

Ferroelectric control of magnons in BiFeO₃-based heterostructures

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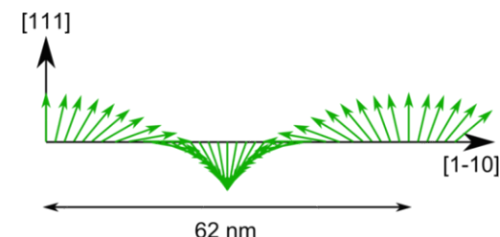
⁹ Laboratoire Albert Fert, CNRS, Thales, Université Paris-Saclay, Palaiseau, France

BiFeO₃ : a room-temperature multifunctional oxide

Magnetic properties

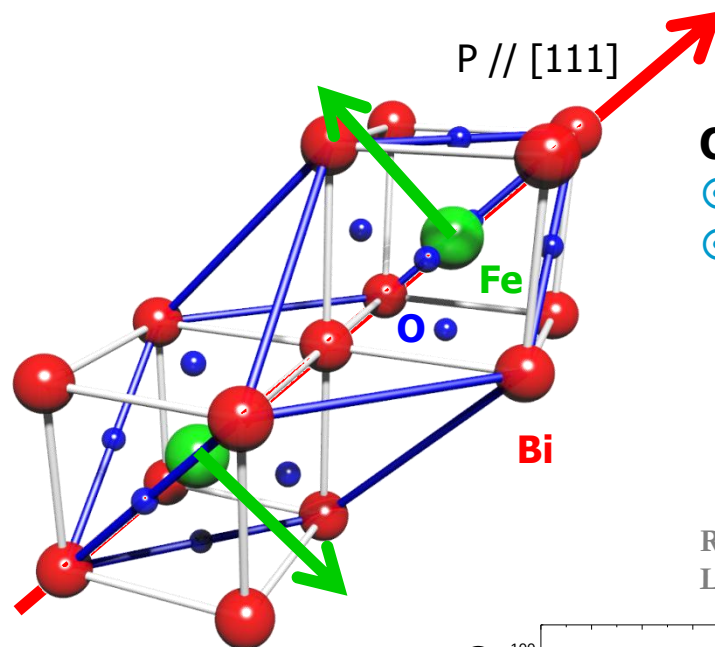
- ⦿ G-type antiferromagnetic + cycloidal modulation ($\lambda=62$ nm)
- ⦿ $T_N \approx 640$ K

Sosnowska et al., J. Phys. C, 15, 4835 (1982)



Structural properties

- ⦿ ABO₃ Perovskite
- ⦿ Space group $R\bar{3}c$
- ⦿ Pseudocubic $a = 3.96$ Å
- ⦿ Coexistence of polar shifts & octahedra rotations



Optical properties

- ⦿ Low band gap $E_g \approx 2.7$ eV
- ⦿ Highly birefringent $\Delta n \approx 0.25$



Rivera et al, Ferroelectrics 204, 33 (1997)

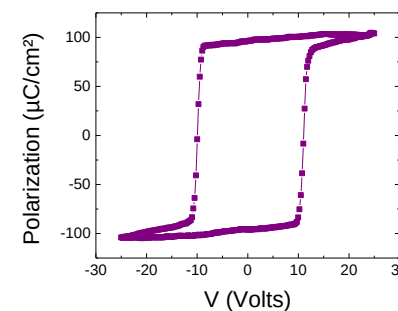
Lebeugle et al., PRB 76, 024116 (2007)

Ferroelectric properties

- ⦿ Very high $T_C \approx 1100$ K
- ⦿ Very large $P=100$ $\mu\text{C}/\text{cm}^2$

Fisher et al., J. Phys. C, 13, 1931 (1980)

Béa et al, APL 93, 072091 (2008)

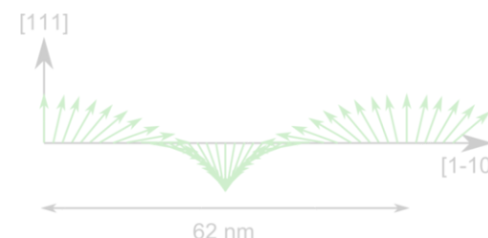


BiFeO₃ : a room-temperature multifunctional oxide

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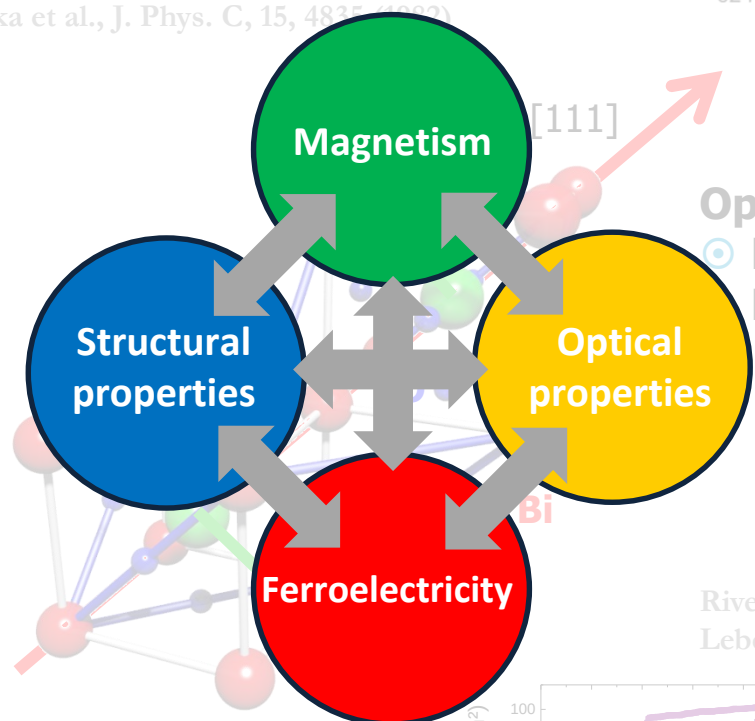
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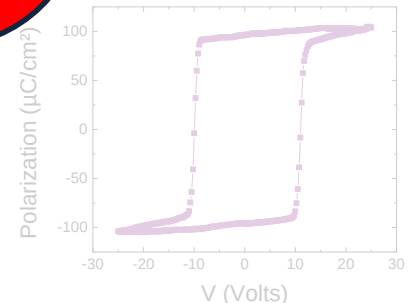


