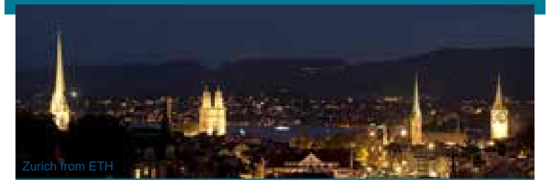
ETH zürich



Magnetoelectric phase control in antiferromagnets

Manfred Fiebig (Department of Materials, ETH Zürich)
SPICE Workshop on Antiferromagnetic Spintronics
Mainz, Germany, 7 – 10 October 2019

ETH zürich

Main contributors

ETH Zürich, Multifunctional Ferroics
A. Bortis, E. Hassanpour, T. Lottermoser, S. Manz, M. Matsubara, M. Weber
ETH Zürich, Materials Theory
N. A. Spaldin et al.
Paul Scherrer Institute, Villigen
N. Leo, M. Kenzelmann, J. White
U. Grenoble
A. Cano
U. Tokyo
T. Kimura et al.
U. Trondheim
D. Meier, S. Selbach
Tohoku U. Sendai
M. Matsubara

Funding:

ETH Eidgenössische Technische Hochschule Zürich Swiss Federal Institute of Technology Zurich

FNSNF Fonds national suisse Schweizerischer Nationalfonds Fondo nazionale svizzero Swiss National Science Foundation

erc European Research Council

ETH zürich

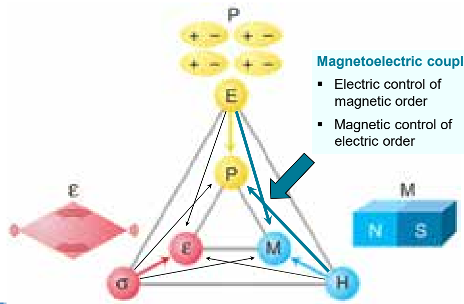
Table of contents

ETH zürich

Table of contents

ETH zürich

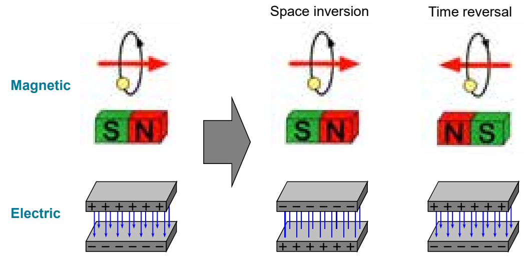
Magnetoelectric coupling



Magnetoelectric coupling
- Electric control of magnetic order
- Magnetic control of electric order

ETH zürich

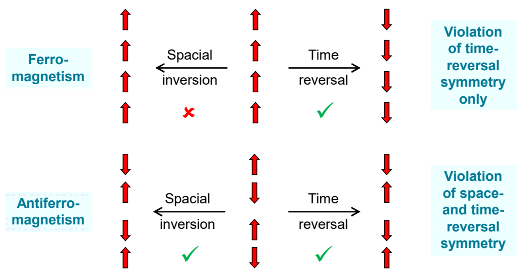
Magnetoelectric coupling and symmetry



Magnetoelectric coupling ↔ breaking of space- <u>and</u> time-reversal symmetries

ETH zürich

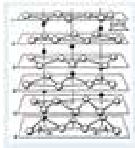
Magnetoelectric coupling in ferro-/antiferromagnets



For magnetoelectric coupling antiferromagnetism is more favourable

ETH zürich

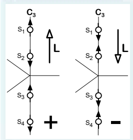
1: Electric field → magnetization



Cr_2O_3

Magnetoelectric antiferromagnet

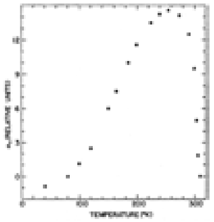
T_N = 307.6 K



Magnetic structure

Two 180° domain states (+/-)

