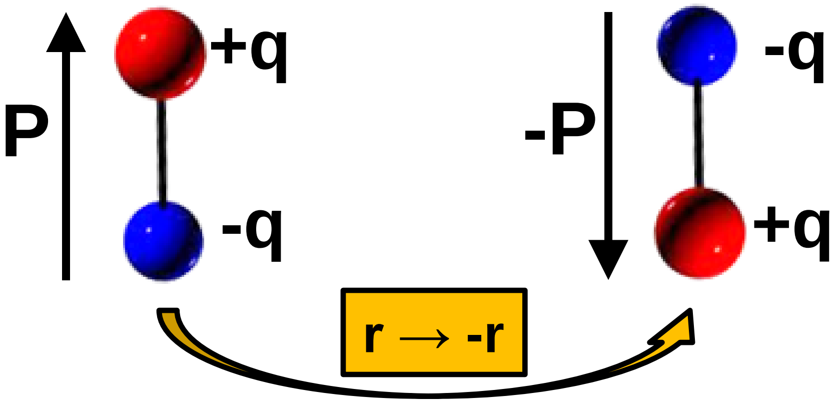


Target compounds: $Ba_2CoGe_2O_7$, $LiCoPO_4$

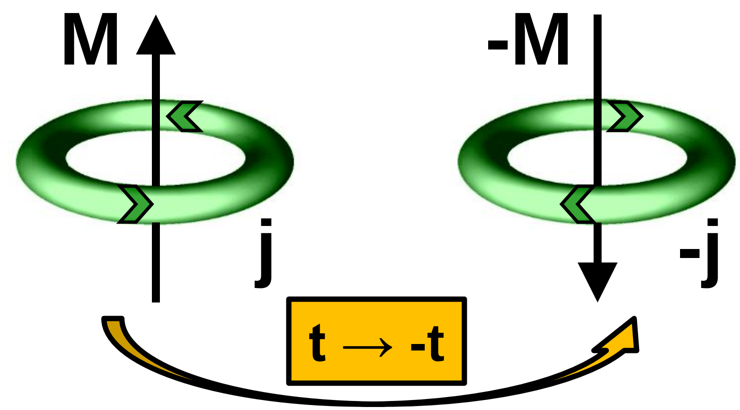


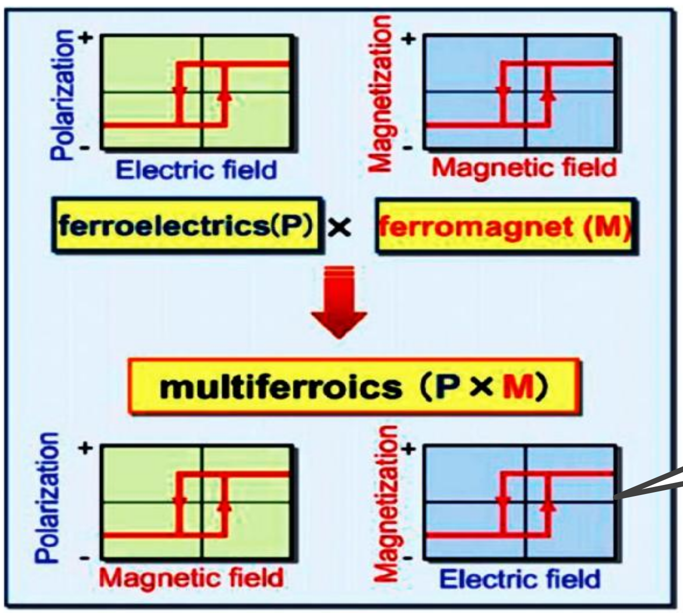
Multiferroics & magnetoelectric effect

Ferroelectricity



Ferromagnetism



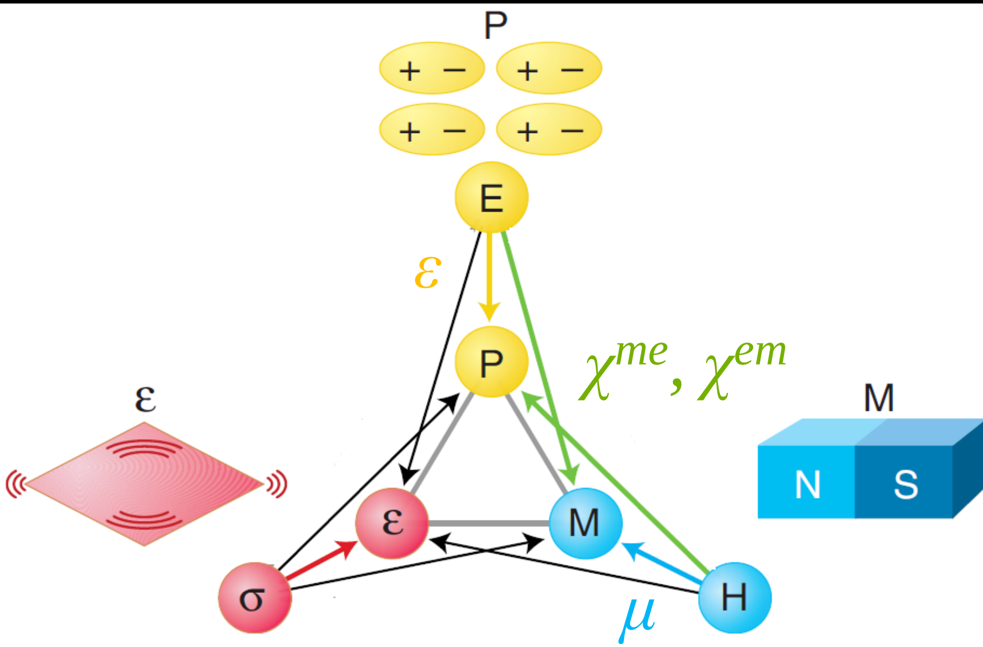


Holy Grail of the field:
Magnetoelectric
memory devices

„Materials should exist, which can be polarized by a magnetic field and magnetized via an electric field."

P. Curie, Journal de Physique **3**, 393 (1894)

Multiferroics & magnetoelectric effect



Spaldin and Fiebig, Science (2005)

Generalized constituve relations

$$\begin{bmatrix} D \\ B \end{bmatrix} = \begin{bmatrix} \hat{\varepsilon} & \hat{\chi}^{em} \\ \hat{\chi}^{me} & \hat{\mu} \end{bmatrix} \begin{bmatrix} E \\ H \end{bmatrix}$$

$$\chi_{ij}^{me}(\omega) = \frac{2}{\hbar N V} \sum_n \frac{\overbrace{\omega_{no}\Re\{\langle 0|M_i|n\rangle\langle n|P_j|0\rangle\}}^{\chi'_{ij}(\omega)} + \overbrace{i\omega\Im\{\langle 0|M_i|n\rangle\langle n|P_j|0\rangle\}}^{\chi''_{ij}(\omega)}}{\omega_{no}^2 - \omega^2 - 2i\omega\delta}$$

$\chi'_{ij}(\omega)$
- inversion (I) odd
- time reversal (T) odd
- static magnetoelectric effect
- directional anisotropy

$\chi''_{ij}(\omega)$
- inversion (I) odd
- time reversal (T) even
- vanishes in the static limit
- natural optical activity

L.D. Barron, *Molecular light scattering and optical activity* (Cambridge, 2004)

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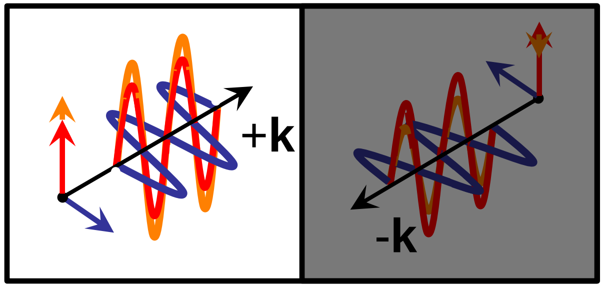

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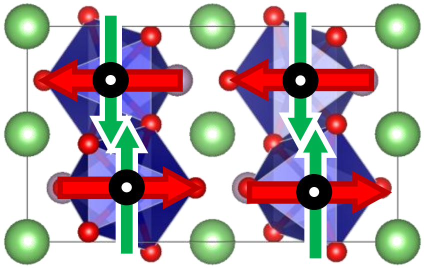
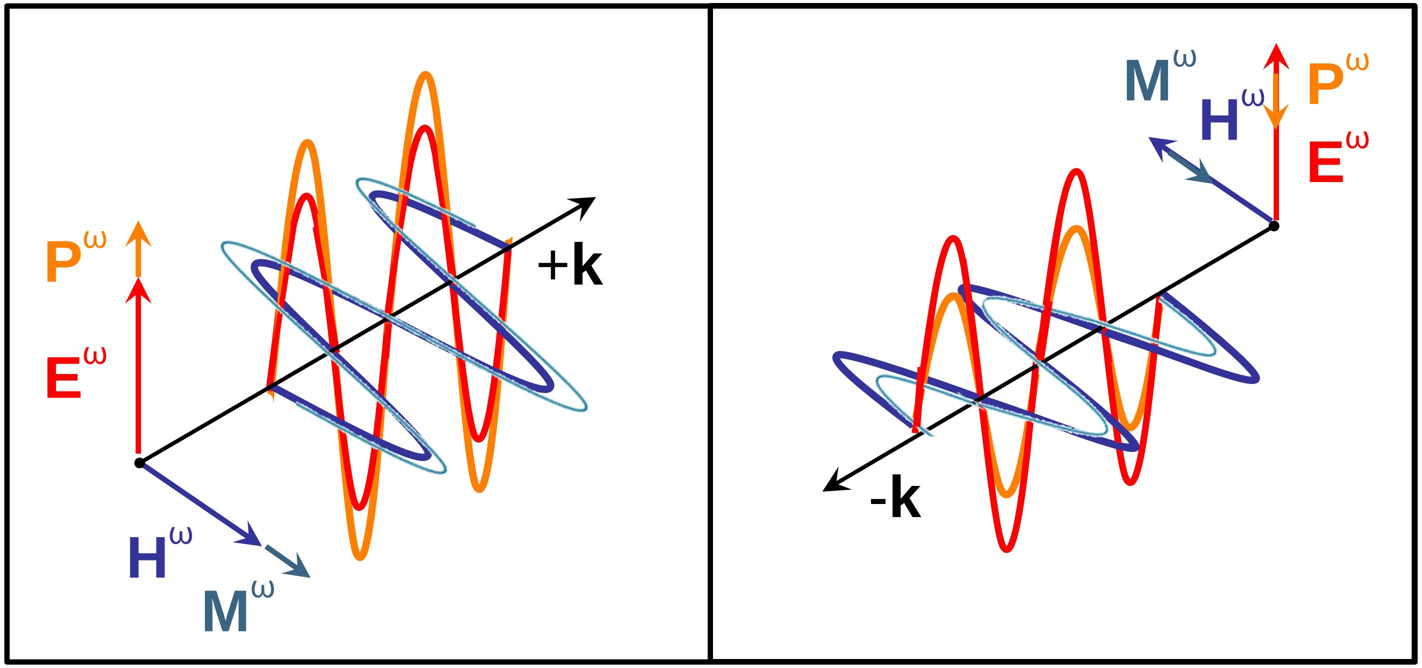


