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Polar textures in multiferroic BiFeO₃-based superlattices

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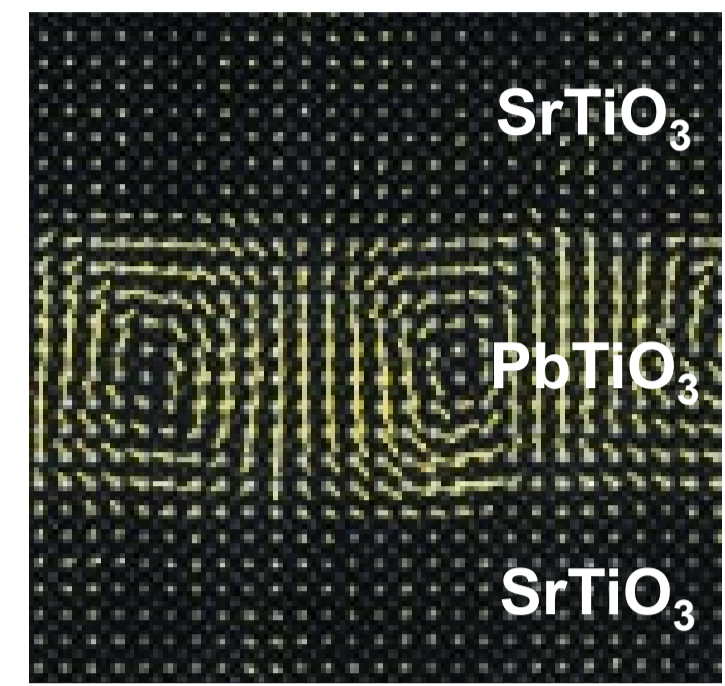
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Context and motivation

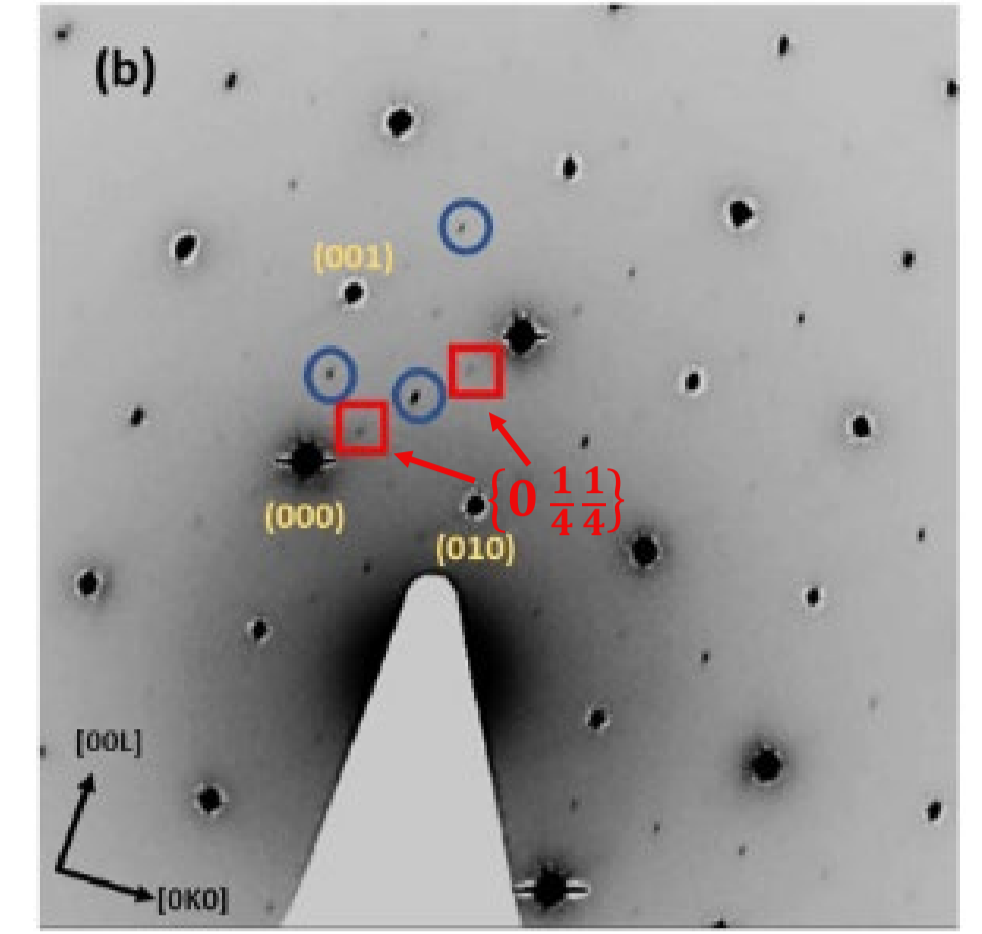
➤ Recently, ferroelectric-based superlattices (SLs) have emerged and led to the discovery of exotic polar textures such as **skyrmionic states** or **vortices** in PbTiO₃/SrTiO₃ SLs [1,2]. Thus, SLs offer a unique and promising new platform to study these novel polar states which result mainly from the effects of strain, confinement, and competing interactions between the degrees of freedom (polar displacements, oxygen tilts) of these perovskite layers.