Distinguished by spin reversal



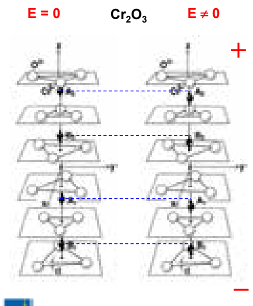
D.N. Astrov, JETP **11**, 708 (1960)

Linear magnetoelectric effect:

$M \propto \alpha E$

ETH zürich

Sources of the linear magnetoelectric effect



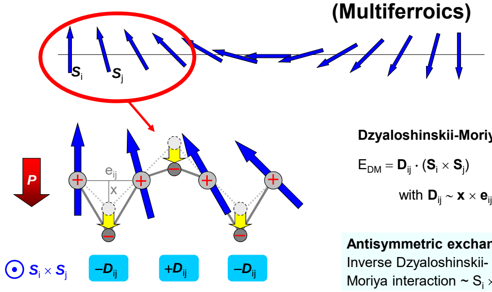
Source in Cr_2O_3:

- Electric field E || z moves ions with respect to ligands
- $A_{1,2}$ and $B_{1,2}$ sites are no longer equivalent → M || z
- In general: many other mechanisms

R. M. Hornreich et al., Phys. Rev. **161**, 506 (1967)
G. A. Gehring, Ferroelectrics **161**, 275 (1994)

ETH zürich

2: Magetic order → spontaneous polarization (Multiferroics)



Dzyaloshinskii-Moriya:

$E_{DM} = \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$

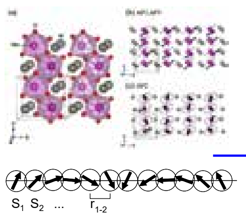
with $\mathbf{D}_{ij} \sim \mathbf{x} \times \mathbf{e}_{ij}$

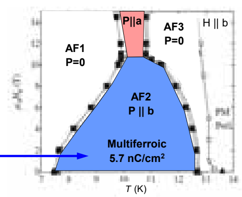
Antisymmetric exchange:
Inverse Dzyaloshinskii-Moriya interaction $\sim S_i \times S_j$

R. E. Newnham et al., J. Appl. Phys. **49**, 6088 (1979)
T. Kimura et al., Nature **426**, 55 (2003)
H. Katsura et al., Phys. Rev. Lett. **95**, 057205 (2005)
M. Mostovoy, Phys. Rev. Lett. **96**, 067601 (2006)

ETH zürich

Magnetically induced ferroelectricity in $MnWO_4$





K. Taniguchi et al., Phys. Rev. Lett. **97**, 097203 (2006)

- **Spiral magnetism** breaks time and space inversion symmetry
- **Electric polarization** allowed according to $P \propto r_{1\text{-}2} \times (S_1 \times S_2)$

ETH zürich

Nonlinear optics: access to (multi-) ferroic order

Electric field in matter:

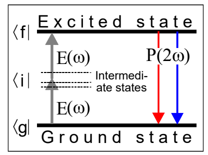
$P(\omega) = \varepsilon_0 \chi E(\omega)$ ($e^{i\omega t}$, $e^{i\omega t}$)

Yet for strong electromagnetic fields (e.g. laser):

$P = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} E E + \chi^{(3)} E E E + \dots)$

With leading-order nonlinear term:

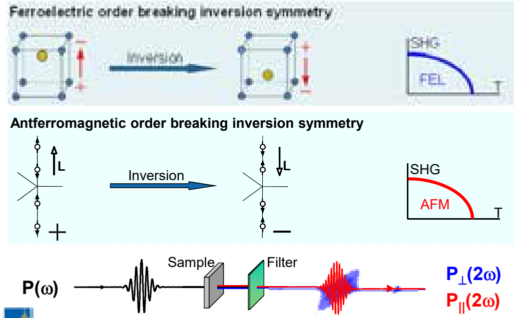
$P(2\omega) = \varepsilon_0 \chi^{(2)} E(\omega) E(\omega)$ ($e^{i2\omega t}$, $e^{i\omega t}$, $e^{i\omega t}$)



