



HAL
open science

Ferroelectric phase transitions of antiferroelectric PbZrO₃ thin films Electrical measurements

Maxime Vallet, Pauline Dufour, Thomas Maroutian, Lluis Yedra, Stéphane Fusil, Brahim Dkhil, Vincent Garcia

► **To cite this version:**

Maxime Vallet, Pauline Dufour, Thomas Maroutian, Lluis Yedra, Stéphane Fusil, et al.. Ferroelectric phase transitions of antiferroelectric PbZrO₃ thin films Electrical measurements. 18ème colloque de la Société française des Microscopies, Jul 2023, Rouen, France. hal-04226974

HAL Id: hal-04226974

<https://centralesupelec.hal.science/hal-04226974>

Submitted on 3 Oct 2023

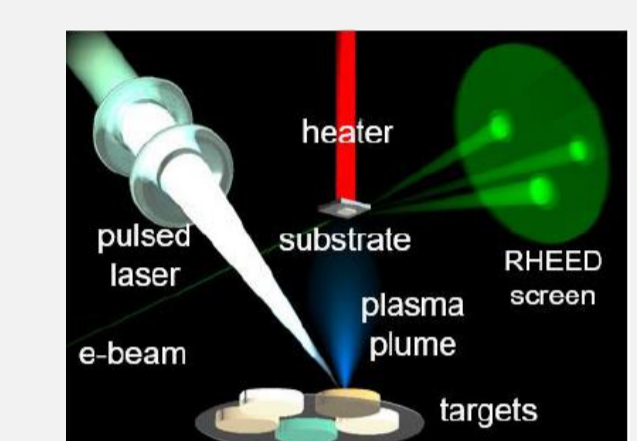
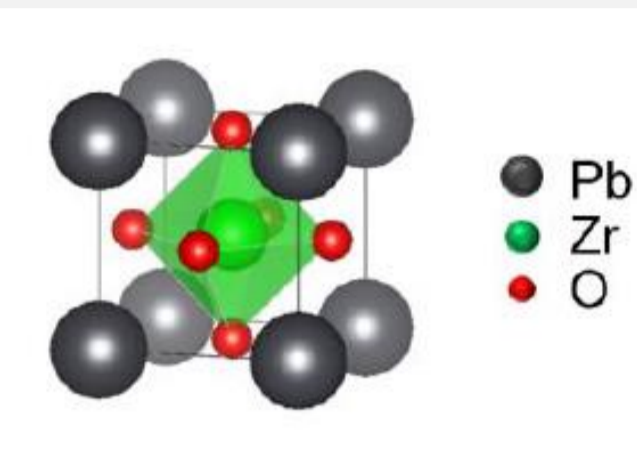
HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