Vortices polar states observed in PbTiO₃/SrTiO₃ SLs [2].

➤ Previous work on BiFeO₃/LaFeO₃ SLs [3]: coexistence of PbZrO₃-like (red squares) and Pnma-like (blue circles) polar states depending on the BFO thickness in the period of SLs.

➤ **Objective:** control the polar states of the BFO by adjusting various parameters: **thickness** of the bilayer (Λ), type of **substrate**, type of **intermediate layer**, etc.



SAED of BFO/LFO SL obtained for $\Lambda = 150$ Å [3].

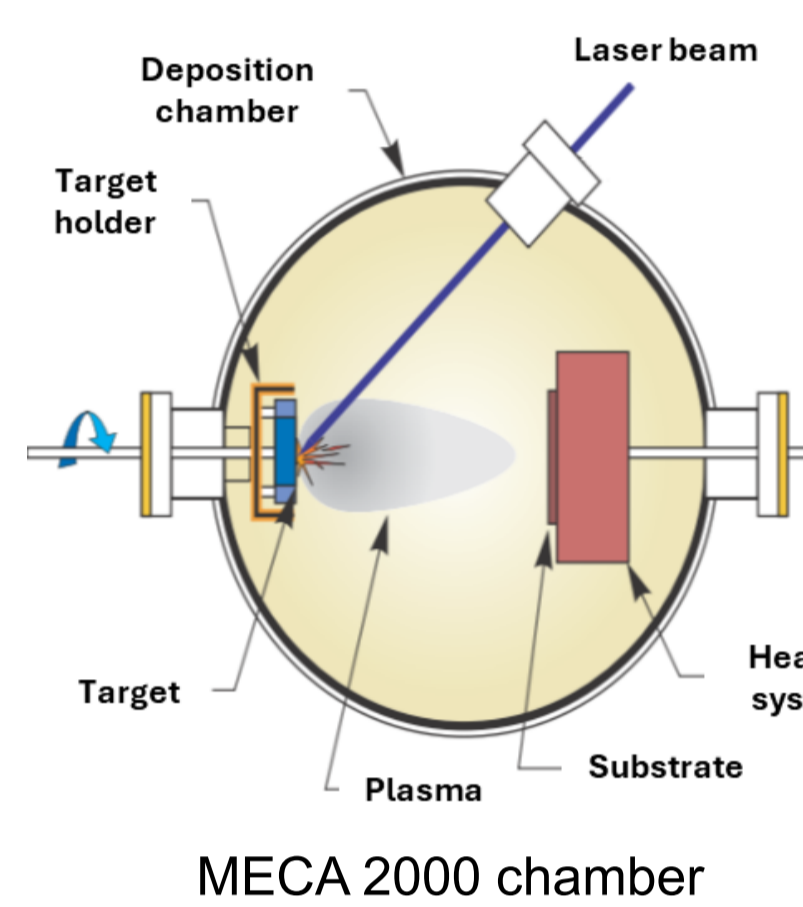
SL system and growth method

➤ SLs grown by PLD on (001) SrTiO₃ and DyScO₃ single crystals.

➤ LaFeO₃ and NdFeO₃ used as intermediate layers.

➤ Various periods Λ (3, 6, 12 and 20 nm) and same BFO/LFO (NFO) thickness ratio.

➤ Deposition characteristics: 0.5 mbar of pO₂ at 740°C, 6 Hz repetition rate using a KrF laser (248 nm).



Characterization methods

➤ **Cross-section TEM** samples prepared on an FEI Thermofisher Helios Nanolab 660.