Rivera et al, Ferroelectrics 204, 33 (1997)
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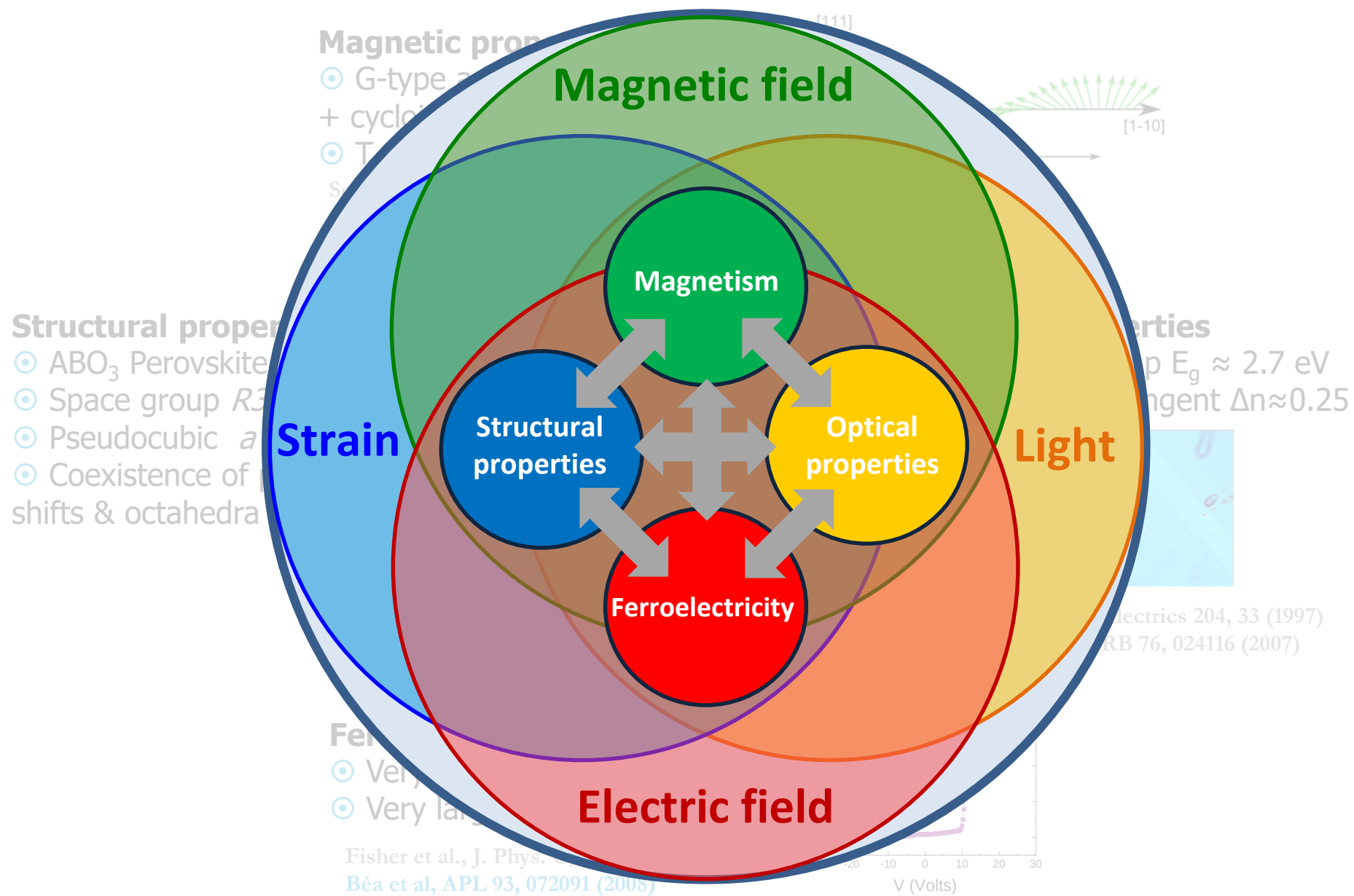
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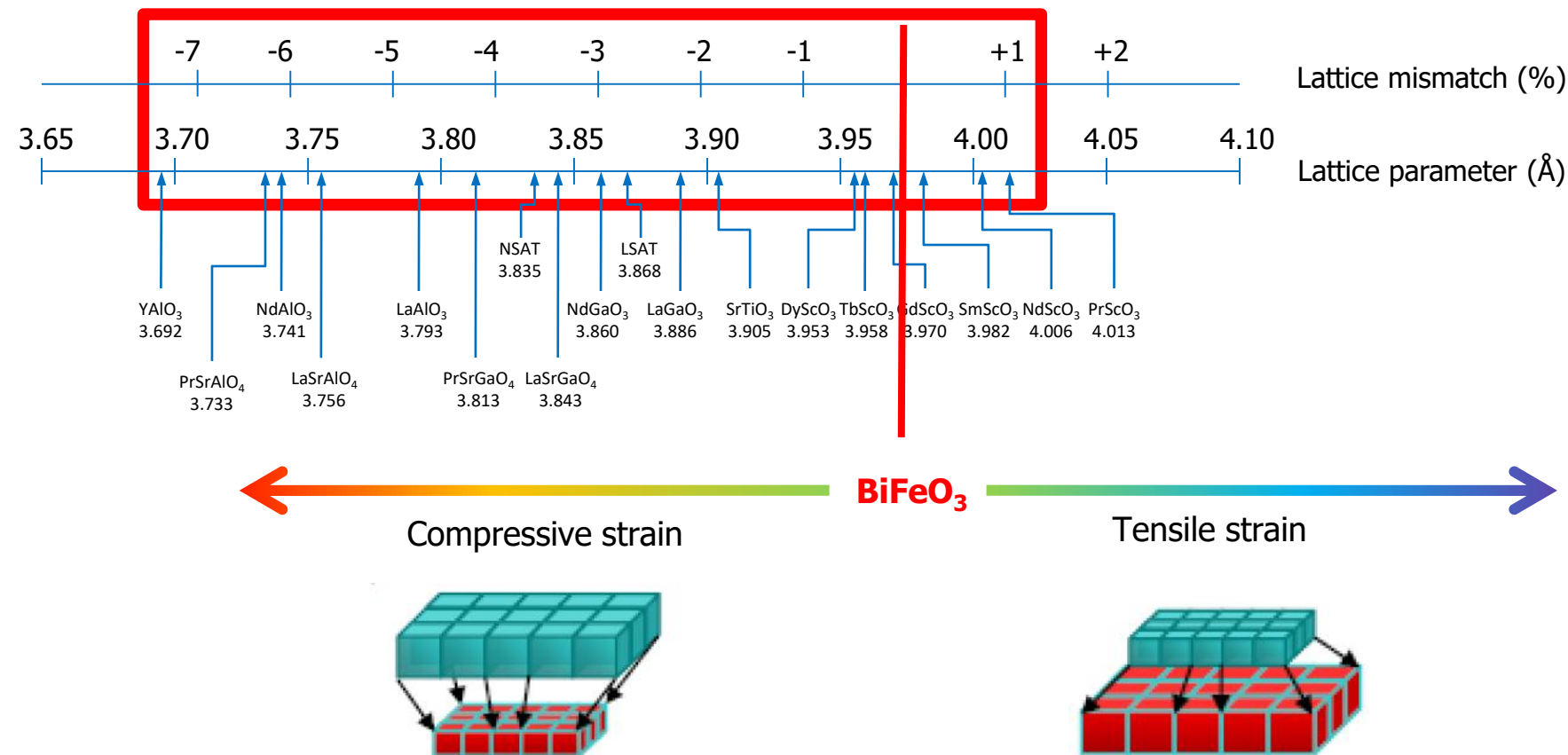
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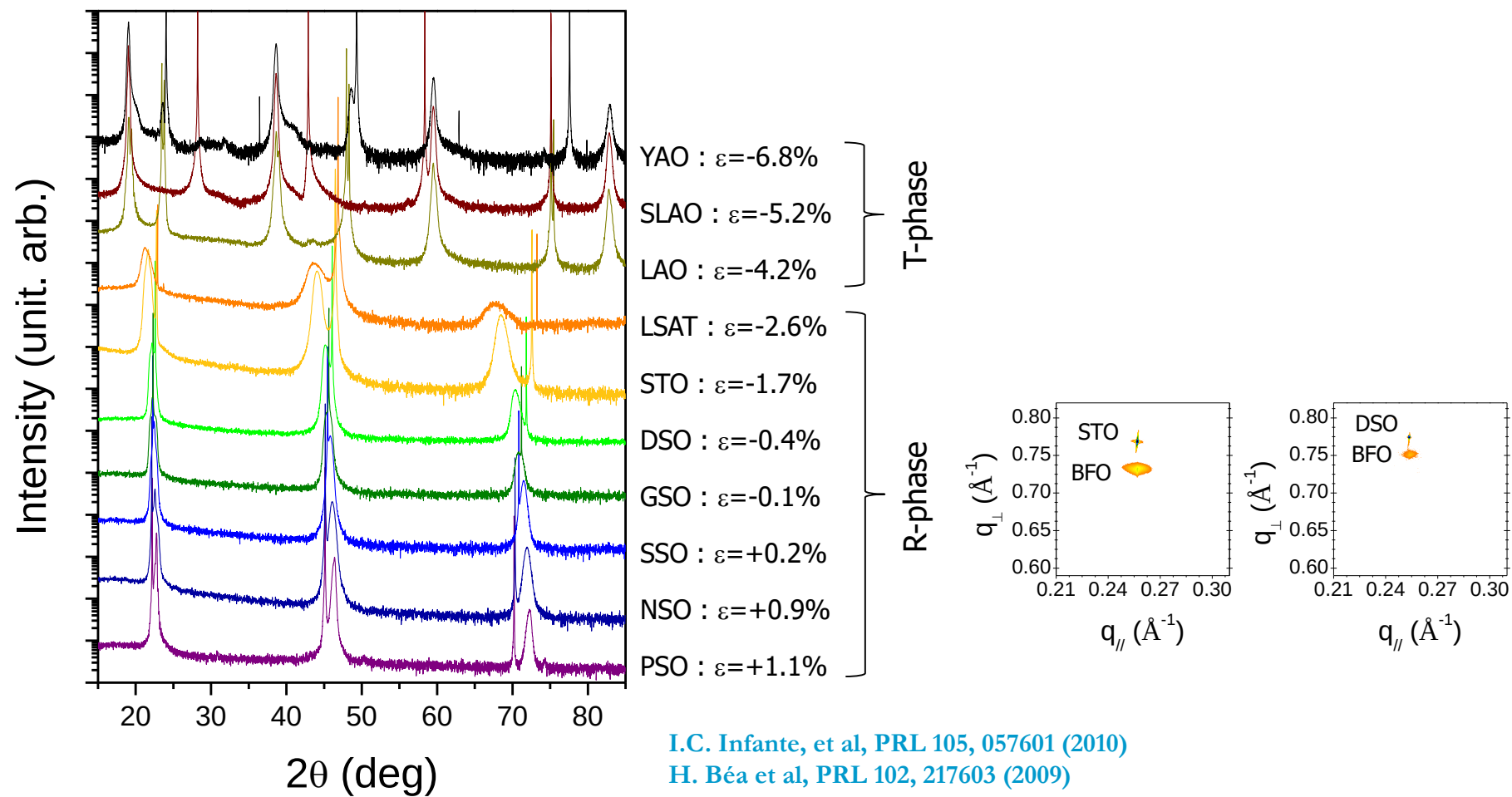


BiFeO₃ : a room-temperature multifunctional oxide

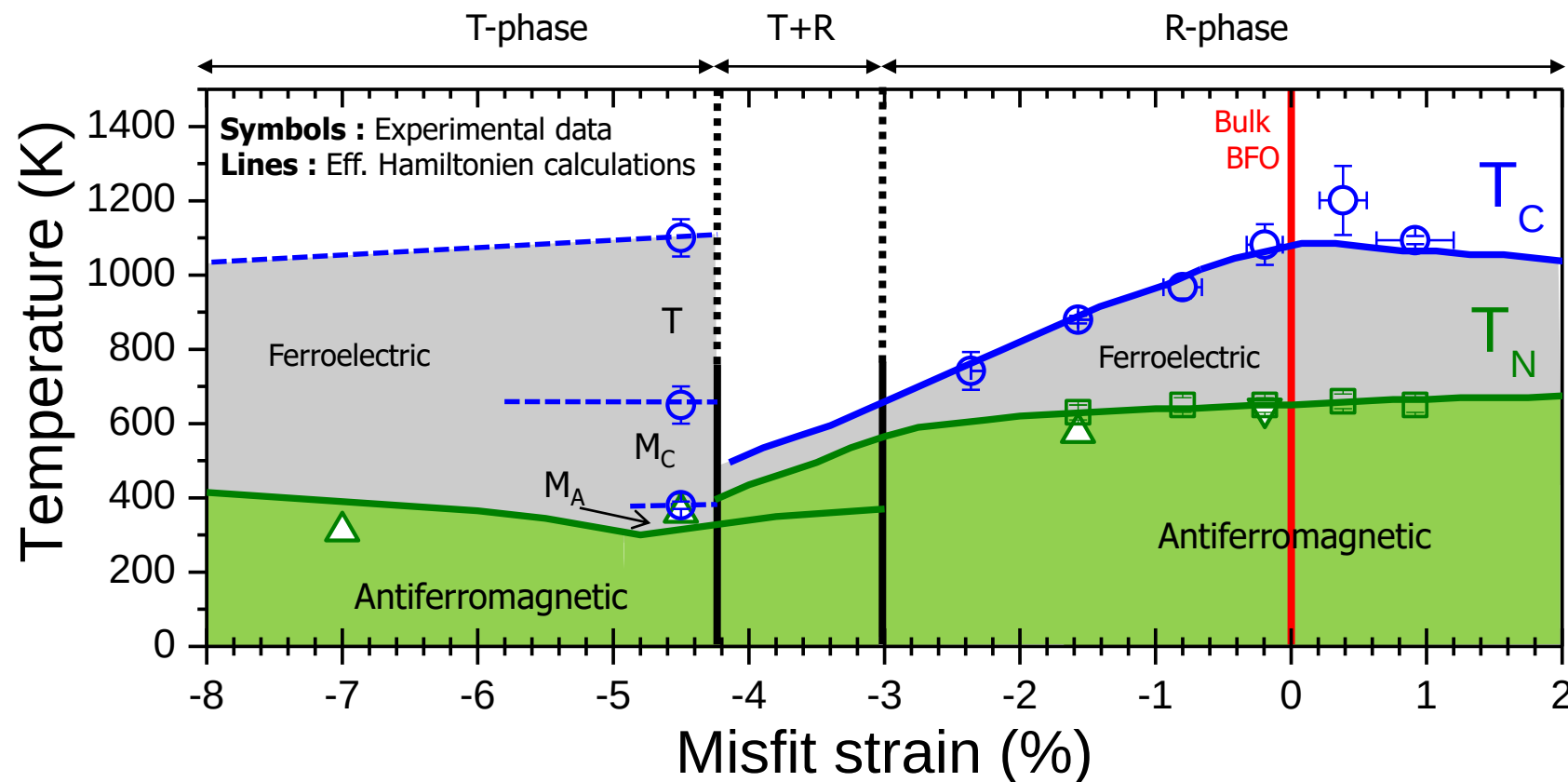


Mismatch with commercial substrates





- Fully strained BFO films are obtained for a **very broad range of lattice mismatch**
- From +1.1% down to nearly -3% : R-type films with monoclinic (Cc) symmetry derived from bulk (R3c)
- Beyond -3% : T-type films with **giant c/a ratio** (1.26 vs 1.05) and Cm symmetry : **new phase** of BiFeO₃



- **R phase:** Decrease of T_C with strain : completely **unexpected** and different from situation in standard ferroelectrics like BaTiO₃ K. Choi et al, Science 306, 1005 (2004)

- Strain increases octahedra rotations, which tends to suppress ferroelectricity

- Néel temperature weakly affected by strain

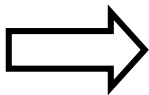
D. Sando et al, J. Phys.: Condens. Matter 26, 473201 (2014)

- **T phase:** Concomitant ferroelectric and magnetic transition just above 300K

H. Béa et al, PRL 102, 217603 (2009) ; I.C. Infante, et al, PRL 107, 237601 (2011) ; I.C. Infante, et al, PRL 105, 057601 (2010) ; Ko et al, Nature Commun. 2, 567 (2011) ; W. Siemons et al, APL 101, 212901 (2012), C. Beekman et al, Adv. Mater. 25, 5561 (2013)

