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Nanoscale Polar Order in Relaxor Ferroelectrics and its response to external stimuli

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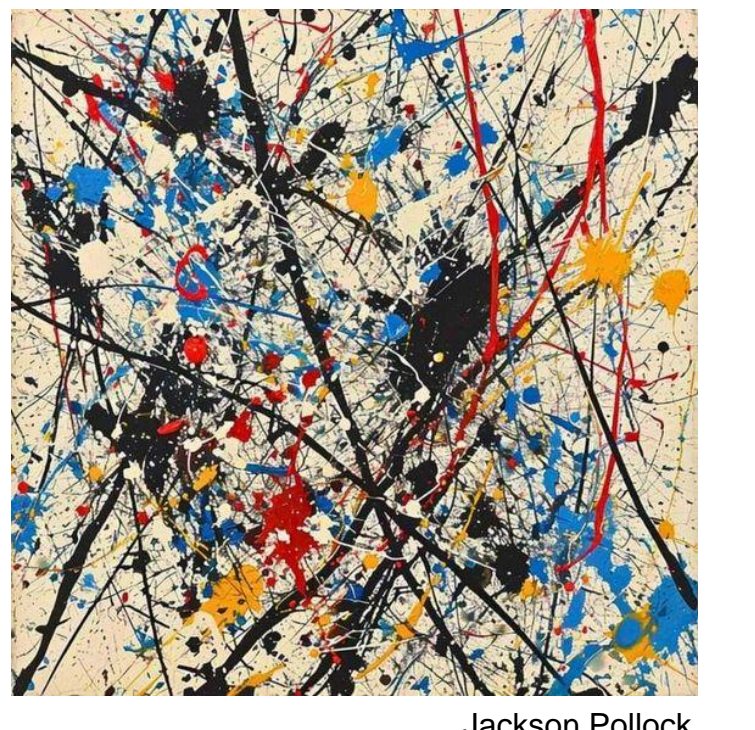
Laboratoire Structures, Propriétés et Modélisation des Solides, CentraleSupélec, Université Paris-Saclay, France

The problem

- **Relaxor Ferroelectric (RFE)** materials exhibit unique characteristics, including a slim polarization-electric field hysteresis loop, diffuse phase transitions, frequency dispersion of maximum permittivity, and exceptional electro-mechanical responses (Li et al., Adv. Funct. Mater., 28, 1801504, 2018).
- These extraordinary properties have been hypothesized to originate from **polar states which correlate at the nanoscale**.
- Although this idea remained speculative for decades, recent advances in high-resolution electron microscopy have enabled the direct detection of polar nanodomains and their spatial correlation with chemically ordered regions (Kumar et al., Nature Materials, volume 20, 2021).
- Despite these advances, the complex polar arrangement and its role in the functionality of RFE are still a matter of debate and especially how the polar disorder responds to external stimuli, such as the electric field (Tadej Rojac, Communication Materials, volume 4, 12, 2023). This thus requires in situ high resolution techniques to directly probe it at the atomic scale.