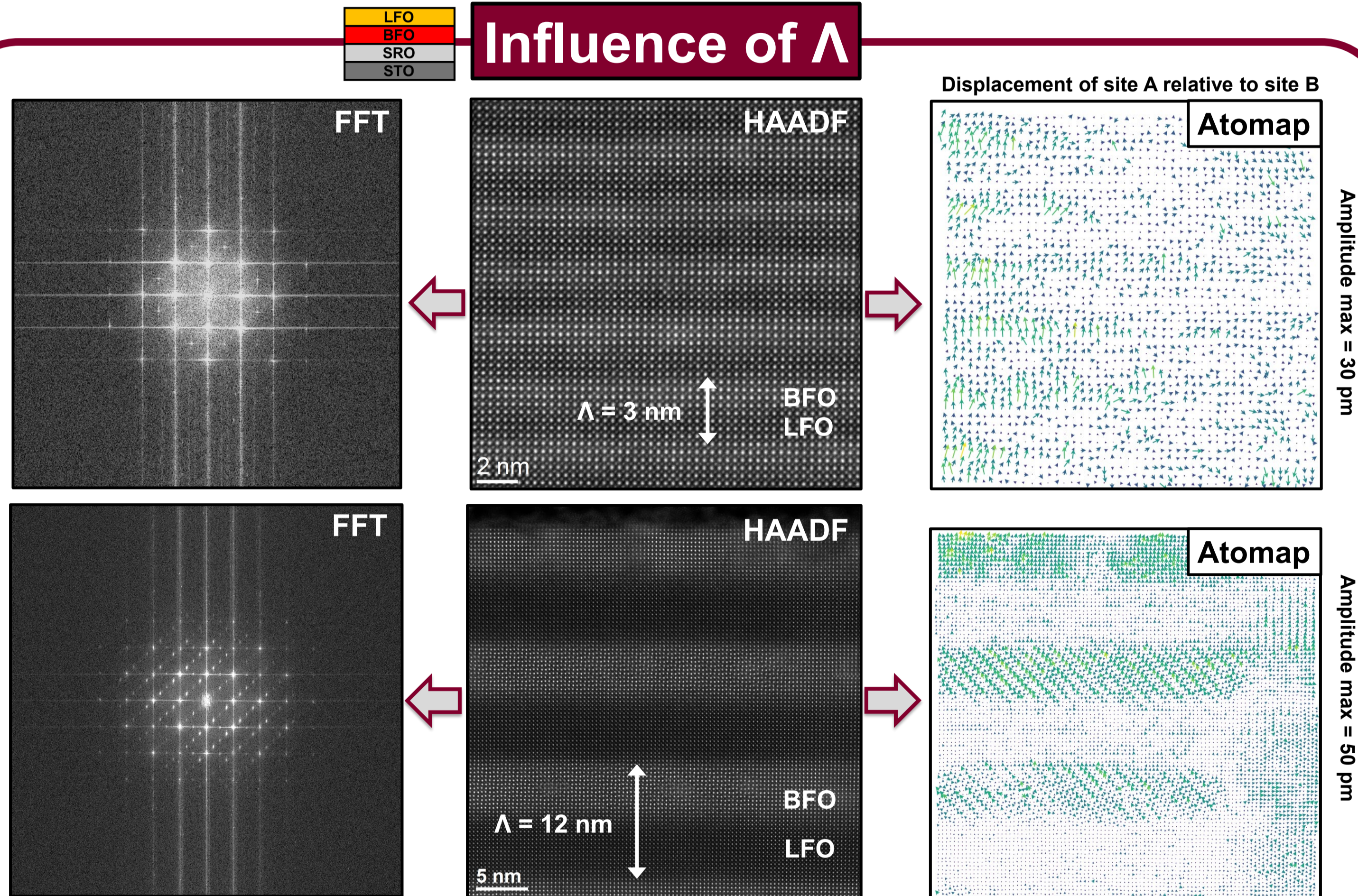
➤ **ABF and HAADF STEM** images acquired on FEI Titan 3 G2 80-300 microscope (Cs probe corrector).

➤ Analysis of atomic displacements by **Atomap** [4,5] and a "render-and-compare" (**RAC**) method based on **Principle Component Analysis (PCA)**.



