First direct observation of the built-in electrical field and oxygen vacancy migration in ferroelectric $Hf_{0.5}Zr_{0.5}O_2$ film during electrical cycling

Supplementary materials

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1. DPC-STEM diagram and the corresponding HRTEM of grain boundary diagram

Below are the DPC (A-C)-STEM and corresponding HRTEM images for the TiN/HZO/TiN samples in the initial, wake-up state and the fatigue status. The borders of different crystalline orientations in HZO clearly reveals the location of the grain boundaries.

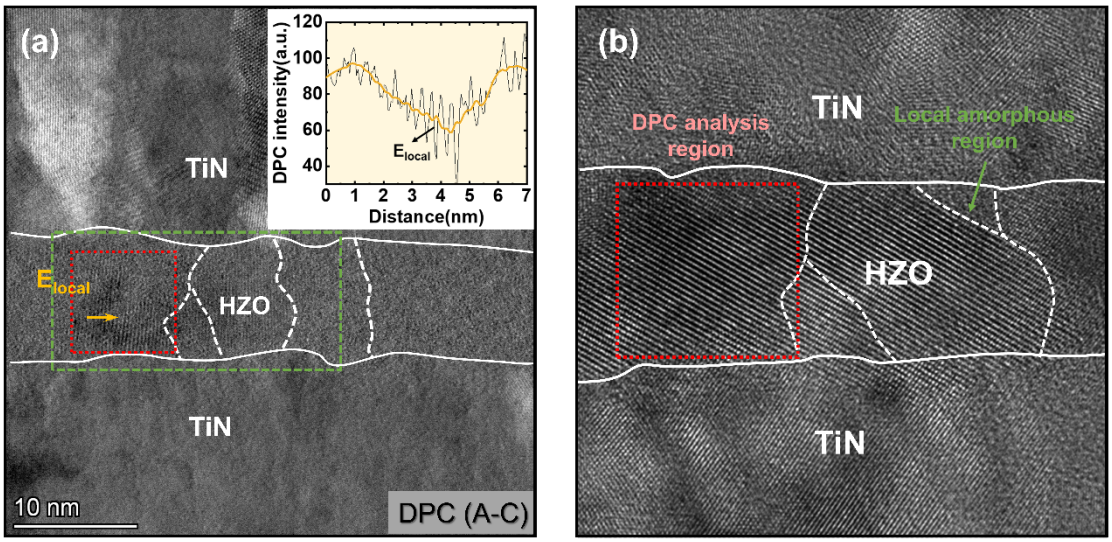

Fig. S1 DPC-STEM image (a) and its corresponding HRTEM image (b) of the sample in the initial status. The red box area represents the DPC analysis region, the green box area is the corresponding HRTEM image, and the white dotted lines represent the grain boundary.

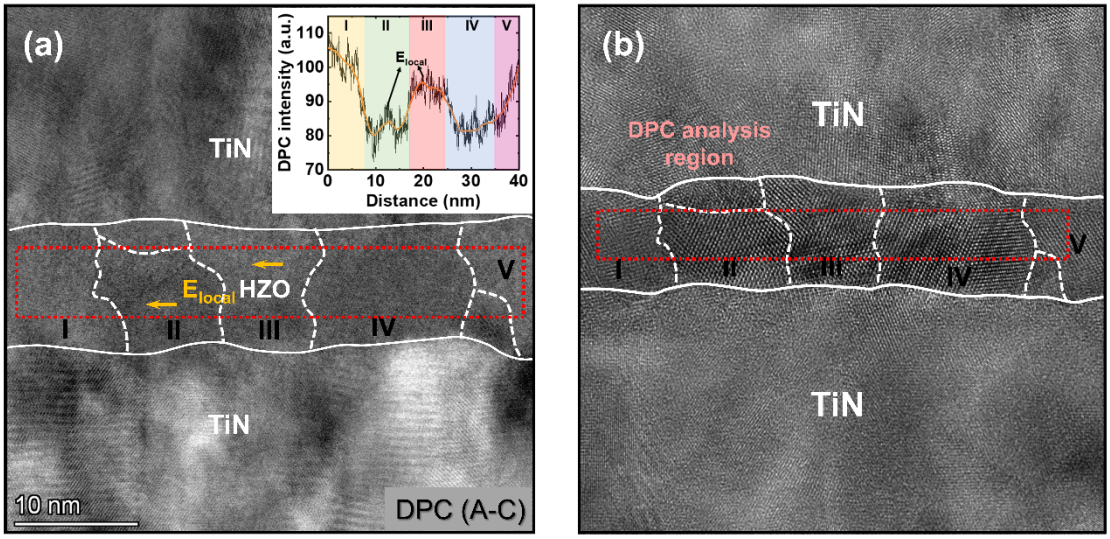

Fig. S2 The DPC-STEM image and its corresponding HRTEM image of the sample in the wake-up status. The red box area represents the DPC analysis region, and the white dotted lines represent the grain boundary.