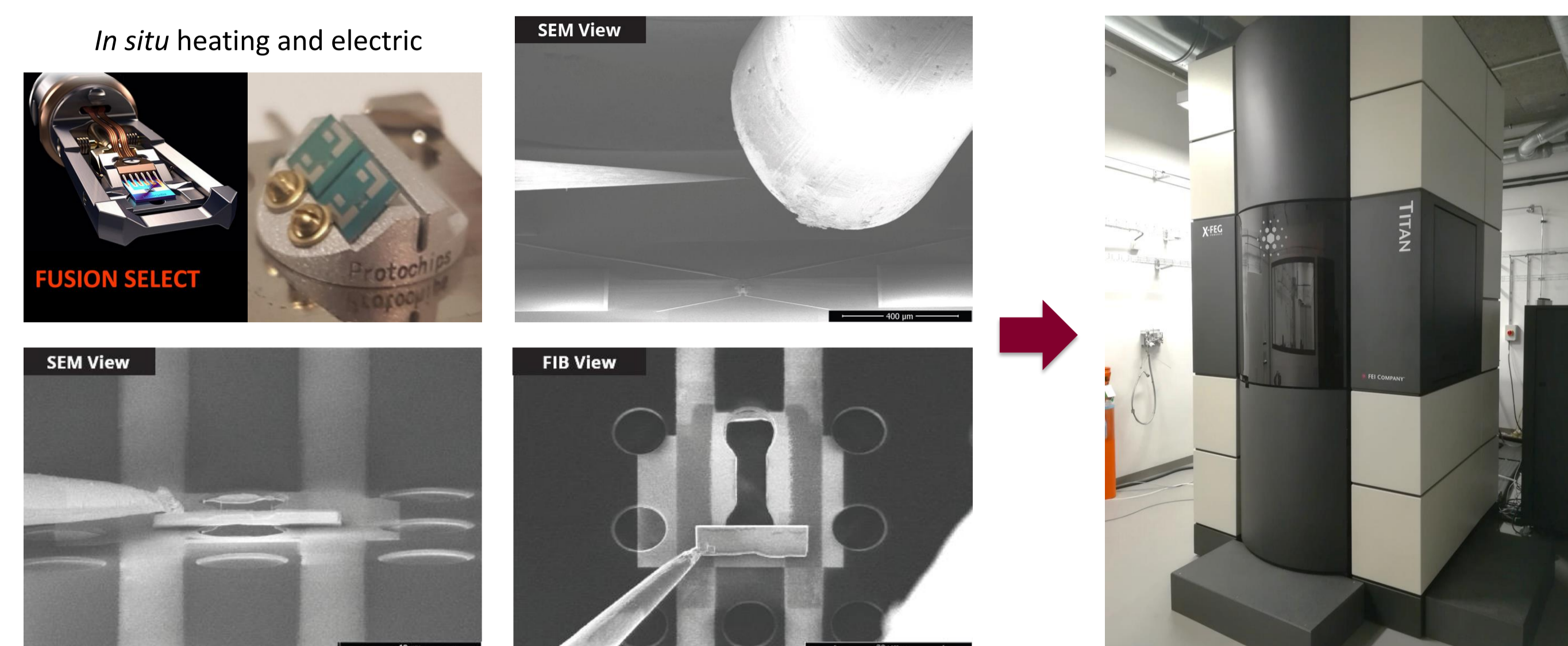
Challenge and growth

The archetypical antiferroelectric, PbZrO₃, is currently attracting a lot of interest, but no consensus can be clearly established on the nature of its ground state as well as on the influence of external stimuli over its physical properties. The debate is even more controversial in epitaxial thin films for which the epitaxial strain can affect the stabilization of the PbZrO₃.

In this work, 45-nm-thick epitaxial PbZrO₃ thin films were grown by pulsed laser deposition on orthorhombic GdScO₃ (110)_o. To enable electric-field measurements, a pseudo-cubic Ba_{0.5}Sr_{0.5}RuO₃ thin films was grown as bottom oxide electrodes.