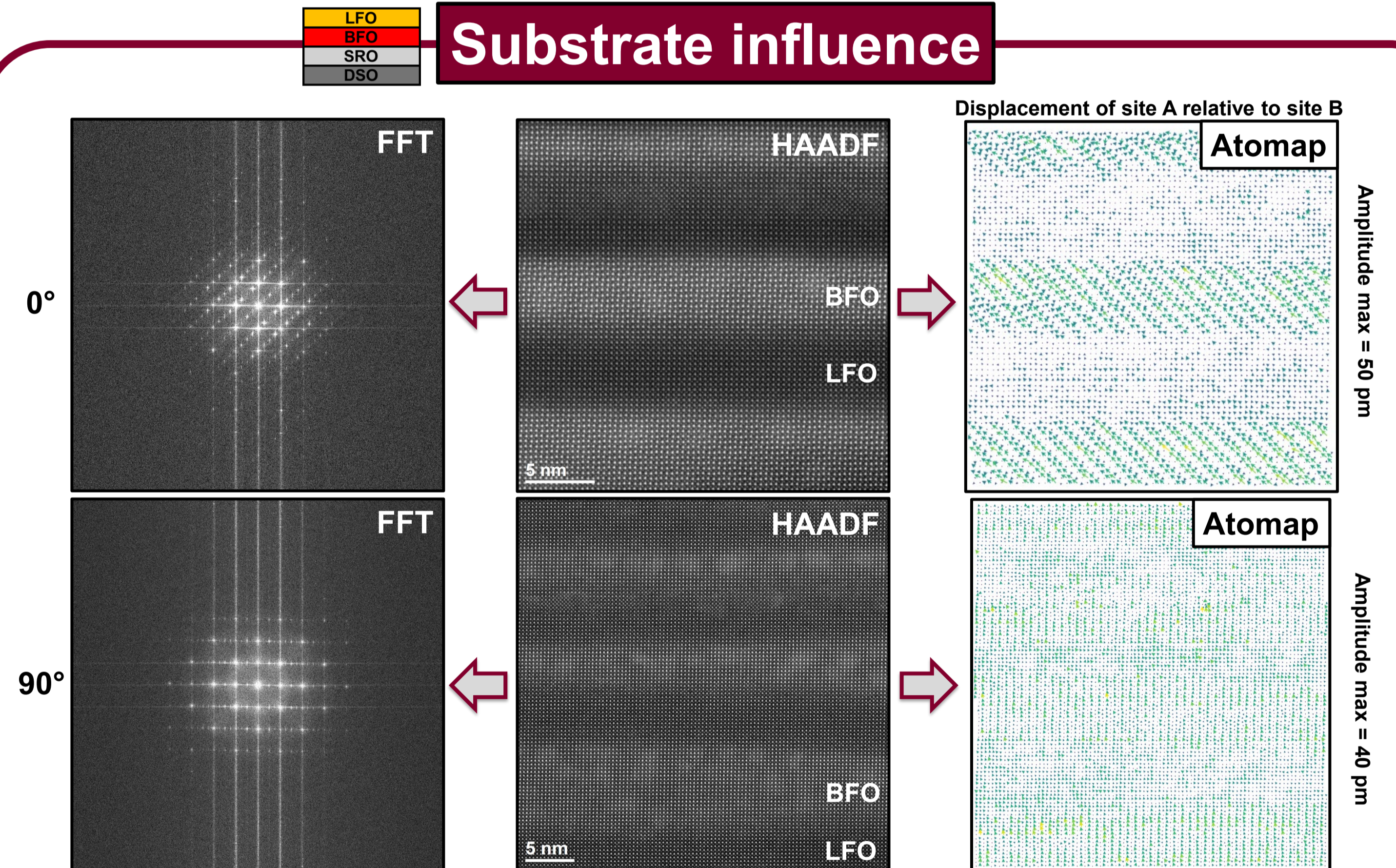
FEI Titan 3 G2

Influence of Λ



➤ Depending on Λ , the BFO polar states can be switched to **antiferro-like** (step-like).

