

# Unveiling Unconventional Ferroelectric Switching in Multiferroic $\text{Ga}_{0.6}\text{Fe}_{1.4}\text{O}_3$ Thin Films Through Multiscale Electron Microscopy Investigations

*Anna Demchenko<sup>a</sup>, Suvidyakumar Homkar<sup>a</sup>, Corinne Bouillet<sup>a</sup>, Christophe Lefèvre<sup>a</sup>, François Roulland<sup>a</sup>, Daniele Preziosi<sup>a</sup>, Gilles Versini<sup>a</sup>, Cédric Leuvrey<sup>a</sup>, Philippe Boullay<sup>b,\*</sup>, Xavier Devaux<sup>c,\*</sup>, and Nathalie Viart<sup>a,\*</sup>*

<sup>a</sup> Université de Strasbourg, CNRS, IPCMS, UMR 7504, 67000 Strasbourg, France

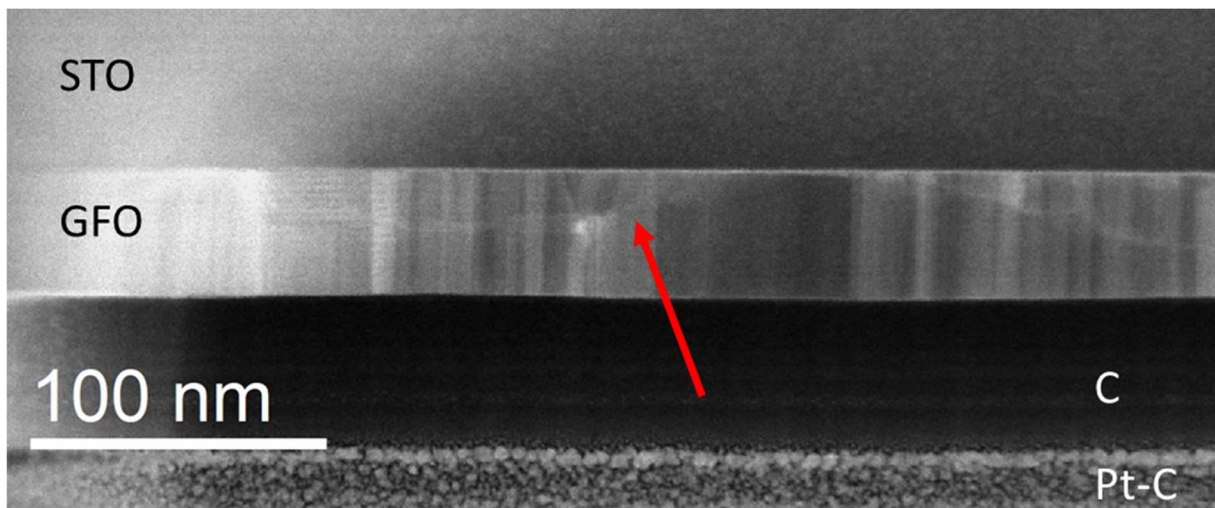
<sup>b</sup> Normandie Université, ENSICAEN, UNICAEN, CNRS, CRISMAT, 14050 Caen, France

<sup>c</sup> Université de Lorraine, CNRS, IJL, 54000 Nancy, France

Corresponding authors: viart@unistra.fr, philippe.boullay@ensicaen.fr, xavier.devaux@univ-lorraine.fr

## Supplementary Information

SI 1



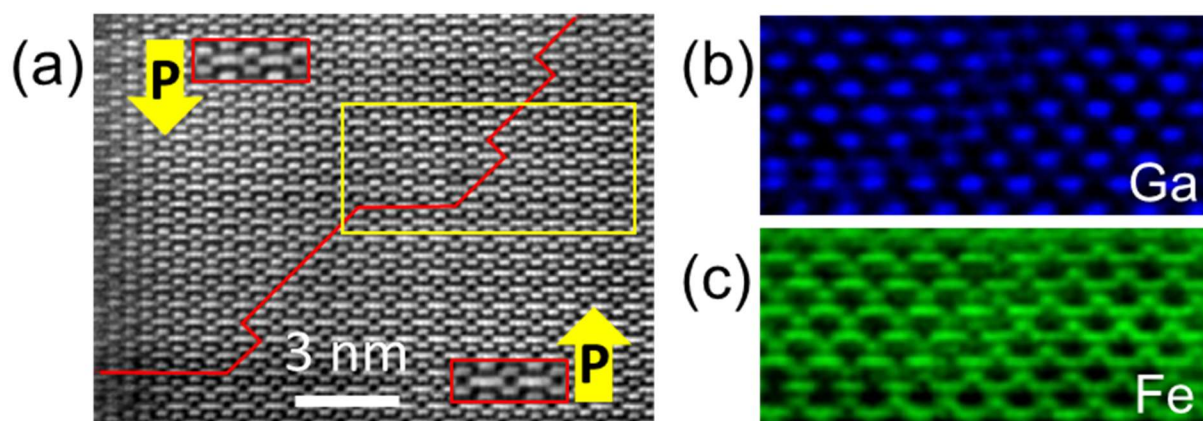
**Figure S1.** Low magnification STEM-ADF image recorded with a collection half-angle range of 30-68 mrad. The arrow points to the area where the images presented in Figures 7 and 8 were recorded.

Polarisation reversal walls, similarly to stacking faults or dislocations, can easily be imaged at low magnification in STEM because a white extra contrast appears on STEM annular dark-field when the area of interest is weakly misoriented as in conventional TEM weak-beam dark-field

imaging [1]. For GFO, only HR imaging allows understanding the nature of the defects, and distinguishing an extended stacking fault from a polarisation reversal wall. **Figure S1** shows a low magnification STEM-ADF image of GFO grown onto STO (111). C and Pt layers were deposited on top of the GFO film for FIB processing (visible at the bottom of the image). The white line that crosses the GFO layer from left to right is due to a long polarisation-reversal wall that crosses many columnar grains. The arrow points to the area of the face-to-face polarisation domain wall that was studied at high-resolution by STEM.

## SI 2

A second spatially resolved EELS spectrum image (SI) was recorded on the central part of the domain wall (DW), after the recording of the EELS SI presented **Figure 8**, in the area where there was previously a clear shift of the wall (see **Figure 7**). It should be noted that the HAADF survey image presented in **Figure S2a** was also recorded after the one presented in **Figure 8**. No chemical trace of the wall-shift is visible on the chemical maps, as observed for **Figure 8**. The cationic disorder appears only in the DW part oriented at 45° degrees with respect to the growth axis of the layer, as shown in the Ga (Fig. S2b) and Fe (Fig. S2c) maps.



**Figure S2.** a) High resolution STEM-HAADF. The inset magnification of the image shows the local polarisation of the structure. An EELS SI image was recorded in the yellow framed-area. Chemical maps of Ga b) and Fe c) were extracted by processing the EELS-SI.

- [1] P.J. Phillips, M.C. Brandes, M.J. Mills, M. De Graef, Diffraction contrast STEM of dislocations: Imaging and simulations, *Ultramicroscopy*. 111 (2011) 1483–1487. <https://doi.org/10.1016/j.ultramicro.2011.07.001>.