Optical magnetoelectric effect: Four-coloured optics

$$\chi'_{ji}(\omega)$$
$$\varepsilon = 1$$
$$\mu = 1$$



$$N^{\pm}(\omega) \approx \sqrt{\epsilon_{ii}(\omega)\mu_{jj}(\omega)} \pm \underbrace{\tfrac{1}{2}[\chi^{\mathrm{me}}_{ji}(\omega) + \chi^{\mathrm{em}}_{ij}(\omega)]}_{\chi'_{ji}(\omega)}$$

- different refractive indices for ±**k** propagation and two polarizations, termed as quadrochroism
- directional (±**k**) optical anisotropy is generally weak, $\Delta N/N \sim 10^{-2}$-10^{-6} Rikken, Nature (1997)
- BUT can be strong in multiferroics!

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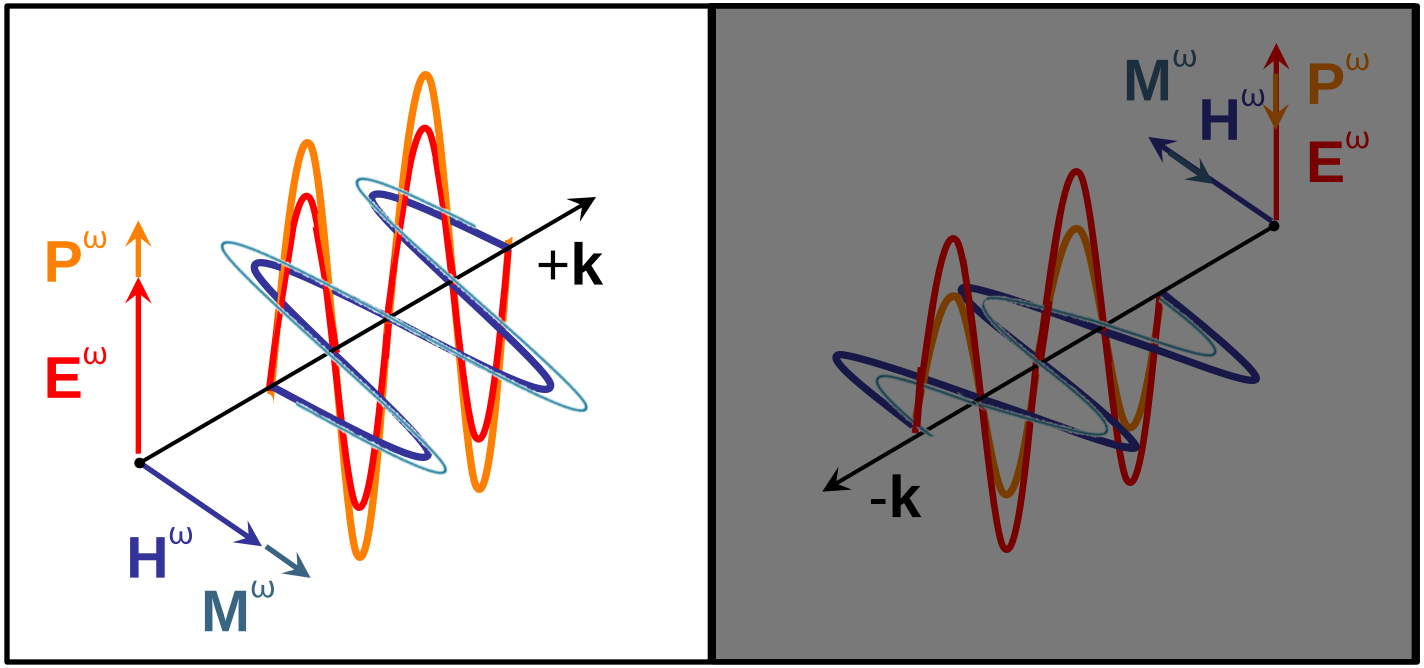

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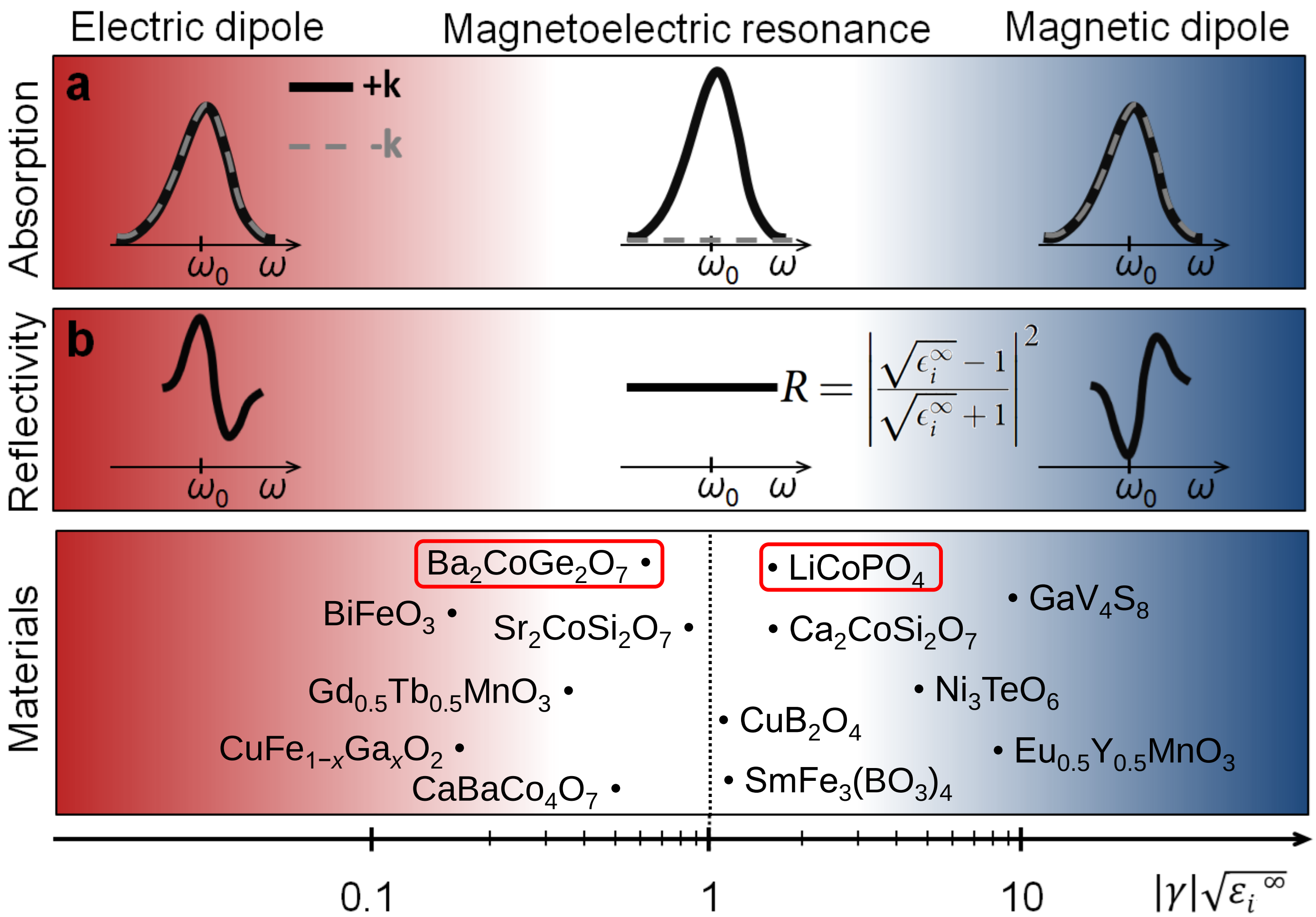
Optical magnetoelectric effect: One-way transparency

Condition for one-way transparency: $\left|\frac{\langle n|\, M_j\, |0\rangle}{\langle n|P_i|0\rangle}\right| \triangleq |\gamma| = \frac{1}{\sqrt{\epsilon_i^\infty}}$ [CGS]

Kézsmárki, NatCommun (2014)



$Ba_2CoGe_2O_7$
Kézsmárki, PRL (2011)
Bordács, NatPhys (2012)

$Eu_{0.5}Y_{0.5}MnO_3$
Takahashi, NatPhys (2012)

$Gd_{0.5}Tb_{0.5}MnO_3$
Takahashi, PRL (2013)

$Sr_2CoSi_2O_7$ & $Ca_2CoSi_2O_7$
Szaller, PRB (2014)
Kézsmárki, NatCommun (2014)

$CuFe_{1-x}Ga_xO_2$
Kibayashi, NatCommun (2014)

$SmFe_3(BO_3)_4$
Pimenov, PRB (2015)