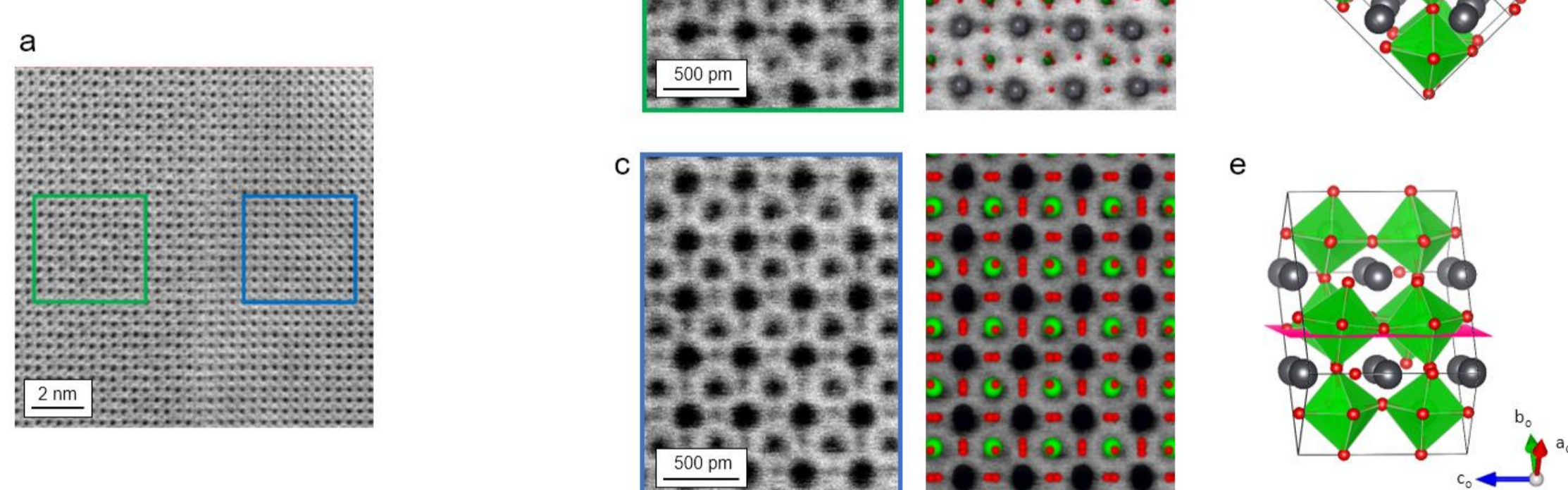
Sample preparation by FIB-SEM



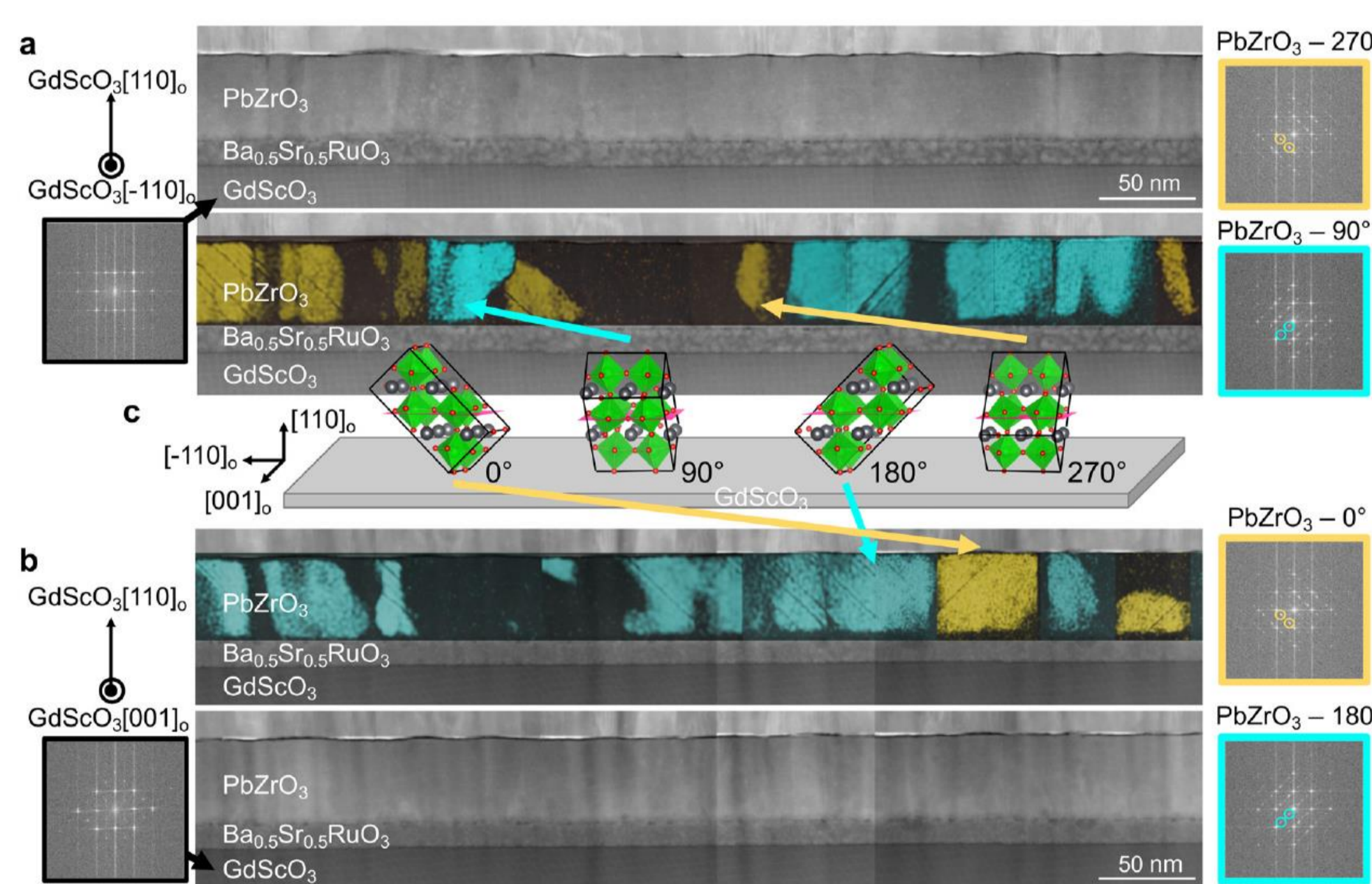
In situ atomic STEM monitoring of phase change as a function of the temperature