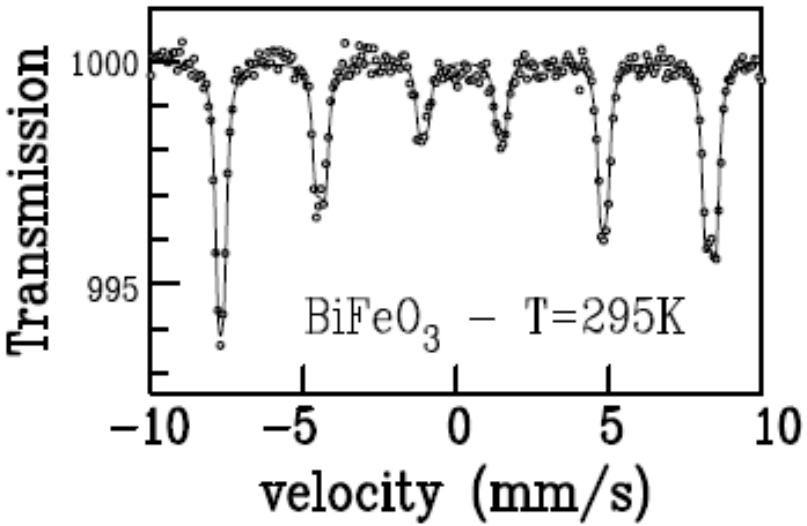
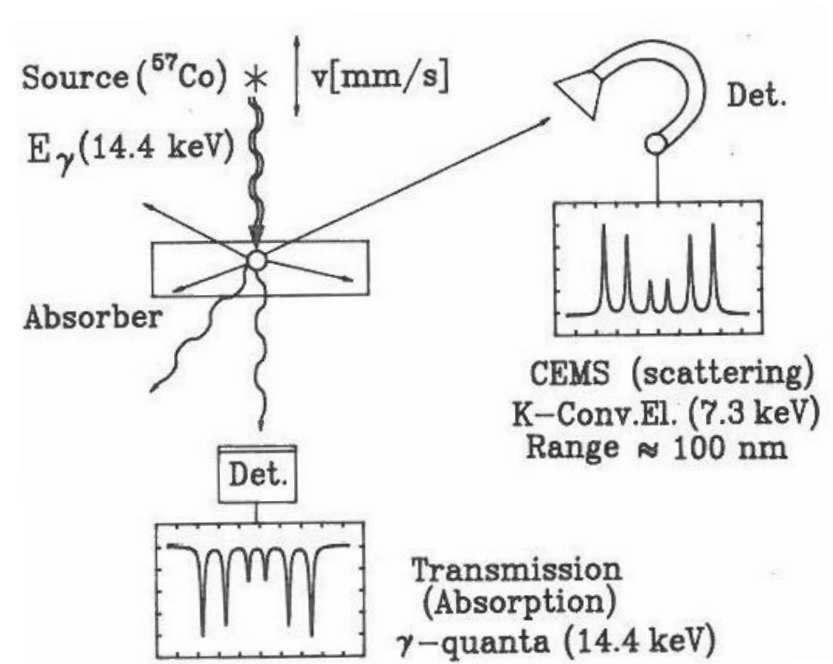
Mössbauer spectroscopy

Mössbauer effect : emission or absorption of a γ -ray photon without loss of energy due to recoil of the nucleus



A probe of hyperfine interactions :

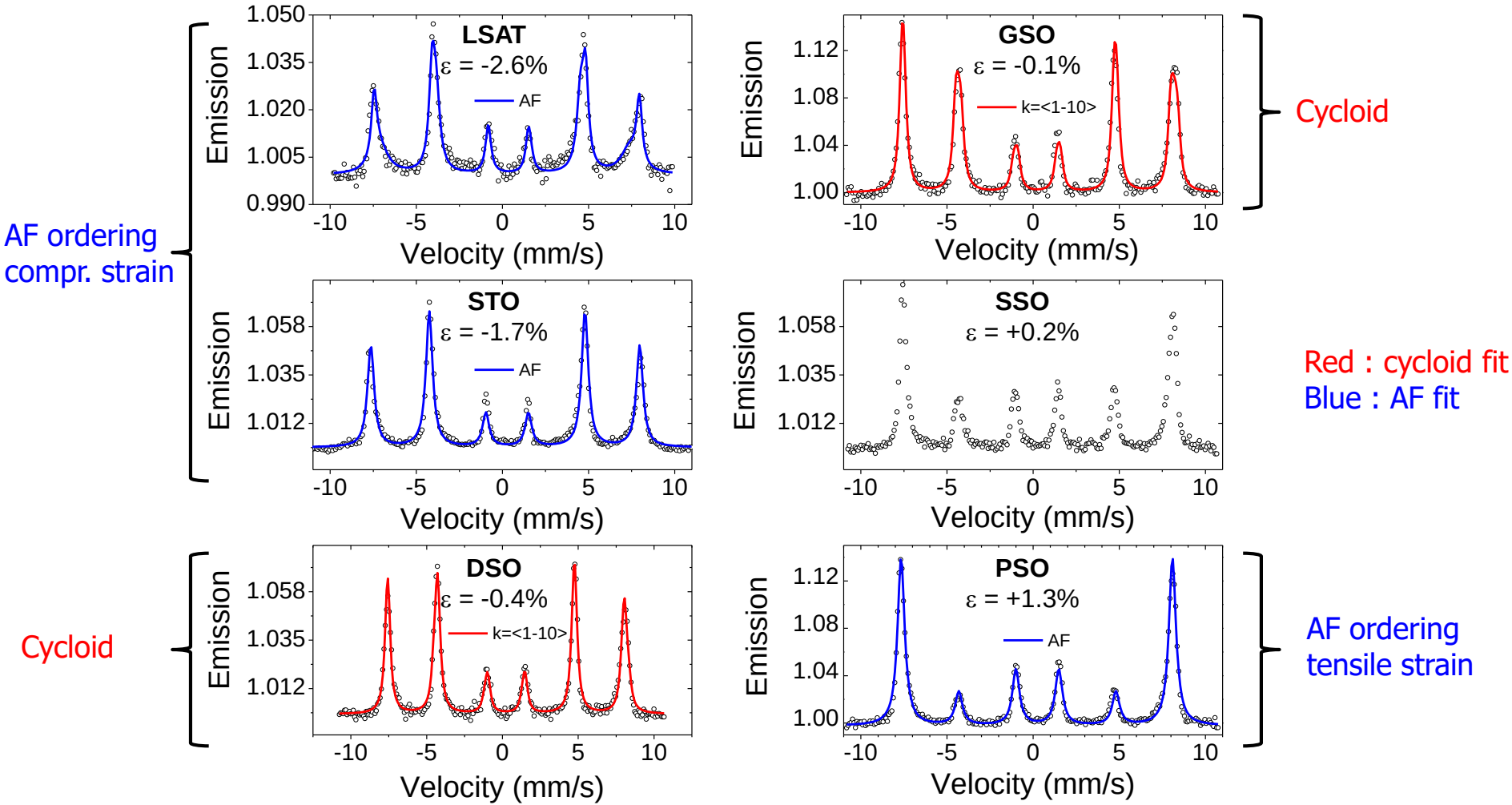
Lebeugle et al, PRB 76, 024116 (2007)



- ⦿ In bulk BFO : **asymmetric sextet**
- ➡ Non collinear order + electric field gradient

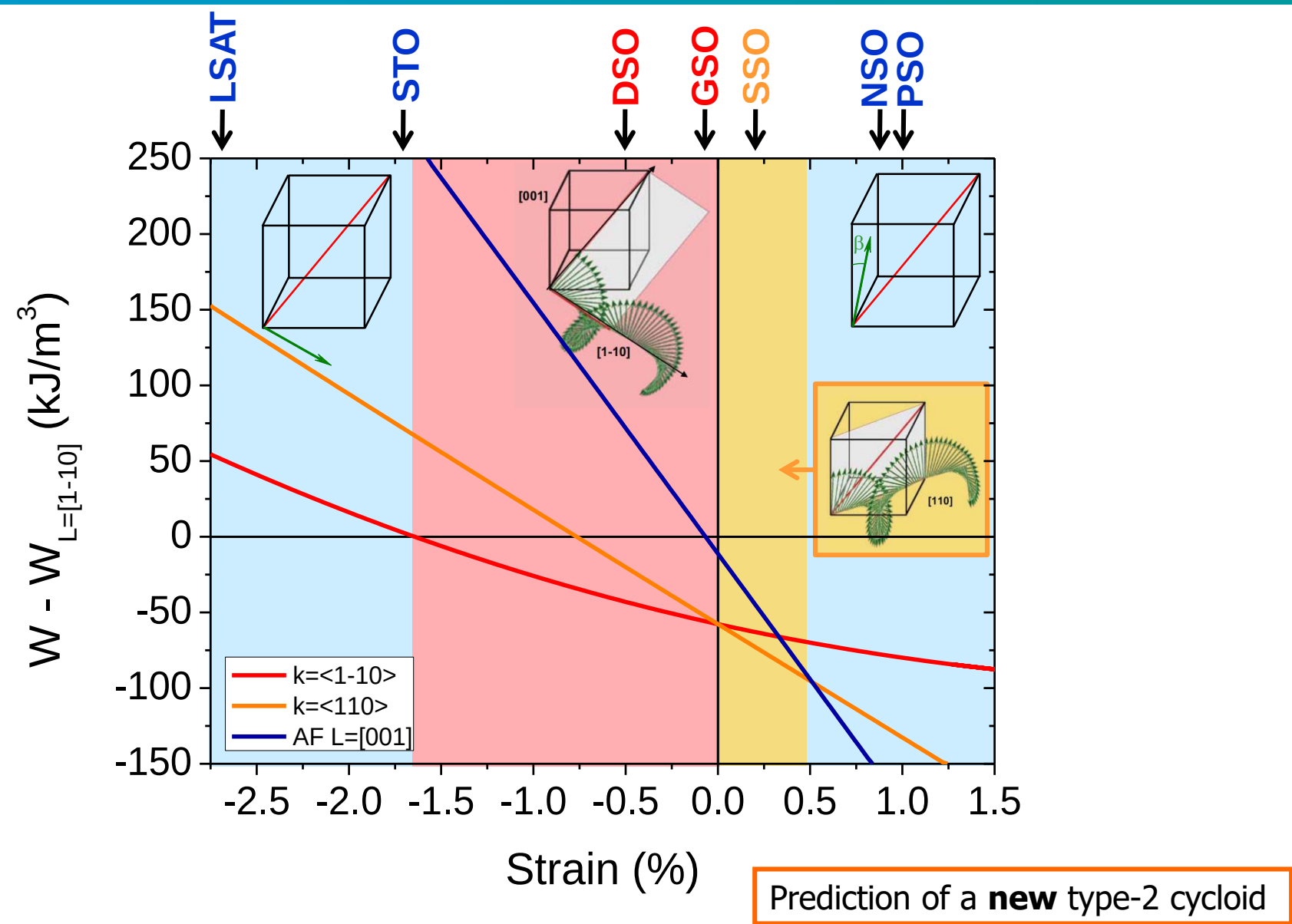
- ⦿ How about it films ?
- ➡ use conversion electron Mossbauer spectroscopy in films ~100% enriched in ⁵⁷Fe