→ Frequency doubling ("second harmonic generation", SHG)
→ Applicable to all ordered structures (symmetry as common basis)

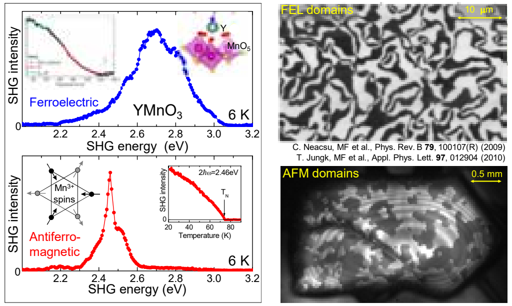
ETH zürich

Nonlinear optics: access to (multi-) ferroic order



ETH zürich

Coexisting electric and magnetic order by SHG



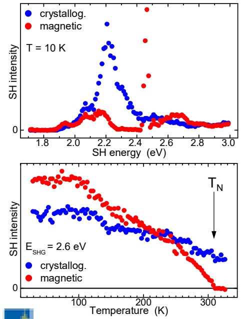
C. Neacsu, MF et al., Phys. Rev. B **79**, 100107(R) (2009)
T. Jungk, MF et al., Appl. Phys. Lett. **97**, 012904 (2010)

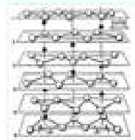
M. Lilienblum, MF et al., Nature Phys. **11**, 1070 (2015)

1996 - 2000: MF et al., Phys. Rev. Lett. **84**, 5620 (2000)

ETH zürich

First antiferromagnetic SHG experiment

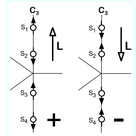




Cr_2O_3

Magnetoelectric antiferromagnet

T_N = 307.6 K



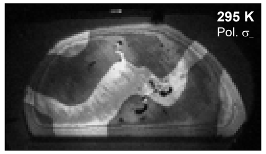
Magnetic structure

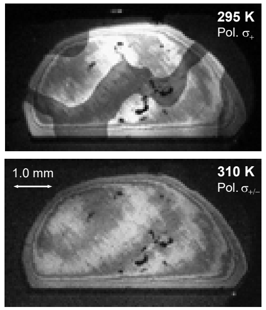
Two 180° domain states (+/-)

Distinguished by spin reversal

ETH zürich

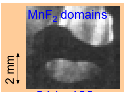
Antiferromagnetic 180° domains in Cr_2O_3





With SHG:
Exposure time:
~ 1 min
Resolution:
~ 1-10 µm

MF et al., Appl. Phys. Lett. **66**, 2906 (1995)



~24 h, 100 µm
Polarized neutrons

M. Schlenker et al., Ferroelectrics **162**, 299 (1994)

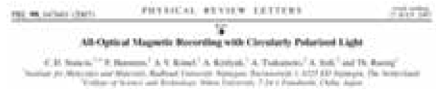
ETH zürich

Table of contents

Thomas Lottermoser
Masakazu Matsubara
Sebastian Manz

ETH zürich

Optical switching of a ferromagnetic state



Optical switching of an antiferromagnetic state?

Useful for
- Exchange bias effect and tight manipulation
- Controlling strong correlation related to antiferromagnetism
- Data storage in antiferromagnetic systems [S. Loth et al., Science **335**, 196 (2015)]

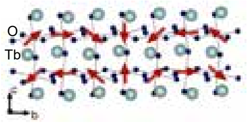
Challenge
- No macroscopic magnetization
- Requires mechanisms for visualization and optical control