Room temperature analyses

Atomic scale of the antiferroelectric feature of PbZrO₃ across a domain boundary observed in ABF-STEM mode

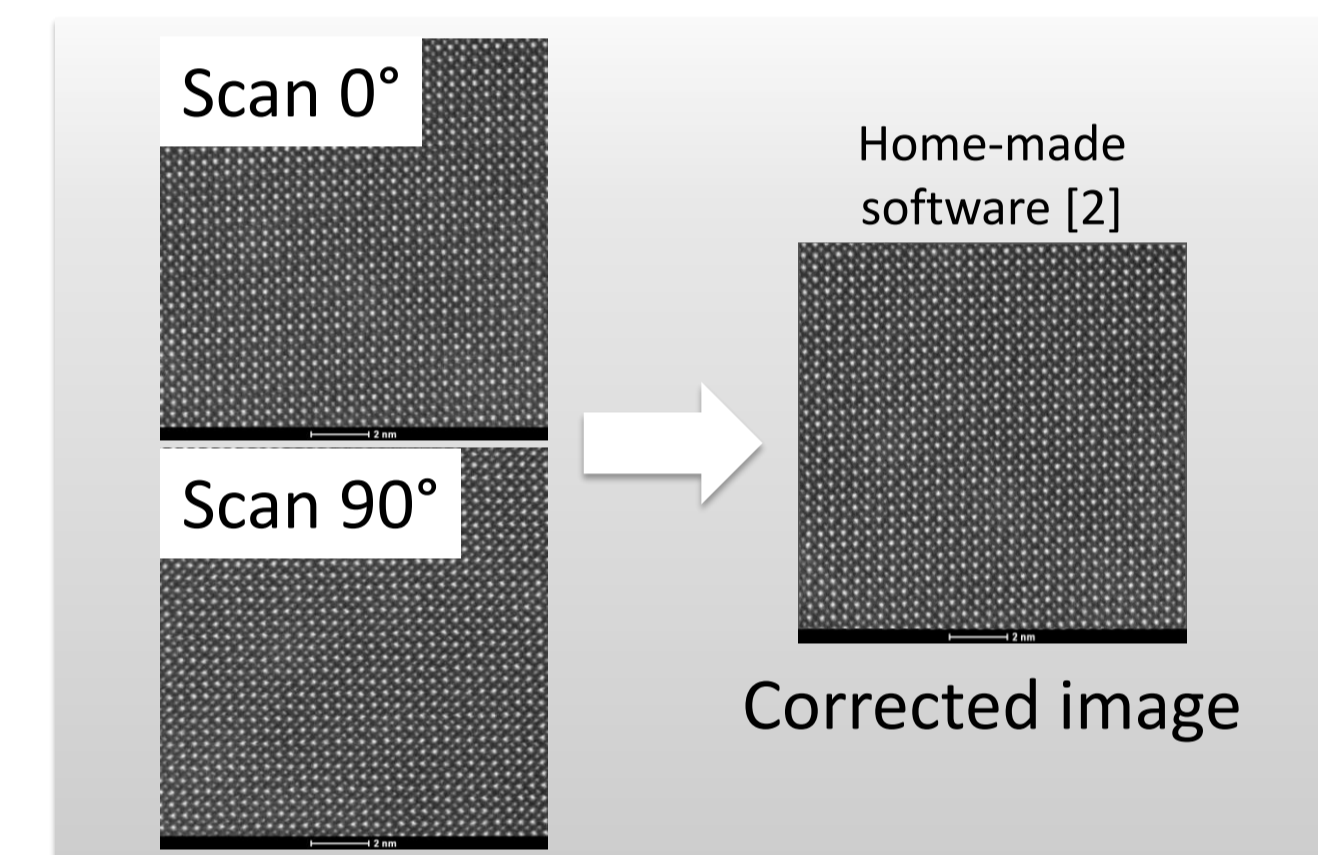
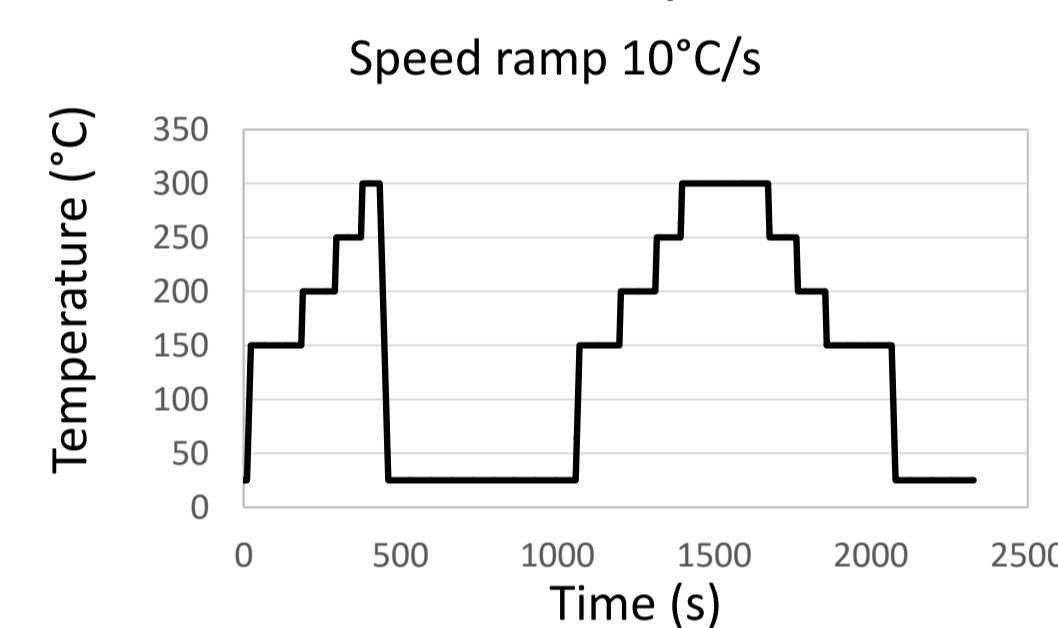


