Supplement Information: Evolution of Grain Boundaries during Phase Transitions in Barium Titanate Nanoparticles

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1. Robustness of fringe cropping and its influence on final reconstruction

We studied the influence of fringes between twin peaks (e.g. Figure 1b) on the quality of the final reconstruction.

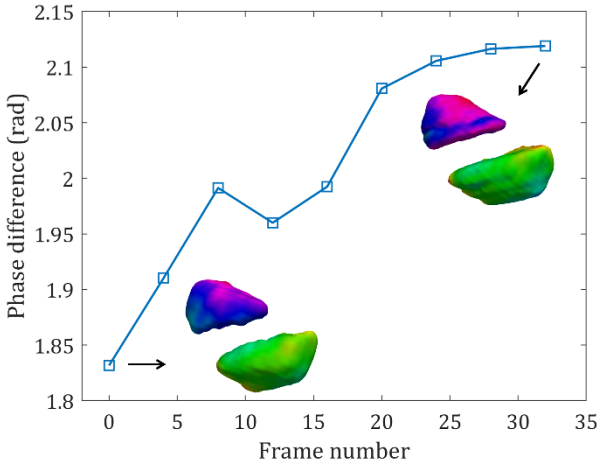

Figure S1. Phase difference of two domains after adding different numbers of frames containing intermediary fringe.

In this case, by adding more of the intermediary fringe frames into the reconstruction, the shape of the crystal, which is defined by Bragg density, remains largely unchanged. However, the relative displacement between the two domains was found to increases gradually and approaches a maximum of 0.2873 rad difference when adding all the fringes. The resolution is expected to improve slightly with more fringes. In this case, however, the resolution was found to remain around 8.4nm.

A redrawing of Figure 1, after taking all the fringes into the reconstruction, is shown in Figure S2.

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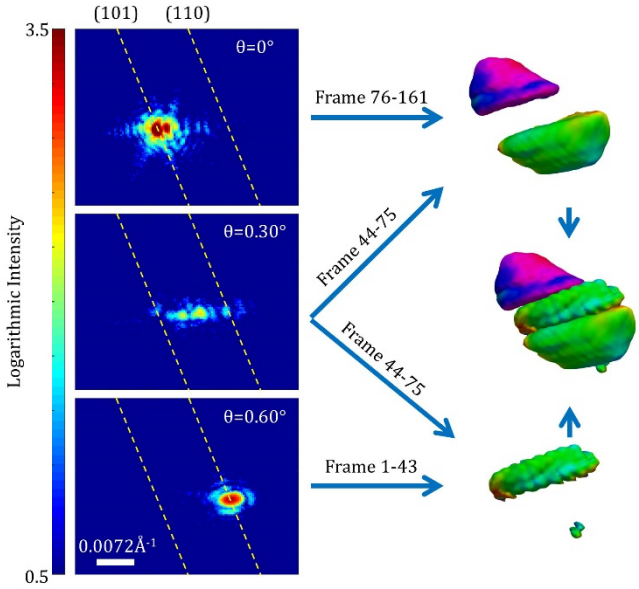

Figure S2. Reconstruction of the separated Bragg peaks with all the intermediary fringes added to both peaks.

2. Combine reconstruction of crystal in Figure 1.

The diffraction patterns in Figure 1 were reconstructed by separating the two Bragg peaks and manually merging the resulting images. If instead, the reconstructions were carried out on the full data without cropping or separating the peaks, the results are shown in Figure S3. For crystal 1 in Figure S3a, the middle domain is misaligned and moved to far side.

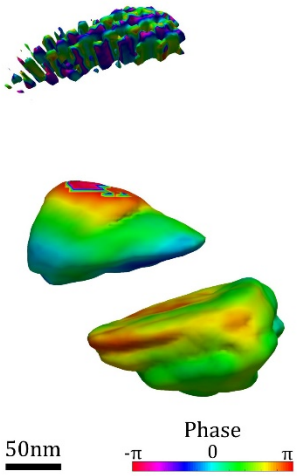

Figure S3. Full-data reconstructed images of the nanoparticles shown in Figure 1.

3. Displacement field difference of the nanoparticle in Figure 3.

To calculate the average displacement field of each grain, the nanoparticle is split at its twin domain boundary, as shown in Figure S4. Because of the discontinuity near the twin boundary, so the volume within 30nm of the boundary is not counted. The average displacement of each region and the displacement difference is listed in Table S1 along with its standard error.

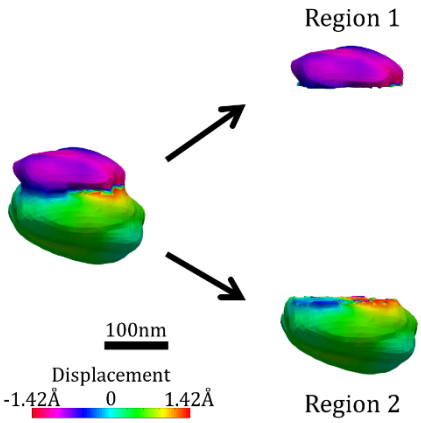

Figure S4. Splitting the nanoparticle into two regions at its twin boundary. The region near the twin boundary is omitted when calculating the average displacement of that region.

Table S1. Measured average displacement of the grains and their difference of the crystal in Figure 3.

	Temperature (K)	Average displacement of region 2 (Å)	Average displacement of region 1 (Å)	Displacement difference (Å)
Heating up	387.2	0.381±0.001	-0.764±0.003	1.145±0.003
	389.9	0.085±0.002	-0.612±0.003	0.697±0.003
	392.6	0.105±0.002	-0.072±0.005	0.177±0.005
Cooling down	390.2	0.113±0.002	-0.055±0.008	0.167±0.008
	387.6	0.137±0.003	-0.163±0.005	0.300±0.005
	384.9	0.075±0.003	-0.347±0.003	0.423±0.003
	379.7	0.240±0.002	-0.435±0.003	0.675±0.003
	371.7	0.316±0.002	-0.726±0.002	1.042±0.002
	366.9	0.201±0.003	-0.881±0.002	1.082±0.002

For the heating period, there is a linear relationship with slope k=-0.48±0.26 and coefficient of determination R^2=0.9982.

For the cooling period, the linear fitting gives k=-0.21±0.10 with R^2=0.9427.