Mössbauer spectroscopy



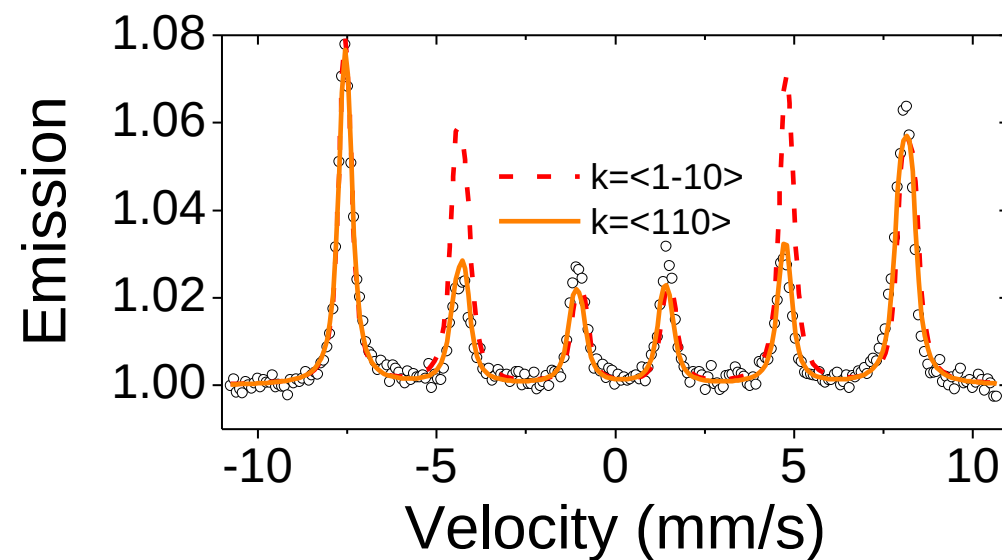
→ Direct evidence for the suppression of the cycloid by epitaxial strain

BiFeO₃ thin films: magnetic phase diagram



D. Sando et al, Nature Mater. 12, 641 (2013)

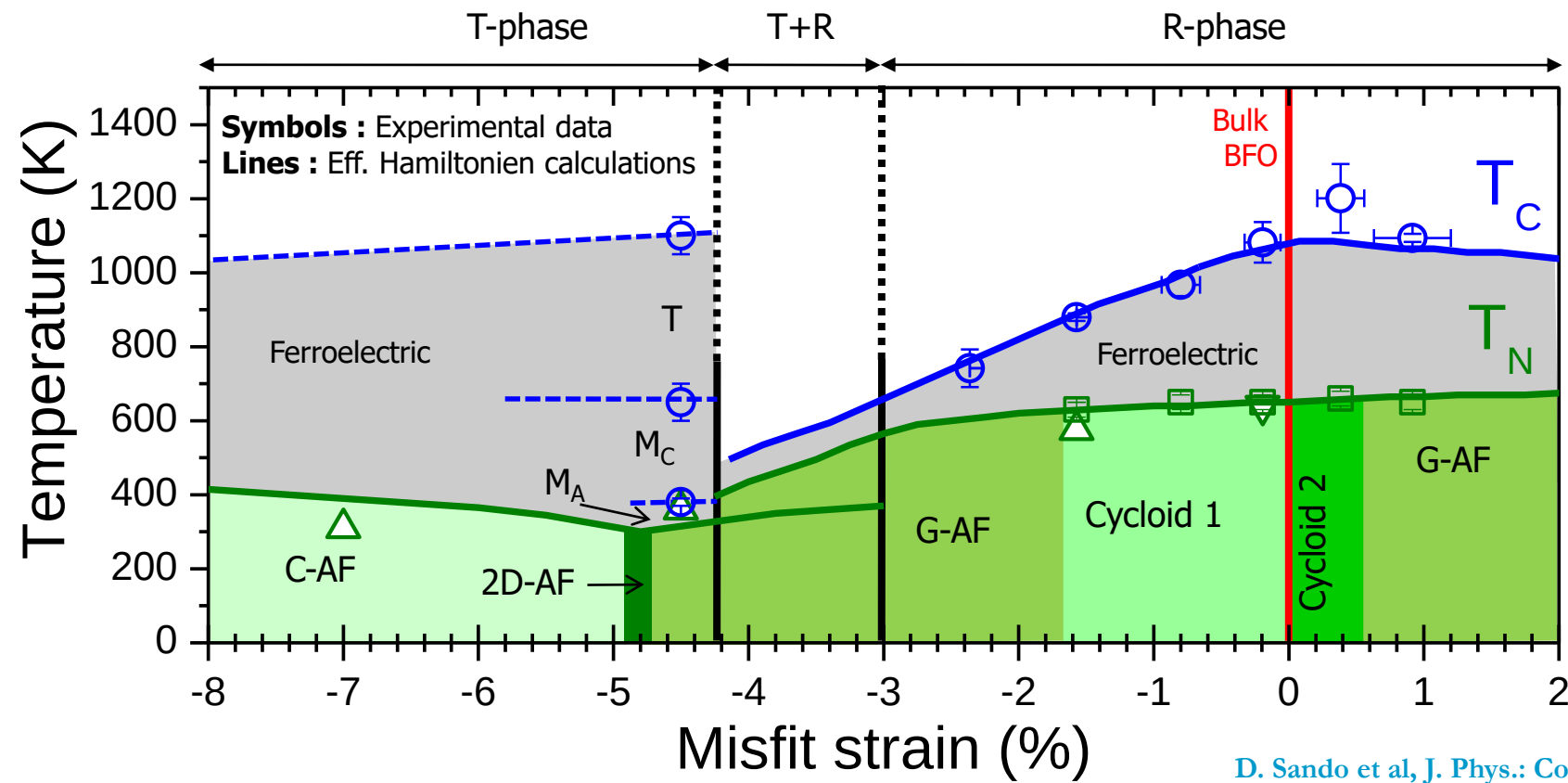
Mössbauer spectroscopy



- ⦿ $\langle 1-10 \rangle$ cycloid model gives poor fit.
- ⦿ Fit with $\langle 110 \rangle$ cycloid yields a far better fit.
- ⦿ **The SSO sample shows evidence of the new cycloid**

D. Sando, MB et al, *Nature Mater.* 12, 641 (2013)

BiFeO₃ thin films: complete phase diagram



D. Sando et al, J. Phys.: Condens. Matter 26, 473201 (2014)

- Extremely **rich material** system
- Strain engineering reveals new structural, ferroelectric and magnetic phases
- Evolution of **non-collinear spin structures** vs T and dimensions ?

H. Béa et al, PRL 102, 217603 (2009) ; I.C. Infante, et al, PRL 107, 237601 (2011) ; I.C. Infante, et al, PRL 105, 057601 (2010) ;
D. Sando et al, Nature Mater. 12, 641 (2013) ; C. Escorihuela-Sayalero et al, PRL 109, 247202 (2012) ;
W. Siemons et al, APL 101, 212901 (2012), C. Beekman et al, Adv. Mater. 25, 5561 (2013)

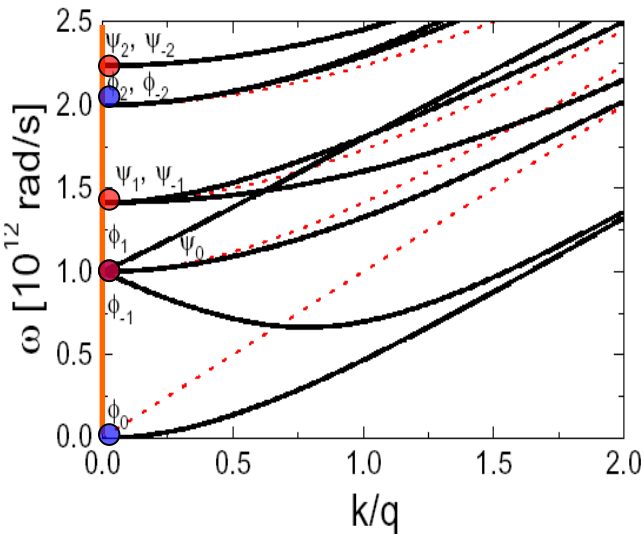
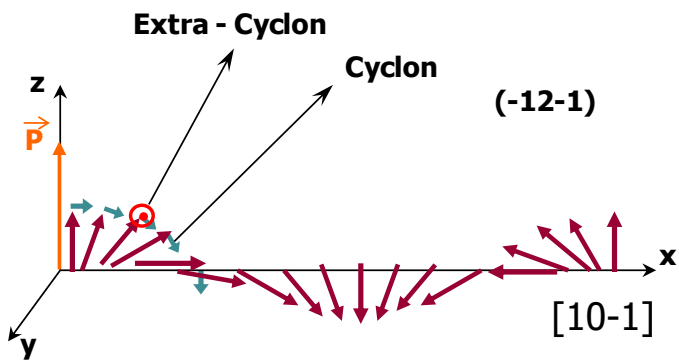
Raman : magnonic response

2 propagating magnon modes :

Φ modes : in the cycloidal plane $E_c(n) = \epsilon_c(q) n$

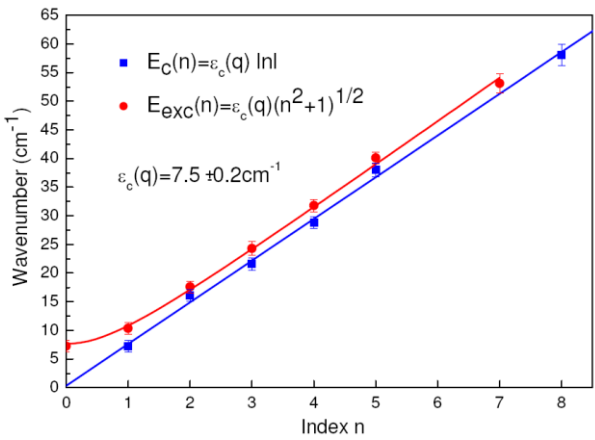
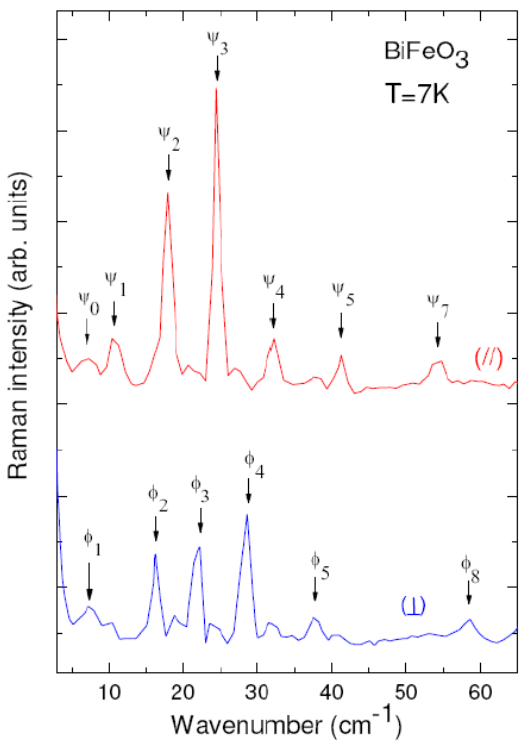
Ψ modes : out of the cycloidal plane $E_c(n) = \epsilon_c(q)(n^2+1)^{1/2}$