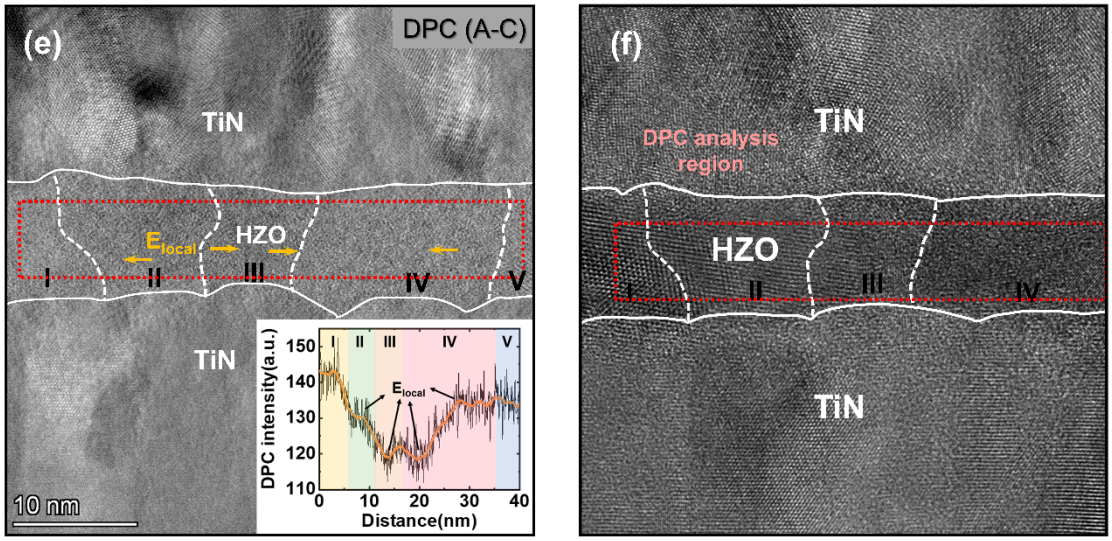

Fig. S3 The DPC-STEM image and its corresponding HRTEM image of the sample in the fatigue status. The red box area represents the DPC analysis region, and the white dotted lines represent the grain boundary.

2. Determination of field-induced phase transition

In order to determine whether the crystalline phase of the sample changes after a certain number of electric cycling pulses, we made statistics on the number of o-phase, t-phase and m-phase in several regions of the sample using FFT, and calculated the percentage of each phase, with the result shown in the main paper file (Fig. 3 (d) and Fig.

5 (d)). Below are the typical high-resolution images of o-, t- and m-phases and their corresponding FFT images.