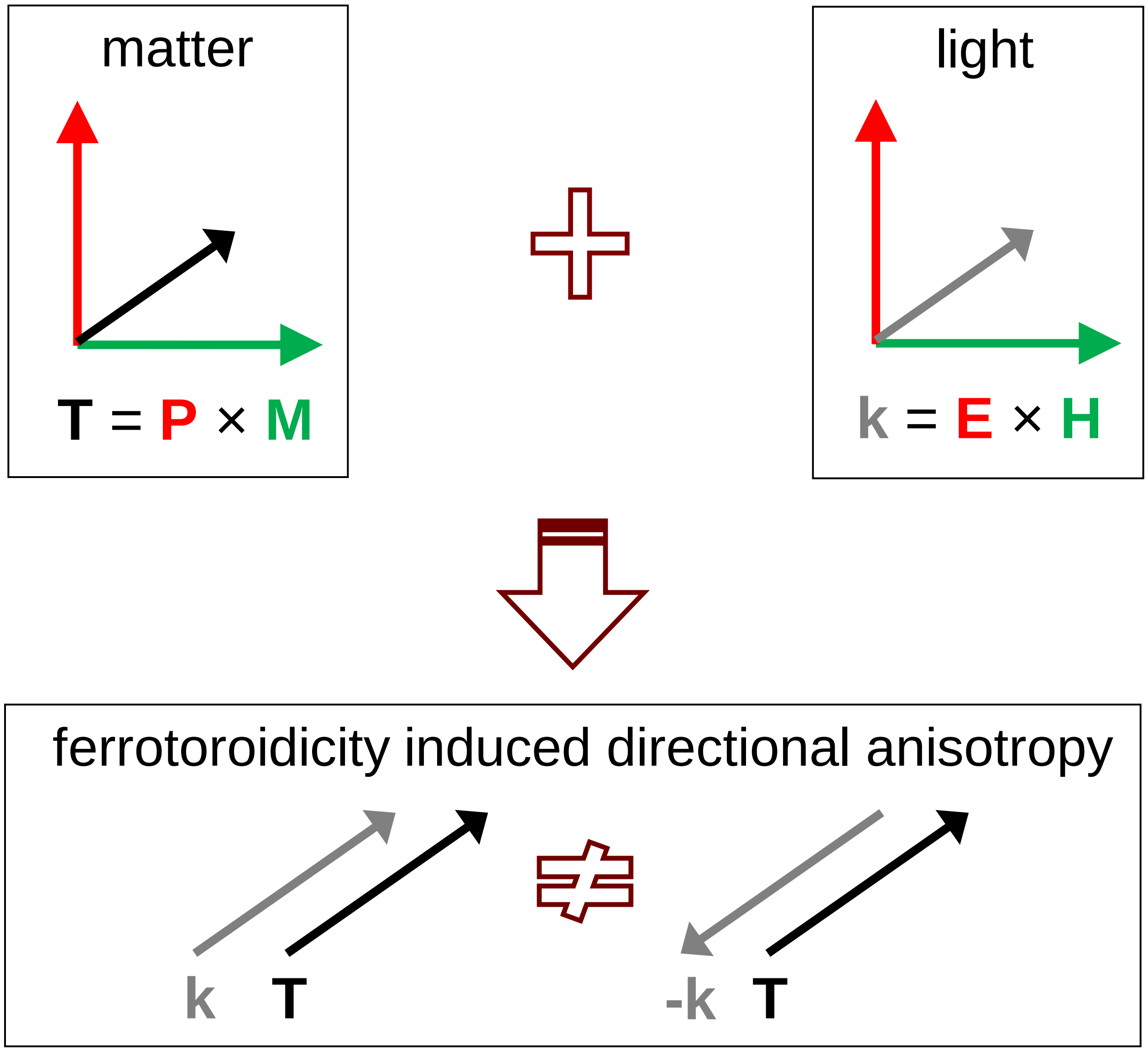
CuB_2O_4
Arima, PRL (2015)

$CaBaCo_4O_7$
Bordács, PRB (2015)

$BiFeO_3$
Kézsmárki, PRL (2015)

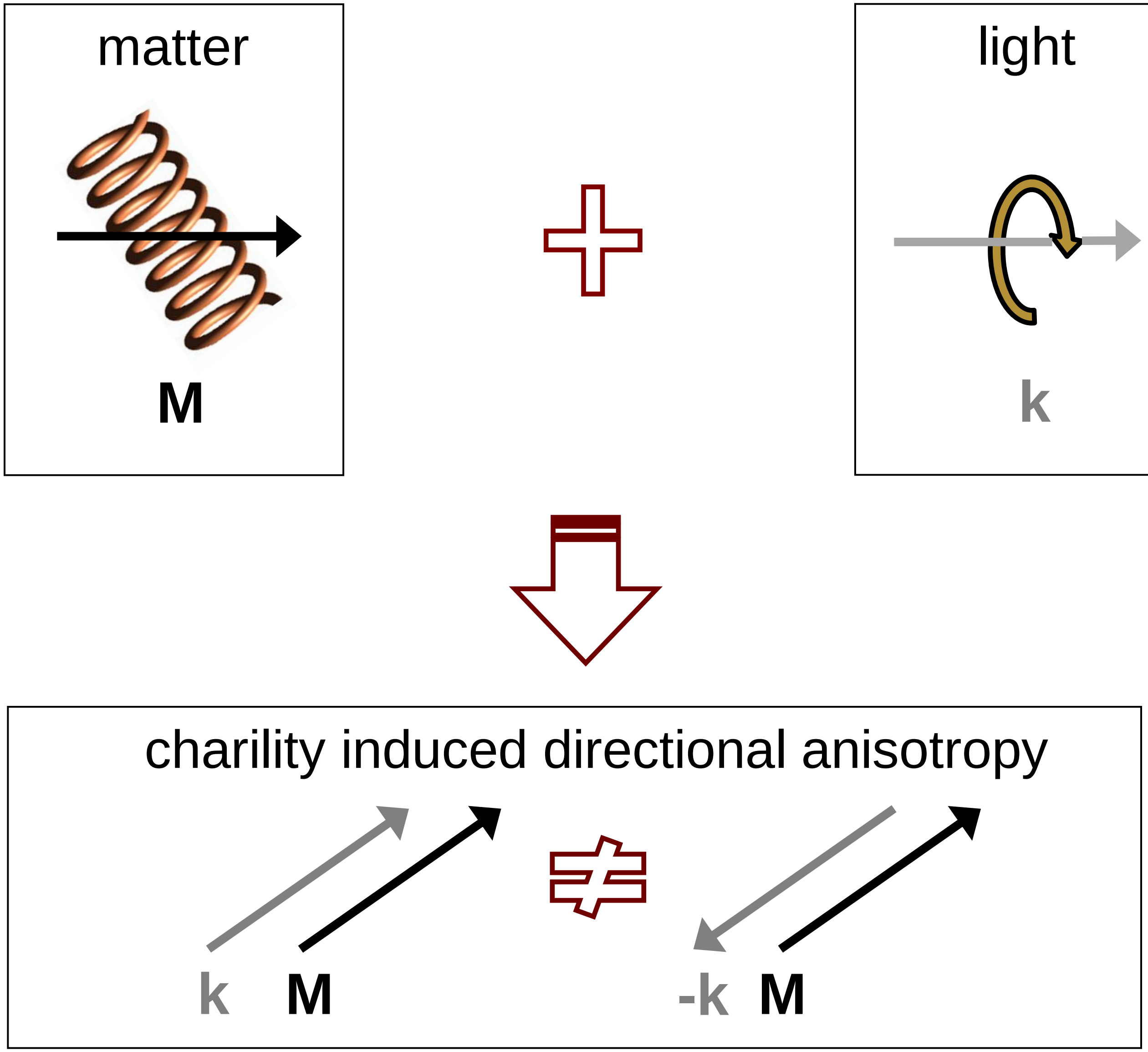
$SmFe_3(BO_3)_4$
Kuzmenko, PRL (2018)

Optical magnetoelectric effect: One-way transparency



Kézsmárki, PRL (2011)

Optical magnetoelectric effect: One-way transparency



Bordács, NatPhys (2012)

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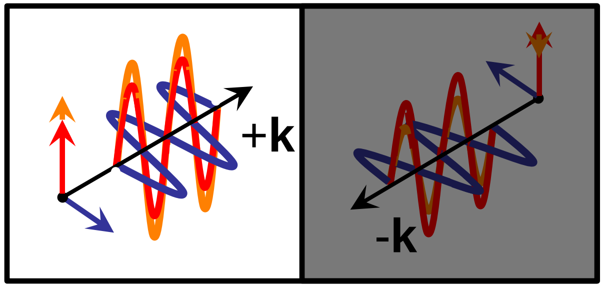