$\epsilon_c(q)$ cyclon energy



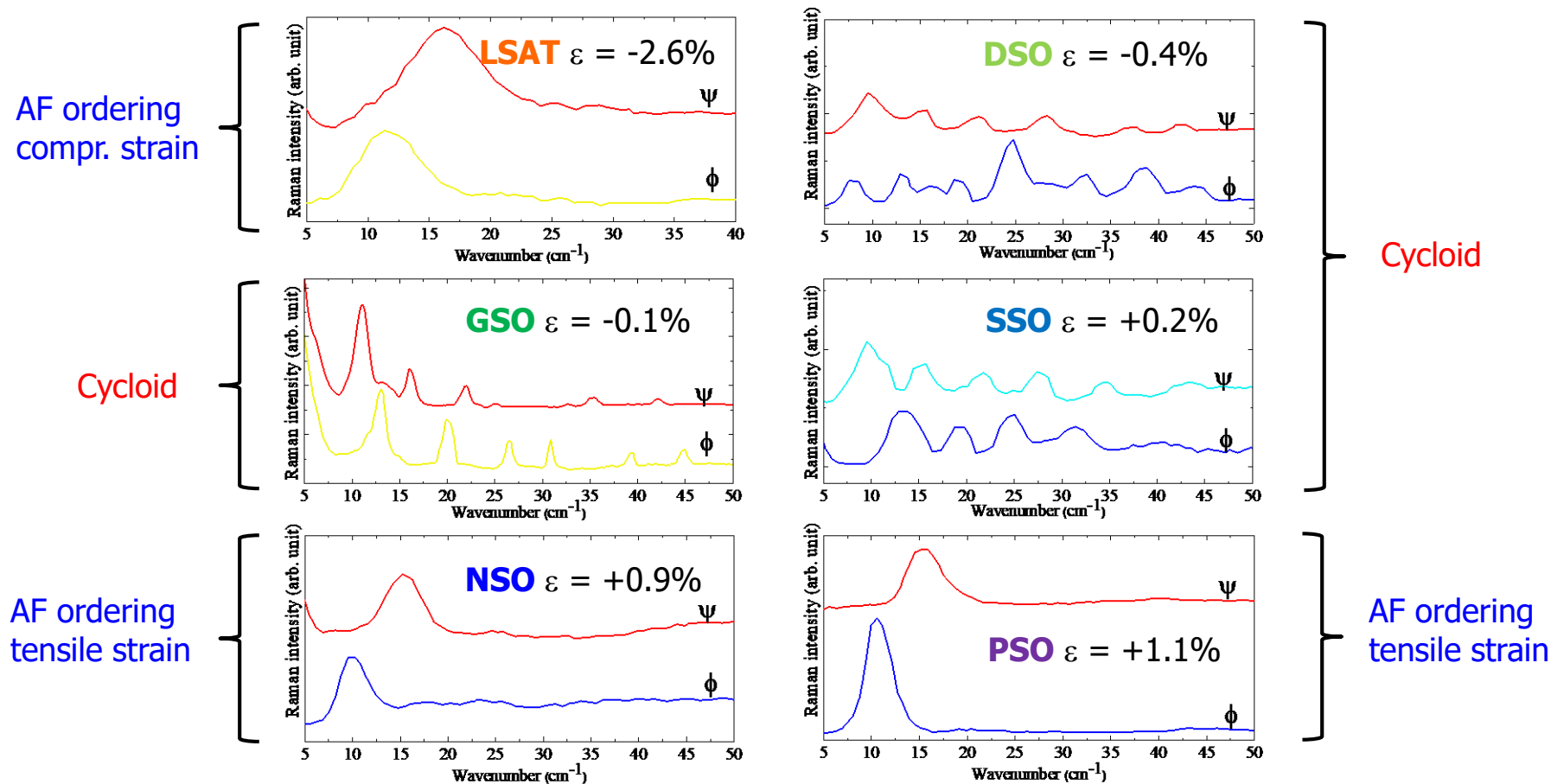
Magnon zone folding

R. de Sousa and J. E. Moore
PRB 77, 12406 (2008)



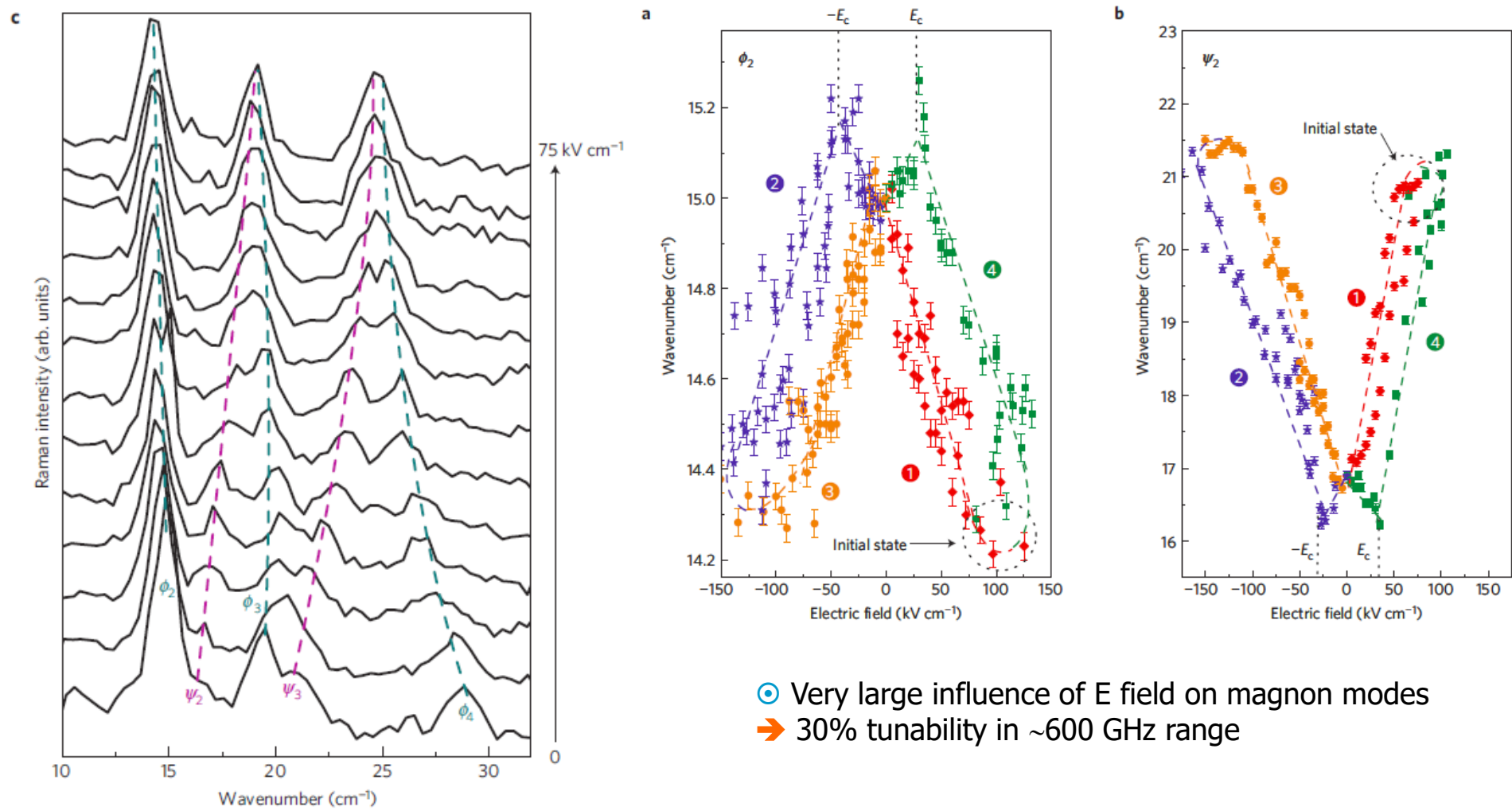
M. Cazayous et al
PRL 101, 037601 (2008)

Raman : magnonic response



- ➔ Confirmation of cycloid at low strain and antiferromagnetic order at large strain
- ➔ Strain controls spin excitations

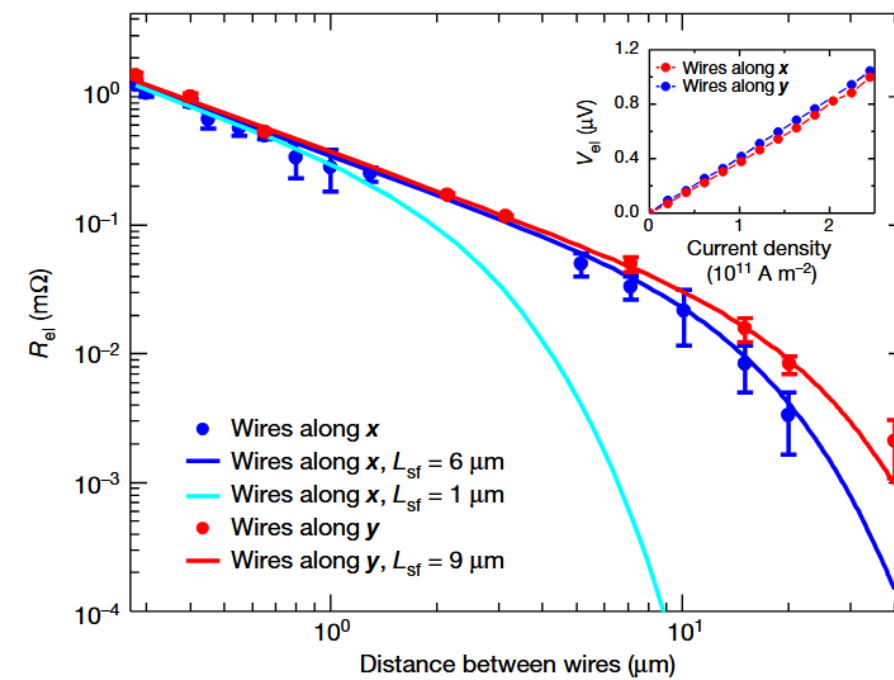
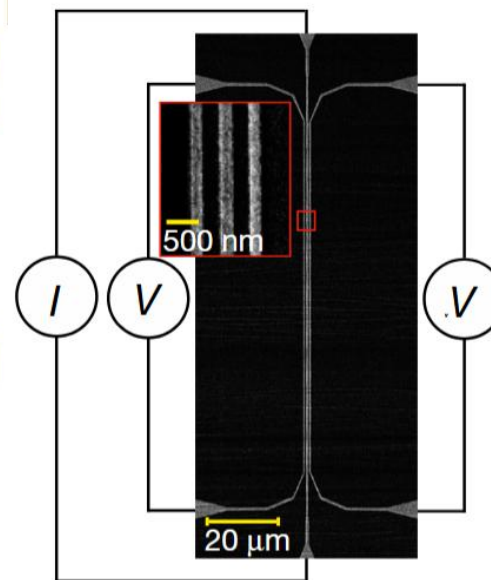
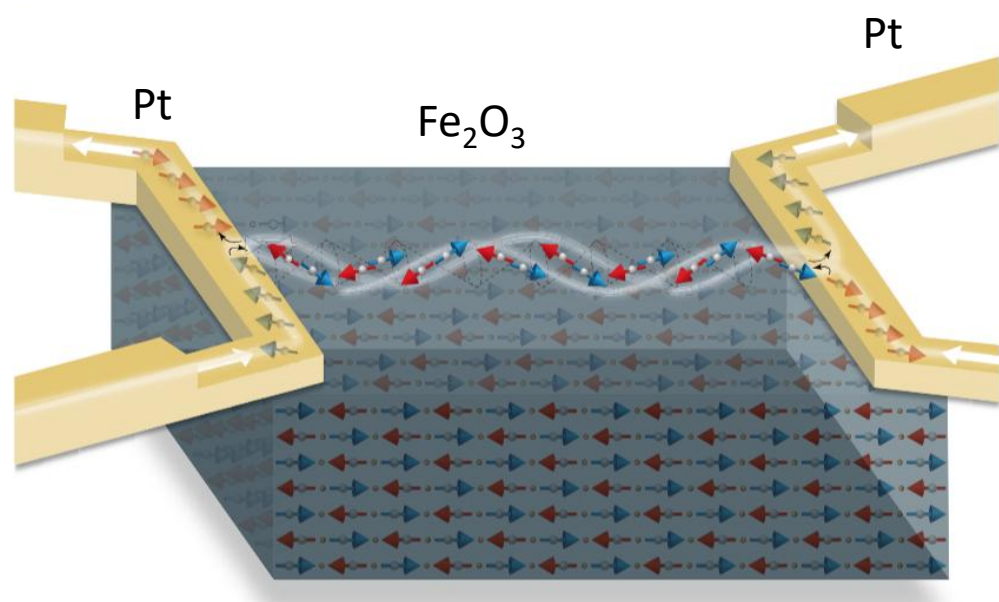
Raman vs Electric field in BiFeO₃ single crystals