Statistical distribution of antiferroelectric domains along the two zone axes of GdScO₃(110)_o substrate obtained from HAADF-STEM images

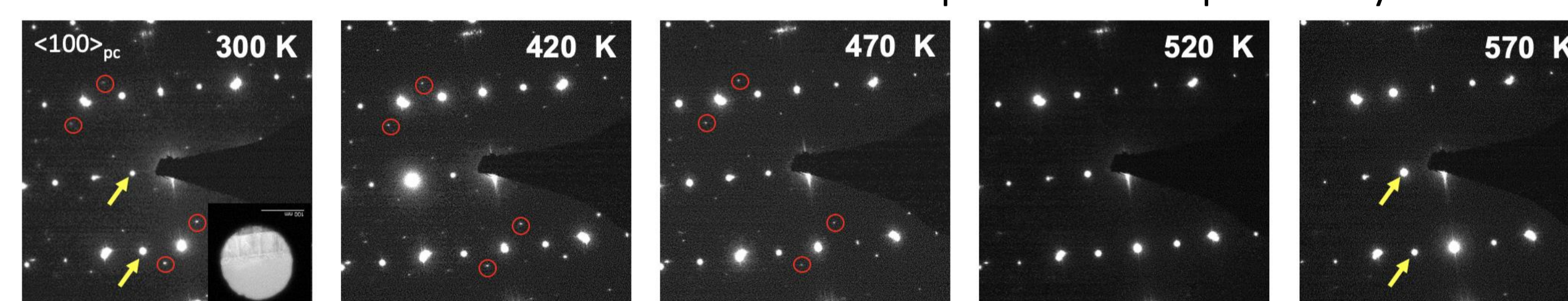


Temperature dependence on one domain

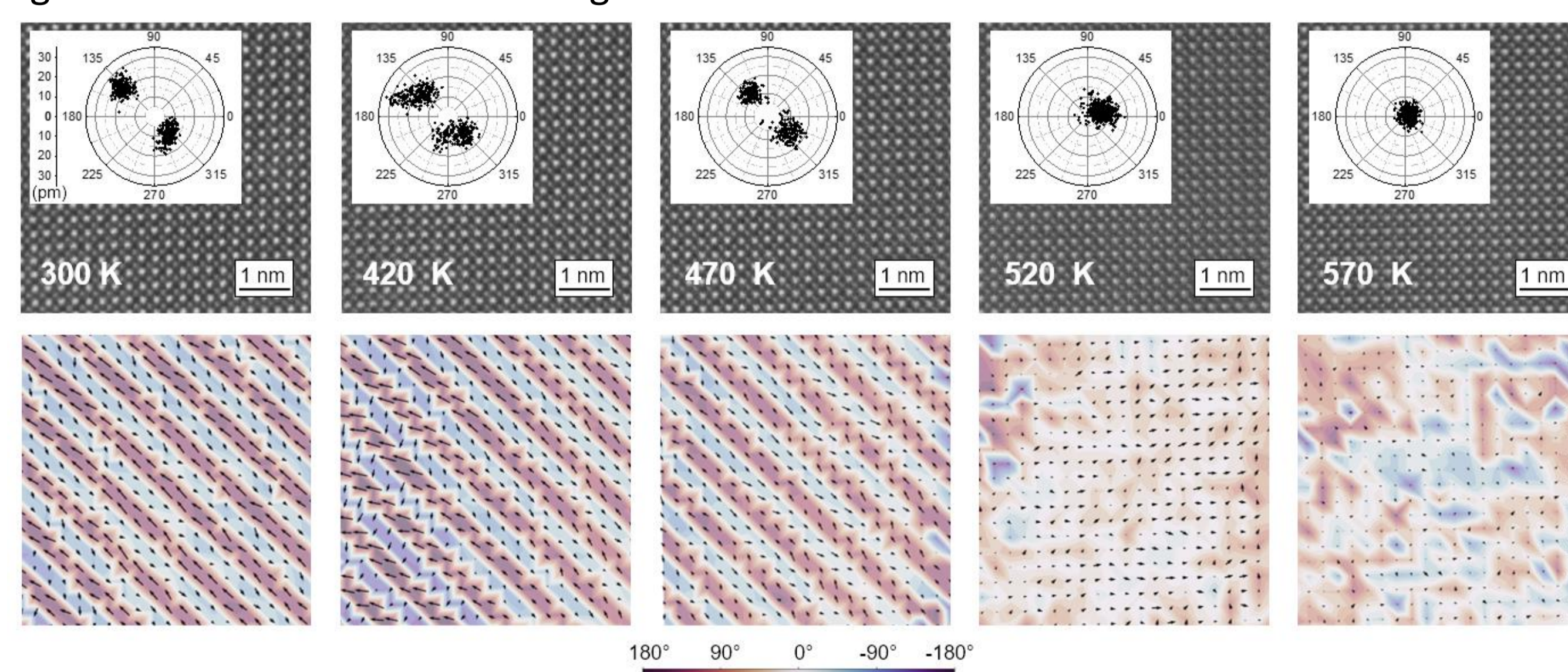
Heating up to 300°C along multiple cycles and different samples



Selected area electron diffraction: loss of diffraction spots from AFE periodicity