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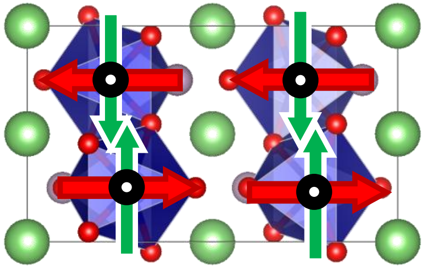
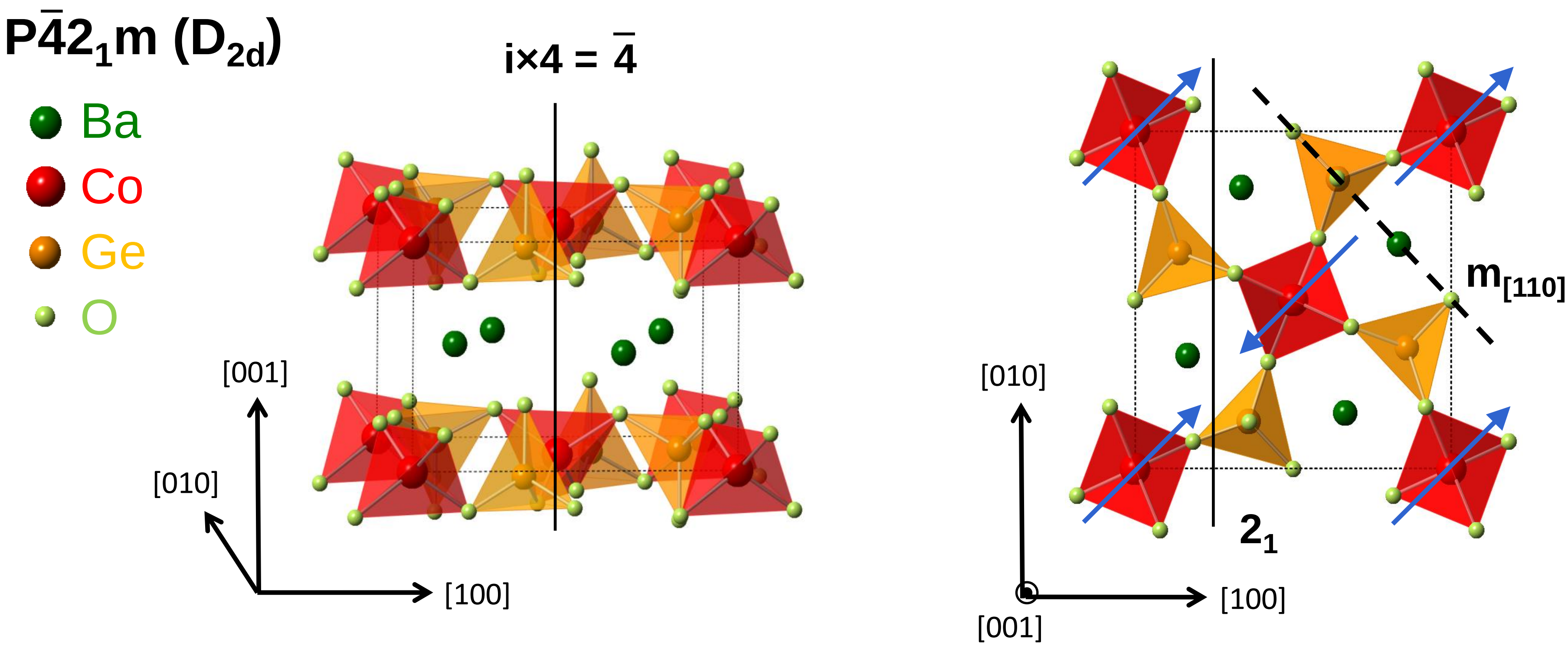
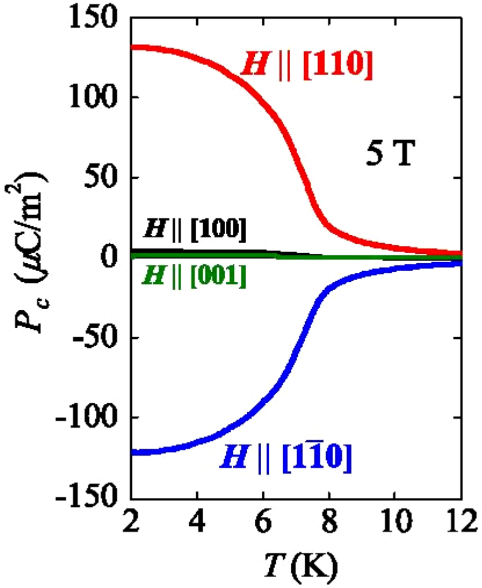


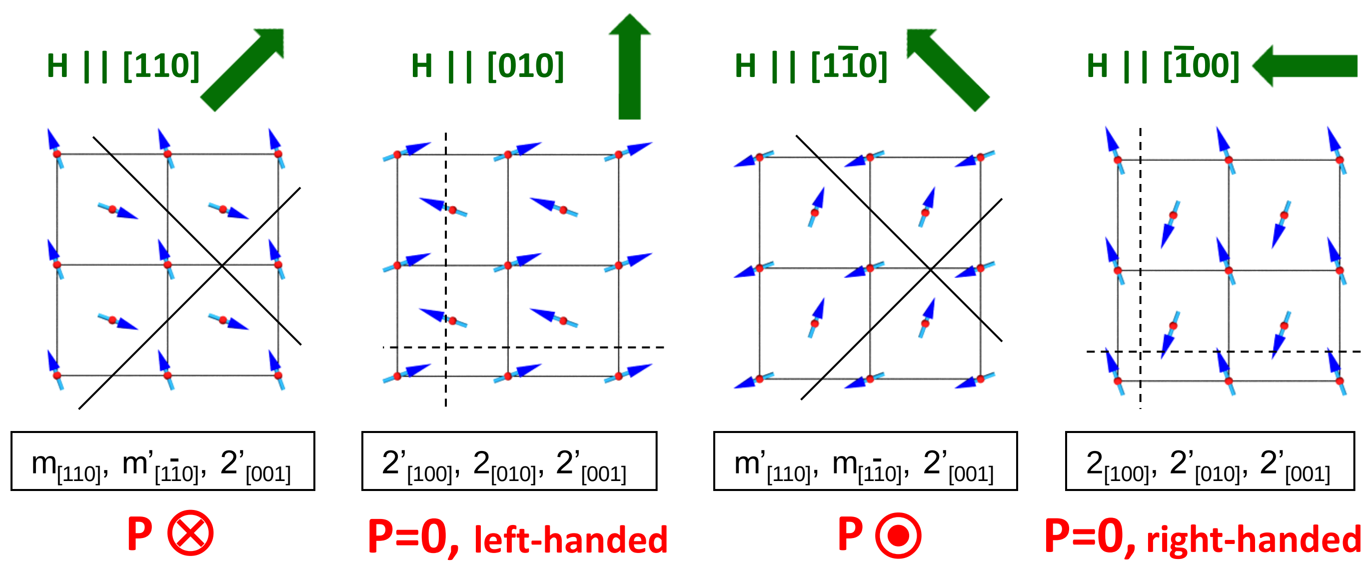
Multiferroic $Ba_2CoGe_2O_7$

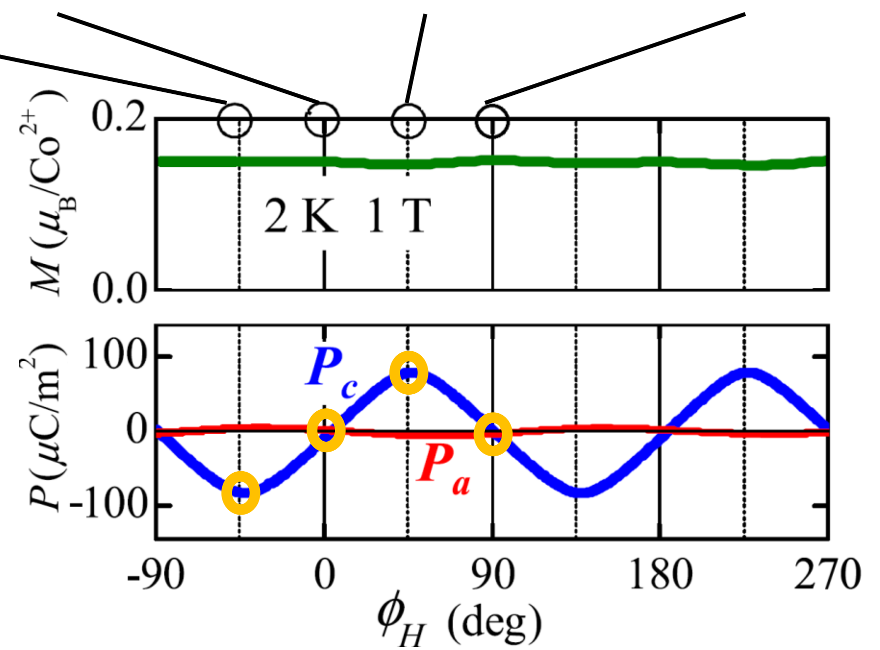


- Tetragonal noncentrosymmetric crystal structure   Hutanu, PRB (2011)
- Magetic Co^{2+} ions with S=3/2 in tetrahedral oxygen cages
- Easy-plane Néel antiferromagnet   Hutanu, PRB (2012)

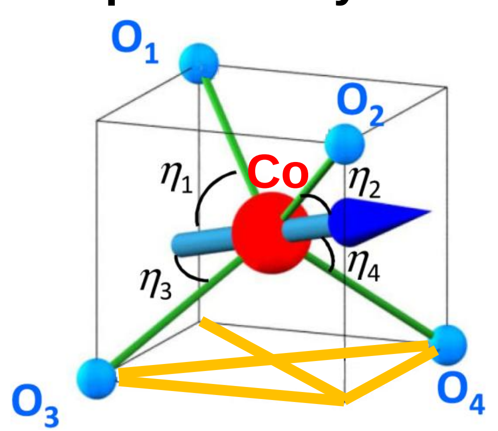
Multiferroic $Ba_2CoGe_2O_7$







Spin-dependent hybridization:

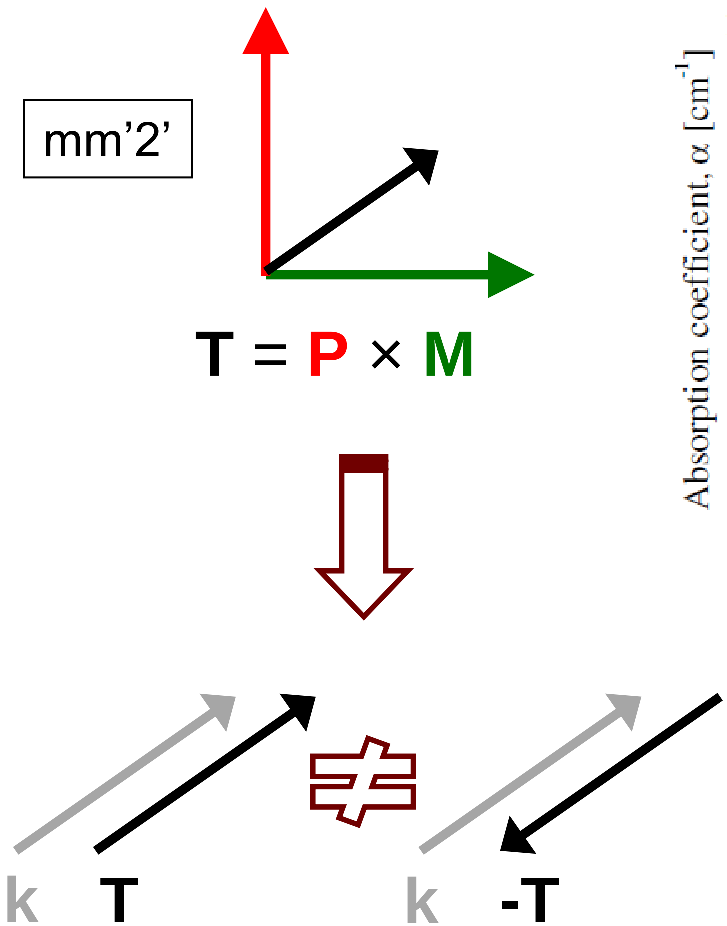


$$P \propto \sum_{i=1}^{4} (S \cdot e_i)^2 e_i \propto \sum_{i=1}^{4} (S \cos\eta_i)^2 e_i$$

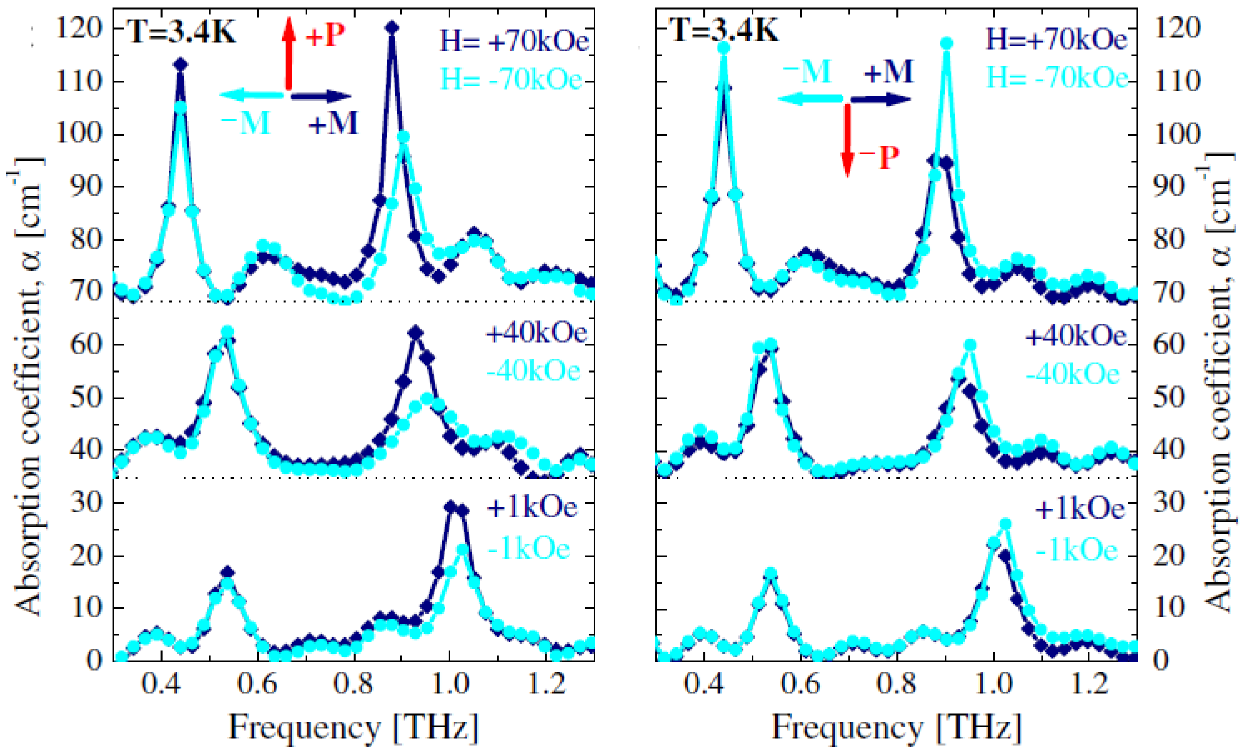
Arima, JPSJ (2007)

Murakawa, PRL (2010)

Multiferroic $Ba_2CoGe_2O_7$

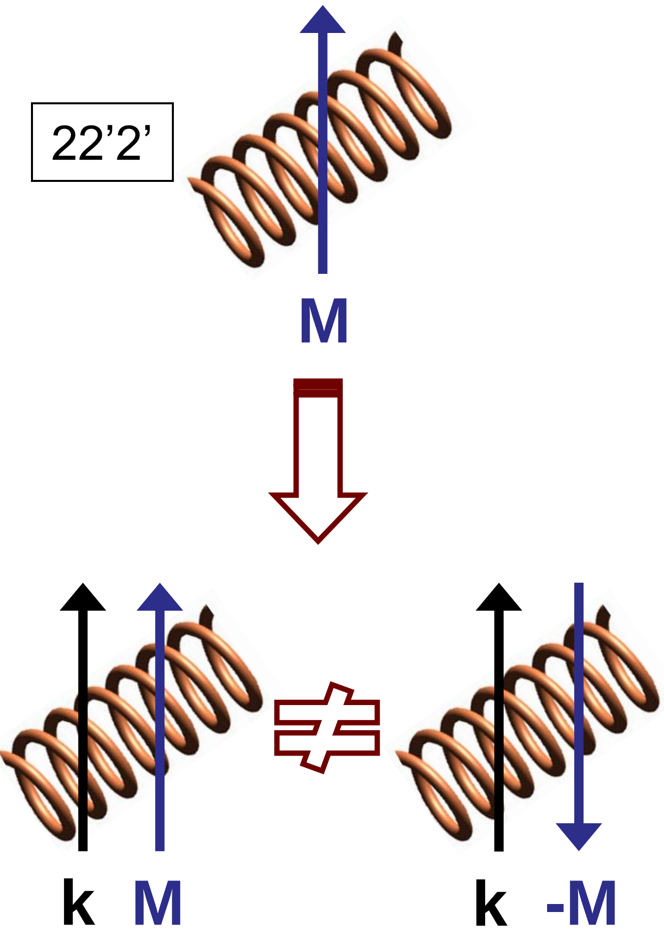


T-type DA

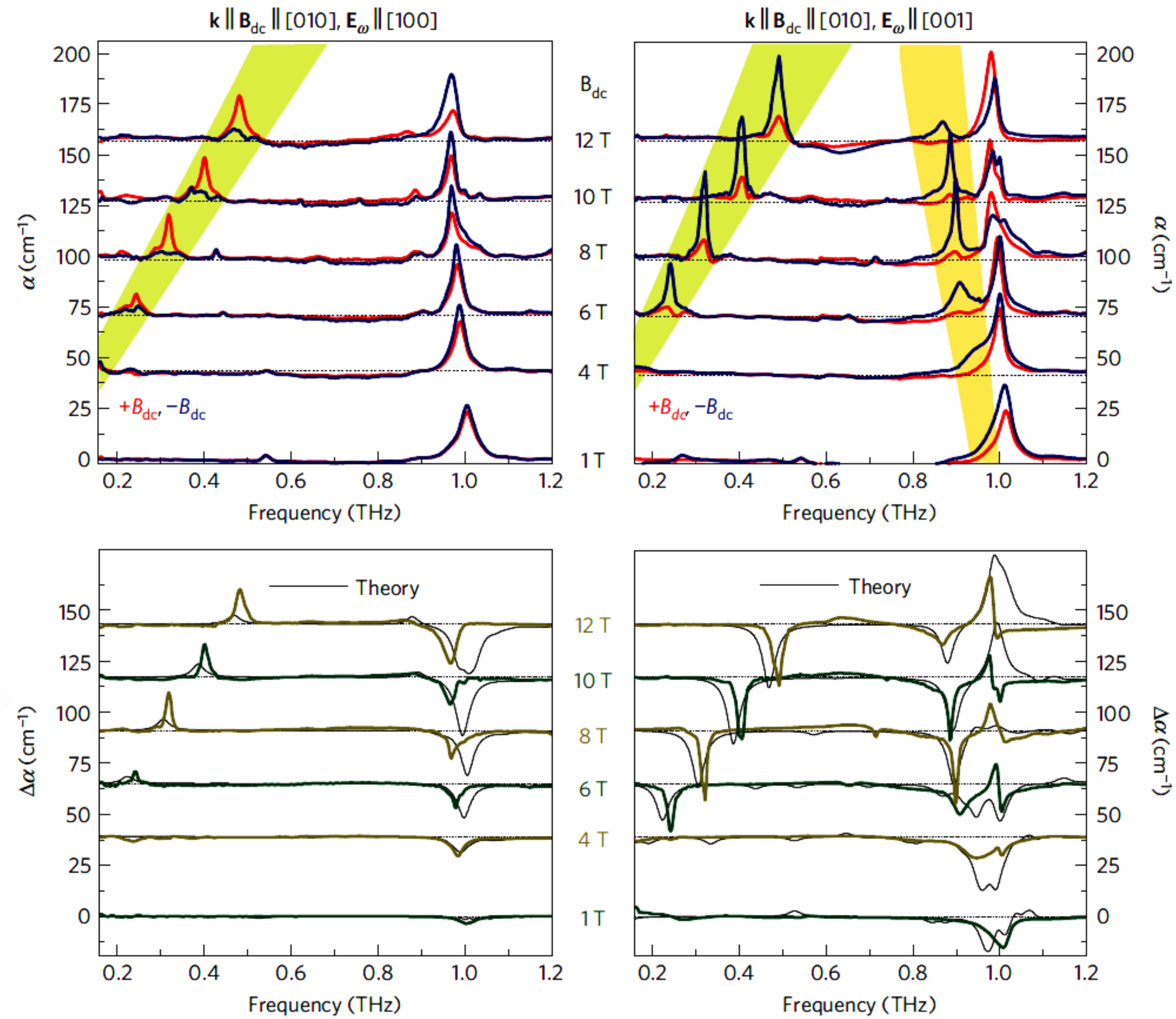


Kézsmárki, PRL (2011)