- Very large influence of E field on magnon modes
- 30% tunability in ~600 GHz range

P. Rovillain et al, Nature Mater. 9, 975 (2010)

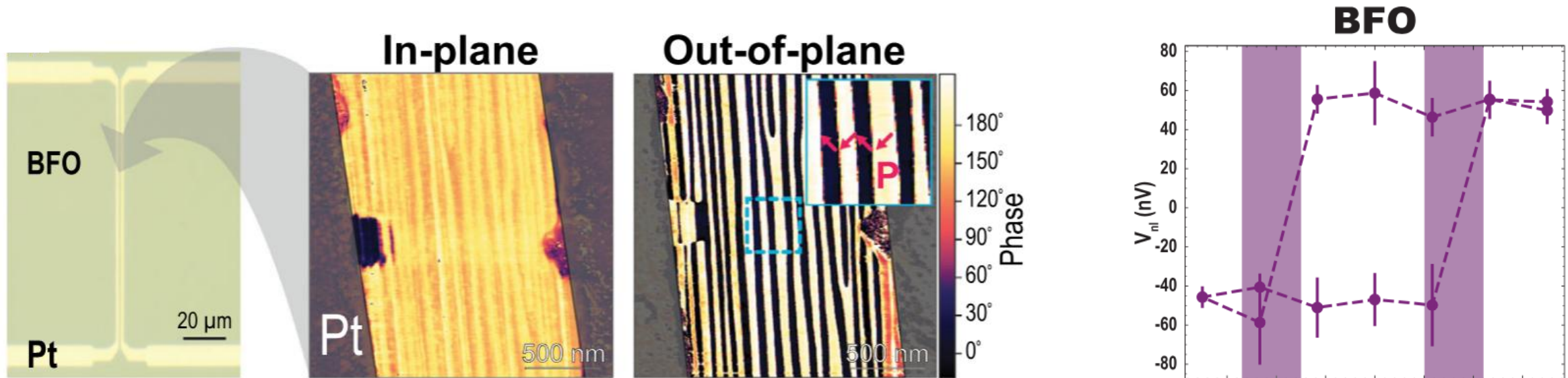
Long-distance magnon transport in antiferromagnets



- ⦿ Pt injector creates spin accumulation via Spin Hall Effect.
- ⦿ Spin accumulation excites antiferromagnetic magnons.
- ⦿ Magnons diffuse through hematite.
- ⦿ Pt detector converts spin current back into voltage.
- ⦿ Diffusion length in the 1-10 μm range

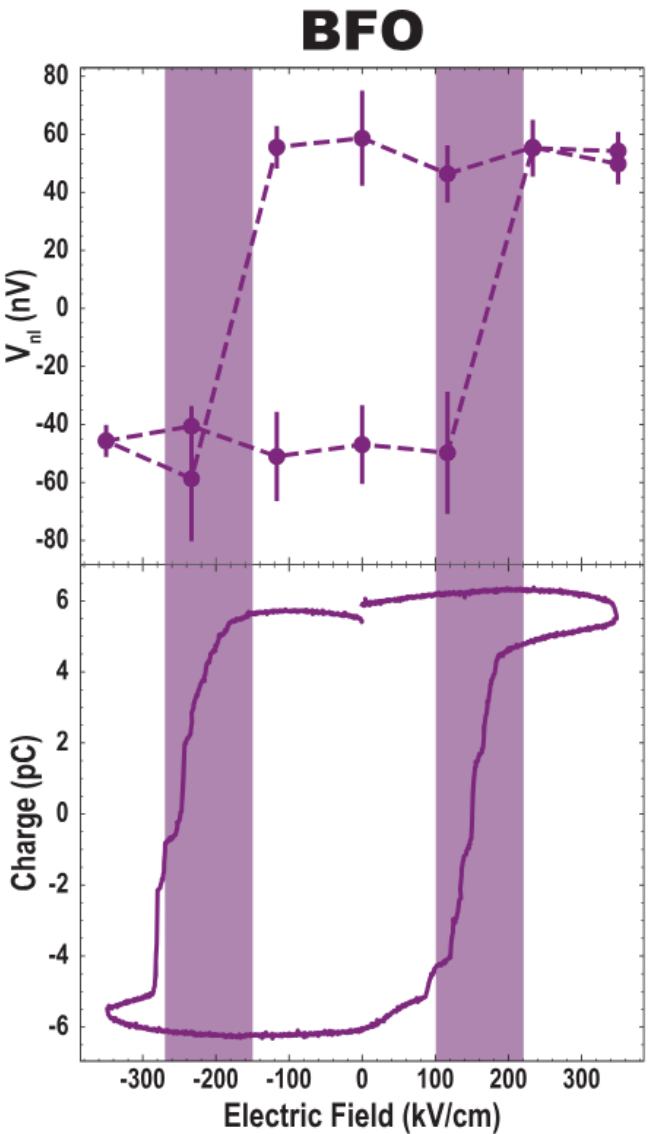
R. Lebrun et al, Nature 561, 222 (2018)

Non volatile electrical control of magnon transport with BiFeO₃ planar structures

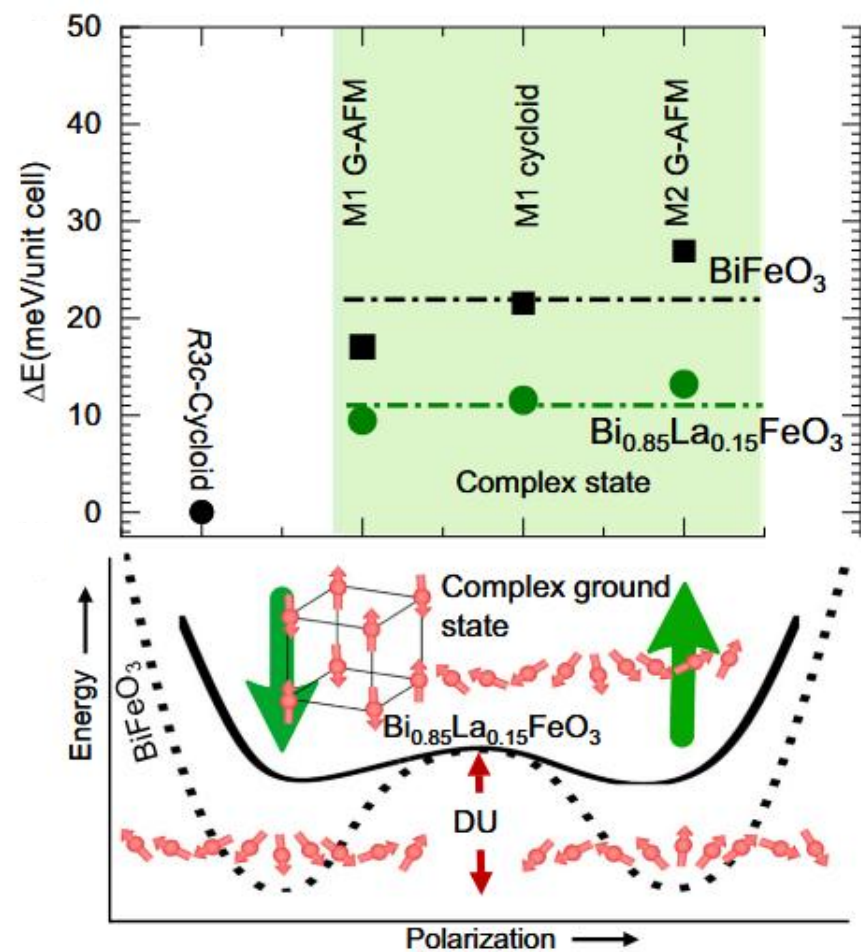


- ⦿ BFO used as magnonic medium
- ⦿ As in Fe₂O₃ experiment Pt wires are used to inject and detect spin
- ⦿ BFO is in multidomain FE and magnetic state
- ⦿ Ferroelectric polarization modifies magnon transport
- ⦿ Nonvolatile electrical switching
- ⦿ Room-temperature operation

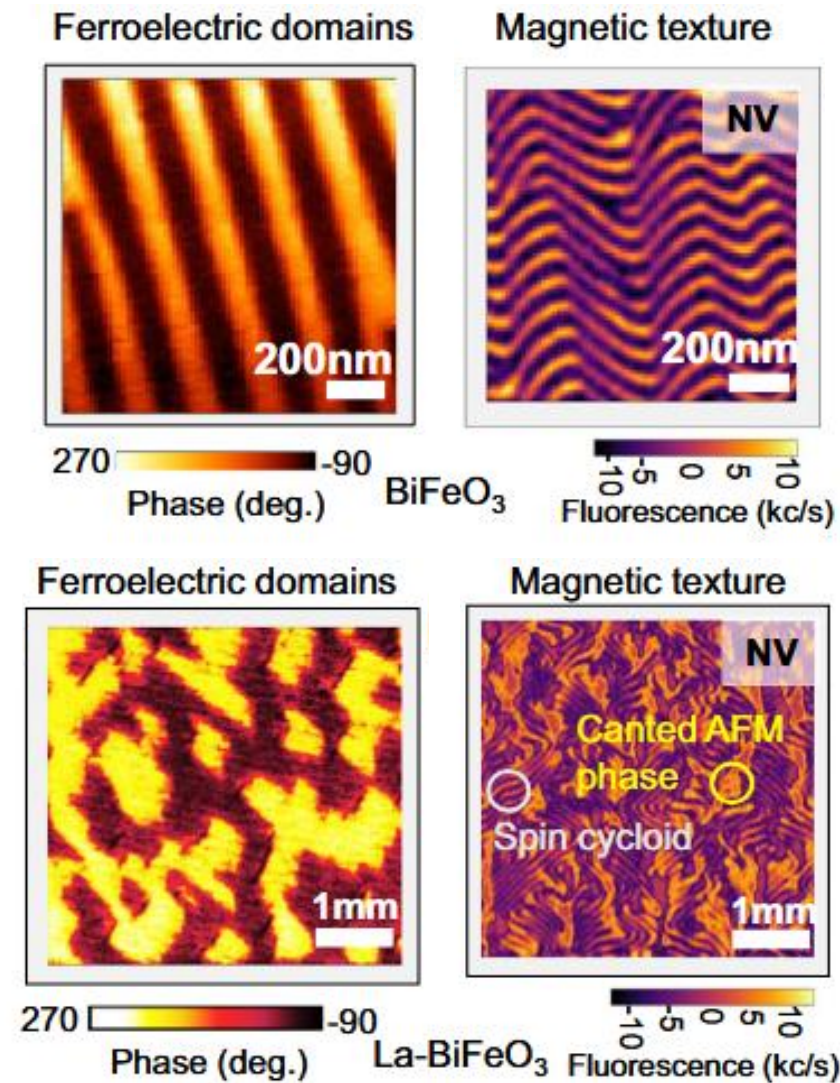
E. Parsonnet et al,
PRL 129, 087601 (2022)