Ferroelectric Relaxors a hopeless mess

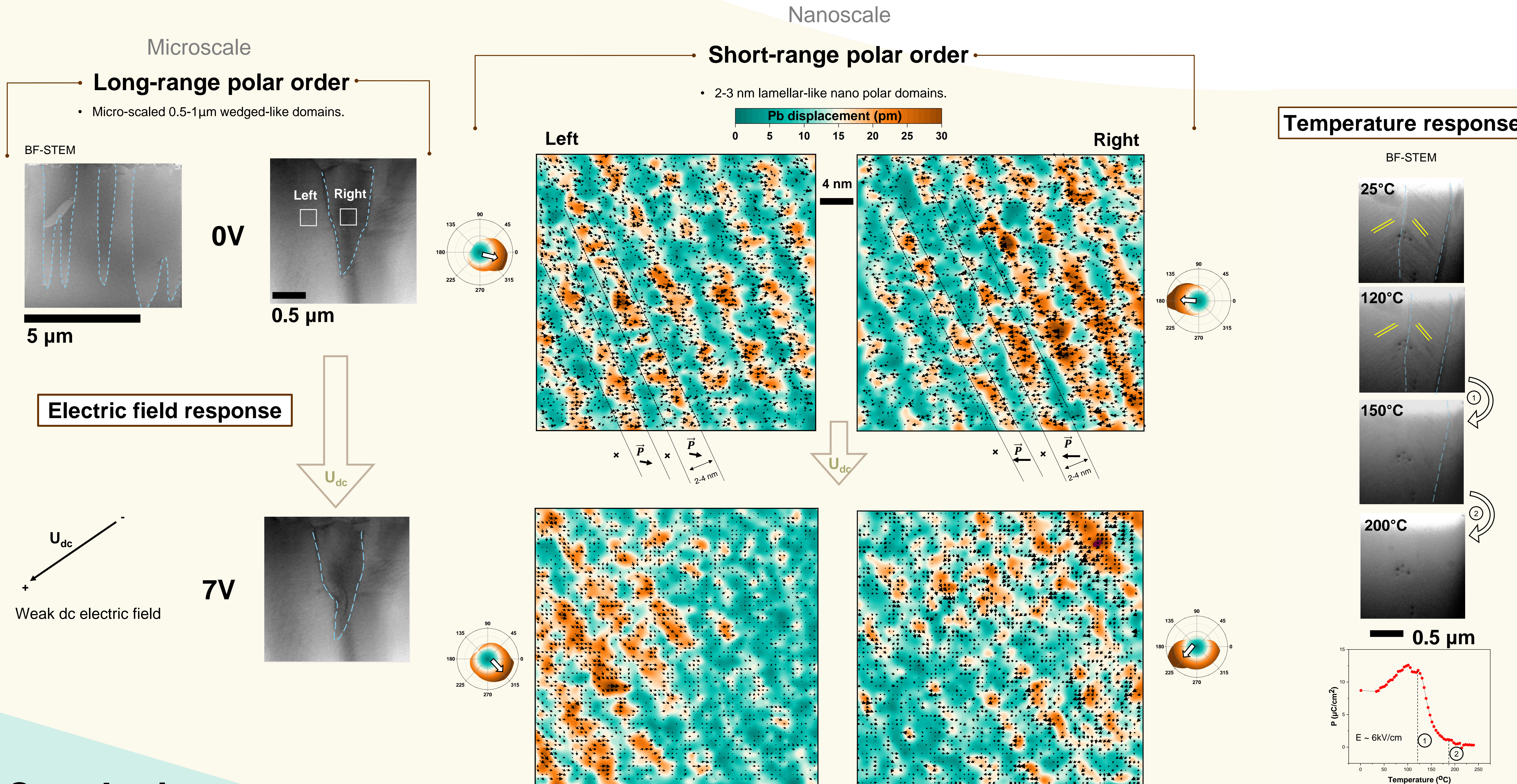
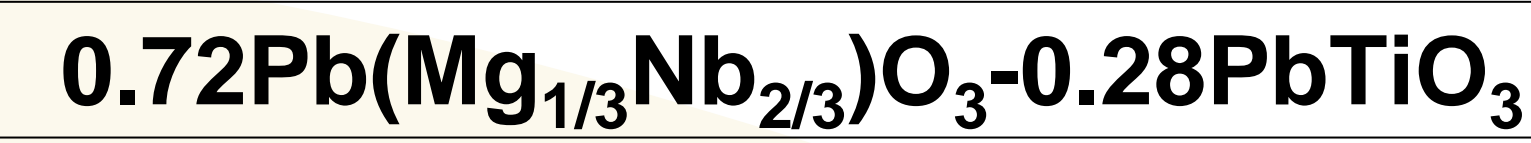
Cohen, Nature 441, 941, 2006



Jackson Pollock



Results



*The Pb displacement maps were determined from HAADF images, as described in the Methods.