Multiferroic $Ba_2CoGe_2O_7$



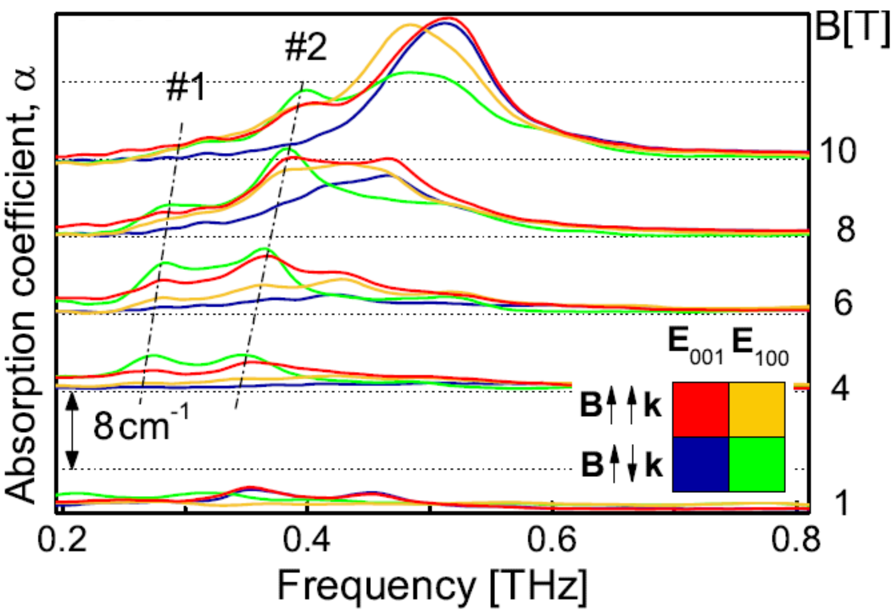
MC-type DA



Bordács, NatPhys (2012)

Multiferroic $Ba_2CoGe_2O_7$

$Ca_2CoSi_2O_7$



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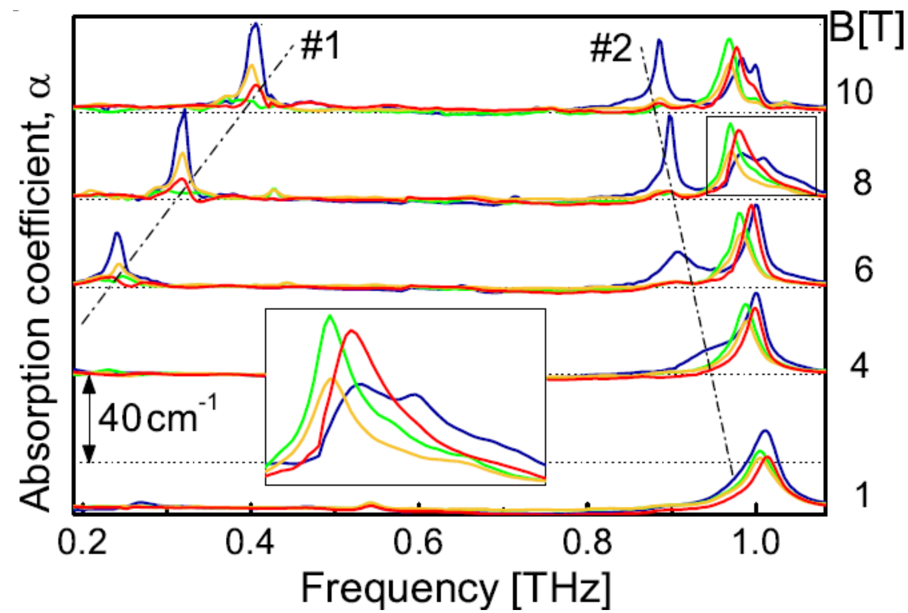


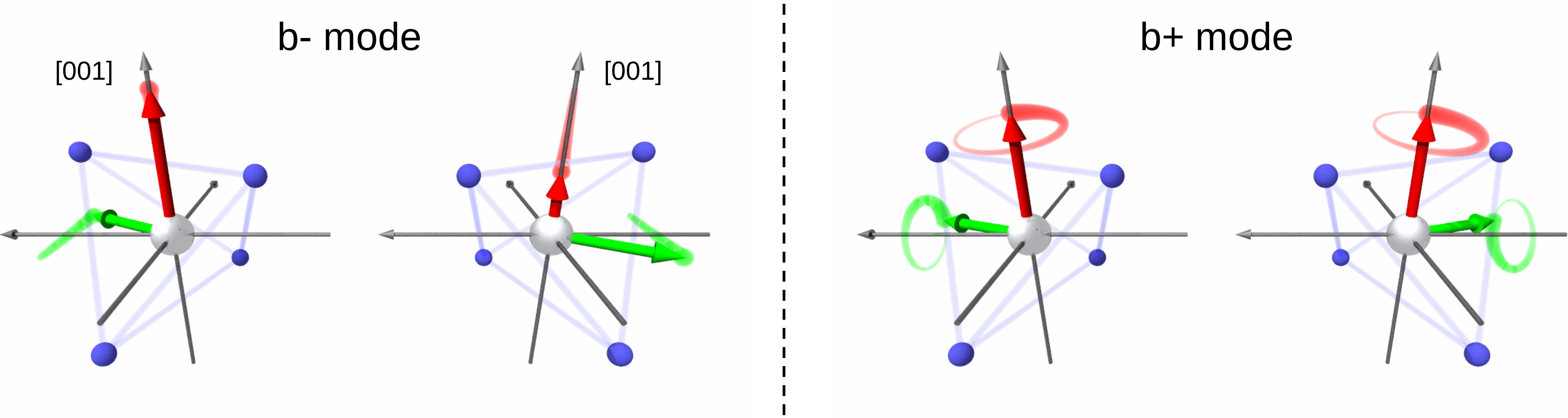
- Four different values of the refractive index for a given axis of propagation: forward and backward (±k) propagation and two orthogonal polarizations,
- Magnons with nearly optimal magnetoelectric ratio, $|\gamma| = \frac{c}{\sqrt{\epsilon_i^\infty}}$,
- Connection with dc ME effect: $\chi_{ij}^{me}(0) = \frac{c}{2\pi} \cdot \int_0^\infty \frac{\Delta\alpha(\omega)}{\omega^2} \mathrm{d}\omega \quad \Leftarrow \quad \Re\chi(\omega) = \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty} \frac{\Im\chi(\omega')}{\omega' - \omega}\mathrm{d}\omega'$

Kézsmárki, NatCommun (2014)

Multiferroic $Ba_2CoGe_2O_7$

S=3/2 spin: $$\mathcal{H} = J \sum_{\langle i,j \rangle} \left(S_i^x S_j^x + S_i^y S_j^y\right) + J_z \sum_{\langle i,j \rangle} S_i^z S_j^z + \sum_i \Lambda \left(S_i^z\right)^2$$

J, J_z exchange interaction — Λ single-ion anisotropy



- b- mode is the Goldstone mode (ω=0) ↔ dc magnetoelectric effect
- b+ would be the other Goldstone mode in the lack of magnetic anisotropy

Penc, PRL (2012)

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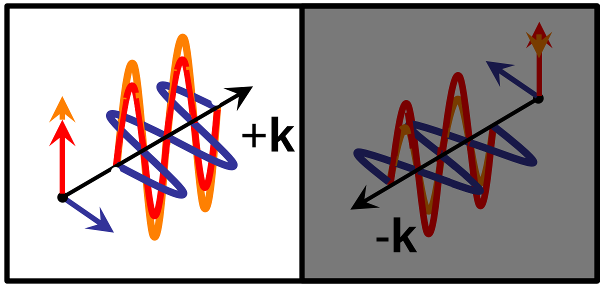

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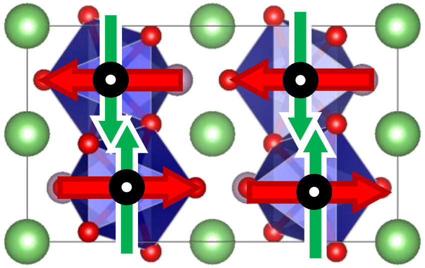
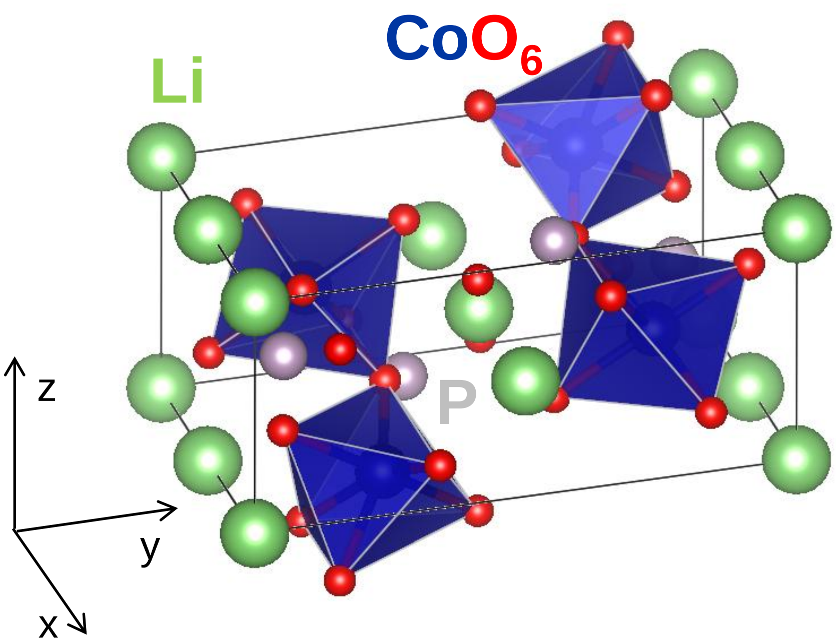
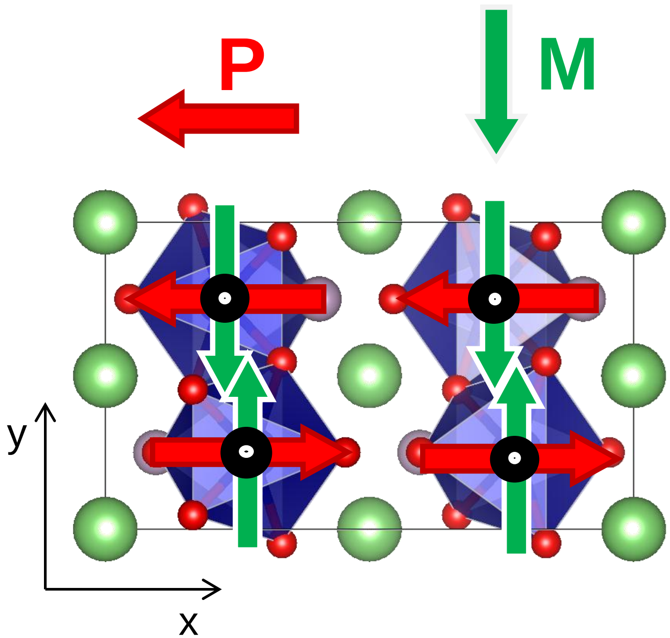