High resolution HAADF-STEM images of the same domain



Electrical measurements

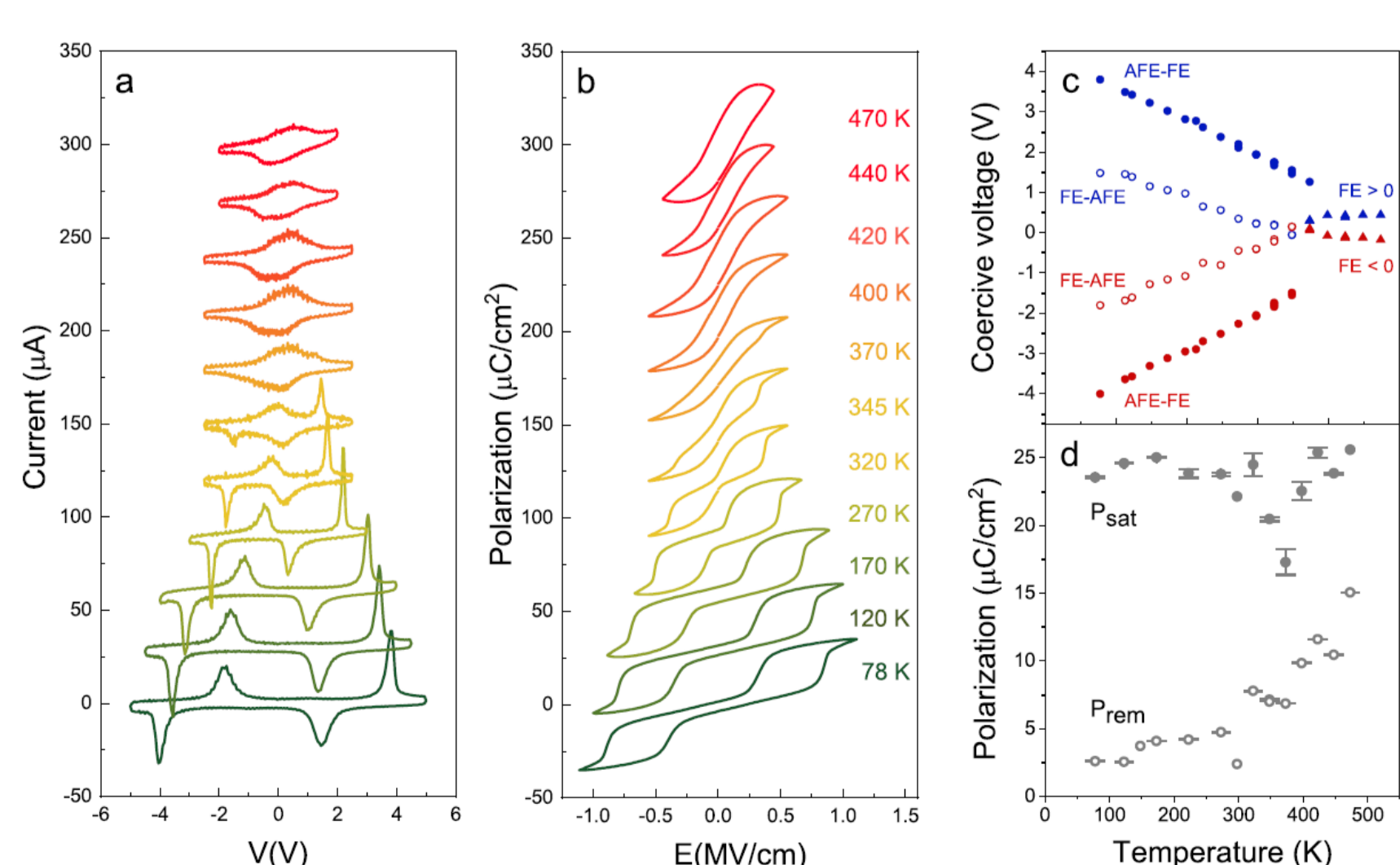


FIG. 6. Electric-field-induced phase transitions with increasing temperature. (a) I-V and (b) P-E loops of a 45-nm thick PbZrO₃ film acquired at several temperatures ranging from 78 to 470 K at 1 kHz. The I-V and P-E loops are vertically shifted. (c) Evolution of the coercive voltages as a function of temperature for bulk (top) and an epitaxial film with fixed in-plane parameters in the (001)_{pc} plane (bottom). The blue circles correspond to the coercive voltages of the transition from an AFE to a FE state (solid circle) and its associated back-switching (open circle) for positive voltages. The red dots are their equivalent for negative bias. The triangles are the coercive voltages of the ferroelectric-like phase. (d) Temperature evolutions of the remnant polarization (P_{rem}) in open symbols and of the saturation polarization (P_{sat}) in solid symbols.