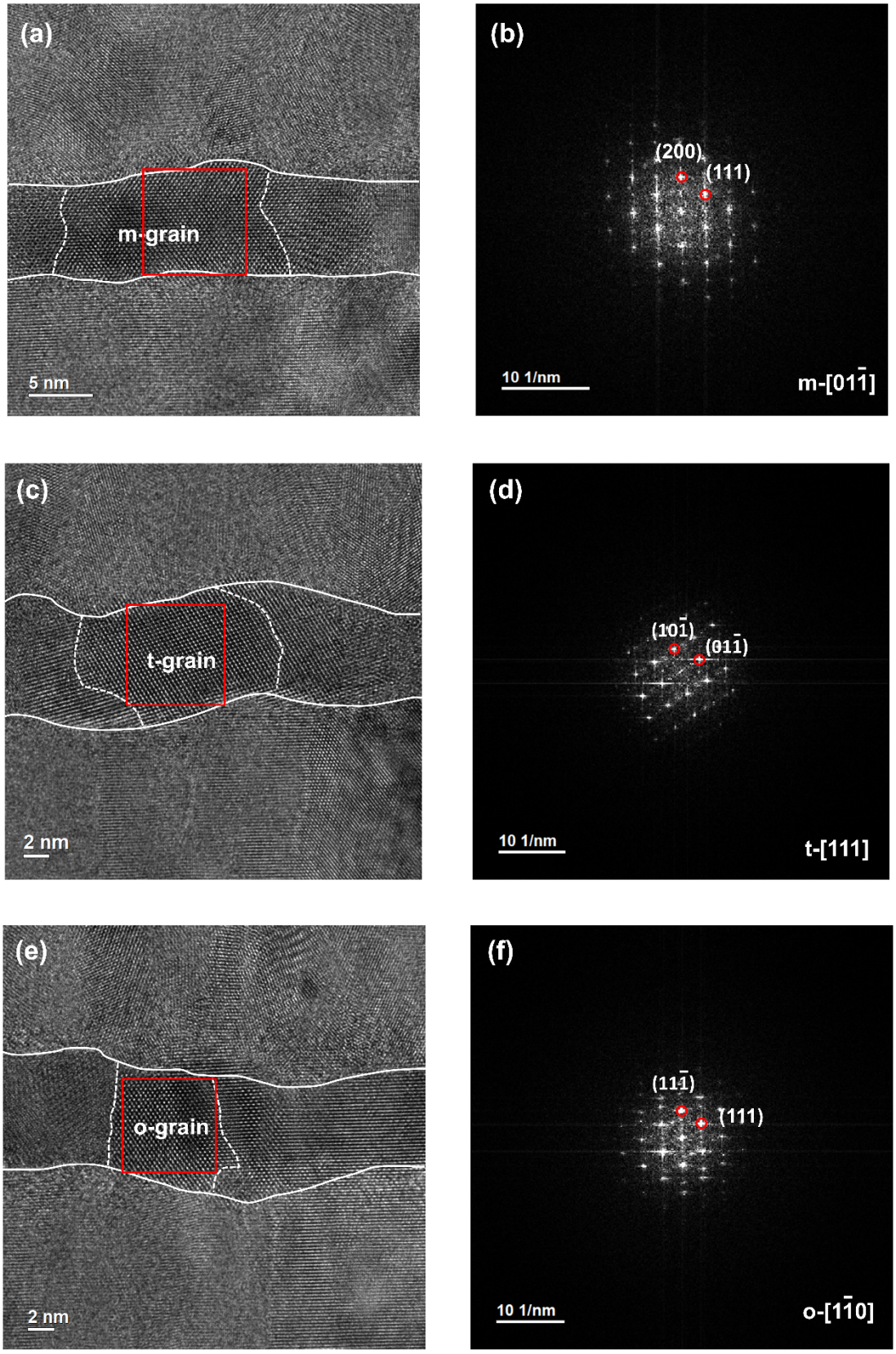

Fig. S4 Typical high-resolution images of m (a), t (c)and o (e) phases and their corresponding FFT images (b) (d) (f).

3. DPC signal intensity of TiN

To further demonstrate that the contrast in the TiN layer originates from grains with different orientations, below we provide LAADF-STEM images corresponding to DPC-STEM analysis region. The LAADF-STEM imaging is known to be sensitive to grains with strong diffraction effect, especially at the grain boundaries. We chose the green boxed area where the DPC contrast is very obvious and compared the same area in the LAADF-STEM plot for comparison. From the sharp contrast shown by the blue dashed lines, it can be clearly determined that the DPC signal in TiN is mainly contributed by the grains with different orientations.

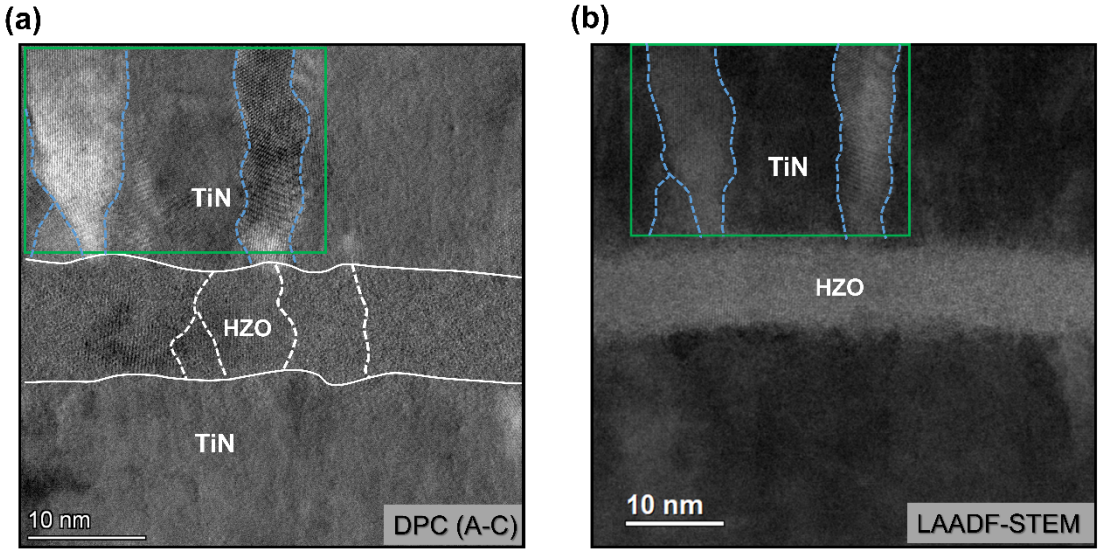

Fig. S5 DPC (A-C)-STEM image (a) and the corresponding LAADF-STEM image (b).