P. Němec, MF et al., upcoming review

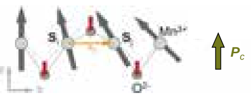
ETH zürich

Antiferromagnetic spiral order in $TbMnO_3$

Orthorhombic $TbMnO_3$



Spiral order below 27 K



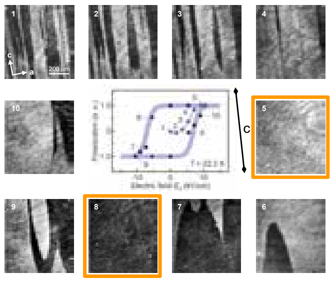
Mn spins forming cycloidal spiral
→ antiferromagnetism

T. Kimura et al., Phys. Rev. B **71**, 224425 (2005)

- Spiral breaks inversion symmetry
- Dzyaloshinskii-Moriya interaction induces "parasitic" electric polarization: $P \propto e_{ij} \times (S_i \times S_j)$

ETH zürich

Hysteretic poling of multiferroic $TbMnO_3$

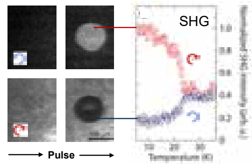


- Antiferromagnetic domains with secondary ferroelectric polarization
- SHG is only existing technique to resolve these
- Single-domain poling at ±10 kV/cm

M. Matsubara, S. Manz, MF et al., Science **348**, 1112 (2015)

ETH zürich

Optical switching of the antiferromagnetic state

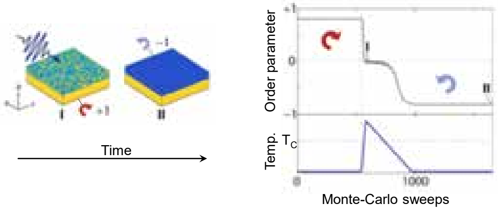


- Switching independent of light pulse duration (120 fs to cw)
- Points to thermal origin
- Verified by Monte-Carlo simulations

S. Manz, MF et al., Nature Phot. **10**, 653 (2016)

ETH zürich

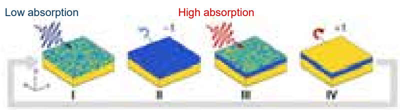
Monte-Carlo simulation of thermal switching



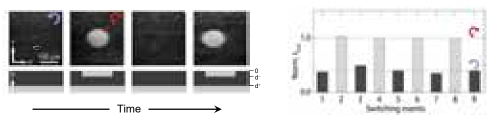
Antiferromagnetic order parameter reversal mediated by reversal of coupled electric polarization in depolarization field during cooling

ETH zürich

Concept for reversible antiferromagnetic switching



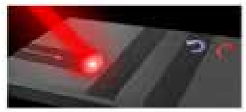
S. Manz, MF et al., Nature Phot. **10**, 653 (2016)

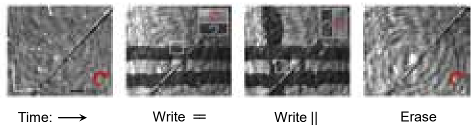


- Full antiferromagnetic reversibility in the top layer 0 … d⁻
- Light color ↔ direction of antiferromagnetic order parameter

ETH zürich

Spatial correlations





Sequential writing and erasing without crosstalk or anisotropy effects

S. Manz, MF et al., Nature Phot. **10**, 653 (2016)

ETH zürich

Table of contents

- Magnetoelectric coupling and antiferromagnetism
- All-optical switching of antiferromagnetic order
- Inversion of domain patterns
- Conversion of multiferroics between 2D and 3D
- Summary

ETH zürich

Complexity of order parameters in Mn_2GeO_4

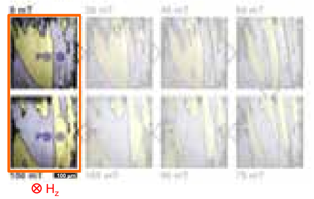
Ordered state to achieve M || P with magnetoelectric coupling is complex!