DFT-based calculations

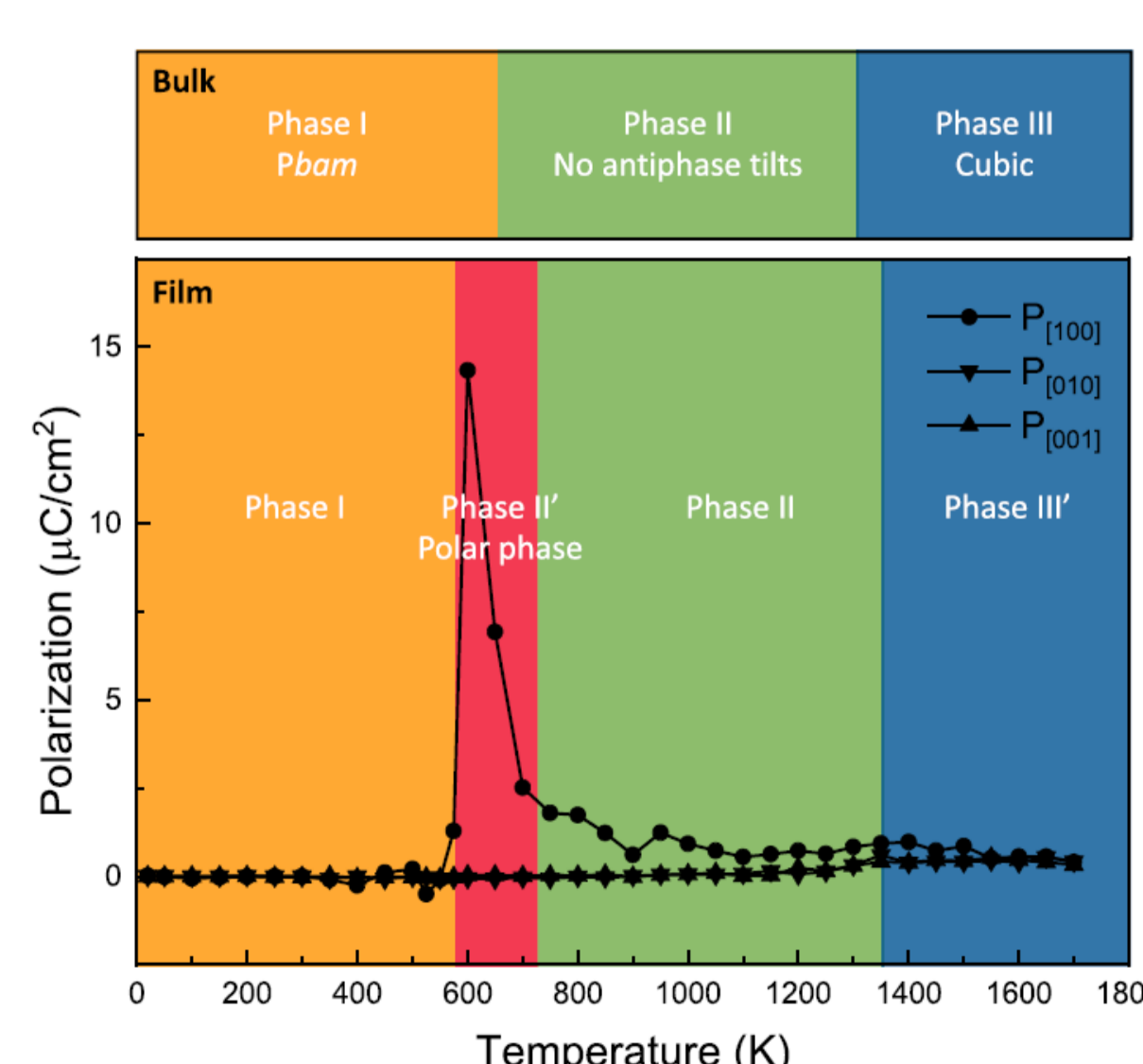


FIG. 8. Computed phase diagram of PbZrO₃ using atomistic simulations. Calculated structural phases of PbZrO₃ as a function of temperature for bulk (top) and an epitaxial film with fixed in-plane parameters in the (001)_{pc} plane (bottom). The computed polarizations vs temperature along the in-plane (100)_{pc}, (010)_{pc} and out-of-plane (001)_{pc} directions are displayed for the film case.

Prediction of a ferroelectric phase with the epitaxial biaxial strain

Summary

- Direct evidence for the antiferroelectric behavior in 45-nm-thick epitaxial PbZrO₃ films at room temperature
- In situ STEM observation of phase transition at the atomic scale up to 300°C.
- Observation of a ferroelectric-like phase in a large temperature window (100°C) in agreement with electrical measurements