Enhancing signal by engineering BiFeO₃

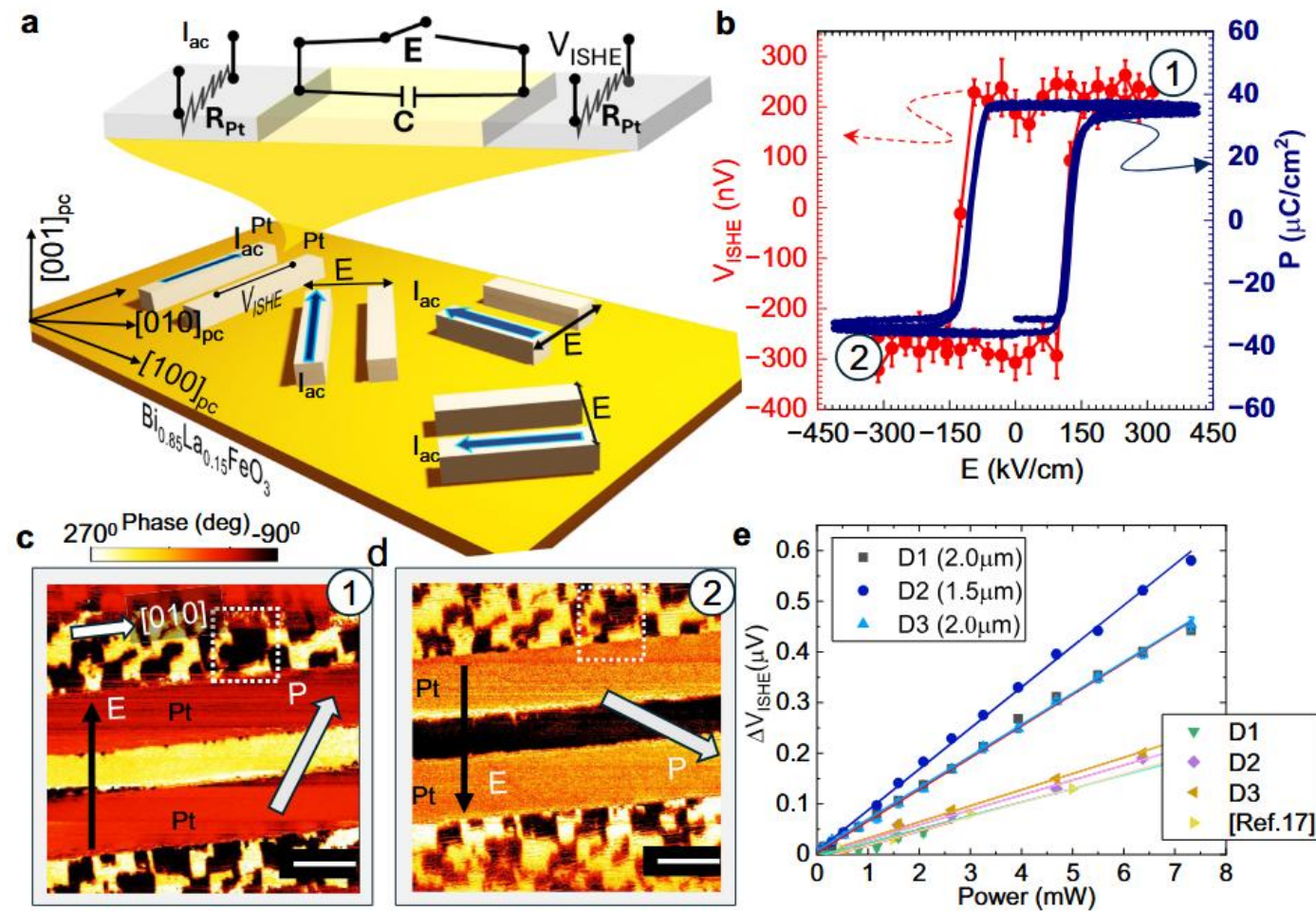


- BFO is engineered by substituting La for Bi (La-BFO)
- Stronger competition between cycloid and canted AFM
- NV microscopy indicates AF + cycloid coexistence



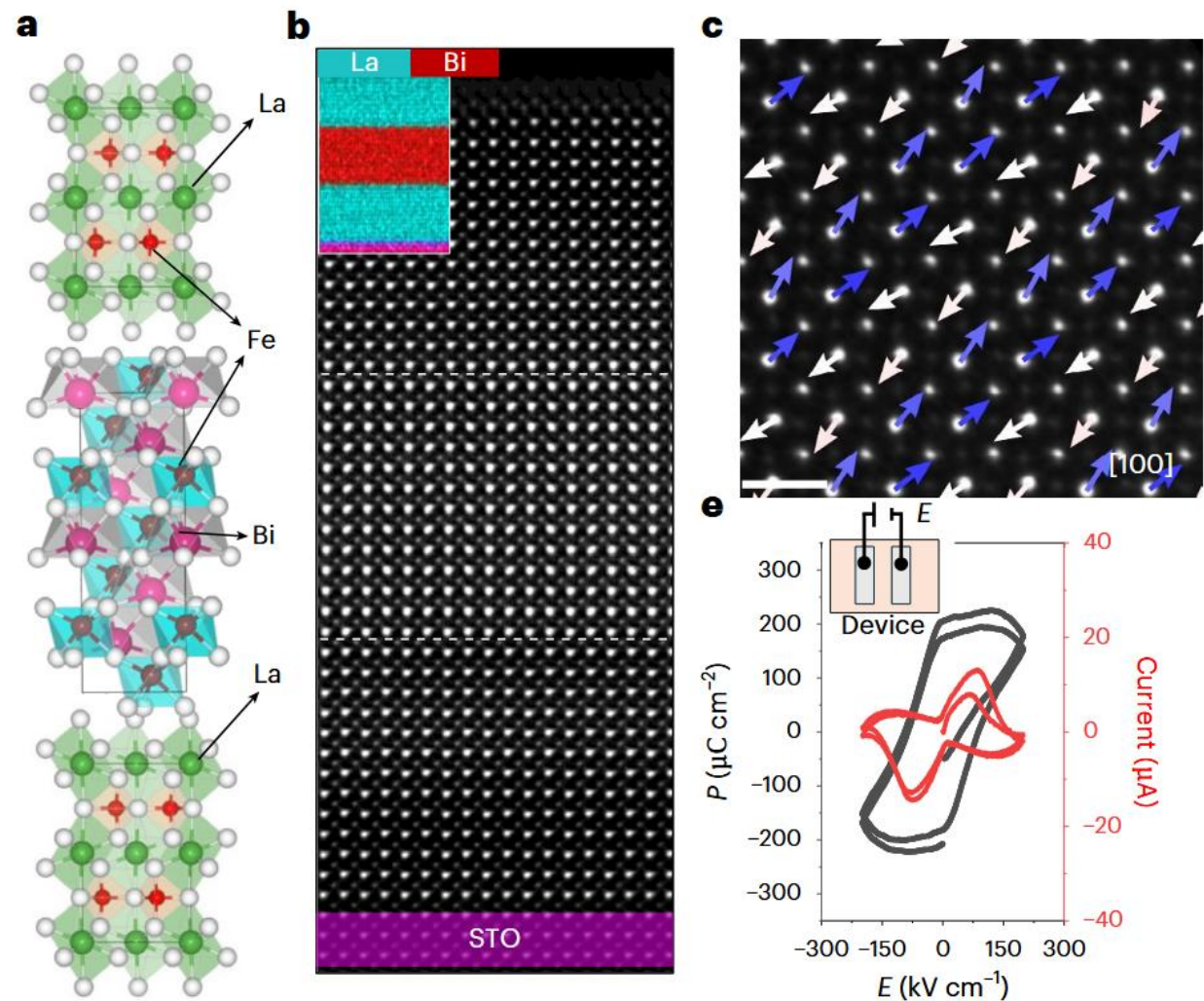
S. Husain et al, Nature Comm. 15, 5966 (2024)

Enhancing signal by engineering BiFeO₃



- ⦿ E field stabilizes single domain state
- ⦿ FE control of spin signal, enhanced over pure BFO case

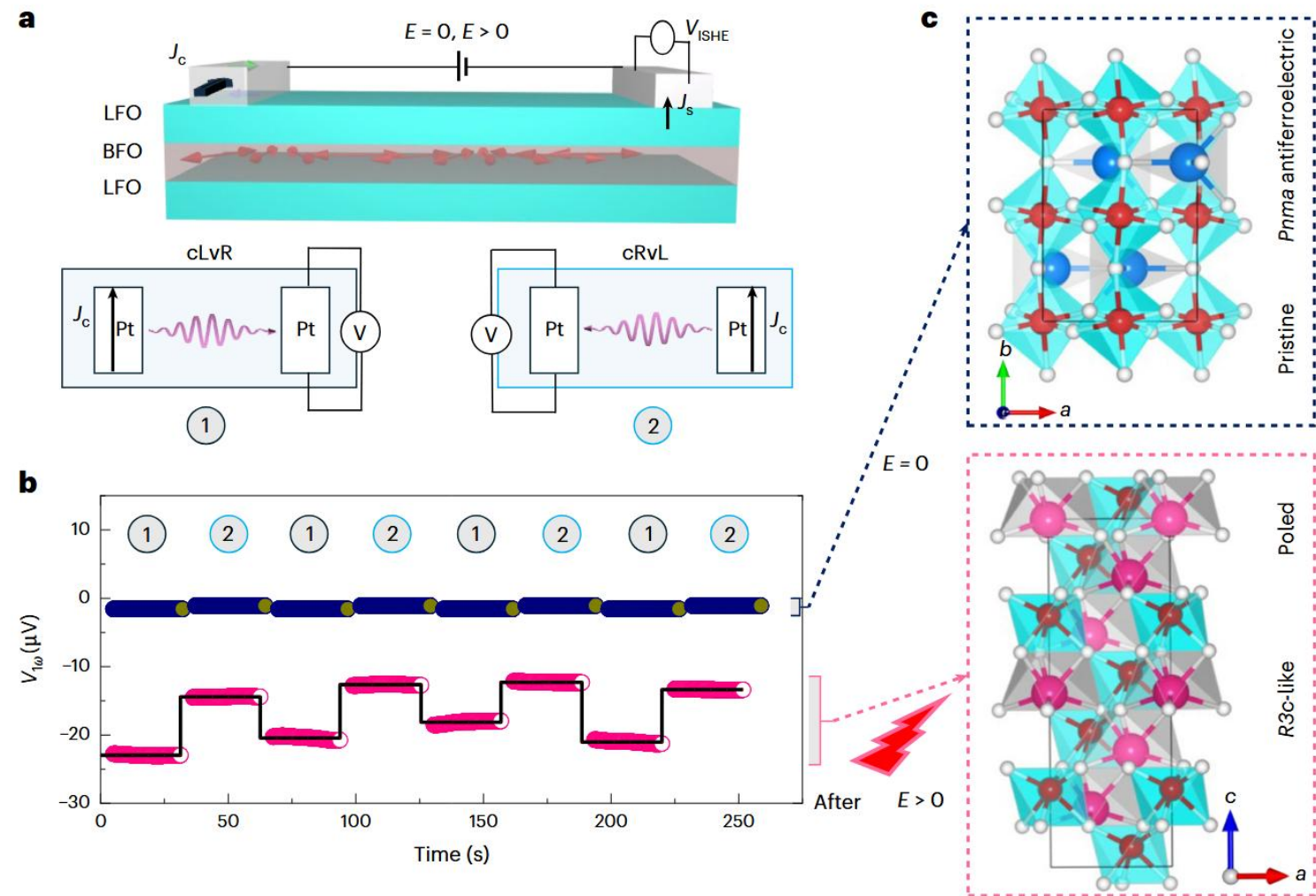
Heterostructuring BiFeO₃ to enhance signal



- ⦿ Sandwiching BFO between 2 LFO layers :
 - Changes BFO ground state from FE to AFE
 - Enables stabilizing FE state after first poling
 - In FE state, stabilizes G-type AFM state