- Two commensurate order parameters at $k_O = (0, 0, 0)$
- Four incommensurate order parameters at $k_\varnothing = (\pm 0.136, \pm 0.211, 0)$

Certainly magnetoelectric multiferroicity is not the only functionality exhibited by such a rich order parameter landscape ?!?

ETH zürich

Inversion of a ferroelectric domain pattern



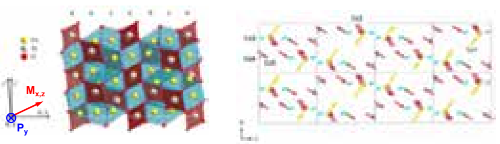
- Magnetically induced ferroelectric $\pm P_z$ domains tuned in a magnetic field H_z (observation by SHG)
- Polarization reversal in each domain, yet without destroying domain pattern
- Inversion of ferroelectric Mn_2GeO_4 domain pattern

Invariant free-energy term mediating the coupling: $F_{inv} \propto C(k_O) \cdot P_z \cdot M_z(H_z)$

Reversal of magnetization M_z by field H_z enforces reversal of P_z in each domain for minimizing F_{inv} — the domain pattern is preserved

ETH zürich

Complexity of order parameters in Co_3TeO_6



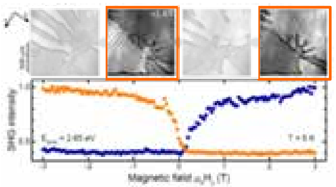
Ordered state to achieve M ⊥ P with magnetoelectric coupling is complex!

- One commensurate one-dimensional order parameter at $k_O = (0, 0, 0)$
- One commensurate four-dimensional order parameter at $k_\varnothing = (0, 0.5, 0.25)$

Does the complexity of the order parameters generally allow the existence of a domain inversion mechanism?

ETH zürich

Inversion of a ferromagnetic domain pattern



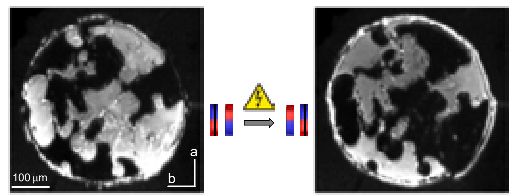
- Ferromagnetic $\pm M_{x,z}$ domains tuned in magnetic field H_y (observation by SHG)
- Magnetization reversal in each domain, yet without destroying domain pattern
- Inversion of ferromagnetic Co_3TeO_6 domain pattern

Invariant free-energy term mediating the coupling: $F_{inv} \propto C(k_\varnothing) \cdot M_{x,z} \cdot M_y(H_y)$

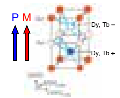
Reversal of magnetization M_y by field H_y enforces reversal of $M_{x,z}$ in each domain for minimizing F_{inv} — the domain pattern is preserved