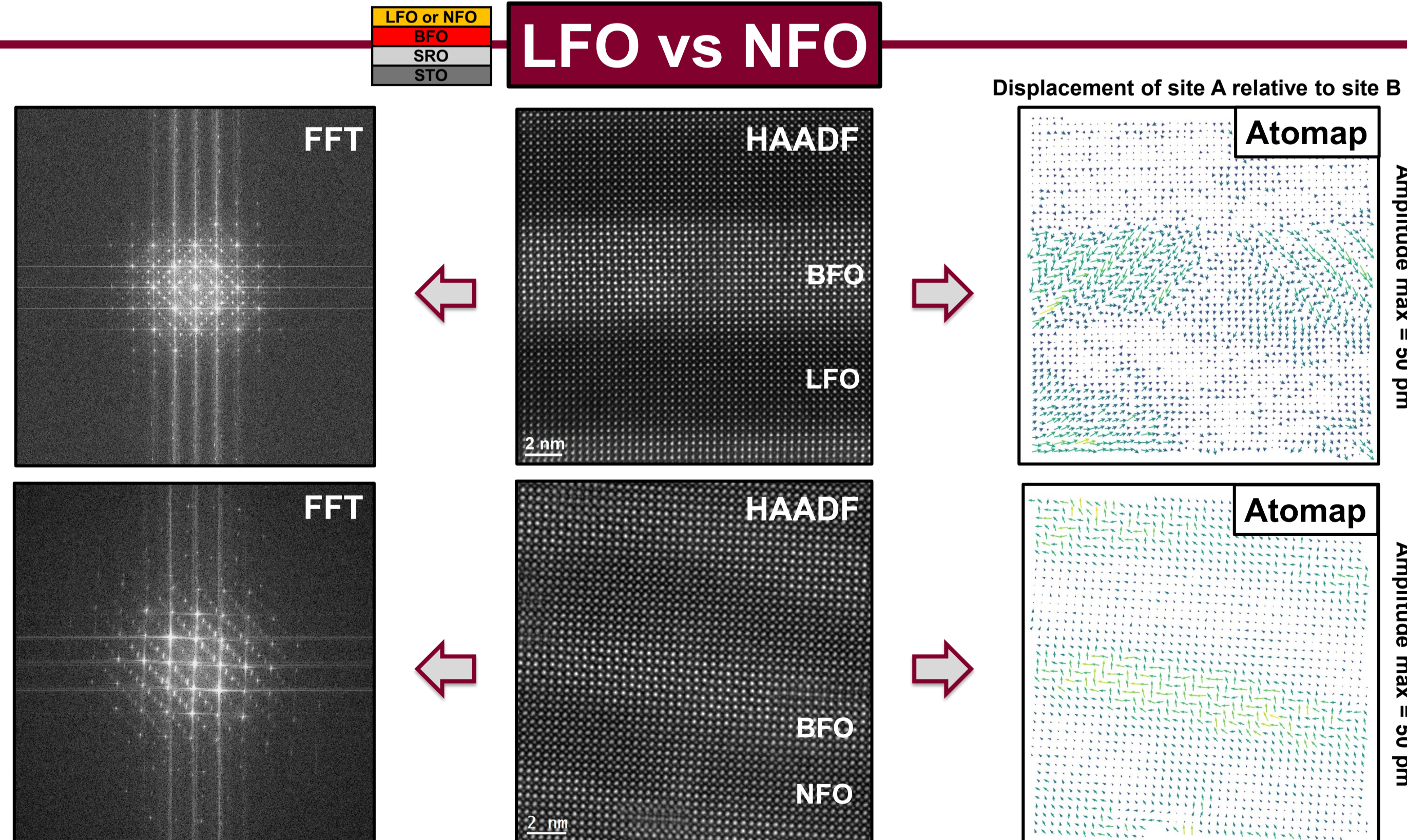
Substrate influence



➤ **Antiferro-like** polar states are still visible using **anisotropic DyScO₃** as a substrate.