Multi-antiferroic $LiCoPO_4$



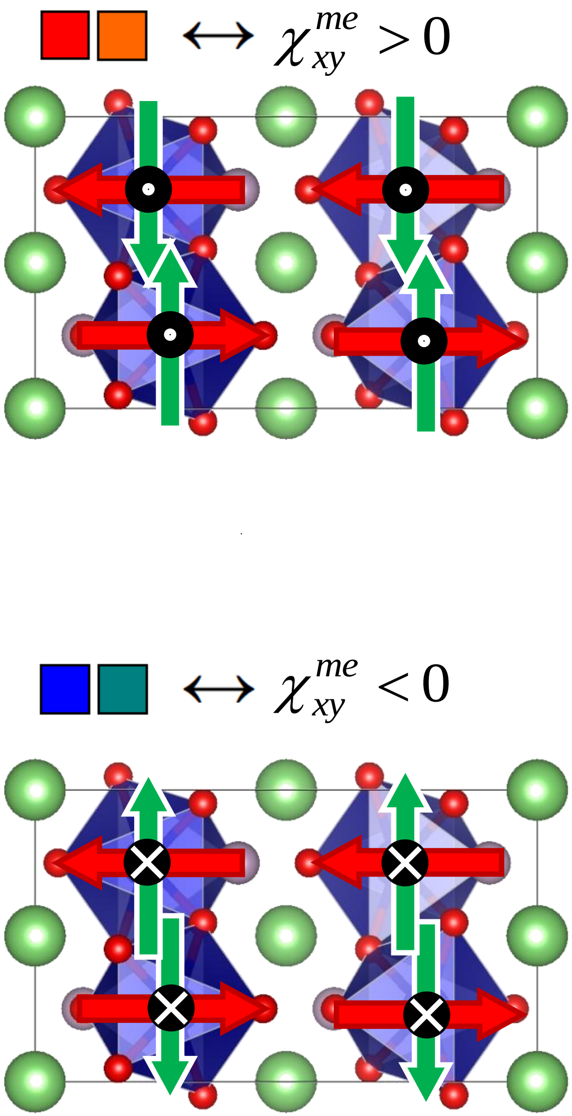
- orthorhombic Pmna (point group: mmm)
- distorted chessboard layers of **CoO_6** octahedra
- highly distorted **CoO_6** octahedra $\Rightarrow$
- **antiferroelectricity** (along x)



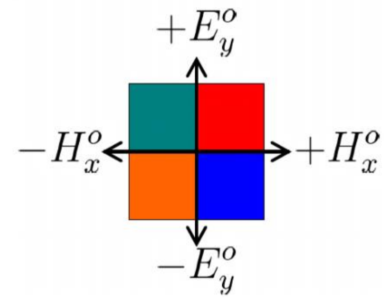
- magnetic order develops below T_N=21K
- **antiferromagnetism** (along y)
- orthorhombic Pmna' (point group: mmm')

antiferroelectricity$_x$ × **antiferromagnetism$_y$** $\Rightarrow \pm \chi_{xy}^{me}$

Multi-antiferroic $LiCoPO_4$

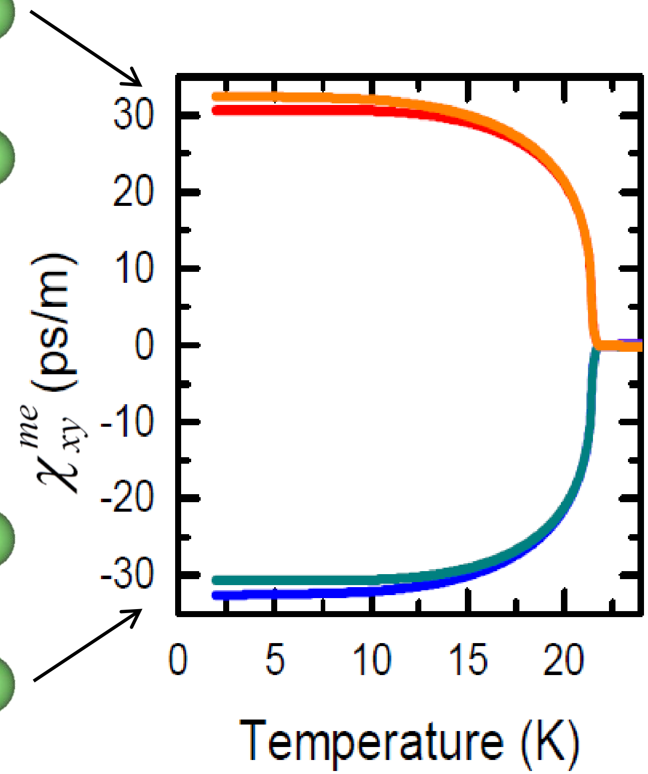


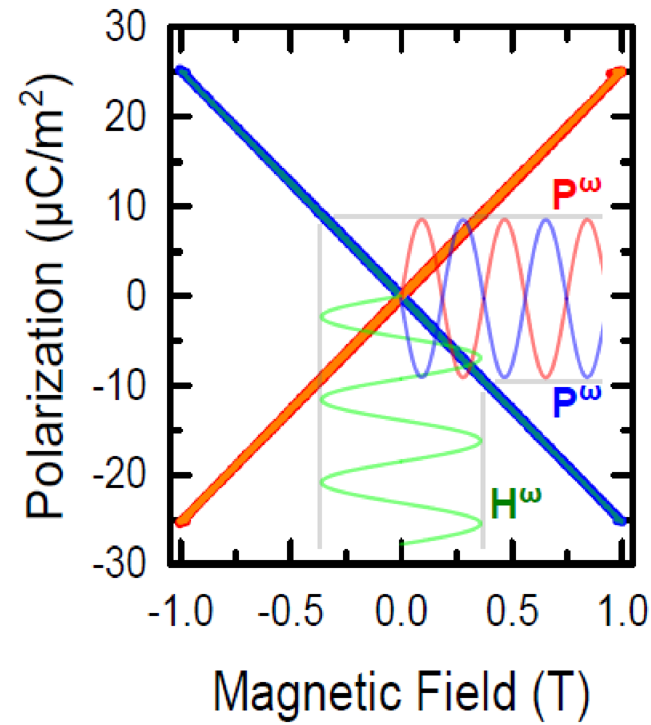
Magnetoelectric poling:



E^0=2 kV/cm

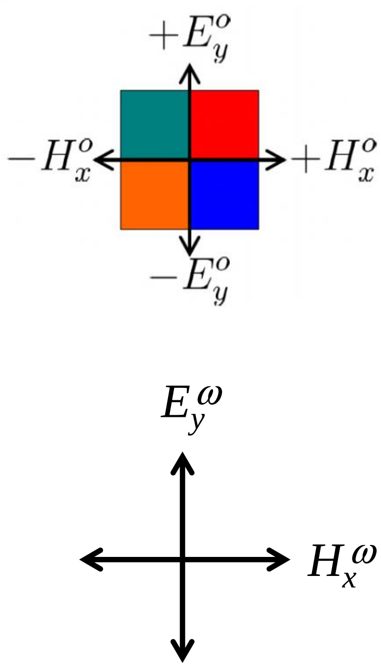
$\mu_0 H^0$=1 T

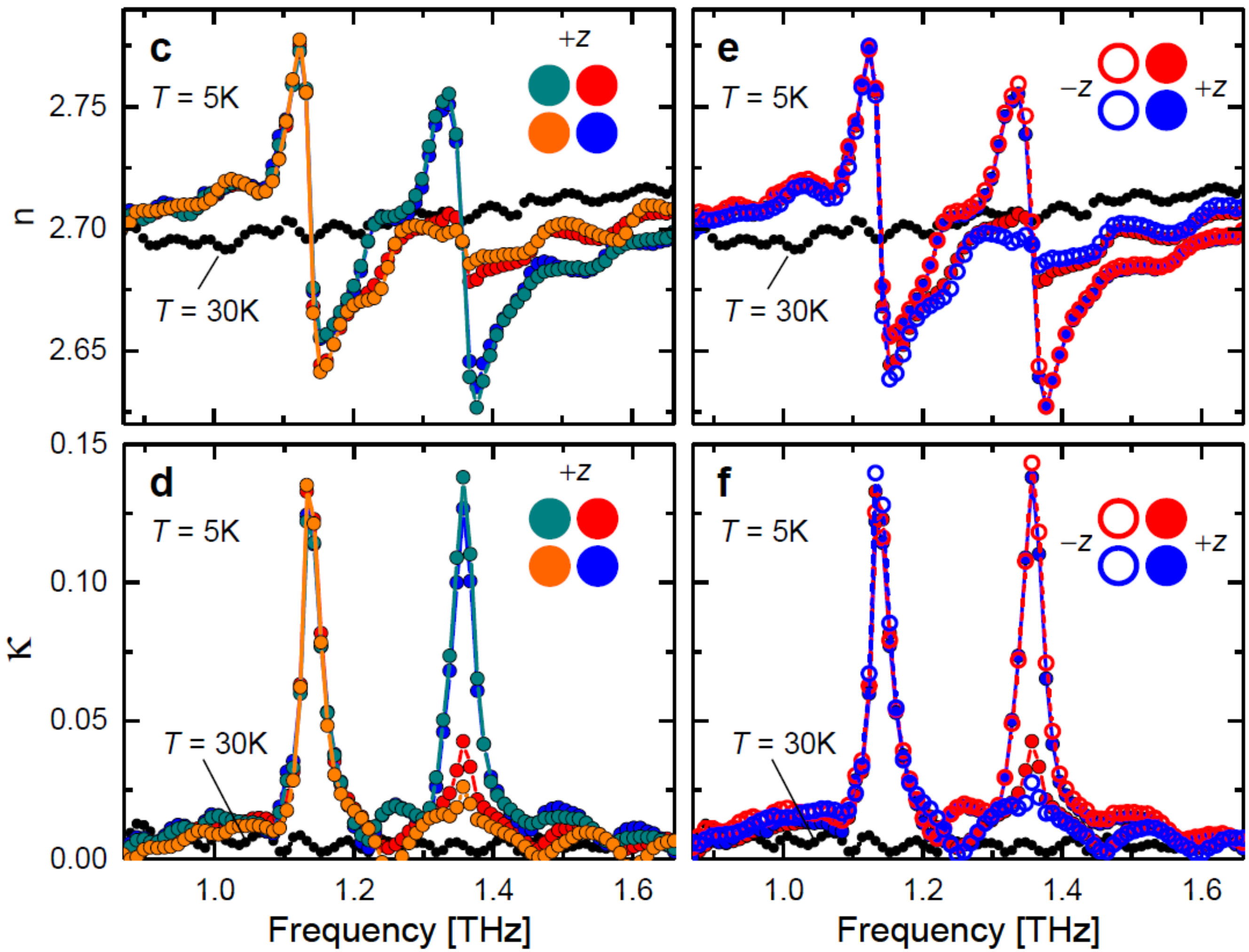




- Sign of χ_{xy}^{me} depends on the sign of the poling $E^0 \times H^0$ field✓

Multi-antiferroic $LiCoPO_4$





- Sign of $\chi_{xy}^{me}(\omega)$ depends on the sign of the poling $E^0 \times H^0$ field✓
- Remnant directional anisotropy in an antiferromagnet✓
- Contrast between AFM domains via simple absorption✓

Kocsis, arXiv:1711.08124

Multi-antiferroic $LiCoPO_4$

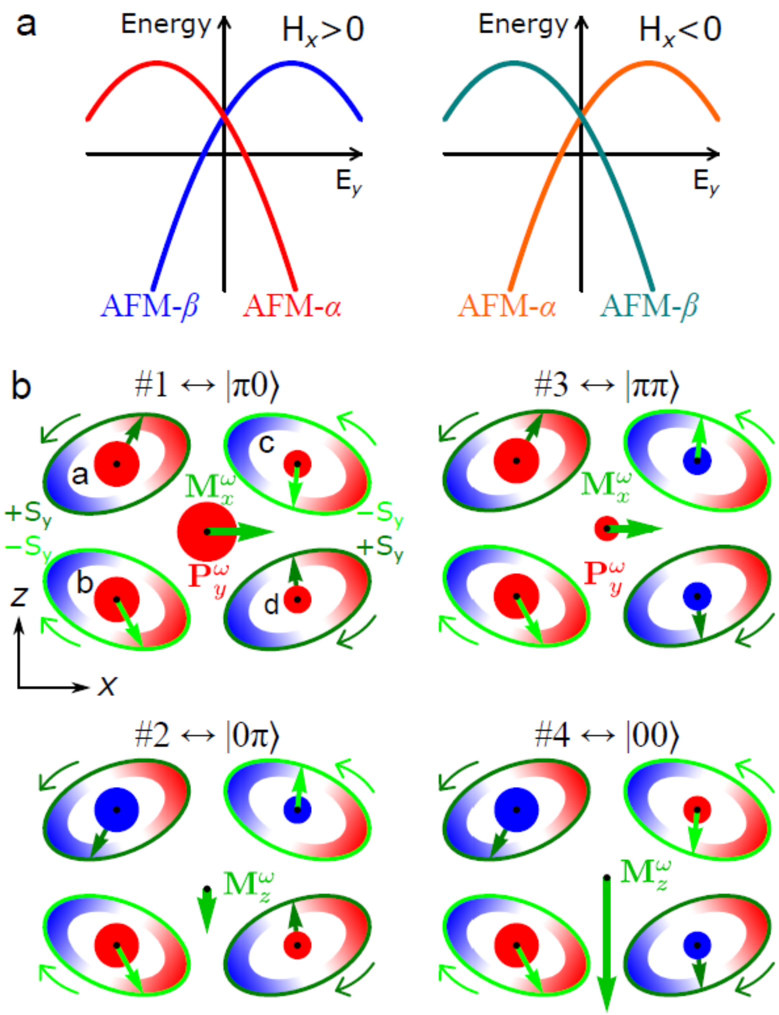
$$P_x = b_{y^2}\left[(S_a^y)^2 - (S_b^y)^2 - (S_c^y)^2 + (S_d^y)^2\right] + b_{x^2-z^2}\left(Q_a^{x^2-z^2} - Q_b^{x^2-z^2} - Q_c^{x^2-z^2} + Q_d^{x^2-z^2}\right) + b_{2xz}\left(Q_a^{2xz} + Q_b^{2xz} - Q_c^{2xz} - Q_d^{2xz}\right),$$

$$P_y = c_{2xy}\left(Q_a^{2xy} - Q_b^{2xy} - Q_c^{2xy} + Q_d^{2xy}\right) + c_{2yz}\left(Q_a^{2yz} + Q_b^{2yz} - Q_c^{2yz} - Q_d^{2yz}\right),$$

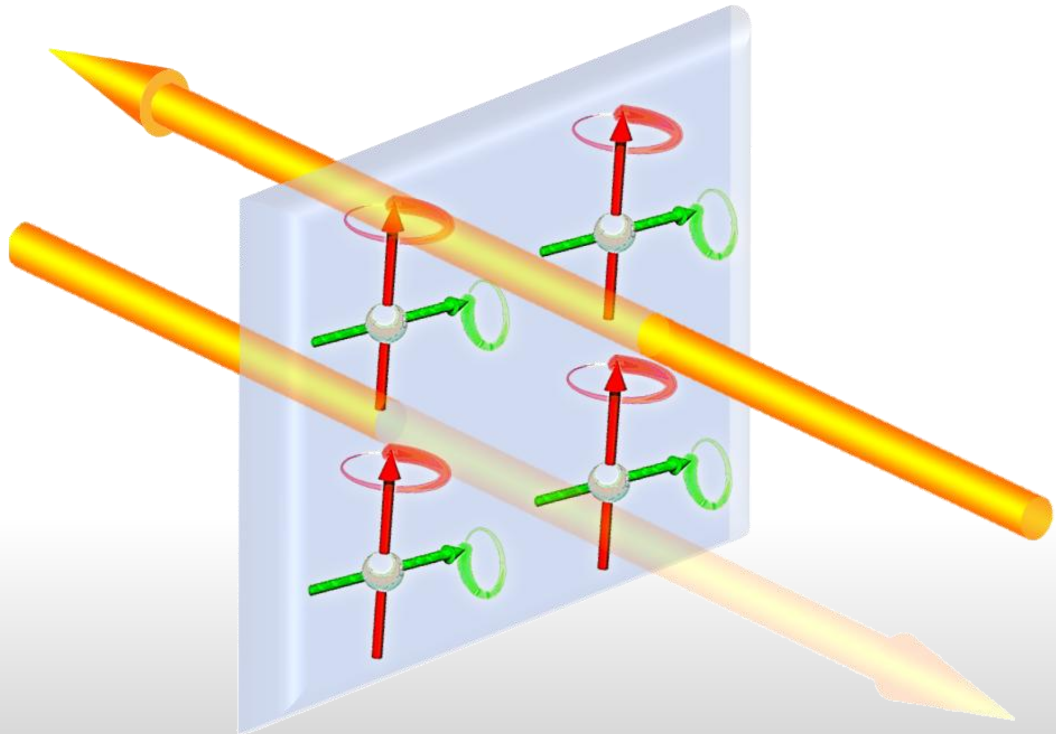
$$P_z = d_{y^2}\left[(S_a^y)^2 + (S_b^y)^2 - (S_c^y)^2 - (S_d^y)^2\right] + d_{x^2-z^2}\left(Q_a^{x^2-z^2} + Q_b^{x^2-z^2} - Q_c^{x^2-z^2} - Q_d^{x^2-z^2}\right) + d_{2xz}\left(Q_a^{2xz} - Q_b^{2xz} - Q_c^{2xz} + Q_d^{2xz}\right),$$

where

$$Q_a^{x^2-z^2} = S_a^x S_a^x - S_a^z S_a^z,$$
$$Q_a^{2xz} = S_a^x S_a^z + S_a^z S_a^x,$$
$$Q_a^{2yz} = S_a^y S_a^z + S_a^y S_a^x$$



Kocsis, arXiv:1711.08124



Thank you for your attention!

PhD and postdoc positions open

in Department of Experimental Physics V, Center for Electronic Correlations and Magnetism of University of Augsburg

istvan.kezsmarki@physik.uni-augsburg.de