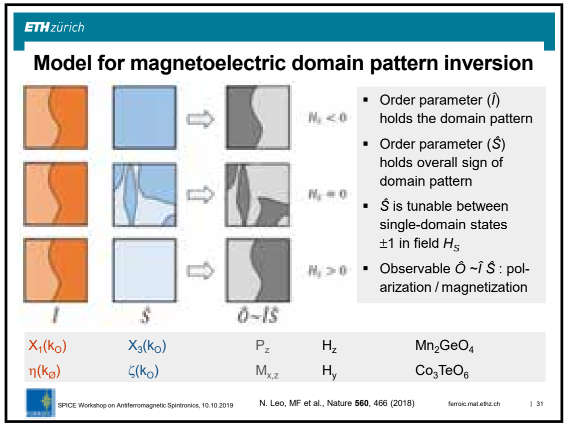
ETH zürich [YT]³ Collaboration

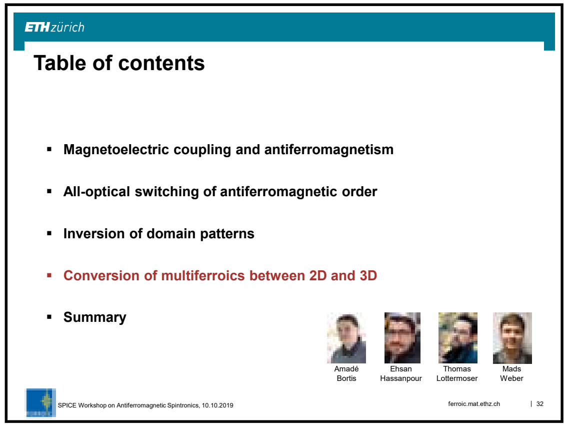
Antiferromagnetic domain inversion by electric field

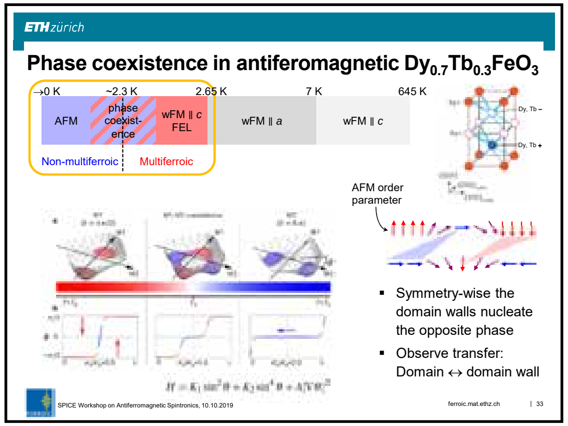


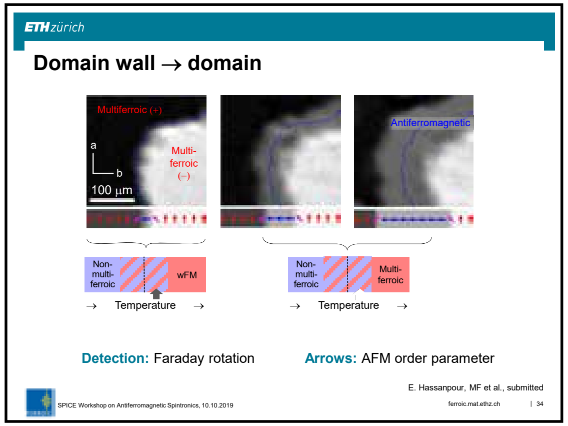
- $Dy_{0.7}Tb_{0.3}FeO_3$ in multiferroic phase at 2.5 K
- M || c (Faraday rotation) reversed by E || c
- Coupling term $\propto M_{Fe} P_s L_{Dy,Tb}$

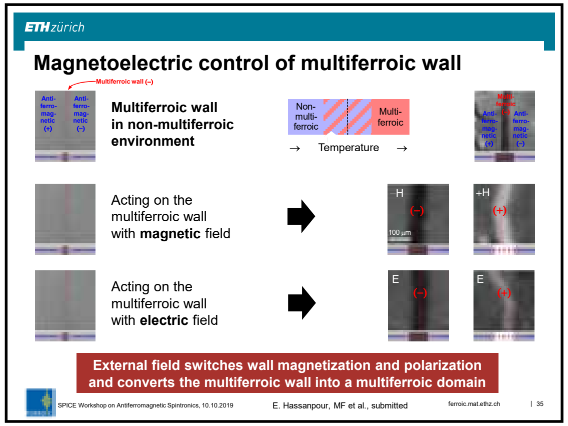



ETH zürich
Model for magnetoelectric domain pattern inversion
Order parameter (Î) holds the domain pattern
Order parameter (Ŝ) holds overall sign of domain pattern
Ŝ is tunable between single-domain states ±1 in field H_S
Observable Ô ~Î Ŝ : polarization / magnetization
X1(kO) X3(kO) Pz Hz Mn2GeO4
η(kO) ζ(kO) Mx,z Hy Co3TeO6
SPICE Workshop on Antiferromagnetic Spintronics, 10.10.2019
N. Leo, MF et al., Nature 560, 466 (2018)
ferroic.mat.ethz.ch
31