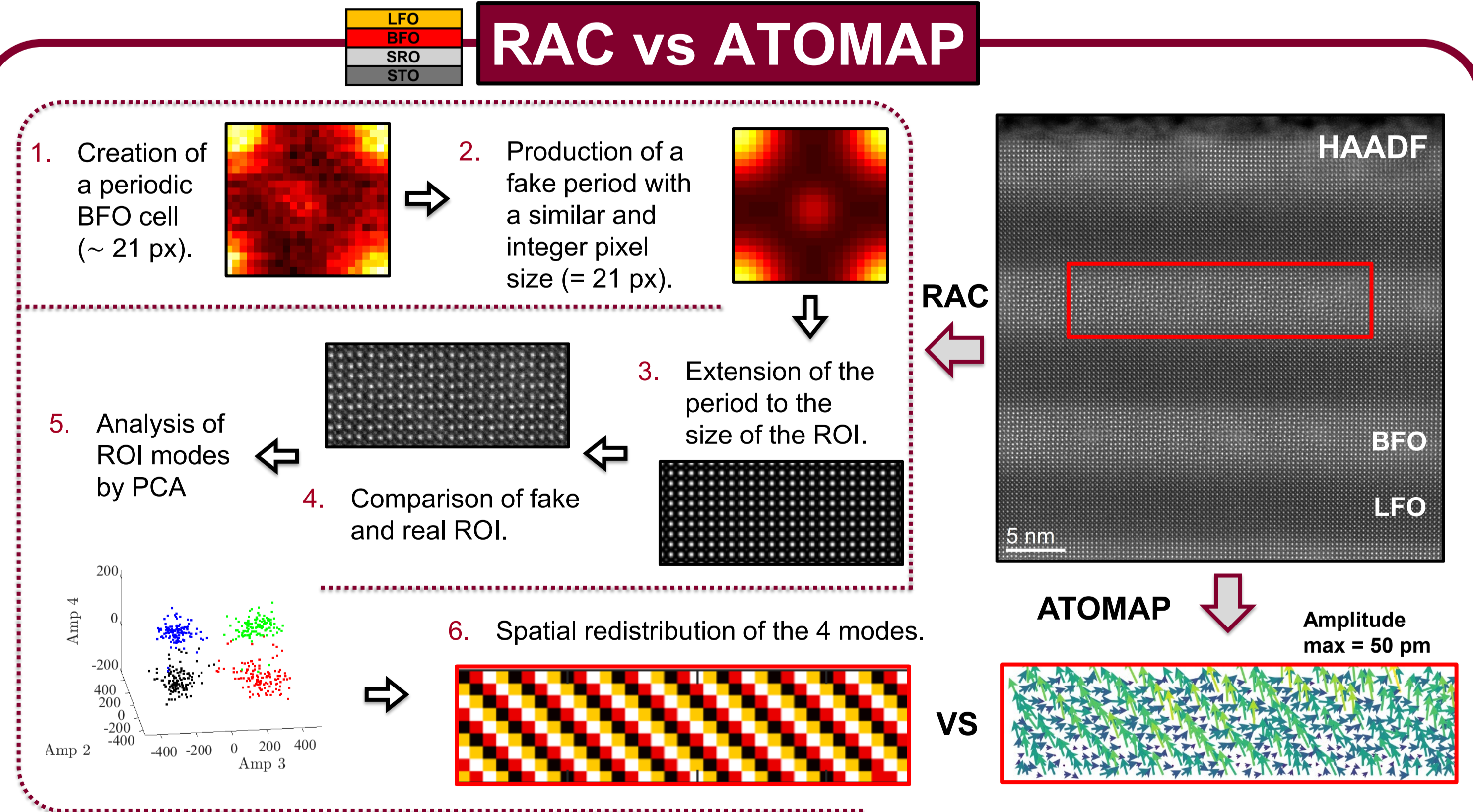
➤ The BFO appears to be **more ordered**, with a **single variant** of polar step-like states.

LFO vs NFO



➤ **Antiferro-like** polar states are observed **regardless** of the intermediate layer used.

RAC vs ATOMAP



➤ In this example, **RAC** shows a more **complex polar distribution** (4 different displacements).

Conclusion

➤ **BFO** exhibits **antiferro-like polar states** regardless of the substrate or intermediate layer used, provided that Λ is **sufficiently high** (> 3 nm).

➤ The use of **DyScO₃** as a substrate seems more appropriate for **controlling** the polar states of the BFO.

➤ **RAC** can be considered as a complementary **analysis tool** for understanding the complexity of polar states.

Outlooks

➤ Understand the crystalline orientation relationships between the BFO and the intermediate layers (**observation of O sites**).

➤ Continue to **develop and optimize** the **Matlab code** for the **RAC** method (analysis of oxygen sites, etc).

References

- [1] Das, S. et al. Nature 568, 368–372 (2019).
[2] Yadav, A. et al. Nature 530, 198–201 (2016).
[3] B. Carcan, H. Bouyanfif, et al. J. Appl. Phys. 124(4), 044105 (2018).
[4] Nord, M. et al. Adv Struct Chem Imag 3, 9 (2017).
[5] E. O'Connell, M. Hennessy, and E. Moynihan, "PinkShnack/TEMUL: Version 0.1.3." (2021) 10.5281/zenodo.4543963.