Conclusions

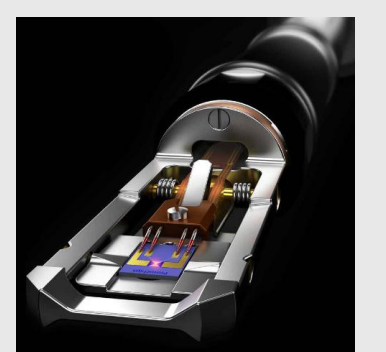
- **Polar Order:** Hierarchical structure, with micro-scaled wedge-like domains (0.5-1 μm) coexisting with nanoscale lamellar domains (2-4 nm).
- **Electric field response**
 - The micro-scaled wedged-like domains undergo domain boundaries movement.
 - The Nano Domains lose the lamellar correlation, shifting towards a "nano-island" distribution. The direction of the Pb displacement vector is changing in accordance to the direction of the electric field.
- **Temperature response:** initially, the short-range order (nanodomains) responds marking a phase transition between 120-150°C. Further, the long-range order (micro domains) disappear around 200°C, marking the paraelectric phase.

Techniques & Methods

Main technique: Scanning transmission Electron Microscopy



TITAN3 G2 80kV-300kV

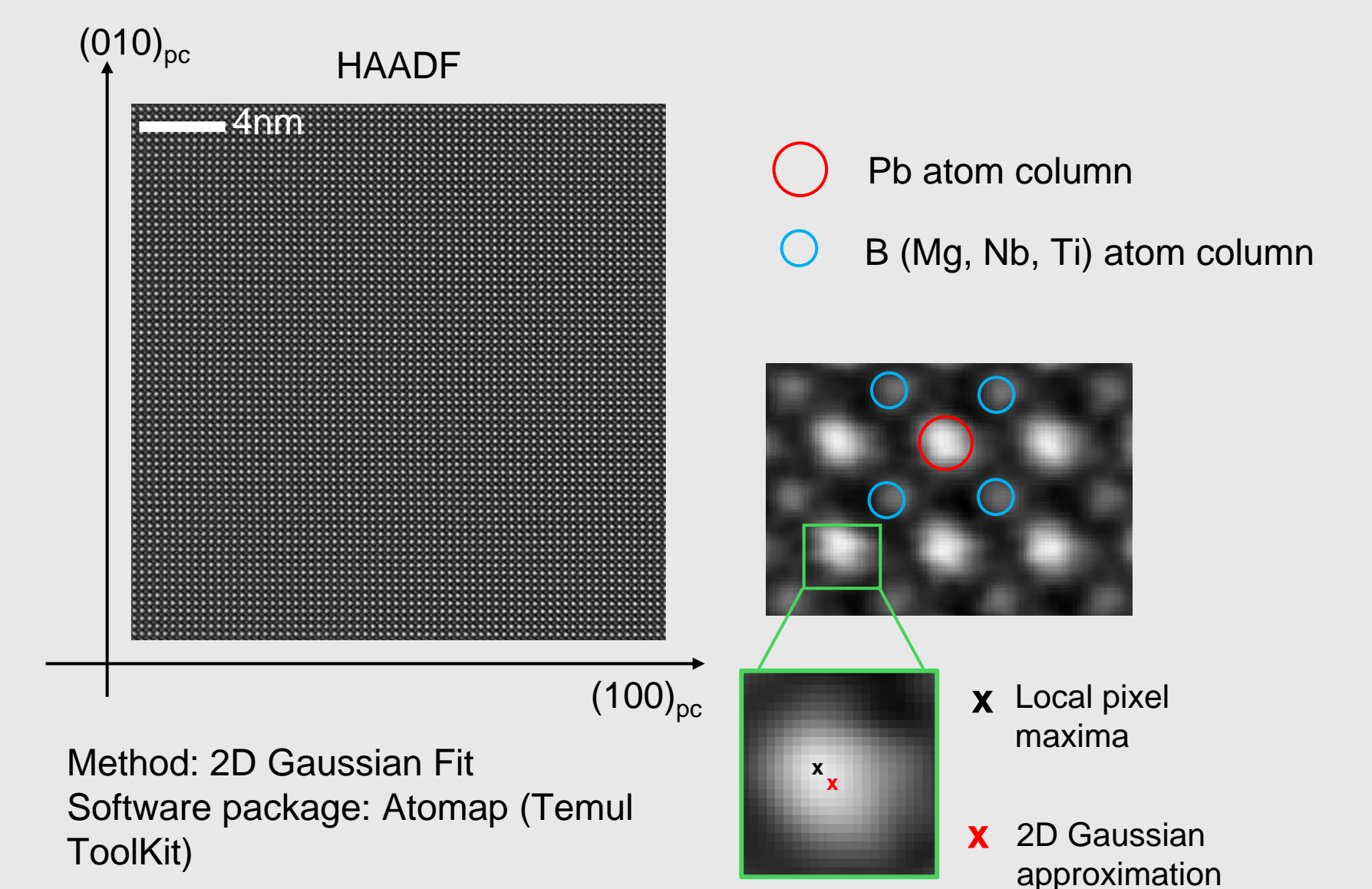


In-situ Protochipts holder: Heating & Electric biasing

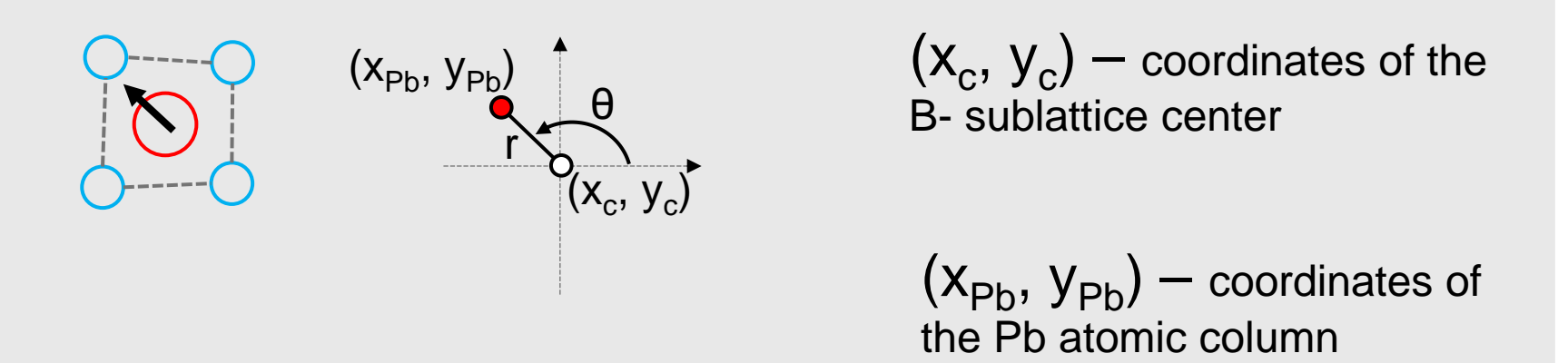
HAADF - High Angle Annular Dark Field
BF - Bright Field

[100]_{pc} Zone axis

Atomic resolution imaging: Coordinates determination



Atomic displacements: Pb-displacement-vector relative to B-sublattice



Magnitude: r $r = \sqrt{(x_{Pb} - x_c)^2 + (y_{Pb} - y_c)^2}$

Direction (Angle) $\theta = \begin{cases} -\arccos\left(\frac{x_{Pb} - x_c}{r}\right), & y_f < y_c \\ \arccos\left(\frac{x_{Pb} - x_c}{r}\right), & y_f > y_c \end{cases}$