ETH zürich
Table of contents
Magnetoelectric coupling and antiferromagnetism
All-optical switching of antiferromagnetic order
Inversion of domain patterns
Conversion of multiferroics between 2D and 3D
Summary
Amadé Bortis
Ehsan Hassanpour
Thomas Lottermoser
Mads Weber
SPICE Workshop on Antiferromagnetic Spintronics, 10.10.2019
ferroic.mat.ethz.ch
32


ETH zürich
Phase coexistence in antiferomagnetic Dy0.7Tb0.3FeO3
→0 K ~2.3 K 2.65 K 7 K 645 K
AFM
phase coexist-ence
wFM ∥ c FEL
wFM ∥ a
wFM ∥ c
Non-multiferroic
Multiferroic
AFM order parameter
Symmetry-wise the domain walls nucleate the opposite phase
Observe transfer: Domain ↔ domain wall
SPICE Workshop on Antiferromagnetic Spintronics, 10.10.2019
ferroic.mat.ethz.ch
33


ETH zürich
Domain wall → domain
Multiferroic (+)
Multi-ferroic (–)
Antiferromagnetic
100 μm
Non-multi-ferroic
wFM
Multi-ferroic
Temperature
Detection: Faraday rotation
Arrows: AFM order parameter
E. Hassanpour, MF et al., submitted
SPICE Workshop on Antiferromagnetic Spintronics, 10.10.2019
ferroic.mat.ethz.ch
34


ETH zürich
Magnetoelectric control of multiferroic wall
Multiferroic wall (–)
Multiferroic wall in non-multiferroic environment
Non-multi-ferroic
Multi-ferroic
Temperature
Acting on the multiferroic wall with magnetic field
–H
+H
Acting on the multiferroic wall with electric field
E
External field switches wall magnetization and polarization and converts the multiferroic wall into a multiferroic domain
SPICE Workshop on Antiferromagnetic Spintronics, 10.10.2019
E. Hassanpour, MF et al., submitted
ferroic.mat.ethz.ch
35

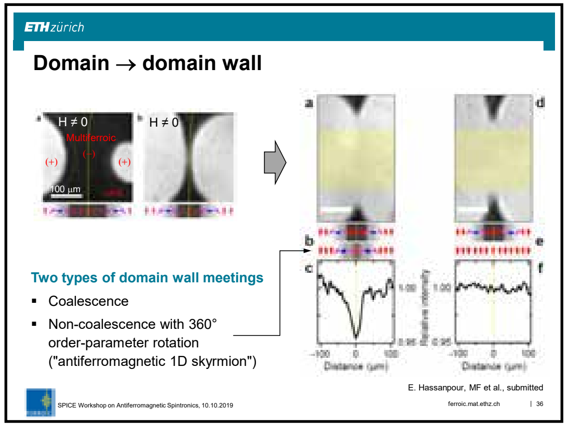

ETH zürich
Domain → domain wall
H ≠ 0
Multiferroic
100 μm
Two types of domain wall meetings
Coalescence
Non-coalescence with 360° order-parameter rotation ("antiferromagnetic 1D skyrmion")
Distance (μm)
E. Hassanpour, MF et al., submitted
SPICE Workshop on Antiferromagnetic Spintronics, 10.10.2019
ferroic.mat.ethz.ch
36

Table of contents

Summary

Magnetoelectric phase control in antiferromagnets

- Magnetoelectric coupling is favoured by broken space- and time-inversion symmetries
- This twofold symmetry violation is predominantly found in antiferromagnets rather than ferromagnets
- A large variety of static and dynamic coupling effects – there is no end in sight
- Nonlinear optics is ideal for probing the symmetry effects around the magnetoelectric coupling, resolve domains, detect dynamical processes down to the sub-picosecond range