S. Husain et al, Nature Mater. 10.1038/s41563-026-02531-8 (2026)

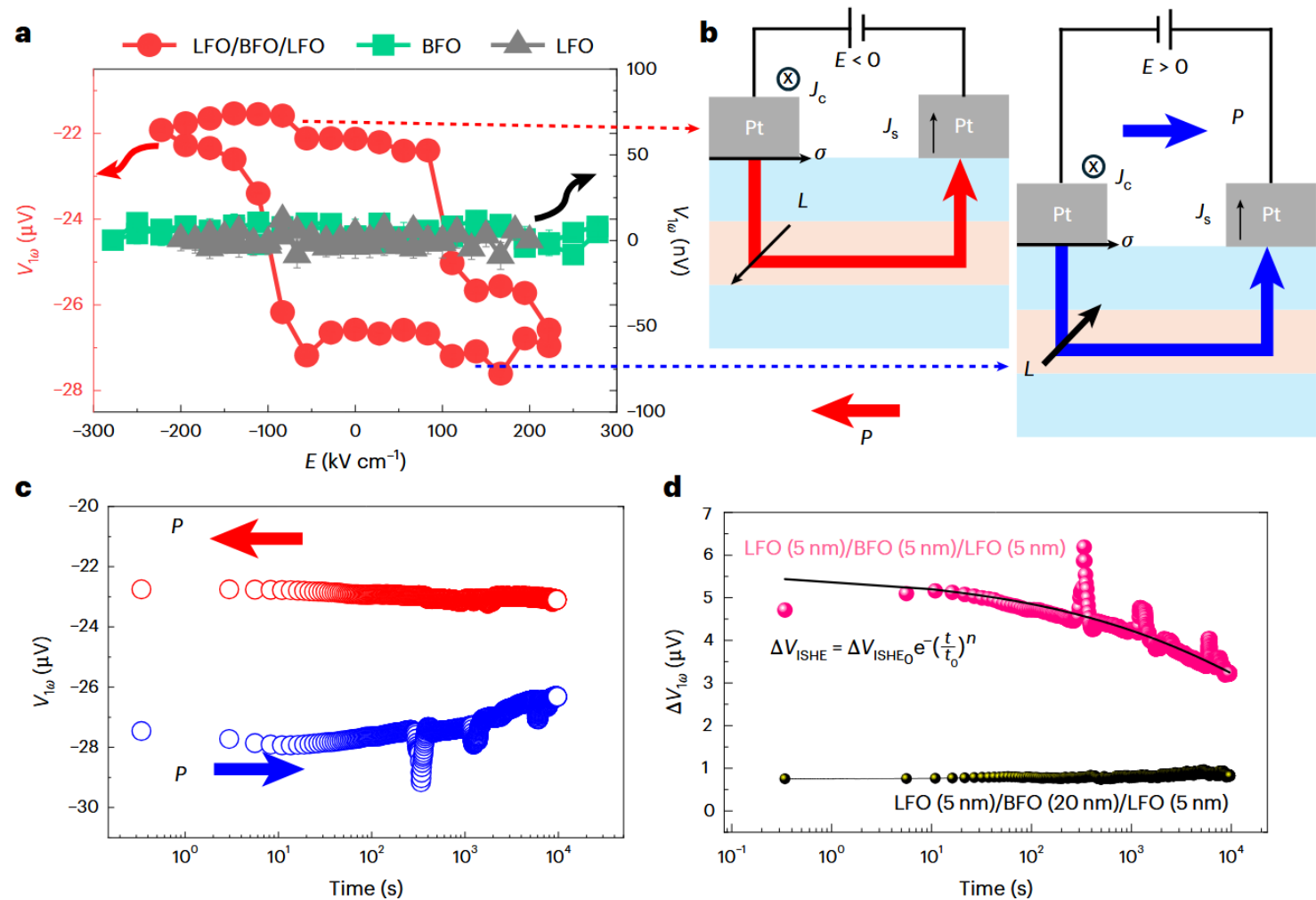
Heterostructuring BiFeO₃ to enhance signal



- ⦿ In virgin state, signal vanishingly small
- ⦿ In poled FE state, signal visible and in μV range

S. Husain et al, Nature Mater. 10.1038/s41563-026-02531-8 (2026)

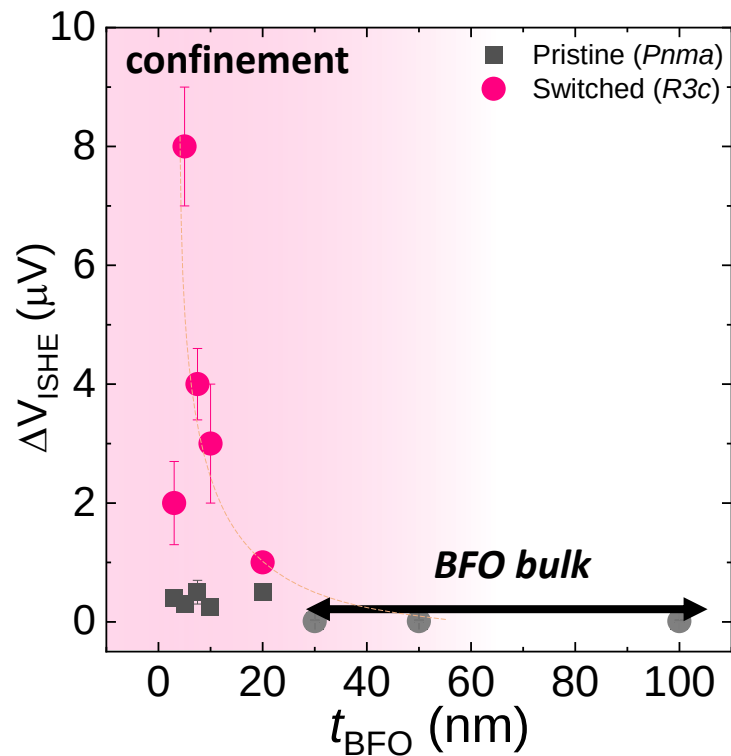
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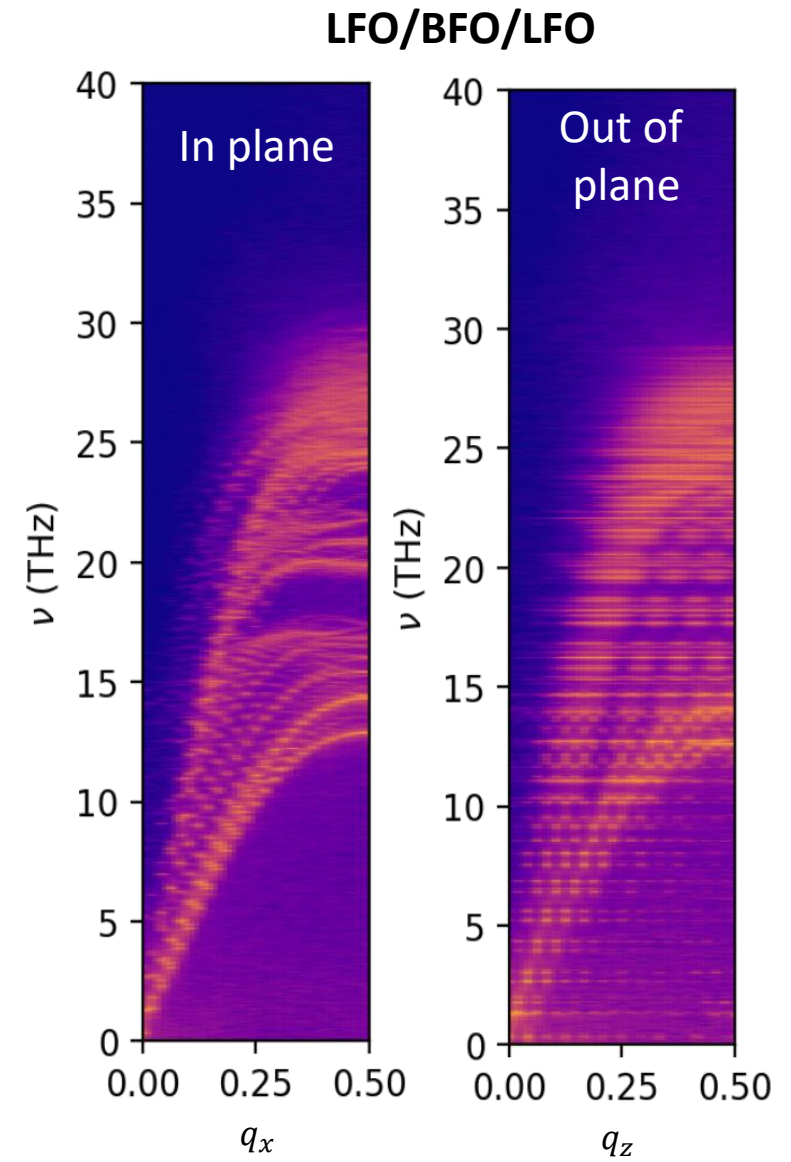
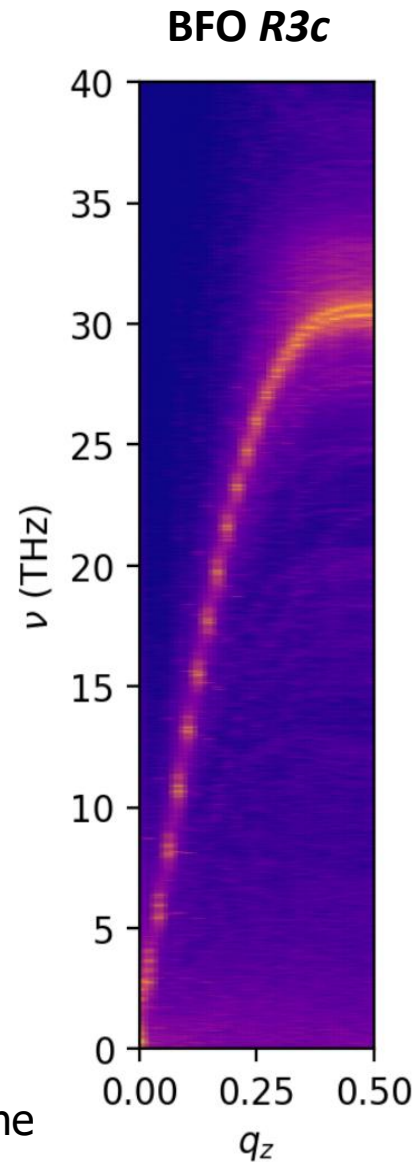
- ⦿ LFO/BFO/LFO trilayers dramatically enhance the spin signal
- ⦿ Nonvolatile electric control is retained

S. Husain et al, *Nature Mater.* 10.1038/s41563-026-02531-8 (2026)

Magnon confinement yields large signal



- ⦿ Signal strongly enhanced at low BFO thickness
- ➔ Magnon confinement ?
- ⦿ Theory shows dispersive in-plane modes but smeared/quantized q_z spectrum
- ➔ Consistent with out-of-plane confinement and in-plane magnon propagation



- ⦿ BiFeO₃ is an extremely rich materials system
- ⦿ Complex magnetic order can be crafted by strain, composition and heterostructuring
- ⦿ Spin wave dynamics can be strongly tuned by electric field
- ⦿ BiFeO₃ is a good medium for long distance magnon transport
- ⦿ Enables non-volatile control of magnon transport

Open questions :

- Exact nature of propagating modes ?
- How to exploit richness of spin textures for ferroelectric magnonics ?
- How to harness hybrid excitations (phonons/magnons/electromagnons/ferrons) ?
- How to couple with the other degrees of freedom available in BFO ?
- etc